ADVANCED ADAPTIVE LIBRARY FOR GAMMA-RAY SPECTROMETERS

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THESIS
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ABSTRACT

The thesis introduces an algorithm to generate customized libraries for automated radio-isotope identifiers using peak searching and library comparison methods. The algorithm has the adaptive feature that could incorporate response curves, such as efficiency-energy curves and full width at half maximum (FWHM)-energy curves, of the detector into the generation process of the library in order to make the generated library to be the best fit for that detector. The suggested algorithm efficiently generates a library that includes centroids and area information of peaks observed in an isotopes’ γ-ray spectrum. The generated centroids are good estimates of the observed peak locations in the γ-ray spectrum and the generated relative area could be taken as reference for the subsequent identification algorithm.
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# TABLE OF CONTENTS

Chapter 1 Introduction .................................................. 1  
1.1 Literature review ........................................................ 1  
1.2 Peak detection algorithm-wavelet analysis .......................... 3  
1.3 Organization of the thesis .......................................... 5  

Chapter 2 Library Material ................................................. 6  
2.1 Library isotopes .......................................................... 6  
2.2 Bateman equation ......................................................... 6  
2.3 Conclusion ................................................................. 10  

Chapter 3 Algorithm .......................................................... 11  
3.1 Mathematical background ............................................ 11  
3.2 Design of the algorithm .............................................. 11  
3.3 Improvement of the algorithm ..................................... 17  

Chapter 4 Validation of the Algorithm ................................. 19  
4.1 290-470 keV complex of $^{239}$Pu ................................ 19  
4.2 Whole $^{239}$Pu spectrum case ....................................... 30  
4.3 $^{152}$Eu case ............................................................... 33  
4.4 Conclusion ................................................................. 37  

Chapter 5 Experiments ......................................................... 38  
5.1 $^{239}$Pu case ............................................................... 38  
5.2 $^{152}$Eu case ............................................................... 42  
5.3 Conclusion ................................................................. 46  

Chapter 6 Summary and Future Work .................................... 47  
6.1 Summary ................................................................. 47  
6.2 Future work ............................................................... 48  

Appendix A Matlab Code ..................................................... 50  
Appendix B Sample Libraries ............................................. 62  
Reference ................................................................. 65
Chapter 1  Introduction

1.1 Literature review

In the field of radioactive isotope identification, $\gamma$-ray spectra have been widely used as the fingerprints of different radioactive isotopes [1]. In recent years, high resolution, high purity germanium (HPGe), detectors have been developed for field use because the energy resolution of the HPGe detector is much better than low resolution sodium iodide (NaI) detectors [2]. However, NaI scintillators are still one of the most widely used commercial detectors for radioactive isotope identification, nuclear emergency response and border examinations in today’s world in order to balance the cost, efficiency and resolution [3]. However, the energy resolution of NaI detectors is considerably worse than HPGe [4]. Several factors, including the poor resolution of NaI, result in the fact that the isotope identification performance of commercial isotope identifiers is much lower than expected [5,6,7]. In order to improve the performance of these detectors, significant development has been made in producing new and improved detector materials [8, 9]. However, algorithm development remains an area that can be enhanced to greatly improve the performance of these detectors [10,11].

Depending on the resolution of the detector, various algorithms have been developed by researchers to achieve radio-isotope identification [1,12,13,14]. Peak searching and library comparison methods are one of the most commonly used radioactive isotope identification algorithms, which uses the centroid of each peak detected from the $\gamma$-ray spectra to compare with a built-in library and identify the isotope [1]. Previous researchers have proposed many peak searching and library comparison methods. However, there is no published work about how to generate the libraries that have been applied in their algorithms [1,10,13,15,16]. Instead, the researchers are focusing on developing algorithms to measure information of the individual peak, such as the peak centroid and peak area, from low-resolution spectra from NaI detectors, and then compare this information with a library in order to identify which isotope generate the spectrum [4,14,17,18]. One possible approach focuses on building up a comprehensive library including as many $\gamma$-ray peaks as possible [19,20]. But this is not ideal because commercial detectors prefer libraries
without too many γ-ray peaks for each isotope since it is very difficult to detect small peaks for each isotope. Research focusing on the library part for these algorithms, such as generating a special library tuned to low-resolution NaI spectra, still remains as a new area.

For example, a $^{239}\text{Pu}$ spectrum has been plotted together with all its γ-ray peaks in Figure 1. It shows that $^{239}\text{Pu}$ has hundreds of γ-ray peaks. However, the peak detection algorithms usually fail to resolve most of these peaks from low-resolution spectra, but rather provide information of peaks observed clearly in the spectra. Hence, rather than a library containing all original γ-ray peaks, a new library containing the observed peaks will be a better choice. Meanwhile, it is important to use more peaks than just the few largest ones since several isotopes have identical peaks. For example, $^{239}\text{Pu}$ and $^{177}\text{Lu}$ have their two most prominent peaks in common at 375 and 414 keV.

![Figure 1 $^{239}\text{Pu}$ spectrum and all its original γ-ray peak locations](image)

Figure 1 $^{239}\text{Pu}$ spectrum and all its original γ-ray peak locations

Therefore, this thesis introduces an algorithm for generating libraries for detectors using peak searching and library comparison methods. The algorithm will have the adaptive feature that can be used to generate a customized library of peaks in the low-resolution spectra collected by NaI detector. Through incorporating the individual detector’s response curves, such as the efficiency-energy curve and full width at half maximum (FWHM)-energy curve, in the algorithm, the generated library
will take the specific features and performance of the detector into consideration. Since the newly-generated library contains the information of detectable peaks in the low-resolution spectra rather than all original peaks information, the identification algorithm will work better because the new library will save the cost of searching for peaks and also eliminate influence from small peaks that have much less possibility being detected. Additionally, the algorithm also provides a good way of extracting main peaks from all original peaks of one isotope, rather than the simple method that incorporate several peaks with big branching ratios in the library. Because the algorithm takes the performance of the peak detection algorithm into consideration, the library generated will contain the peaks that have bigger probability of being observed in the spectrum and detected by the peak detection algorithm.

1.2 Peak detection algorithm-wavelet analysis

Radio-isotope identification algorithms using peak searching and library comparison methods usually contains three parts: the peak detection algorithm, the library, and the identification algorithm. The peak detection algorithm is used to extract peaks information like peak centroid and area information from spectra, and is based on wavelet analysis and non-negative least squares (NNLS) for this work [21,22,23].

The whole peak detection algorithm is designed to finish the jobs of peak detection, measuring the peak’s centroid, and peak quantification, to get the peak’s area and its uncertainty. With wavelet analysis, the first part of algorithm efficiently detects the peak and measures its centroid [22,23,24]. As for peak quantification, linear regression methods including NNLS have been playing an important role for a long time in diverse areas such as mass spectrometry, nuclear magnetic resonance data analysis and molecular spectroscopy [24,25,26]. Combining the ability of peak detection with wavelet analysis and the peak quantification ability of NNLS, the wavelet peak detection algorithm could work very well on providing peaks information of centroid and area for good quality spectra. However, the performance of the peak detection algorithm still needs improvement for spectra with considerably noise.

Generally, the wavelet analysis-based peak detection algorithm takes the spectrum and performs the continuous wavelet transform, shown in Eq. (1), in order
to obtain the wavelet transform coefficient matrix $T(E, s)$, which is called the scalogram, as shown in Figure 2.

$$T(E, s) = \int_{-\infty}^{\infty} \phi \left( \frac{t - E}{s} \right) f(t) dt \quad (1)$$

The algorithm will then find the local maxima in the scalogram, named the wavelet transform modulus maxima (WTMM). By orderly collecting and linking these points, WTMM lines are formed and shown as the black lines in Figure 2. It can be observed that the straight, vertical WTMM lines provide a good estimation of the peak locations in the spectrum. Other unqualified WTMM lines will be filtered out. The while curve shown in Figure 2 is the optimal scale curve, which is the scale of the maximum wavelet transform coefficient along the WTMM line for Gaussian peaks located at the corresponding channel. A WTMM line that passed all filters will indicate the location of a peak. The algorithm will then use the NNLS method to solve Eq. (2) in order to obtain vector $k$, which is the peak’s area information.

$$S = kB \quad (2)$$

where $S$ is the wavelet transform of the signal at optimal scale and $B$ is the basis function matrix. The NNLS method can also resolve the overlapping peaks. This information is captured in the vector $k$. All non-zero values of $k$ are the individual peak areas of peaks located at corresponding channels. A more detailed description of
above material is beyond the scope of this thesis and can be found in reference [21, 23].

1.3 Organization of the thesis

This thesis, which will have 6 chapters, introduces an algorithm for generating a library used by peak searching and library comparison methods. Chapter 2 will answer what isotopes and what kind of information on those isotopes should be included in the library. Chapter 3 is the theory part of the thesis, presenting the design of the algorithm. Chapter 4 will generate some sample libraries of several example isotopes and provide verification that the algorithm suggested in Chapter 3 works properly. In Chapter 5, tests with the peak detection algorithm will be done to test the overall performance of coupling between the peak detection algorithm and libraries generated by this algorithm. Finally, Chapter 6 is a summary of the research done in the thesis and some points for future work.
Chapter 2  Library Material

2.1 Library isotopes

The algorithm suggested in this thesis is designed to generate customized isotope libraries for peak searching and library comparison methods. Hence, the material included in the library for each isotope is the centroid and expected area of each peak in the isotopes’ γ-ray spectrum [1]. Usually, the library size depends on the specific performance of the detector. However, in general, since the library is designed to be used in future commercial detectors, the isotopes included in the library should satisfy the national standard performance criterion [27] and also include isotopes of special interest, such as medical, industrial, special nuclear material (SNM) and naturally occurring radioactive material (NORM) categories, designed for that kind of commercial detector [1]. Hence, the isotopes included in the library can be categorized as,

- Medical isotopes: $^{67}Ga$, $^{51}Cr$, $^{75}Se$, $^{99m}Tc$, $^{103}Pd$, $^{111}In$, $^{123}I$, $^{125}I$, $^{131}I$, $^{201}Tl$ and $^{133}Xe$
- Industrial isotopes: $^{57}Co$, $^{60}Co$, $^{133}Ba$, $^{137}Cs$, $^{192}Ir$, $^{204}Tl$, $^{226}Ra$, and $^{241}Am$
- SNM isotopes: $^{233}U$, $^{235}U$, $^{237}Np$ and $Pu$
- NORM isotopes: $^{40}K$, $^{226}Ra$, $^{232}Th$ and its daughters, and $^{238}U$ and its daughters

2.2 Bateman equation

Since the existence of some isotopes can be inferred from the existence of its daughters, the library also includes the daughters of isotopes listed above in order to improve the performance of isotope identification algorithm. For example, in Figure 3, if we find existence of $^{228}Th$ but no $^{228}Ac$, then it may indicate the existence of $^{232}U$, which should be seen as a threat since $^{232}U$ is a common contaminant in $^{233}U$, which is fissile.
Figure 3 Example of $^{232}\text{Th}$ and $^{232}\text{U}$ decay chain

The time evolution of nuclide concentrations satisfies a set of first order differential equations called the Bateman equation [28]. The radioactive chain decay case of a nuclide, shown in Figure 4, satisfies the following Bateman equation:

$$\frac{dN_i}{dt} = -\lambda_1 N_1 \quad (3)$$

$$\frac{dN_i}{dt} = \lambda_{i-1} N_{i-1} - \lambda_i N_i \quad (i = 2, ..., n) \quad (4)$$

where $N_i$ is the concentration of $i$th nuclide at time $t$, and $\lambda_i$ is the decay constant of $i$th nuclide.

