Physics-based Radionuclide Event Mode Sequence and Radioactive Pattern Analysis

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***Abstract*—The accurate and rapid identification of radionuclide is an extremely important matter of radioactive material control and public security check. The classical detection method using γ-ray spectrum depends on large numbers of γ-ray measurements, which means a long time cost, especially when the radioactivity is low. A physics-based radionuclide γ-ray event modes sequence, representing specific nuclide is introduced, which offers a pattern recognition approach to attack this problem. The development of radionuclide representation in terms of characteristic γ-ray energy along with branching ratio that utilizes the measurement of photon energies by the detector and the probable γ-ray emission is described. The Kalman filtering is used to estimate the underlying characteristic γ-ray energy parameter in specific energy interval related to interested nuclides from the uncertain measurement photon by photon in this paper. The implementation of this approach is applied to experimental data to demonstrate its validity.**

Keywords—radionuclide detection; physics-based approach; event sequence mode; Kalman filtering

I. INTRODUCTION

Radionuclide detection is a key step in radioactive material control and public security check. Traditional approach finds radionuclide(s) by detecting the characterizing full-energy peak as fingerprint in a γ-ray pules height spectrum (PHS) [1~3].

Think about this procedure: After flying across a normally short distance, a γ-ray with specially energy that is emitted from a radioactive material arrives in detector and ionizes the sensitive material. Photons are produced and then collected by photomultiplier tube (PMT) as much as possible. Photoelectrons are excited in the photocathode due to the photoelectric effect by the photons. These photoelectrons are multiplied by 10^4~10^5 times in the PMT, and finally are collected by the anode as an output pulse signal. Because of the statistical fluctuations that derived from photon ionization process in scintillator and the amplifying process in PMT, the pulse amplitude responding to the same energy γ-ray is Gaussian distribution.

Traditional approach needs a large number of γ-ray measurements to make the Gaussian feature of full-energy peak in special energy interval clear and easy to confirm the characterizing energy. Sometimes, potential radioactive material is shielded by package and adjacent facility, these shielding reduce the count rates and attenuate the γ-ray energy. Rapid and accurate detection become more difficult to traditional method since this approach should solve the infection from background radiation, detector energy resolution and low count rates.

In this paper, the underlying radionuclide is treated as a unique union of characteristic energy sources. The γ-ray radiations from underlying radionuclide are represented as a series of pulse sequences containing γ-ray energy information in form of event mode sequence (EMS). Different from the spectrum that characterize γ-ray energy distribution after a period of time measurement, event mode sequence characterize whole structure including every γ-ray's energy, probability and there order based physics theory photon-by-photon [4]. This model provides a statistical signal processing approach that can be used to detect underlying radionuclide in low count rates and uncertain measurement by analysis γ-ray events sequence pattern in interested energy interval (related to underlying radionuclide).

There are three major things discussed in this paper: (1) theoretical derivation of physics-based event mode sequence; (2) characteristic parameter estimate; and (3) application to experiment data.

II. THE PHYSICS-BASED EVENT MODE SEQUENCE MODEL

As well known, a particular radionuclide can be uniquely characterized with feature energy of γ-ray and specific activity. The feature energy γ-ray describes the types of potential nuclides; the specific activity describes the nuclide content in the radioactive materials. In a period of measurement, the number of feature energy γ-ray is depending on the specific activity σ, the mass of the radioactive material M, the emission ratio α_m of the radionuclide, the detection efficiency η_m to the feature energy, and the geometric factor S between the radioactive material and the detector.

In mathematical, we use the pair,$[\{\epsilon_m\}, \{\lambda_m\}]$, to define the feature γ-ray energy level and detection rate of the mth

component of radioactive material. The outputs of the detector is a sequence of events which containing the time and energy information. Sequentially, the γ-ray can be characterized by sets of energy and times: $[\{\epsilon_\mathrm{m}\},\{t_\mathrm{m}\}]$ $\mathrm{m}=1,\dots,\mathrm{M}_\epsilon$. The index m presents the mth γ-ray of the radionuclide with M_ϵ feature energies, and the time t_m is related to the detection rate λ_m. Thinking of each feature energy as a separate source, the underlying radioactive material is presented as a union of individual γ-ray sources. As shown in Fig. 1, a γ-ray events sequence emitted by a radionuclide with three feature energy is depicted with the monoenergetic decomposition.

