Automated Energy Calibration and Fitting of $LaCl_3(Ce)$ γ-spectra using Peak Likelihood and Tabu Search

Timothy P. McClanahan[1,2]
Astrochemistry Division, NASA Goddard Space Flight Center[1]
Greenbelt, MD 20771, USA
timothy.p.mcclanahan@nasa.gov

Murray H. Loew[2]
Electrical and Computer Engineering Dept., George Washington University[2]
Washington D.C 20052, USA
loew@gwu.edu

Jacob I. Trombka[1]
Astrochemistry Division, NASA Goddard Space Flight Center
Greenbelt, MD 20771, USA
jacob.i.trombka@nasa.gov

ABSTRACT

An automated method for γ-emission spectrum calibration and deconvolution is presented for spaceflight applications for a Cerium doped Lanthanum Chloride, ($LaCl_3(Ce)$) γ-ray detector system. This detector will be coupled with a pulsed neutron generator (PNG) to induce and enhance nuclide signal quality and rates, yielding large volumes of spectral information. Automated analytical methods are required to deconvolve and quantify nuclide signals from spectra; this will both reduce human interactions in spectrum analysis and facilitate feedback to automated robotic and operations planning. Initial system tests indicate significant energy calibration drifts (>6%), that which must be mitigated for spectrum analysis. A linear energy calibration model is presently considered, with gain and zero factors. Deconvolution methods incorporate a tabu search heuristic to formulate and optimize searches using memory structures. Iterative use of a peak likelihood methodology identifies global calibration minima and peak areas. The method is compared to manual methods of calibration and indicates superior performance using tabu methods. Performance of the Tabu enhanced calibration method is superior to similar unoptimized local search. The techniques are also applicable to other emission spectroscopy, eg. X-ray and neutron.

Keywords: Spectroscopy, gamma-ray, automation, tabu, elemental

1. Introduction

We describe a methodology for automated spectrum energy calibration and deconvolution for a γ-ray detector system, $LaCl_3(Ce)$ that is currently being studied for use in planetary surface remote sensing. The γ-detector is coupled with a pulsed neutron generator (PNG) to actively induce nuclide signals in the 1-10 MeV energy range. Detected emissions then are used to identify planetary bulk surface and sub-surface elemental compositions as well as to infer surface geophysical properties such as stratigraphy and mineralogy [1]. Considered for on and near surface operations, the combined instrument system significantly increases γ-signal rates as compared to naturally induced γ-ray production. This detector system is currently being considered as a candidate detector material for this application, due to its dual qualities of maintaining high resolution while not requiring active cooling. This effectively provides a high signal/ noise capability, while minimizing spaceflight requirements for power, weight and volume [2]. With high-data-rate operational modes, continuous surface operations can be performed, significantly increasing both total coverage and surface signal resolutions due to the enhanced data rates. Future on-surface operational scenarios also may require automated elemental feedback to on-board processes for automated control of navigation and exploration planning [3][4].

A series of laboratory tests were recently conducted on this PNG-detector system to determine important science and operational factors. In analyzing our initial set of test spectra we found that the detector energy calibration (*gain*) varied by as much as ~6% in the range between 5.19 and 5.5 keV/channel, as seen in *Fig. 3*. This constitutes a large deviation from the expected calibration, which must be mitigated prior to spectrum analysis to yield accurate elemental results. This variation causes spectrum peaks to be shifted electronically, in some cases 10's of channels from their expected positions. Compounding the problem, peaks are variably convolved with other peaks depending on source sample. This requires variably complex spectrum fitting models to derive energy calibration for arbitrary spectra. Further, these issues are significant obstacles for automated energy calibration methods that may have neither appropriate controls nor an easily identified, independent set of standard peaks.

To mitigate these issues we propose the use of a Tabu local-search heuristic used in conjunction with a peak likelihood method for quantifying γ-ray emission spectra. Tabu search is a mathematical optimization heuristic that utilizes memory structures to optimize search formulation for real valued and combinatorial problems [5].