Assuming the concentration of all daughters at time $t=0$ is zero, and the concentration for the parent at time $t=0$ is $N_1(0) \neq 0$, then, the general solution of Bateman equation is given by [28,29].
\[ N_n(t) = \frac{N_1(0)}{\lambda_n} \sum_{i=1}^{n} \lambda_i \alpha_i \exp \left[ -\lambda_i t \right] \quad (5) \]

where

\[ \alpha_i = \prod_{j=1, j \neq i}^{n} \frac{\lambda_j}{(\lambda_j - \lambda_i)} \quad (6) \]

Hence, the activity of \( n \)th nuclide at time \( t \) is given by

\[ A_n(t) = N_1(0) \sum_{i=1}^{n} \lambda_i \alpha_i \exp \left[ -\lambda_i t \right]. \quad (7) \]

The time \( t \) shown in Eq. (7) is the age of the parent. Since the parent isotopes could be the NORM, which may exist thousands and millions of years, or manmade isotopes (MMI), which have only existed less than one hundred years, we can categorize the parent isotopes as NORM and MMI in order to study the decay chain behavior characteristics of these two classes.

From Eq. (7), as for NORM, the calculation result shows that the activity of the parent and daughter isotopes in the decay chain is very similar. This behavior comes from the fact that the activity of the daughter will roughly equal to the activity of the parent if the half-life of the daughter is much smaller than the half-life of the parent, which is a special case result of Bateman equation called secular equilibrium [30]. And it turns out the very first few isotopes shown in the decay chain usually have at least one isotope whose half-life is much bigger than following daughters. Meanwhile, because the time \( t \) could vary from a very small value to a value much larger than the half-life of most isotopes shown in the decay chain, the activity of all daughters could reach steady state result and cannot be ignored compared to the activity of the parent [30]. Hence, the library could include all isotopes in the decay chain for NORM. However, for a library used in a commercial detector, whether or not and how many daughters included in the library highly depends on the design of identification algorithm.

However, as for MMI, the result is different. If the time \( t \) could be any value, the result is the same as NORM since the calculation is the same. Because the time \( t \) for MMI is generally less than 100 years, the activity of the daughter will be very small if the daughter half-life is much larger than time \( t \). The main reason is that there is not enough time for the decay to reach the secular equilibrium, which requires at
least seven half-lives of the daughter [30]. Mathematically, from Eq. (7), if the half-life of the parent is much larger than the daughter, we have

\[ A_2(t) = A_1(t)\left[1 - \exp(-\lambda_2 t)\right]. \] (8)

The relationship between relative activity \(A_2(t)/A_1(t)\) and time \(t\) is shown in Figure 5. From Figure 5, we can find that the activity of the daughter, \(A_2(t)\), will be much smaller than the activity of the parent, \(A_1(t)\), if the time \(t\) is small compared to the half-life of the daughter. Hence, as for MMI, the activity of the daughter in the decay chain could be ignored if the half-life of the daughter is bigger than time \(t\) and much smaller than the half-life of its direct parent. Since the activity of following daughters in the decay chain will be not bigger than the activity of that boundary daughter (BD), the library will only take account of the first few isotopes in the decay chain before the BD and ignore all isotopes after the BD.

![Secular equilibrium decay](image)

**Figure 5** Secular equilibrium decay

The determination of the BD depends on the desired accuracy of the library and the importance of daughter isotopes for identification algorithm. One threshold
can be set up for the relative activity, which can be used to determine the position of the BD.

2.3 Conclusion

This chapter answered the question of what isotopes should be included in the library and how to evaluate their decay daughters for inclusion. Meanwhile, this chapter also stated that the library should include the centroid and area information of the γ-ray peaks because the library is designed for detectors using peak searching and library comparison methods. Since the library is aimed to be applied in commercial detectors, it should include all isotopes required by national. The specific choice of isotopes from medical, industrial, SNM and NORM categories depend on the performance design of the detector. The daughters of radioactive isotopes could be included in the library in order to improve the performance of identification algorithm. All daughters of NORM should be included in the library, but only isotopes before the BD in the decay chain of MMI need to be added to the library for MMI. However, for library used in commercial detector, whether or not and how many daughters included in the library highly depends on the design of identification algorithm.
Chapter 3  Algorithm

The algorithm is designed to generate customized libraries for detectors using peak searching and library comparison methods. Using the information like the efficiency-energy curve and full width at half maximum (FWHM)-energy curve of the detector, the algorithm has the adaptive feature that can be used to provide a customized library for each specific detector in order to improve the identification performance of the detector. The sample libraries for several isotopes can be found in Appendix B.

3.1 Mathematical background

The algorithm assumes that a single photopeak in the γ-ray spectra has a shape of Gaussian function [15,31]. Hence, the parameter of a single photopeak satisfies the Gaussian correlation in Eq. (9).

\[
y = \frac{a_0}{\sqrt{2\pi a_2}} \exp \left( -\frac{(x - a_1)^2}{2a_2^2} \right) \tag{9}\]

where \(a_0, a_1, a_2\) are the area, centroid and width of the peak, respectively. Since the area, \(a_0\), of the peak is the total counts around the peak energy registered by the detector, assuming no background continuum is present, it should be proportional to the product of the branching ratio of the peak energy and efficiency of the detector corresponding to the peak energy. The peak information for a specific isotope including peak centroid \((a_1)\) and branching ratio \((a_0)\) can be found from standard reference databases [32]. Meanwhile, the detector response to a given energy, such as the efficiency-energy curve \((a_0)\) and FWHM-energy curve \((a_2)\) can be measured through experiments. Hence, all parameters for a single photopeak with Gaussian shape are known factors.

3.2 Design of the algorithm

In the area of nuclear isotope identification, sodium iodide (NaI) detectors have played an important role for several years in order to balance cost with efficiency [3]. However, the poor resolution of NaI detectors makes it very difficult to decompose the blurred peaks into the original peak energies [4]. The algorithm
suggested in this paper, instead of trying to recover precise original Gaussian peak information from low-resolution spectra, aims to calculate the centroid and area of what the detector will actually measure in the low-resolution spectra. Peak detection algorithms usually fail to resolve the overlapping peaks from low-resolution spectra, but rather provide information on the resulting convolved peaks. Therefore, it is expected that library comparison methods will work better with customized libraries.

From the above analysis, the algorithm is very straightforward. If there are three original peaks overlapped, such as the case shown in Figure 6, the peak shown in the spectrum will be the overall summation of the three peaks, which is plotted in blue solid curve. If the peak detection algorithm used cannot resolve these three peaks because the peaks are too close, it will instead get the centroid and area information of the summation peak. If the algorithm suggested above can generate a library including the centroid and area of the summation peak rather than the three original peaks, then the identification algorithm can still correctly identify the isotope. Through this method, the new library will only save the information of the summation peak rather than all three original peaks. Hence, the new library will have smaller size, which is beneficial to the identification algorithm. Meanwhile, because the peak detected is the overall summation peak, the new library containing the summation peak information will absolutely work better than library containing original peaks.

![Figure 6 Three overlapping peaks and the newly generated peak](image-url)
However, there are two key problems before the algorithm can work properly. One problem is how to determine whether the peak detection algorithm can resolve the overlapped peaks or not. The other problem is how to get the newly-generated peaks, namely the summation peak, with respect to a series of given peak energies from original peaks for one specific isotope. After solving these two problems, the algorithm can calculate the centroid and area information of newly-generated peaks and create the new library.

The minimum energy gap that the peak detection algorithm can resolve was determined through a series of tests of the peak detection algorithm. As expected, the MRE will vary for different peak detection algorithms. Through the tests of wavelet peak detection algorithm [21], the test results showed that the minimum resolvable energy (MRE) is not a constant, but depends on the relative amplitude of the adjacent peaks. An MRE of 28.8 keV is found to be the minimum energy difference that the wavelet peak detection algorithm can resolve under the case of two same amplitude Gaussian peaks with signal to noise ratio (SNR) equaling to 10. Meanwhile, the tests of any arbitrary relative amplitude of two overlapped Gaussian peaks (SNR=10) show that the MRE value increases as amplitude ratio increases. However, it will not increase much over the test region. The test also shows that MRE will reach a saturated value of 36 keV as the amplitude ratio approaches 10 or higher. The exact tests results are shown in Table 1.