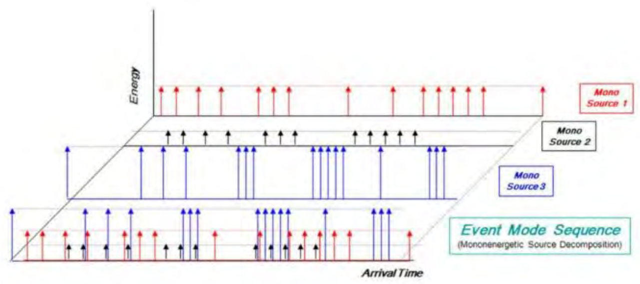


Fig. 1. Ideal radionuclide monoenergetic source decomposition:union EMS and three monoenertetic source components

We define $\epsilon_\mathrm{m}(\mathrm{n};\ \epsilon_\mathrm{m},\ \mathrm{t_m})$ to represent the nth γ-ray of the mth feature energy ϵ_m, by using pulse function $\delta(\mathrm{t})$, it could be represented as

$$\epsilon_\mathrm{m}(n;\epsilon_\mathrm{m},t_\mathrm{m})=\epsilon_\mathrm{m}\delta(t-t_\mathrm{m}(n)) \tag{1}$$

where $\mathrm{t_m}$ is arrival time of the nth γ-ray with associated detection rate, λ_m. On this basis, we introduce the symbols, $\tilde{\mathrm{t}}_\mathrm{m}:=\{\mathrm{t_m}(1),\dots,\mathrm{t_m}(\mathrm{N}_\epsilon(\mathrm{m}))$ with $\mathrm{N}_\epsilon(\mathrm{m})$ is the total γ-ray number of the mth energy in time, $\mathrm{t_m}(\mathrm{N}_\epsilon(\mathrm{m}))$. Then the mth energy γ-ray sequence in time $\mathrm{t_m}(0)$ to $\mathrm{t_m}(\mathrm{N}_\epsilon(\mathrm{m}))$ could be characterized as

$$\begin{aligned}&\epsilon(N_\epsilon(m);\epsilon_\mathrm{m},\tilde{t}_m)\\&=\sum_{n=1}^{\mathrm{N}_\epsilon(\mathrm{m})}\epsilon_\mathrm{m}(n;\epsilon_m,t_m)=\sum_{n=1}^{\mathrm{N}_\epsilon(\mathrm{m})}\epsilon_m\delta(t-t_m(n))\end{aligned} \tag{2}$$

Finally, we represent the potential radionuclide sources which could be decomposed into M_ϵ monoenergetic sources from the assumption that it is extremely impossible that any two γ-rays of monoenergetic source will be measured simultaneously by detector like

$$\begin{aligned}&\mathrm{R}(\mathrm{N}:\underline{\epsilon},\underline{\mathcal{T}}):\\&=\sum_{\mathrm{m}=1}^{\mathrm{M}_\epsilon}\sum_{\mathrm{n}=1}^{\mathrm{N}_\epsilon(\mathrm{m})}\epsilon(\mathrm{n};\epsilon_\mathrm{m},\mathrm{t_m})\\&=\sum_{\mathrm{m}=1}^{\mathrm{M}_\epsilon}\sum_{n=1}^{\mathrm{N}_\epsilon(\mathrm{m})}\epsilon_m\delta(t-t_m(n))\end{aligned} \tag{3}$$

for $\underline{\epsilon}:=\{\epsilon_1,\dots,\epsilon_{\mathrm{M}_\epsilon}\}$ is the set of underlying feature energy of R, and $\underline{\mathcal{T}}:=\{\tilde{\mathrm{t}}_1,\dots,\tilde{\mathrm{t}}_{\mathrm{M}_\epsilon}\}$ is the set of detected times. The index $\mathrm{N}=\sum_{\mathrm{m}=1}^{\mathrm{M}_\epsilon}\mathrm{N}_\epsilon(\mathrm{m})$ is the total number of all energy γ-rays.