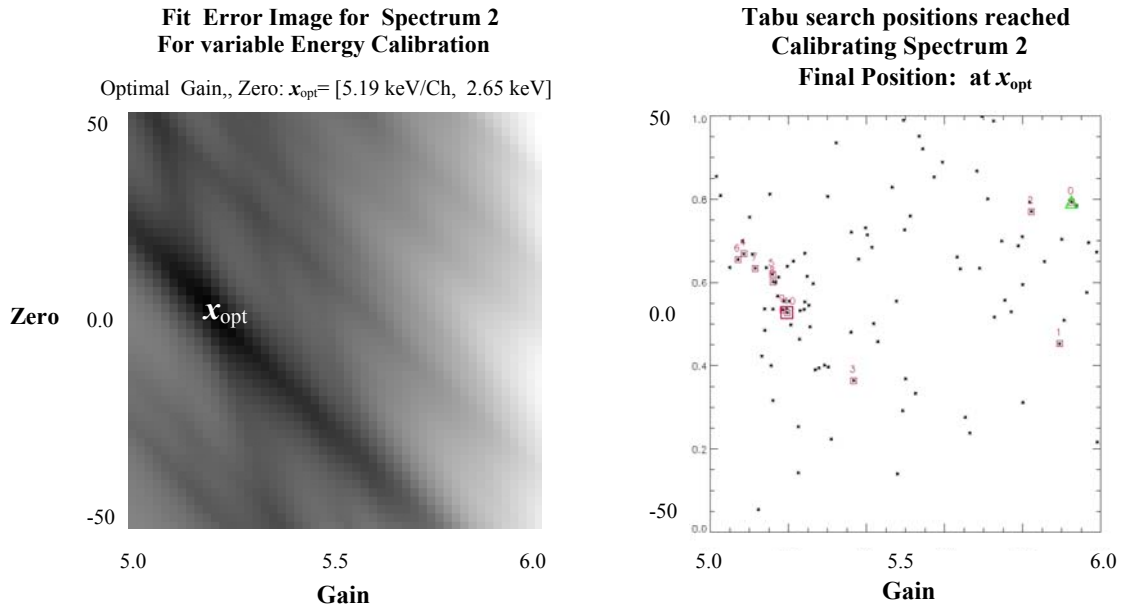

Figure 1: (*left*) Error image for library fits as a function of two parameters (*gain, zero*). Error image for a gridded search of a fixed range of gain and zero settings: Gain = [5.0, 6.0], Zero = [50,-50]. (*right*) plot points indicate the set of search positions visited by tabu search system. Green triangle with 0, indicates the starting position for the tabu procedure. Squared points indicate the numbered sequence and positions of local minima. Large square indicates final position at $\boldsymbol{x}_{opt}$. Calibration points $\boldsymbol{x}$' initially are spread across feature space to perform global search and converge for fine tuning search at the end of process.

In this context, memory list structures guide the positioning of energy calibration points by preventing cycling (repeated evaluation of previously visited positions) thereby, optimizing search formulations. This approach effectively identifies global energy calibration minima and addresses the problems posed by local minima to similar gradient descent type process optimization, *Fig. 1*. Methods are also shown to provide better calibration performance than standard two-point calibration techniques and improve convergence times vs unoptimized local search.

Section 2, describes background related to design of the present detector system. It addresses related issues for energy calibration and deconvolution of γ-ray emission spectra. Section 3, provides a high level flow diagram of major system components and discussion of tabu and peak likelihood methodologies. Section 4, provides results of the evaluation of the proposed system. This includes comparisons vs manual (gold standard) results as well as the measurements of the improvements over unoptimized local search processes provided by tabu search heuristics. Section 5, states paper conclusions.

2. Background

Recent developments in detector materials for γ-ray detection $LaX_3(Ce)$ (Lanthinum Halide) show advantages compared to both inorganic scintillator, (Sodium Iodide (NaI) [5]) and actively cooled High purity Germanium (HPGe) detector systems [6]. The new materials provide improved light yield and resolution as compared to NaI and do not require active cooling as does HPGe. Given these attributes, the new material is considered a strong candidate for use in spaceflight applications [2][3].

In high-data-rate modes of operation automated analytical methods are required to handle potentially large collections of spectral information. Detailed knowledge of the detector system is required, including expected nuclide signals and energy calibration standards to accurately quantify signals and infer elemental quantities. In laboratory settings, detector designs are optimized for maintaining high resolution by tightly controlling instrumental and calibration factors. These environmental and electronic controls minimize the degrees of freedom required to perform quantitative spectrum analysis. In less tightly controlled operational configurations being considered, e.g. planetary remote sensing, other instrument and detector design factors such as power, weight and volume are prioritized to meet stringent spaceflight requirements. In those cases, calibration and other factors, such as detector temperature can induce variations in the quality of spectral information. As a result a more complex analytical process is required to automatically 1) reduce the number of degrees of freedom and 2) mitigate the possibility of the deconvolution process falling into a local minimum [7].