<table>
<thead>
<tr>
<th>Amp1/Amp2</th>
<th>MRE/Channel</th>
<th>MRE/ keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>8-10</td>
<td>28.8-36.0</td>
</tr>
<tr>
<td>2</td>
<td>8-10</td>
<td>28.8-36.0</td>
</tr>
<tr>
<td>3</td>
<td>8-10</td>
<td>28.8-36.0</td>
</tr>
<tr>
<td>4</td>
<td>8-10</td>
<td>28.8-36.0</td>
</tr>
<tr>
<td>5</td>
<td>8-10</td>
<td>28.8-36.0</td>
</tr>
<tr>
<td>6</td>
<td>10</td>
<td>36.0</td>
</tr>
<tr>
<td>7</td>
<td>10</td>
<td>36.0</td>
</tr>
<tr>
<td>8</td>
<td>10</td>
<td>36.0</td>
</tr>
<tr>
<td>9</td>
<td>10</td>
<td>36.0</td>
</tr>
<tr>
<td>10-20</td>
<td>10</td>
<td>36.0</td>
</tr>
</tbody>
</table>
However, the above result is only an approximation of the true MRE of the wavelet peak detection algorithm [21]. The tests also show that the performance of the wavelet peak detection algorithm highly depends on the SNR of the spectra. If the SNR is approaching infinity, the wavelet algorithm can perform quite well, identifying any two adjacent peaks with accuracy of one channel under any relative amplitude condition. Meanwhile, when the SNR equals to 5, the MRE is roughly same with the result for SNR=10 shown in Table 1, which indicates that the performance of wavelet peak detection algorithm will not change much in the region of \(\{10 \geq SNR \geq 5\}\). As for typical spectra collected by NaI detector over a few minutes collection time, the SNR value varies case by case, but the above result can still be used as the reference MRE value. The optimal MRE relationship will be tested and analyzed in Chapter 4 and Chapter 5. Hence, approximately, the MRE could be modeled as:

\[
MRE = \begin{cases} 
36 \text{ keV} & \text{if } \frac{Amp(peak1)}{Amp(peak2)} \geq 5 \\
28 \text{ keV} & \text{if } \frac{Amp(peak1)}{Amp(peak2)} < 5
\end{cases}
\]  

The above step function is defined in the domain of \(Amp1/Amp2 \geq 1\), because of the assumption that only peaks with amplitude bigger than adjacent peaks (on at least one side) have the possibility of standing out in the final library as will be discussed in Section 3.3. In Chapter 4 and Chapter 5, a new constant MRE and functional MRE, referring to Eq. (10), will be proposed to test the performance of the algorithm suggested in Chapter 3. Meanwhile, in Chapter 5, the locally optimal MRE value can also be analyzed by comparing the overall performance of the total identification system by coupling the peak detection algorithm to the newly generated libraries.

The algorithm will then use the MRE as an energy bin and check whether there are peaks located within the energy bin for every \(\gamma\)-ray energy peak emitted by the isotope. As illustrated in Figure 7, for each peak in the series, the algorithm will generate a new peak, which can be the same as original one if there is only one peak within the energy bin around that peak, or different from the original one if there are more than one peak within the energy bin. The new peak is generated from summing all peaks located in the same energy bin around the original peak. Since the original peak satisfies the Gaussian relationship in Eq. (9), the new peak centroid could be
simply calculated from adding several known Gaussian peaks together and finding the location of highest point of the new peak. The area of the new peak is calculated from summing up areas of original peaks locating at the same energy bin. Then, the algorithm will check the newly-generated peaks series and make sure that each peak energy only shows up once by deleting any duplicates. The algorithm will then use the MRE to check the newly-generated peaks and iterate the above process until the final peak series satisfies the condition that the difference of any two adjacent peaks in the series is bigger than MRE.

<table>
<thead>
<tr>
<th>Original peaks</th>
<th>$E_1$</th>
<th>$E_2$</th>
<th>$E_3$</th>
<th>$E_4$</th>
<th>..........</th>
<th>$E_N$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st iteration</td>
<td>1</td>
<td>$E_1'$</td>
<td>2</td>
<td>1</td>
<td>$E_2'$</td>
<td>3</td>
</tr>
<tr>
<td>2nd iteration</td>
<td>1</td>
<td>$E_1''$</td>
<td>3</td>
<td>1</td>
<td>$E_2''$</td>
<td>4</td>
</tr>
<tr>
<td>\vdots \</td>
<td>\vdots \</td>
<td>\vdots \</td>
<td>\vdots \</td>
<td>\vdots \</td>
<td>\vdots \</td>
<td>\vdots \</td>
</tr>
<tr>
<td>nth iteration</td>
<td>1</td>
<td>$E_1^{(n)}$</td>
<td>4</td>
<td>3</td>
<td>$E_2^{(n)}$</td>
<td>8</td>
</tr>
</tbody>
</table>

Figure 7 Algorithm iteration example

In Figure 7, one isotope has a series of original $\gamma$-ray energy peaks, $E_1, E_2, E_3, E_4, ..., E_N$, in its $\gamma$-ray spectrum. As for those original peaks, they satisfy the Gaussian correlation shown in Eq. (9). With the known value of MRE, the algorithm will check whether there are peaks located in the region of $[E_i - MRE, E_i + MRE]$ for each peak in the series ($1 \leq i \leq N$). Here, if using a non-constant MRE, the algorithm will first determine which MRE value should be used by checking the relative amplitude of adjacent peaks and comparing with Eq. (10). By summing up Gaussian peaks that are located in the same energy bin around $E_i$ ($1 \leq i \leq N$), the new peak series, $E_1', E_2', E_3', E_4', ..., E_N'$, is generated. In Figure 7, the 1st iteration results in the new peak series, $E_1', E_2', E_3', E_4', ..., E_N'$. And the numbers around the newly generated peak energy $E_i'$ are the lower and upper limits from which the new peak energy $E_i'$ has been generated by summing up original Gaussian peaks located in the lower and upper limits region. For example, $[1, E_1', 2], [1, E_2'', 4]$ represent $peak(E_1') = peak(E_1) + peak(E_2)$ and $peak(E_2'') = peak(E_1) + peak(E_2) + peak(E_3) + peak(E_4)$, respectively. As for the newly generated peak series, the
algorithm will check whether there are duplicate peaks and only keep one in the series. For example, in Figure 7, the 2nd iteration of the algorithm obtains [1, $E_2''$, 4] and [1, $E_3''$, 4] in its new peak series. However, those two peaks $E_2''$ and $E_3''$ have the same lower and upper limits, which represents the same peak energy appearing twice in the same peak series. Hence, the algorithm will delete $E_3''$ before it enters the third iteration. Through deleting duplicate peaks in each iteration, the total peak number in the new peak series will decrease as the iteration number increases. Finally, the algorithm will converge when the energy difference of any adjacent peaks in the new peak series is greater than the MRE.

For each nuclide included in the library, the algorithm will converge after $n$ iterations and the final newly-generated peak series is recorded. Through tests of real spectra, $n = 10$ is a good choice to ensure that the algorithm will converge after $n$ iterations. The newly-generated peak series will not only have many fewer peaks than original peak series, but also will have the benefit that it took the MRE of peak detection algorithm into consideration and will ultimately improve the performance of the identification algorithm.

The new library generated by the algorithm described above will include the peak information of peak centroid and area. The centroid is calculated by finding the location of biggest amplitude of newly-generated peaks and the area is obtained by directly summing up areas of original peaks that constitute the generated peak in the new library. However, the absolute value of the area information in the library has no practical meaning since the area of peaks in the spectra depends on strength of the source, detector’s efficiency and the time to collect the spectrum. Instead, the relative value of the area has the meaning of relative strength of peaks.

Meanwhile, the algorithm suggested does not take the Compton scattering of gamma rays into consideration since the peak detection code based on wavelet analysis eliminates the influence of the Compton continuum. For future work, incorporating the influence from Compton scattering in the algorithm could provide improvements since the wavelet algorithm cannot always completely eliminate influence from Compton continuum.
3.3 Improvement of the algorithm

The application of the algorithm introduced above turns out to have a problem that two resolvable peaks become unresolvable in the new library because of the existence of several much smaller peaks located in the region between the two peaks. In Figure 8, Peak1 and Peak2 are resolvable to the peak detection algorithm. However, the library generated by the algorithm failed to resolve Peak1 and Peak2, and only provided one peak by summing up all peaks between Peak1 and Peak2. The cause of this phenomenon is that the algorithm equally weights all peaks in the peak series. In Figure 8, Peak1 and Peak2 become unresolvable in the library because any two adjacent peaks in the region between Peak1 and Peak2 are unresolvable. In other words, the energy difference of any two adjacent peaks is smaller than MRE. To solve this problem, a small improvement was made to the algorithm. The modified algorithm weights the peaks in the peak series by their amplitudes when generating new peak series. The specific modification is that the algorithm will not count the peak whose amplitude is smaller than both two adjacent peaks, when generating new peak series, even though the peak and its adjacent peaks are not resolvable. For example, in Figure 8, Peak3 will not be counted when generating new peaks since its amplitude is smaller than both its adjacent peaks. Through this approach, Peak4 and Peak5 become resolvable because Peak3 will not be counted. Eventually, Peak1 and Peak2 are resolvable in the generated library. The modification depends on the understanding that the original peak with larger area occupies more in the final newly-generated peaks and only original peaks with bigger amplitude than its adjacent peaks (on at least one side) has the possibility of standing out in the final peak series.

Figure 8 Example of algorithm improvement
Incorporating the modification described above, the algorithm could then work properly and generate the library used by peak searching and library comparison methods. Because the idea of the algorithm is based on summing up unresolvable original Gaussian peaks and correspondingly generating a new peak, the algorithm can incorporate the specific features of the detector by using efficiency-energy curve and FWHM-energy curve of the detector in the Gaussian combination. Hence, the generated library is actually designed for that detector and could be customized by changing the specific choice of the MRE function.
Chapter 4  Validation of the Algorithm

This section will select $^{239}\text{Pu}$ and $^{152}\text{Eu}$ $\gamma$-ray spectra as examples to test the practical application of the algorithm. The algorithm will be used to generate new library from original $\gamma$-ray energy peaks of each isotope. The specific result of tests from $^{239}\text{Pu}$ and $^{152}\text{Eu}$ spectra will verify that the algorithm suggested in Chapter 3 can work properly. In order to precisely analyze the performance of the algorithm, tests were carried out with multiple choices of MRE rather than only one MRE relationship shown in Eq. (10), since the MRE in Eq. (10) is only an approximation of the true MRE for the wavelet peak detection code. Generally, a larger MRE will result in fewer peaks for each isotope and it is difficult to judge whether the peak information obtained from the algorithm make sense or not for very few peaks. The choice of MRE will then have roughly the same approximate MRE in Eq. (10) but be smaller in value.

4.1 290-470 keV complex of $^{239}\text{Pu}$

In order to quantitatively and precisely analyze the performance of the algorithm, one part of the $^{239}\text{Pu}$ spectrum has been taken as the test for the algorithm. Within the energy range of [290,470] keV, $^{239}\text{Pu}$ has 37 original $\gamma$-ray energy peaks in its spectrum, which is shown in Figure 9 [32].

![Figure 9 Original peak series of part of $^{239}\text{Pu}$ spectrum]
In Figure 9, the relative amplitude is normalized to the largest peak in the region. Hence, the largest peak has a relative amplitude of 1 and other peaks have relative amplitude values smaller than 1. Since a larger MRE will result in fewer peaks for each isotope and it is difficult to judge whether the peak information obtained from the algorithm make sense or not for very few peaks, a new test functional MRE in Eq. (11) is proposed for testing, as shown in Figure 10. The test only aims to verify that the algorithm can work properly in generating libraries containing correct peak information, which is independent of the peak detection algorithm. Hence, using a different MRE is acceptable. Meanwhile, the MRE in Eq. (10) is only an approximation of true MRE for the wavelet peak detection algorithm and the main function of the MRE shown in Eq. (10) is to propose the order of MRE that should be chosen for the applied peak detection algorithm. Hence, choosing a new MRE with the same order is meaningful and can also be used to find the locally optimal MRE in Chapter 5. In this chapter, besides the functional MRE shown in Eq. (10) and Eq. (11), a constant MRE \( MRE = 10 \text{ keV}, 20 \text{ keV} \) and \( 30 \text{ keV} \) will also be tested. However, \( MRE = 30 \text{ keV} \) and the MRE in Eq. (10) will only be used for full spectrum tests of \(^{239}\text{Pu}\) rather than test in energy range of [290,470] keV since the results will be very similar to those using \( MRE = 10 \text{ keV}, 20 \text{ keV} \) and the MRE in Eq. (11).

As shown in Figure 10, the MRE satisfies Eq. (11).

\[
MRE = \begin{cases} 
20 \text{ keV} & \text{if} \quad \frac{\text{Amp(peak1)}}{\text{Amp(peak2)}} \geq 5 \\
10 \text{ keV} & \text{if} \quad \frac{\text{Amp(peak1)}}{\text{Amp(peak2)}} < 5 
\end{cases} \tag{11}
\]

**Figure 10 Functional MRE example with smaller values**

As shown in Figure 10, the MRE satisfies Eq. (11).
In Figures 11-13, with $MRE = 10 \text{ keV}$ and $20 \text{ keV}$ and the functional MRE shown in Eq. (11), there are 8 peaks, 5 peaks and 8 peaks respectively remaining in the final library generated by the algorithm. Larger MRE values should result in fewer peaks in the final library, which is verified by the results shown in Figures 11-13. Intuitively, the results match the expected performance of the algorithm. The newly-generated peaks are closer to the locations of original peaks with larger amplitude. Also, if the detection algorithm cannot resolve two adjacent peaks, the newly-generated peak is located in the middle region of those two peaks.

![Figure 11 Original peaks and newly-generated peaks of part of $^{239}\text{Pu}$ spectrum](image1)

($MRE = 10 \text{ keV}$)

![Figure 12 Original peaks and newly-generated peaks of part of $^{239}\text{Pu}$ spectrum](image2)

($MRE = 20 \text{ keV}$)
Compared to 37 original peaks in the same energy range, the newly-generated peak series have many fewer peaks, which means the new library will have smaller size and correspondingly the identification algorithm will have lower computation cost while applying library comparison methods. The result obtained from functional MRE in Eq. (11) is very similar with result from smaller constant MRE shown in Figure 11. The major difference of the three results is the peak centroid and area information obtained from the algorithm. A summary of the peak information in Figures 11-13 is shown in Table 2.

![Figure 13: Original peaks and newly-generated peaks of part of $^{239}$Pu spectrum](image)

*(functional MRE, Eq. (11))*

<table>
<thead>
<tr>
<th>Table 2 Peaks centroid and relative amplitude information corresponding to Figure 9, Figure 10 and Figure 11</th>
</tr>
</thead>
<tbody>
<tr>
<td>$MRE = 10 \text{ keV}$</td>
</tr>
<tr>
<td>Relative Amplitude</td>
</tr>
<tr>
<td>$MRE = 20 \text{ keV}$</td>
</tr>
<tr>
<td>Relative Amplitude</td>
</tr>
<tr>
<td>Functional MRE, Eq. (9)</td>
</tr>
<tr>
<td>Relative Amplitude</td>
</tr>
</tbody>
</table>
In Table 2, the peak’s centroid information shows that the algorithm converged since the final peak series satisfies the criterion that the energy difference of any two adjacent peaks in the final peak series is greater than the MRE. A larger MRE will result in fewer peaks in the new library, which is the reason that the tests are conservatively done by using smaller MRE rather than large MRE shown in Eq. (10) in this test energy region.

However, the validation of the algorithm needed more sophisticated tests to verify the peak information in Table 2 is meaningful. Hence, a real NaI detector spectrum of $^{239}$Pu in the same energy region was chosen to conduct further tests. The example spectrum, shown in Figure 14, was provided by collaborators at Los Alamos National Laboratory. The part of spectrum we are interested in is shown in Figure 15. However, the specific detector response curves (efficiency-energy curve, FWHM-energy curve) of the NaI detector used by the researcher are unknown. In the following tests, the detector response curves used were obtained for a 2-in diameter by 2-in length NaI detector produced by ORTEC, catalog number is 905-3 [33]. The efficiency-energy curve and FWHM-energy curve of the detector are shown in Figure 16 and Figure 17 respectively.

![Pu239 spectrum](image)

**Figure 14** $^{239}$Pu spectrum (NaI detector)
Figure 15 Featured part of $^{239}Pu$ spectrum

Figure 16 Efficiency-energy curve of the detector

<table>
<thead>
<tr>
<th>Model</th>
<th>Exponential</th>
</tr>
</thead>
<tbody>
<tr>
<td>Equation</td>
<td>$y = y_0 + A \exp\left(R_0^2 x\right)$</td>
</tr>
<tr>
<td>Reduced Chi-Adj. R-Square</td>
<td>4.66596E-7</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>B</th>
<th>Value</th>
<th>Standard Er</th>
</tr>
</thead>
<tbody>
<tr>
<td>$y_0$</td>
<td>0.0016</td>
<td>4.301E-4</td>
</tr>
<tr>
<td>$A$</td>
<td>0.5210</td>
<td>0.00296</td>
</tr>
<tr>
<td>$R_0$</td>
<td>-0.007</td>
<td>6.83843E-5</td>
</tr>
</tbody>
</table>
The efficiency-energy curve and FWHM-energy curve shown in Figures 16 and 17 will be used in the algorithm to calculate the parameters of the Gaussian shaped peak in the original peak series. In the $^{239}$Pu test case, the algorithm with the functional MRE in Eq. (11), was used to generate new peaks in the energy range shown in Figure 15. The algorithm-generated values of centroid and relative area are shown in Table 3 and the library peak locations with respect to $^{239}$Pu spectrum are shown in Figure 18.

Table 3 Newly-generated peaks information: centroid and relative area

<table>
<thead>
<tr>
<th>Functional MRE</th>
<th>Centroid/keV</th>
<th>Relative Area</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>298.6</td>
<td>0.03</td>
</tr>
<tr>
<td></td>
<td>318.8</td>
<td>0.07</td>
</tr>
<tr>
<td></td>
<td>332.6</td>
<td>0.3</td>
</tr>
<tr>
<td></td>
<td>343.5</td>
<td>0.31</td>
</tr>
<tr>
<td></td>
<td>376.2</td>
<td>1.00</td>
</tr>
<tr>
<td></td>
<td>389.9</td>
<td>0.34</td>
</tr>
<tr>
<td></td>
<td>414.3</td>
<td>0.68</td>
</tr>
<tr>
<td></td>
<td>451.3</td>
<td>0.08</td>
</tr>
</tbody>
</table>
Similarly, the results of $MRE = 20 \text{ keV}$ are shown in Table 4 and Figure 19. In Figure 18 and Figure 19, the left vertical axis is the counts of real $^{239}\text{Pu}$ spectrum in the energy region of interest and the right vertical axis is the relative amplitude of newly-generated peaks. Intuitively, the newly-generated peaks’ centroid matches the expected location. In particular, in Figure 19 the three new main peaks’ centroid is the estimated location of observed peaks in the spectrum. Hence, the algorithm does have the ability to calculate the centroids of newly-generated peaks. Additionally, the algorithm also outputs relative area information of the peaks in the new library. In Figure 19, the relative amplitude given by the library matches the real amplitude trend of the observed peaks in the spectrum. In other words, larger peaks observed in the spectrum have larger relative amplitude in the library.

Table 4 Newly-generated peaks information: centroid and relative area

($MRE = 20 \text{ keV}$)

<table>
<thead>
<tr>
<th>$MRE = 20 \text{ keV}$</th>
<th>Centroid/keV</th>
<th>$303.03$</th>
<th>$337.43$</th>
<th>$377.53$</th>
<th>$414.23$</th>
<th>$451.03$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative Area</td>
<td>$0.06$</td>
<td>$0.66$</td>
<td>$1.00$</td>
<td>$0.46$</td>
<td>$0.04$</td>
<td></td>
</tr>
</tbody>
</table>
Figure 19 $^{239}\text{Pu}$ spectrum and newly-generated peaks under $MRE = 20 \text{ keV}$

Figure 20 Gaussian and linear background fit of the $^{239}\text{Pu}$ spectrum

$(MRE = 20 \text{ keV})$
However, the absolute value of relative amplitude (or area) obtained by the algorithm has little difference with the values obtained from the fit of the spectrum. In Figure 20, the spectrum is fit by a linear continuum and three Gaussian peaks located at the centroids suggested by the algorithm. The fitting results provide the area of three fit peaks, namely, $A_1, A_2$ and $A_3$. From Figure 20, a linear continuum and three Gaussian peaks fit of the spectrum work well. Using fixed Gaussian peaks, centroids obtained from the new library and width calculated from method of net-width, the fit was carried out to obtain the optimal value of the peak area. Since the three newly-generated peaks are not the original Gaussian peaks, the standard deviation (FWHM/2.355) of peaks does not satisfy the FWHM-energy curve shown in Figure 17. Intuitively, the new peaks will have larger widths than the width obtained by applying FWHM-energy relationship because the new peak is the overall summation of several overlapping Gaussian peaks. The overlapping of peaks will construct a new peak with broader width, referred to as the net-width. The value of fixed net-width applied in the fit is calculated by fitting the new peak with a standard Gaussian relationship. For example, in Figure 21, the second peak width is obtained by first summing up all the original peaks contributing to the new peak in order to determine the peak data and then fit the data by a Gaussian relationship.