Automated quantitative analysis of a γ-ray emission spectrum determines the net area of peaks in the spectrum and the removal of the background contribution [8][9]. This is a critical step as errors in background estimation propagate to all subsequent processing steps [10]. Once subtracted, the principal objective is to identify the discrete combination of peaks in the spectrum as well as their relative areas. Difficulties in this process include the ability to identify discrete combinations of nuclide peaks, while quantifying peak parameters. In Eq. 1, a prior nuclide library $\boldsymbol{A}$ is developed, containing a superset of the expected nuclide peaks described as a function of energy. The analytical objective is to determine a model of the observed

spectrum peaks by determining relative peak scales $\boldsymbol{A\beta}$, and identification of background $\boldsymbol{M}$.

$$\boldsymbol{S}(\mathrm{i}) = \sum_{\mathrm{i=chan\text{-}1}}^{\mathrm{chan\text{-}n}} \sum_{\mathrm{j=nuclide\text{-}1}}^{\mathrm{nuclide\text{-}k}} \boldsymbol{A}(\mathrm{j},\mathrm{i})\boldsymbol{\beta}(\mathrm{j}) + \boldsymbol{M}(\mathrm{i}) \quad \text{Eq (1)}$$

Where, spectrum channel $\boldsymbol{S}$(i) is the integral of the product of the 1-D nuclide library element $\boldsymbol{A}$(j,i) with its scaled quantity $\boldsymbol{\beta}$(j) added to the derived background contribution at channel *i*, $\boldsymbol{M}$(i) [8].

In the case of arbitrary spectra, peak identities, and quantitiies are unknown. Spectrum deconvolution processes such as peak fitting can fall into local minima preventing these methods from converging to an accurate solution. The issue is exacerbated in miscalibrated conditions, where library peaks become misaligned, yielding erroneous results. This issue is illustrated as light and dark bands in the left error image of *Fig. 1*. The error image was generated by automatically determining the spectrum fit error for a range of energy calibration points using a laboratory $LaCl_3$(Ce) spectrum and library of known nuclides (peaks). Spectrum fit error is recorded at each discrete energy calibration position on the grid. Image (left) position at $\boldsymbol{x}_{\mathrm{opt}}$ is at the global fit error minimum and represents the optimal energy calibration parameters. Local minima constitute dark positions in the feature space image brighter than at $\boldsymbol{x}_{\mathrm{opt}}$. Existence of local minima precludes the use of gradient descent methods for optimizing the energy calibration of an arbitrary spectrum [9].

Energy calibration is often performed to determine the energy-to-spectrum-channel relationship, Eq. 2. Where, the energy in a given spectrum channel *ch*, is related by the following linear function:

$$\mathrm{Energy} = zero + (gain * ch) \quad \text{Eq (2)}$$

Where, *zero* and *gain* terms are used to describe a linear energy calibration model and constitute the dominant calibration terms. *gain* is energy step per spectrum channel; *zero* is the energy offset. When spectra are miscalibrated, assumptions of *gain* and *zero* vary from their expected value, affecting positions of spectrum peaks, thereby, reducing the ability to identify and quantify spectrum peaks. Recalibration is required in cases, where calibration drift has occurred, e.g. temperature variation [7].

Tabu search is a class of mathematical optimization meta-heuristic techniques that employs local search processes to solve real valued and combinational problems. Developed by Glover [11], the tabu process is termed from the Polynesian word "tabu" or "taboo", which is to '*ban on the grounds of morality or taste or as a continuing risk …*". Its meaning in the context of numerical optimization refers to the use of tabu memory structure's to bias the formulation of local search strategies. The primary benefit of tabu search, which is linked to artificial intelligence methodologies, lies in the use of a flexible memory structures which are used as quick reference for search formulation. In this process, a transition from the current state of the program $\boldsymbol{x}$ to $\boldsymbol{x}$' is made if and only if, the functional evaluations *f*, of $\boldsymbol{x}$ and $\boldsymbol{x}$' result in lower error error values at $\boldsymbol{x}$', Eq 3.

$$\boldsymbol{x} \rightarrow \boldsymbol{x}' \; iff \; f(\boldsymbol{x}') \langle \; f(\boldsymbol{x}) \quad \text{Eq (3)}$$

Locally optimal solutions encountered in the search are classified with their solution attributes (*tabu-active*) vs the sequence of local minima which are entered into a memory structure. Tabu lists then used during search formulation define feasible and unfeasible regions in the solution space. The result is a strategy that makes efficiently evaluates solution spaces by biased selection search position.

3. Methods

Figure 2. contains an overview of this method, which determines which determines energy calibration and linear spectrum deconvolution automatically. The system iteratively combines feedback from peak likelihood spectrum quantification to tabu search processes to define global optima. In the first initialization process #1, constraints for possible parameter ranges are defined for the $LaCl_3$(Ce) detector system. In this case, we empirically determined operational bounds for the system and set upper and lower bounds for the *gain* = [5.0, 6.0] KeV/ch and *zero* = [-50,50] keV. The objective function *f*, is continuous and a decision $\boldsymbol{x}_{\mathrm{opt}}$ must exist within this space. By definition, this guarantees satisfaction of the *extreme value theorem* which defines the existence of maxima and minima within closed intervals of continuous functions.

A method of position and range ratios provides a transformation between real and normalized coordinates. This representation also has implications in the use of distance functions, where the warping between real and normalized spaces can yield elliptical realizations of distance functions. Two memory structures are implemented in this process. The first is the 2 dimensional tabu list $\boldsymbol{T}$. Entries in $\boldsymbol{T}$ are from sub-optimal searches $\boldsymbol{x}$' encountered in the process. $\boldsymbol{L}_s$ contains the locally optimal sequence of positions visited during the course of the search. Contents of each list are determined as a function of evaluation by *f*.

Critical neighborhood parameters are required for the method to operate effectively. These parameters guide the process towards a global solution by a fixed method of parameter aging described by process #6. At issue, is the balance between operational objectives of required time dedicated to uniform exploration of the search space to identify global optima, and the desire to perform focused searches of local minima to fine tune the result. Initial neighborhood parameters are defined by radii around each point in the lists providing zones of inclusion and exclusion to the search formulation process. By starting with large radii, the process initially implements broad searches of the solution space to identify local minima which are entered in $\boldsymbol{L}_s$. Tabu points or sub-optimal points entered in $\boldsymbol{T}$ preclude searches in respective neighborhoods.

Once configured process #2 formulates the next search position, $\boldsymbol{x}$' by checking its intersection with the regions defined in $\boldsymbol{L}_s$, $\boldsymbol{T}$, lists. First, distances from positions $\boldsymbol{x}$' are checked for proximity to the sub-optimal list $\boldsymbol{T}$ to define rejected search points. Distance tests are subsequently performed to accept regions around presently local optima in $\boldsymbol{L}_s$. By sequentially performing position rejection and acceptance in sequence realizes a reinstatement

of the regions defined by the list intersection, $L_s \cap T$. This is described using the following process:

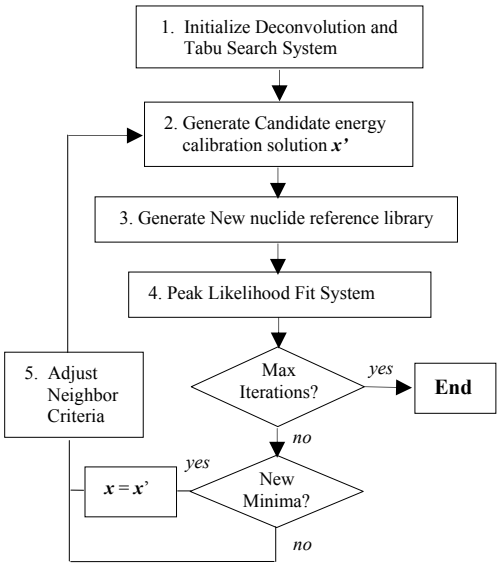

Figure 2: Flow diagram of Automated Spectral Energy Calibration and fitting system

As the knowledge of the solution space increases due to successive calibration search formulation and evaluation, an aging parameter reduces the influence of neighborhoods by gradual reduction in neighbor radii. Results of calibration point selection is a new linear energy calibration point, ***x'***. Process #3 consists of generating an instrument nuclide library that is a function from the energy calibration point, ***x'***. This entails the generation of a library of new 1-dimensional probability distribution functions that describes nuclide-specific energy interactions of gamma-rays with the detector that are used for spectrum analysis.

In process #4 an automated spectrum analysis program is implemented to identify and quantify spectrum peaks given the current nuclide library defined using the energy calibration at ***x***'. This process is performed using an iterative deconvolution method that determines peak areas. The result of this process is a scalar error estimate of the spectrum fit as a function of the energy calibration. Checks are then made of the number of iterations performed to determine end of process as well as whether a new minima has been achieved. If so, the present optimum is updated, **x** = **x**'. Step #5 updates neighbor criteria.