![Figure 21 Example of getting net-width of newly-generated peaks](image-url)
The comparison between relative area obtained from the fitting of the spectrum and the relative area obtained by the library algorithm is shown in Table 5. In Figure 22, the relative area ratio with respect to energy of each peak is plotted.

Table 5 Comparison of relative area between fitting and library results

\[(MRE = 20\, \text{keV})\]

<table>
<thead>
<tr>
<th>Peaks centroid/keV</th>
<th>Fitting Area</th>
<th>Relative area from fitting (A_f)</th>
<th>Relative area from library (A_l)</th>
<th>Ratio (A_f/A_l)</th>
</tr>
</thead>
<tbody>
<tr>
<td>337.4</td>
<td>17493.3</td>
<td>0.52</td>
<td>0.66</td>
<td>0.78</td>
</tr>
<tr>
<td>377.5</td>
<td>33795.6</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>414.2</td>
<td>16318.8</td>
<td>0.48</td>
<td>0.47</td>
<td>1.03</td>
</tr>
</tbody>
</table>

Figure 22 Relative area ratios between fitting and library results

The three points in Figure 22 show that the ratio does not equal to the expected value of 1, but has a trend of increasing as the energy of the peak increases. The reason for this may be that the detector response curves applied were not good estimates of the detector used to collect the spectrum, or that the algorithm does not take shielding effects into consideration. Since the detector response curves of the detector used to collect the spectrum of \(^{239}\text{Pu}\) are not determined, the calculation of relative area values in the library algorithm should be influenced. On the other hand, the shielding will decrease the possibility of \(\gamma\)-rays reaching detector. However, this
affects the lower energy γ-rays more than the higher energy γ-rays. Meanwhile, the self-shielding of materials has the same influence. Hence, the lower energy peaks should have a smaller ratio because the library result does not take the shielding into consideration.

From above analysis, it is clear that the algorithm has the ability to generate new libraries containing the peaks that having the largest possibility of being detected by a peak detection algorithm. The algorithm succeeds in generating new peaks whose centroids are estimations of the peaks that will be observed in the spectrum. The similarity between the peak detection algorithm performance and the library results will make the overall identification algorithm work better. The absolute value of the relative area suggested by the algorithm still needs some improvement to precisely match the peak area information of the real spectrum.

4.2 Whole \(^{239}\text{Pu}\) spectrum case

In this section, the overall performance of the algorithm on the whole spectrum of \(^{239}\text{Pu}\) will be tested and not just an energy region of interest. The final library generated by using the functional MRE in Eq. (11), constant \(MRE = 20 \text{ keV}\), the functional MRE in Eq. (10), and constant \(MRE = 30 \text{ keV}\) are shown in Figure 24-27. Compared to original peaks case shown in Figure 23, there are significantly fewer peaks in the new libraries shown in these figures, which will decrease the computational cost in the identification algorithm and improve overall performance.

![Pu239 spectrum and all its original γ-ray peak locations](image)
In Figure 23-27, the horizontal axis, left vertical axis and right vertical axis represent the energy, counts of the spectrum and “Log(Area)+30”, respectively. The blue solid curve is the real \(^{239}Pu\) spectrum. The red dash line illustrates the final peak locations generated by the algorithm. The amplitude of the red lines corresponds to the area of the new peaks calculated from the algorithm, and has been modified by
taking their logarithm values and adding 30 in order to clearly show all of the peaks. Figures 24-27 demonstrate that a larger MRE will result in fewer peaks in the final library as expected. Comparing the results in Figures 24, 26, and 27 shows that the final library changes significantly for these three MRE cases, especially for the peak centroid. The new peaks in the final library tend to correlate well with the location where peaks are obvious with the naked eye.

The overall performance of the algorithm through the whole test of $^{239}$Pu case using a constant MRE and the functional MRE in Eq. (11) shows that the algorithm has the ability to accurately generate a new library with peak centroids and relative areas. The library peak centroids are well matched to the peak locations in the original spectrum of $^{239}$Pu, which verifies that the algorithm works well on predicting the new peak locations. Meanwhile, the relative area in the new library generally matches the relative area of peaks in the spectrum. However, the absolute value of relative area in the new library has some differences from the relative area value obtained from a fit of the spectrum. The reason may come from the fact that the exact detector response curves and shielding configurations were unknown.

![Figure 26 $^{239}$Pu spectrum and newly-generated peaks (MRE = 30 keV)](image)

The library generated is much better than a simple library that chooses several biggest peaks from original peak series since the simple library cannot insure that the peaks in it are separated enough to be resolvable by the peak detection algorithm.
However, because the algorithm took the MRE of peak detection algorithm into consideration, the library generated will contain the peaks that have a higher probability of being observed in the spectrum and detected by the peak detection algorithm. Hence, the library generated is much more efficient and meaningful.

Figure 27 $^{239}\text{Pu}$ spectrum and newly-generated peaks (Functional $MRE$ in Eq. (10))

In practical application of the algorithm, the library should be generated using one optimal MRE valid for the specific peak detection algorithm applied. Hence, the performance of the libraries generated by using different MREs will be tested in Chapter 5 through coupling with a specific wavelet peak detection algorithm and different libraries in order to analyze the optimal MRE value and evaluate the overall performance of the two algorithms.

4.3 $^{152}\text{Eu}$ case

In this section, the algorithm suggested in Chapter 3 is tested with $^{152}\text{Eu}$ and compared with a real $^{152}\text{Eu}$ spectrum obtained with the same NaI detector, whose response curves are the same curves used in the $^{239}\text{Pu}$ case shown in Figure 16 and Figure 17. Since $^{152}\text{Eu}$ also has many peaks in its $\gamma$-ray spectrum, it is another good example to show the real performance of the algorithm. The libraries are generated using constant $MRE = 20 \text{ keV}$ and $30 \text{ keV}$ and the functional MRE in Eq. (10) and Eq. (11). In Figure 28, the $^{152}\text{Eu}$ spectrum and all its $\gamma$-ray peaks have been plotted.
Figure 28 $^{152}Eu$ spectrum and original peak locations

Figure 28 demonstrates that the $^{152}Eu$ has numerous peaks in its $\gamma$-ray spectrum. Some peaks are too close to be resolved by most peak detection algorithms. Additionally, most nearby peaks’ areas have an order of magnitude difference. Because peaks with smaller area have much less possibility to be detected by a peak detection code, it is meaningful to find a method that could extract useful peaks from the hundreds of original peaks shown in Figure 28. Hence, the algorithm suggested in Chapter 3 is applied to create new libraries.

Figure 29 $^{152}Eu$ spectrum and newly-generated peaks ($MRE = 20 \text{ keV}$)
In Figures 29-32, the $^{152}$Eu spectrum and new library peaks generated by using the different MRE relationships have been plotted. They show that the newly-generated peaks are located very close to the main peaks in the original peak series, which is just as we expect how the algorithm should work. The relative area information, shown in Figures 29-32, indicates that the newly-generated peaks have the correct relative area as derived from original peaks’ information shown in Figure 28. Compared with Figure 28, the new library preserves the large peaks that have more probability of being detected and eliminates the small peaks that have little possibility to be recognized. Generally, the results show that the algorithm can work for extracting main peaks from hundreds of original peaks.

Comparing the libraries in Figure 29 to Figure 32, the library generated using the larger MRE will have fewer peaks compared to the library generated using smaller MRE. From Figures 29-32, either library could be the best library, which really depends on how well the peak detection code works. If the peak detection algorithm can resolve more peaks, the library generated using smaller MRE will be better. On the other hand, if peak detection algorithm could only find very clear peaks, a library generated using bigger MRE is enough to provide needed peaks information.
Meanwhile, in Figures 29-32, they also show that the newly-generated peaks may show up at locations that seem no peaks showing up. The reason for this behavior comes from the fact that the spectrum is got from low-resolution NaI detector. The poor resolution of the detector will blur small peaks and induce that it is hard to recognize the peaks with naked eyes.
4.4 Conclusion

From above three test cases, the results show that the algorithm suggested in Chapter 3 does have the ability to generate a new library from the original standard $\gamma$-ray peaks of each isotope. The new library will contain the new peak centroids and area information. Sample libraries can be found in Appendix B. As expected, the new peaks generated by the algorithm correlate well with the main peaks in the original peak series of that isotope. The algorithm also provides the new peaks’ area, which will be used in the isotope identification algorithm. The $^{239}Pu$ test shows that the relative value of new peaks’ area matches the desired performance, namely, peaks with larger branching ratios have larger areas. Additionally, the new library is much better than a simple library that chooses several large peaks from original peak series because the algorithm took the MRE of the peak detection algorithm into consideration and the new library peaks have a higher probability of being observed in the spectrum and detected by the peak detection algorithm. Therefore, the overall performance of the algorithm reaches the design criterion.

In this chapter, the algorithm has used several different MRE relationships to generate new libraries for each isotope. In Chapter 5, the generated libraries will be tested with a wavelet peak detection algorithm in order to evaluate the overall performance of both algorithms and to analyze the optimal MRE.
Chapter 5  Experiments

In this chapter, $^{239}\text{Pu}$ and $^{152}\text{Eu}$ will be taken as examples to test both the wavelet peak detection algorithm and the libraries generated in Chapter 4. The main purpose is to compare the peaks detected by wavelet analysis algorithm with the new peaks in libraries. The optimal MRE to be studied in this chapter is the one that will generate an optimal library for a library comparison method applied in following identification algorithm. In this thesis, since the identification algorithm is the subject of on going research, the exact optimal MRE will not be determined but analyzed in Chapter 5.