4. Results

Laboratory tests of the detector system generated 34 $LaCl_3(Ce)$ γ-ray spectra under a variety of experimental conditions. Gold standards for the true energy calibration were derived using manual 2-point peak fitting, where peaks are identified and ratios of the peak channel and energy centroids provide *gain* and *zero* terms. Results of the manual calibration determined energy *gain* to vary between ~5.16 and 5.5 keV/channel, *Figure 3*. Test sequences indicate slowly increasing gain during the first 27 spectra, with a large gain shift 0.25 keV/chan to ~5.49 keV/channel occured. *Zero* terms vary significantly in the bottom plot from -1 to 8 keV. These are however minor terms in the energy calibration model and secondary in consideration.

Fig. 1. illustrates an example of the tabu energy calibration execution for a laboratory spectrum, where the manual energy calibration was determined using a 2 point method at [5.19 KeV/ch, 2.67 KeV] described above. The *x,y* scales and ranges are identical for right and left images and plot (right) indicates the local search positions (*stars*) that are visited during the search process. The green triangle indicates an initial calibration search at visit position 0, $\boldsymbol{x}_0$ at ~[5.8 KeV/ch, 35 KeV]. Local minima encountered during the search process are indicated by bounded squares (*red*) and numbered by their visit sequence. The process terminal position is indicated by the large square (*red*) which coincides with convergence at the global minimum (left image) at 100 evaluations.

A subsequent test was performed to quantify the benefit of the tabu search heuristics for energy calibration vs simple local search strategy, Eq 1, *Table 2*. Hypothesis is that the reference to memory structures precludes search cycling, so tabu infrastructure should more efficiently converge. In this test, each of the 34 LaCl3 spectra was processed 5 times using 4 different number of search settings [60,80,100,120]. Total spectra for each setting = 34x5=170. Start position was identical for both Local and Tabu enhanced tests. In each run, candidate search position collisions with the tabu list resulted in the following solution candidate rejection averages: Results also indicate the average mean error for tabu enhanced methods is less than the local search method for all 4 tests. Only the 80 search mean was significant 2.48 > 1.65. Average errors for a given search was however consistently lower.

A comparison of tabu to manual methods converged fit error variance indicates the tabu search has lower error variance. i.e., *the gold standard does not facilitate a better spectrum energy calibration than the tabu method*. Given both methods objectives of identifying a global calibration error minimum for each spectrum, the distributions of both results are most likely Poisson processes with longer error tails in the high error regions and not symmetric about a minimum error position. F-test of the distributions for each test indicate the tabu method maintains lower error variance for each method but were not significant at α=0.05, *Table 3*. The lower tabu variances are

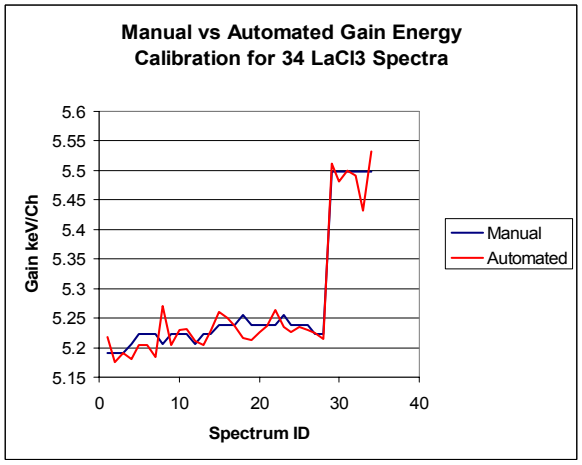

Figure 3: Manual vs Automated Gain Calibration

Process Search	μ	σ	Extra Tabu Searches
60	0.283699	0.106366	**17.02193**
80	0.399044	0.131573	**31.92353**
100	0.520965	0.150104	**52.09649**
120	0.675343	0.184776	**81.04118**

Table 1: Mean fraction of calibration searches rejected by tabu system as a function of search length.

Process Searches	t-value	α= 0.05 dof = 169
60	1.269018	
80	***2.480788***	
100	1.163907	
120	1.154402	
t-critical	**1.65**	

Table 2: t-values for tabu vs local search. Only search length =80 was the null Hypothesis rejected. Probability rejection [60,100, 120] < 0.13

Process Searches	F-Value	α=0.05 dof =169
60	1.084	
80	1.186	
100	1.06	
120	1.2	
F-critical	**1.288**	

Table 3: F-test values for Local search vs Tabu method results. Failed to reject null hypothesis in all tests.

likely due to the tabu method likely converging to the global optimum earlier than the unoptimized system in most cases.