5.1 $^{239}\text{Pu}$ case

The $^{239}\text{Pu}$ spectrum was analyzed by the wavelet peak detection code, which will provides the peaks’ centroid and area information. Due to the present limitations of the wavelet code, in this chapter, the comparison between peaks detected by wavelet code and the new peaks in libraries will be focused on the comparison of peaks’ centroid information. In Figure 33, the peaks of $^{239}\text{Pu}$ spectrum detected by the wavelet peak detection algorithm are plotted. In Figures 34-37, the peaks detected by wavelet peak detection algorithm and the peaks in the different libraries have been plotted. They show that the wavelet code fails to detect several peaks across the spectrum, such as peaks around 110 keV and 350 keV. The lowest energy peak detected by the wavelet code was not present in the libraries of $^{239}\text{Pu}$. The reason may be that there are contamination or daughter isotopes existing in the $^{239}\text{Pu}$ source. An isotope identification algorithm based on library comparison method will then compare the results from the peak detection algorithm with built-in libraries and output the final result.

In Figures 34-37, the blue curve, the black dash line and the red dash line represent the real $^{239}\text{Pu}$ spectrum, the location of peaks detected by wavelet algorithm and the peaks’ centroid location in the corresponding libraries. It shows that there is one peak in each library locating very close to each peak detected by wavelet peak detection algorithm except for the lowest energy peak.
Figure 33 $^{239}$Pu spectrum and peaks detected by wavelet peak detection algorithm

Figure 34 Comparison between peaks detected by wavelet algorithm and peaks in library generated using $MRE = 20\text{ keV}$

Through comparing peaks detected by wavelet algorithm with peaks in libraries, an isotope identification algorithm will evaluate which isotope library matches the detected peaks best and conclude that the spectrum is from that isotope. Hence, a better match between the detected peaks’ centroid and peaks’ centroid in the library will result in better isotope identification.
Figure 35 Comparison between peaks detected by wavelet algorithm and peaks in library generated using $MRE$ in Eq. (11)

Figure 36 Comparison between peaks detected by wavelet algorithm and peaks in library generated using $MRE = 30 \text{ keV}$
Figure 3 Comparison between peaks detected by wavelet algorithm and peaks in library generated using $MRE$ in Eq. (10).

Table 6 Errors between library peak centroids and peaks detected

<table>
<thead>
<tr>
<th>Peaks detected/keV</th>
<th>MRE = 20 keV</th>
<th>MRE = 30 keV</th>
<th>MRE in Eq.(9)</th>
<th>MRE in Eq.(8)</th>
</tr>
</thead>
<tbody>
<tr>
<td>61.5</td>
<td>-16.0%</td>
<td>-16.0%</td>
<td>-16.0%</td>
<td>-16.0%</td>
</tr>
<tr>
<td>205.1</td>
<td>-1.6%</td>
<td>-1.6%</td>
<td>-1.6%</td>
<td>-1.6%</td>
</tr>
<tr>
<td>240.2</td>
<td>5.2%</td>
<td>5.2%</td>
<td>5.2%</td>
<td>5.2%</td>
</tr>
<tr>
<td>377.9</td>
<td>-0.1%</td>
<td>-0.2%</td>
<td>-0.5%</td>
<td>-0.2%</td>
</tr>
<tr>
<td>410.2</td>
<td>1.0%</td>
<td>-0.4%</td>
<td>1.0%</td>
<td>-0.4%</td>
</tr>
<tr>
<td>621.1</td>
<td>0.0%</td>
<td>4.0%</td>
<td>-0.6%</td>
<td>4.0%</td>
</tr>
<tr>
<td>764.6</td>
<td>1.2%</td>
<td>1.1%</td>
<td>-1.1%</td>
<td>1.1%</td>
</tr>
<tr>
<td>884.8</td>
<td>0.2%</td>
<td>0.2%</td>
<td>0.2%</td>
<td>0.2%</td>
</tr>
<tr>
<td>1037.1</td>
<td>1.9%</td>
<td>1.9%</td>
<td>1.9%</td>
<td>1.9%</td>
</tr>
</tbody>
</table>

From Figures 34-37, library peak centroid errors with respect to the peaks detected by the current wavelet peak detection algorithm have been quantitatively shown in Table 6. It shows that the library generated by using $MRE = 20$ keV matches a little better than other three libraries. However, the above conclusion is only valid when the identification algorithm equally weights all detected peaks instead of weighting more for peaks with larger area and smaller area uncertainty. On
the other hand, if the peak detection algorithm weights more on peaks with larger area and better area uncertainty, the best library choice may be different. From tests of the wavelet algorithm, it was observed that the performance of the algorithm is very sensitive to noise, especially for the peak area calculation. Hence, the four peaks detected between 600 keV and 1100 keV where the spectrum has a low signal to noise ratio (SNR) should be given a smaller weight when applying the identification algorithm. The optimal MRE highly depends on the specific identification algorithm, which will not be determined in this thesis.

In Figures 33-37, the plots also show the peak area information for both peaks detected by wavelet algorithm and peaks in the libraries. However, simply comparing absolute values of area obtained with the wavelet code and the libraries themselves makes no sense because the area of peaks depends on the strength of the source, detector’s efficiency and the time to collect the spectrum. On the other hand, comparing the relative area, such as comparing two detected peaks’ area ratio with corresponding ratio calculated from peak area in the library, is much meaningful. In Figures 33-37, the relative area from the wavelet code and the libraries match better in the lower energy region, such as 50 keV to 400 keV, but worse in region between 600 keV and 1100 keV. The reason comes from the fact that current wavelet peak detection algorithm is sensitive to noise and can have relatively large errors between 600 keV and 1100 keV when calculating the peaks’ centroid and area. Generally, in the lower energy region where the wavelet peak detection algorithm is more accurate in calculating peaks’ centroid and area, the libraries demonstrate good performance in providing the main peaks’ centroid and area information.

5.2 $^{152}$Eu case

In this section, $^{152}$Eu spectrum was analyzed by the wavelet peak detection code and then the peak detection results were compared with the libraries generated in Chapter 4. In Figure 38, the peaks detected by the wavelet algorithm have been plotted together with $^{152}$Eu spectrum. Similarly, the wavelet algorithm also detected several peaks in the low energy region that are x-ray peaks or back scattering and will not be present in the libraries. Additionally, the wavelet algorithm also indicated that the three peaks between 300 keV and 600 keV are overlapping peaks, such as the peak around 350 keV, which actually consists of two overlapping peaks.
Figure 38 $^{152}$Eu spectrum and peaks detected by wavelet peak detection algorithm

Figure 39 Comparison between peaks detected by wavelet algorithm and peaks in library generated using $MRE = 20$ keV
In Figures 39-42, the peaks detected by the wavelet peak detection algorithm and peaks in the different libraries have been plotted. For each peak detected by wavelet peak detection algorithm, the libraries, shown in Figures 39-42, have corresponding peaks located at very close peak energy. For some peaks, the library perfectly matched the peak location, such as the peaks around 120 keV, 250 keV, 780 keV and 970 keV in Figures 39-42. However, for overlapping peaks, the peak detection algorithm result is a little different from the libraries. For example, in Figures 39 and 40, the detected overlapping peaks around 350 keV have one library peak very close to the detected lower energy peak. But the detected higher energy peak is a little far from library record. This difference may come from the performance of peak detection algorithm, namely, how well the peak detection algorithm could resolve overlapping peaks and how accurate the result is. Also, the library peak locations are a little shifted to the left around the overlapping peaks located around 350 keV and 1100 keV in Figures 39 and 40. The linear assumption of energy calibration may contribute to this behavior.

In Figures 41-42, because of the larger MRE, the algorithm suggested in Chapter 3 will sum up some “close” peaks and generate a new peak in the middle region compared to the libraries in Figures 39 and 40. However, it turns out the wavelet algorithm succeeded in identifying the two overlapping peaks around 1100 keV. Hence, from this point of view, the libraries in Figures 39 and 40 are better.
However, the wavelet peak detection algorithm may fail to resolve those two overlapping peaks when the noise is bigger. In conclusion, the optimal library should be generated using an MRE that could take the performance of the peak detection algorithm over the whole spectrum into consideration.

**Figure 41** Comparison between peaks detected by wavelet algorithm and peaks in library generated using $MRE = 30$ keV

**Figure 42** Comparison between peaks detected by wavelet algorithm and peaks in library generated using $MRE$ in Eq. (10)
5.3 Conclusion

In $^{239}\text{Pu}$ case, the wavelet algorithm works well over 50 keV to 500 keV but could not output high quality results over region between 600 keV and 1100 keV because of the much smaller SNR in that region. Because $^{239}\text{Pu}$ doesn’t have important overlapping peaks in the region between 50 keV to 500 keV, the library generated by relatively larger MRE is better choice since the library could eliminate small peaks which have much less possibility being detected, but may have big influence on the isotope identification algorithm if the identification algorithm equally weights all peaks. In the $^{152}\text{Eu}$ case, the spectrum has several important overlapping peaks and they are located in the region where the wavelet algorithm could work well enough to resolve them. Hence, the smaller MRE is better choice for $^{152}\text{Eu}$ since a larger MRE will result in the generating a library missing one of the overlapping peaks. On the other hand, if the spectrum has a small SNR and the wavelet peak detection algorithm can’t resolve the two overlapping peaks, a larger MRE will be better.

In practical application of the library algorithm, because of the expected relatively small SNR of spectra collected by detectors in real world applications, the library generated by using a relatively larger MRE maybe the better choice. The optimal MRE still needs researching and there are many factors that could influence the choice of MRE, such as the specific identification algorithm, the SNR of the spectra, the specific feature of the isotope spectra and so on. The easiest method to determine the optimal MRE is to generate several libraries and apply the isotope identification algorithm to evaluate which library work best. The optimal MRE will be the one that generates the library with best performance when applying the isotope identification algorithm.
Chapter 6 Summary and Future Work

6.1 Summary

This thesis aims to suggest an algorithm to generate customized libraries for commercial radioactive isotope identifiers. Through incorporating response curves, such as the efficiency-energy curve and the FWHM-energy curve of the specific detector, the algorithm generates a library that is custom designed for that detector. As for future application, the thesis lists isotopes that should be included in the library in Chapter 2 and discussed the Bateman equation concerning whether the daughters of those radioactive isotopes should also be included in the library. In Chapter 3, the algorithm is proposed and explained. Chapters 4 and 5 showed the tests of the algorithm suggested in Chapter 3 in order to verify that the algorithm performance works as expected, and analyze the optimal settings for the algorithm.

The minimum resolvable energy (MRE), the minimum energy difference of two adjacent peaks that the peak detection algorithm could resolve, is the most important factor that could have direct impact on final library outputs. In Chapters 4 and 5, several libraries were generated using different MRE relationships and then tested with real spectra and the wavelet peak detection algorithm. The test results show that the optimal MRE is difficult to determine because it highly depends on the SNR of spectrum to be analyzed and specific \( \gamma \)-ray peak features of the isotope. Generally, for good quality spectra with a large SNR, the wavelet peak detection algorithm works sufficiently for resolving overlapping peaks. Hence, under this condition, a relatively small MRE is better since the library generated will have enough peaks to match with the peak detection result. On the other hand, if the SNR of the spectrum is small, a relatively large MRE will be the better choice since the peak detection code cannot effectively resolve overlapping peaks. The optimal MRE is also associated with the identification algorithm. A smaller MRE will generate a library with more peaks that will have a bad influence on the isotope identification if the identification algorithm equally weights all peaks regardless of their area, since some isotopes have very close characteristic peaks. On the other hand, if using a larger MRE, the generated library will combine small peaks with the adjacent bigger peaks. Hence, even though two isotopes have close characteristic peaks in the original
peak series, the newly-generated peaks in the library of one isotope may eliminate that “close” peaks because of their small area.

The algorithm was demonstrated to generate a new library with a new series of peaks whose centroid is good estimation of the observed peaks in the real spectrum. It also provides the new peaks’ area information. Since the absolute value of peak area in the spectrum depends on the strength of the source, detector’s efficiency and the time to collect the spectrum, the absolute value of area for the new peaks obtained by the algorithm has no meaning, but the relative area could be used as reference for the isotope identification algorithm. However, the tests also showed that more work is needed with the algorithm in order to measure a highly precise value of relative area.

Meanwhile, the new library was shown to be much better than a simple library that chooses the several largest peaks from the original peak series. Because the algorithm took the MRE of the peak detection algorithm into consideration, the library generated will contain the peaks that have the highest probability of being observed in the spectrum and detected by the peak detection algorithm.

In conclusion, the algorithm suggested in this thesis worked well in extracting the main peaks from the original peak series of one isotope. The newly-generated peaks in the library are good estimates of those observed peaks in the real spectrum. Knowing the response curves of the detector and determining the MRE of the peak detection algorithm applied, the algorithm could be used to generate a customized library for the specific combination of detector and peak detection algorithm used. However, the algorithm still needs improvements with area calculation of the newly-generated peaks in order to make it more precise. The optimal MRE to generate the best applicable library depends on several factors and cannot be determined currently.

6.2 Future work

The current performance of the algorithm demonstrates promise. More work needs to be done on the calculation of area for newly-generated peaks, and the optimal MRE to generate library is still not determined. Even though it depends on the specific identification algorithm applied, a quantitative analysis of the effect from the SNR of the spectrum on the MRE should be done in future. Meanwhile, the algorithm suggested does not take the Compton scattering of gamma rays into consideration since the peak detection code based on wavelet analysis eliminates the
influence of the Compton continuum. For future work, incorporating the influence from Compton scattering in the algorithm could provide improvements since the wavelet algorithm cannot always completely eliminate influence from Compton continuum.
Appendix A  Matlab Code

function [centroid,LibArea]=newcompeak_minE(Energy,BrR,minE,maxE,thresh)

%Generate a library for a isotope that has gamma-ray energy, Energy, and
%branching ratio, BrR, using minimum resolvable energy (MRE).
%MRE=minE if Amp2/Amp1<thresh
%MRE=maxE if Amp2/Amp1>=thresh
%where Amp2, Amp1 are amplitude of two adjacent peaks.
%example:[cen,area]=newcompeak_minE(Pu239_Energy,Pu239_BR,10,20,5);

% clear all;
% load('Pu239.mat');
% Energy=Pu239_Energy;
% BrR=Pu239_BR;
nonzero=find(BrR>0);   %Deleting branching ratio data being empty
E=Energy(nonzero)';
BR=BrR(nonzero)';
num=size(E,2);  %number of peaks in original peak series
maxnum=num;

% minE=28;
% maxE=36;
% thresh=5;

Emin=min(E)-50;
Emax=max(E)+50;
Ex=Emin:0.1:Emax;
num2=size(Ex,2);
opeaks=zeros(num,num2);
area=zeros(1,num);
Amp=zeros(1,num);

%Incorporating detector response curves, efficiency-energy & FWHM-energy
%curve, in the peak parameters. Assuming original peak has a shape of
%Gaussian, whose area is proportional to efficiency*Branching Ratio.

for i=1:num
    ieff=0.52109*exp(-E(i)/142.70732)+0.00161;
iwidth=(2.32647e-5*(E(i).^2)+0.08463*E(i)+1.83083)/2.355;
area(1,i)=BR(i)*ieff;
oparks(1,:)=(area(1,i)*1/(sqrt(2*pi)*iwidth)*exp(-((Ex-E(i)).^2)/(2*iwidth.^2)));
Np=opeaks(i,:);
res=find(Np==max(Np),1,'first');
Amp(1,i)=Np(1,res);
end

%Using the MRE given, iteratively find left&right limits of peaks that
%cannot be resolved by peak detection algorithm. Generally, the
<algorithm aims to sum up the peaks located in the same energy window of
%MRE since they are not likely to be resolved.

Inamp=Amp;

j=1;
Epeak=zeros(num,num2);
Area=zeros(1,num);
dlim=zeros(10,num); %left&right limits matrix, saving left & right limits
ulim=zeros(10,num); %of peaks during each iteration

%first iteration: find left&right limits of each peak in original peak
%series
for i=1:num
    N=zeros(1,num2);
tempA=0;
    if i==1
        dlim(1,j)=1;
    else
        dlim(1,j)=llim_minE(E,Amp,i,minE,maxE,thresh); %left limit
    end
    if i==num
        ulim(1,j)=maxnum;
    else
        ulim(1,j)=rlim_minE(E,Amp,i,minE,maxE,thresh); %right limit
    end
    if dlim(1,j)<=ulim(1,j) %changed on Feb 24 from '<' - '=='
        for k=dlim(1,j):ulim(1,j)
            Nk=opeaks(k,:);
            N=N+Nk;
tempA=tempA+area(1,k);
        end
        Epeak(j,:)=N;
        Area(1,j)=tempA;
    j=j+1;
    end
end

nump=j-1;
Ep=zeros(1,nump);
Amp=zeros(1,nump);
for i=1:nump %find centroid&area of new summation peaks
    Np=Epeak(i,:);
    res=find(Np==max(Np),1,'first');
    Ep(1,i)=(res-1)*0.1+Emin;
    Amp(1,i)=Epeak(i,res);
end
iasum=zeros(100,nump);
icsum=zeros(100,nump);
Asum=zeros(100,nump);
[Ep2,ia,ic]=unique(Ep);  %delete any duplicates in the new peak series

pdlim=zeros(100,nump);
pulim=zeros(100,nump);

%Several iterations after the first iteration, taking small step toward  
%convergence through only comparing adjacent peaks rather than all peaks
%that cannot resolved depending on MRE

%Through tests, 10 iterations are enough to converge
for n=1:10
    iasum(n,1:length(ia))=ia;
    icsum(n,1:length(ic))=ic;
    Asum(n,1:length(Area))=Area;
    pdlim(n,1:length(ia))=dlim(n,ia);
    pulim(n,1:length(ia))=ulim(n,ia);
    num=size(Ep2,2);
    Epeak=zeros(num,num2);
    Area=zeros(1,num);
    j=1;
    upcheckno=0;
    pn=0;
    for i=1:num
        N=zeros(1,num2);
        flag1=1;
        flag2=1;
        nopeak_left=0;  %flag for judging whether the peak has adjacent left peak
        nopeak_right=0;  %flag for judging whether the peak has adjacent right peak
        tempA=0;
        if i==1
            dlim(n+1,j)=1;
            flag1=0;
            nopeak_left=1;
        else
            if Ep2(i-1)<Ep2(i)-maxE  %No left adjacent peak within MRE
                dlim(n+1,j)=pdlim(n,i);
                flag1=0;
                nopeak_left=1;
            else
                ratio=Amp(i)/Amp(i-1);
                %Only count peaks whose amplitude is bigger than adjacent
                %peaks(at least one side)
                if ratio>=1
                    if ratio<thresh
                        dE=minE;
                %end
            else
                %end
        end
end
end
else
dE=maxE;
end

if Ep2(i-1)>Ep2(i)-dE %Left adjacent peak cannot resolve
   if pdlim(n,i-1)<=pdlim(n,i)
      dlim(n+1,j)=pdlim(n,i-1);
   else
      dlim(n+1,j)=pdlim(n,i);
   end
else %Left adjacent peak can resolve
   dlim(n+1,j)=pdlim(n,i);
end
else
dlim(n+1,j)=pdlim(n,i);
flag1=0; %indicate the peak has left adjacent peak, but amplitude is smaller than its left adjacent peak
end
end

%Right adjacent peak case
if i==num
   ulim(n+1,j)=maxnum;
   flag2=0;
   nopeak_right=1;
else
   if Ep2(i+1)>Ep2(i)+maxE %no adjacent right peak
      ulim(n+1,j)=pulim(n,i);
      flag2=0;
      nopeak_right=1;
   else
      ratio=Amp(i)/Amp(i+1);
      if ratio>=1
         if ratio<thresh
            dE=minE;
         else
            dE=maxE;
         end
      end
      if Ep2(i+1)<Ep2(i)+dE
         if pulim(n,i+1)>=pulim(n,i)
            ulim(n+1,j)=pulim(n,i+1);
         else
            ulim(n+1,j)=pulim(n,i);
         end
      else
         ulim(n+1,j)=pulim(n,i);
      end
   else
      ulim(n+1,j)=pulim(n,i);
   end
else
   ulim(n+1,j)=pulim(n,i);
end
ulim(n+1,j)=pulim(n,i);
flag2=0; %indicate the peak has right adjacent peak, but amplitude is smaller than its right adjacent peak
end
end
end