Results indicate there is a search range over which the tabu method maximizes the performance. At small search numbers few candidate solutions are regenerated due to intersection with the tabu list, yielding little benefit from tabu infrastructure. At large search numbers, the distribution of searches in the solution space is saturated and both methods perform well indicated by a convergence of means. Results vary significantly at high search numbers probably due the tabu methods more efficient search formulation.

5.0 Conclusions

This paper introduced a method for automated energy calibration and fitting of pulse height spectra using an iterative peak likelihood heuristic and tabu search process. Methods are shown to effectively calibrate data a database of 34 $LaCl_3$(Ce) γ-ray emission spectra, where the gain calibration was shown to significantly vary by ~6%. In this process a potential energy calibration point is selected and evaluated using the peak likelihood heuristic, which determines an error term from fast linear approximation of peak areas from arbitrary spectra. Local search of the calibration solution space is implemented as an optimized state transition methodology using a Tabu meta-heuristic.

Method evaluations on a database of 34 $LaCl_3$(Ce) spectra obtained calibration results that closely match the manual gold standard results. Hypothesis testing of gains and zeros using t and F testing were ambiguous in concluding the experimental results exceeded the expected (manual gold standard results). However, further hypothesis testing of the method using a comparison of the final fit errors indicated that significant statistical differences were achieved with the described methods. Results indicate that the experimental fitting method achieved lower error, μ=5.8% than the 2-point manual method. An added benefit is that this approach is that the energy calibration and linear fit are performed simultaneously for a linear fit system.

Evaluation of methods using local search vs tabu search heuristics, indicated that the tabu implementation has merit in precluding searches of previously searched spaces for non-saturated solution spaces. Mean errors using tabu enhanced methods for 4 tests were consistently lower than unoptimized local search strategies, though hypothesis testing indicates results are not significant at α=0.05.

Potential method improvements include dynamic parameter tuning of neighborhood radii for both optimal and sub-optimal lists. Secondly, method testing used random initialization of calibration points. Using either expected or most temporally recent energy calibration points, the method for sequences of spectra, can be seeded in close proximity to solution spaces increasing performance efficiencies, yield a faster results using fewer searches. Further optimization can be implemented by minimization of the ranges /dimensionality of the solution space. Method's may also be extensible to a third dimension of energy calibration facilitating a quadratic fit of the calibration system.

6. References

[1] R. Starr et. al., **Combined Gamma-ray Spectrometer and Pulsed Neutron Generator for In-Situ Planetary Geophysical Analysis**, Lunar and Planetary Science Conference XXXVIII, 2007

[2] J. Trombka et. al., **A Pulsed Neutron Gamma-Ray System for Mars Rover Missions**, IEEE Astrobiol. Conf. , #1060

[3] H. Akkurt et. al., **Pulsed Neutron Generator System for Astrobiological Exploration of Planetary Bodies**, Nucl. Inst. and Meth. in Phys. Res. B. Appl. of Accel. Res. and Ind., pg 232-237, Dec. 2005

[4] P. Backes et. al., **Automated Rover Positioning and Instrument Placement**, IEEE Aerospace Conference, March 5-12, 2005

[5] J. Trombka et. al., **Compositional mapping with the NEAR X-ray /Gamma-ray spectrometer**, J. Geophy. Res., vol. 102, no. e10, pg 729-750. 1997

[6] Boynton et. al, **The Mars Odyssey Gamma-ray Spectrometer Instrument Suite**, Space Science Reviews, vol 110., no. 1-2, Jan 2004

[7] G. Gilmore, **Practical Gamma-ray Spectrometry**, John Wiley and Sons, ISBN 0 471 95150 1, 1995

[8] D. Marquardt, **An Algorithm for Least-Squares Estimation of Non-Linear Parameters**, SIAM Journal of Applied Mathematics, vol. 11, pg 431-441, 1963

[9] J. Trombka, **A Numerical Least Square Method for Resolving Pulse Height Spectra,** NASA Off. of TechUtil., SP-3044, 1968

[10]Schulze et. al, **Investigation of Selected Baseline Removal Techniques as Candidates for Automated Implementation, Applied Spectrometry**, vol. 59 5, pg 545-573, 2005

[11]F. Glover, **Future Paths for Integer Programming and links to Artificial Intelligence**, Computers and Operational Research, vol. 5, pg 533-49