%sum up unresolvable peaks
if flag1+flag2>0 || nopeak_left*nopeak_right==1
%if peak has adjacent peaks, it’s amplitude should be bigger than one adjacent peak;
if peak doesn’t have adjacent peaks within Emin, accept it.
    for k=dlim(n+1,j):ulim(n+1,j)
Nk=opeaks(k,:);
    N=N+Nk;
tempA=tempA+area(1,k);
end
Epeak(j,:)=N;
Area(1,j)=tempA;
end
end

%finding peak centroids and areas
nump=j-1;
Ep=zeros(1,nump);
Amp=zeros(1,nump);
fwhm=zeros(1,nump);
for i=1:nump
    Np=Epeak(i,:);
    res=find(Np==max(Np),1,'first');
    Ep(1,i)=(res-1)*0.1+Emin;
    Amp(1,i)=Epeak(i,res);
end

[Ep2,ia,ic]=unique(Ep);
if nump==1
    break;
end
end

centroid=Ep';
LibArea=Area';
function rminE=rlim_minE(E,Amp,i,minE,maxE,thresh)
%Find left limit of unresolvable peaks
if E(i+1)>E(i)+maxE
    rminE=i;
    return;
else
    n=i+1;
    while(n<=size(E,2))
        ratio=Amp(i)/Amp(n);
        if ratio>1
            if ratio<thresh
                if E(n)<E(i)+minE
                    rminE=n;
                else
                    rminE=n-1; return;
                end
            else
                if E(n)<E(i)+maxE
                    rminE=n;
                else
                    rminE=n-1; return;
                end
            else
                rminE=n-1;return;
            end
        end
        n=n+1;
    end
end

function lminE=llim_minE(E,Amp,i,minE,maxE,thresh)
%Find right limit of unresolvable peaks
if E(i-1)<E(i)-maxE
    lminE=i;
    return;
else
    n=i-1;
    while(n>0)
        ratio=Amp(i)/Amp(n);
        if ratio>1
            if ratio<thresh
                if E(n)>E(i)-minE
                    lminE=n;
                else
                    lminE=n+1; return;
                end
            else
                lminE=n+1; return;
            end
        else
            if E(n)>E(i)-maxE
                lminE=n;
            else
                lminE=n;
            end
        end
        n=n-1;
    end
end
else
    lminE=n+1; return;
end
else
    lminE=n+1; return;
end
n=n-1;
end
end

function [centroid,LibArea]=Cons_minE(Energy,BrR,dE)

%Generate a library for a isotope that has gamma-ray energy, Energy, and
%branching ratio, BrR, using constant minimum resolvable energy (MRE).
%MRE=dE
%example:[cen,area]=Cons_minE(Pu239_Energy,Pu239_BR,20);

% clear all;
% load('Pu239.mat');
% Energy=Pu239_Energy;
% BrR=Pu239_BR;
nonzero=find(BrR>0);
E=Energy(nonzero)';
BR=BrR(nonzero)';
num=size(E,2);
maxnum=num;

%dE=30
Emin=min(E)-50;
Emax=max(E)+50;
Ex=Emin:0.1:Emax;
num2=size(Ex,2);
opeaks=zeros(num,num2);
area=zeros(1,num);
Amp=zeros(1,num);
for i=1:num
    ieff=0.52109*exp(-E(i)/142.70732)+0.00161;
iwidth=(-2.32647e-5*(E(i).^2)+0.08463*E(i)+1.83083)/2.355;
area(1,i)=BR(i)*ieff;
    opeaks(i,:)=area(1,i)*1/(sqrt(2*pi)*iwidth)*exp(-(Ex-E(i)).^2/(2*iwidth.^2));
    Np=opeaks(i,:);
    res=find(Np==max(Np),1,'first');
    Amp(1,i)=Np(1,res);
end
Inamp=Amp;
j=1;
Epeak=zeros(num,num2);
Area=zeros(1,num);
dlim=zeros(10,num);
ulim=zeros(10,num);
for i=1:num
    N=zeros(1,num2);
tempA=0;
    if i==1
        dlim(1,j)=1;
    else
        dlim(1,j)=llim_conE(E,Amp,i,dE);
    end
    if i==num
        ulim(1,j)=maxnum;
    else
        ulim(1,j)=rlim_conE(E,Amp,i,dE);
    end
    if dlim(1,j)<=ulim(1,j)
        %changed on Feb 24 from '<' - '=<'
        for k=dlim(1,j):ulim(1,j)
            Nk=opeaks(k,:);
            N=N+Nk;
            tempA=tempA+area(1,k);
        end
        Epeak(j,:)=N;
        Area(1,j)=tempA;
        j=j+1;
    end
end
nump=j-1;
Ep=zeros(1,nump);
Amp=zeros(1,nump);
fwhm=zeros(1,nump);
for i=1:nump
    Np=Epeak(i,:);
    res=find(Np==max(Np),1,'first');
    Ep(1,i)=(res-1)*0.1+Emin;
    Amp(1,i)=Epeak(i,res);
end
iasum=zeros(100,nump);
icsum=zeros(100,nump);
Asum=zeros(100,nump);
[Ep2,ia,ic]=unique(Ep);
pdlim=zeros(100,nump);
pulim=zeros(100,nump);
maxE=dE;

for n=1:10
    iasum(n,1:length(ia))=ia;
    icsum(n,1:length(ic))=ic;
    Asum(n,1:length(Area))=Area;
    pdlim(n,1:length(ia))=dlim(n,ia);
    pulim(n,1:length(ia))=ulim(n,ia);
    num=size(Ep2,2);
    Epeak=zeros(num,num2);
    Area=zeros(1,num);
    j=1;
    upcheckno=0;
    pn=0;
    for i=1:num
        N=zeros(1,num2);
        flag1=1;
        flag2=1;
        nopeak_left=0;  %flag for judging whether the peak has adjacent left peak
        nopeak_right=0;  %flag for judging whether the peak has adjacent right peak
        tempA=0;
        if i==1
            dlim(n+1,j)=1;
            flag1=0;
            nopeak_left=1;
        else
            if Ep2(i-1)<Ep2(i)-maxE  %No left adjacent peak within MRE
                dlim(n+1,j)=pdlim(n,i);
                flag1=0;
                nopeak_left=1;
            else
                if Ep2(i-1)<Ep2(i)-dE  %Left adjacent peak cannot resolve
                    if pdlim(n,i-1)<=pdlim(n,i)  %changed from < to <=
                        dlim(n+1,j)=pdlim(n,i-1);
                    else
                        %left adjacent peak being merged & deleted
                        dlim(n+1,j)=pdlim(n,i);
                        end
                    else
                        %Left adjacent peak can resolve
                        dlim(n+1,j)=pdlim(n,i);
                        end
                    else
                        dlim(n+1,j)=pdlim(n,i);
                    end
                end
            end
        end
        flag1=0;
        %indicate the peak has left adjacent peak, but amplitude is smaller than its left adjacent peak
    end
end
%Right adjacent peak case
if i==num
    ulim(n+1,j)=maxnum;
    flag2=0;
    nopeak_right=1;
else
    if Ep2(i+1)>Ep2(i)+maxE %no adjacent right peak
        ulim(n+1,j)=pulim(n,i);
        flag2=0;
        nopeak_right=1;
    else
        ratio=Amp(i)/Amp(i+1);
        if ratio>=1
            if Ep2(i+1)<Ep2(i)+dE
                if pulim(n,i+1)>=pulim(n,i)
                    ulim(n+1,j)=pulim(n,i+1);
                else
                    ulim(n+1,j)=pulim(n,i);
                end
            else
                ulim(n+1,j)=pulim(n,i);
            end
        else
            ulim(n+1,j)=pulim(n,i);
        end
    end
end
end

%sum up unresolvable peaks
if flag1+flag2>0 || nopeak_left*nopeak_right==1
    %if peak has adjacent peaks, it's amplitude should be bigger than one adjacent peak;
    %if peak doesn't have adjacent peaks within Emin, accept it.
    for k=dlim(n+1,j):ulim(n+1,j)
        Nk=opeaks(k,:);
        N=N+Nk;
        tempA=tempA+area(1,k);
    end
    Epeak(j,:)=N;
    Area(1,j)=tempA;
    j=j+1;
end
%finding peak centroids and areas
nump=j-1;
Ep=zeros(1,nump);
Amp=zeros(1,nump);
fwhm=zeros(1,nump);
for i=1:nump
    Np=Epeak(i,:);
    res=find(Np==max(Np),1,'first');
    Ep(1,i)=(res-1)*0.1+Emin;
    Amp(1,i)=Epeak(i,res);
end
[Ep2,ia,ic]=unique(Ep);
if nump==1
    break;
end
centroid=Ep';
LibArea=Area';

function rminE=rlim_conE(E,Amp,i,dE)
%Find left limit of unresolvable peaks
if E(i+1)>E(i)+dE  %changed on Feb 23  E(i)+20 -> E(i)+dE
    rminE=i;
    return;
else
    n=i+1;
    while(n<=size(E,2))
        ratio=Amp(i)/Amp(n);
        if ratio>1
            if E(n)<E(i)+dE
                rminE=n;
            else
                rminE=n-1; return;
            end
        else
            rminE=n-1;return;
        end
    end
    n=n+1;
end
end

function lminE=llim_conE(E,Amp,i,dE)
%Find left limit of unresolvable peaks
if E(i-1)<E(i)-dE  %changed on Feb 23  E(i)-20 -> E(i)-dE
    lminE=i;
    return;
else
    n=i-1;
    while(n>0)
        ratio=Amp(i)/Amp(n);
        if ratio>1
            if E(n)>E(i)-dE
                lminE=n;
            else
                lminE=n+1; return;
            end
        else
            lminE=n+1; return;
        end
        n=n-1;
    end
end
### Appendix B  Sample Libraries

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