Since the probability that one photon is emitted with energy ϵ_m from M_ϵ monoenergetic possibilities is fixed and only one γ-ray is detected once time by electronic system, we could present this probability by a random variable like

$$I_j(m)=\begin{cases}1 & m=j\\0 & m\neq j\end{cases} \tag{4}$$

for $P_r\left(I_j(m)=1\ \mathrm{R}(\mathrm{n}:\underline{\epsilon},\underline{\mathcal{T}})\right)=P_r\left(I_j(m)=1\middle|\Xi_n\right)=\alpha_j$ where α_j is the emission probability of the jth energy. Using this indicator variable, we could write the jth energy γ-ray set as

$$\mathrm{R}_j(\mathrm{N}:\underline{\epsilon},\underline{\mathcal{T}})=\sum_{m=1}^{\mathrm{M}_\epsilon}\sum_{n=1}^{\mathrm{N}_\epsilon(\mathrm{m})}I_j(m)\epsilon_m\delta(t-t_m(n)) \tag{5}$$

To be more precise, we model the noise produced by detector as a White Gaussian noise $\vartheta_{\epsilon_m}\sim N\left(0,R_{\vartheta_{\epsilon_m}\vartheta_{\epsilon_m}}\right)$, the measurement $\xi_m(n)$ coulde be present as

$$\xi_m(n)=\epsilon_\mathrm{m}(n;\epsilon_\mathrm{m},t_\mathrm{m})+\vartheta_{\epsilon_m}$$

for energy variations $\xi_m(n)$ following Gaussian distribution, $\xi_m\sim N(\epsilon_m,{\sigma_{\epsilon_m}}^2)$, ${\sigma_{\epsilon_m}}^2=R_{\vartheta_{\epsilon_m}\vartheta_{\epsilon_m}}$, and then the measurement of jth energy event mode sequence could be rewritten like

$$\xi_j(\mathrm{N}:\underline{\epsilon},\underline{\mathcal{T}})=\sum_{m=1}^{\mathrm{M}_\epsilon}\sum_{n=1}^{\mathrm{N}_\epsilon(\mathrm{m})}I_j(m)\xi_m(n)\delta(t-t_m(n)) \tag{6}$$

This is the final outputs from electronic system what respond to sets of jth energy γ-ray in energy interval $[\epsilon_\mathrm{m}+k\times\sigma_{\epsilon_m},\epsilon_\mathrm{m}-k\times\sigma_{\epsilon_m}]$.

In this section, we introduced the monoenergetic source decomposition of radionuclide, and developed a physics-based event mode sequence model. This radioactive material unique characterization provides a basic signal model on uncertain measurement.

III. EVENT SEQUENCE MODE PARAMETER ESTIMATE

Following the EMS model, a radionuclide can be characterized as a unique union of monoenergetic sources by the energy and the branching ratio of the emitted γ-ray. In order to detect underlying radionuclide timely from low-count γ-ray measurement, our approach is analyzing event sequence mode (energy vs. ratio) in interested energy interval sequentially (photon-by-photon) instead of detecting the full-energy peak structure in spectrum after enough data acquisition. There are 2 key parameters characterizing the event sequence pattern: the energy and the detect rate. The energy distribution implies the underlying radionuclide monoenergetic source and the detect rate suggests the emission probability.

A. Monoenergetic Source Energy Estimate

Because of the finite detector (energy) resolution, the typically responses to same monoenergetic is infected and the electronic system output is a list of event which energy is Gaussian distribution. In another word, events we measured in specific energy interval must be (or a part of) Gaussian distribution if γ-ray emitted by underlying radionuclide is detected.

Suppose the energy can be characterized by a random sliding model as

$$\epsilon_m(n)=\epsilon_m(n-1|n-1)+\omega_{\epsilon_m}(n-1) \tag{7}$$

for $\epsilon_m(n-1|n-1)$ is the optimal estimate of the feature energy in mth energy interval $\left[\epsilon_m + k \times \sigma_{\epsilon_m}, \epsilon_m - k \times \sigma_{\epsilon_m}\right]$ in the (n-1)th γ-ray, and $\omega_{\epsilon_m} \sim N\left(0, R_{\omega_{\epsilon_m}\omega_{\epsilon_m}}\right)$ is the estimate noise caused by the propagation along with estimate error which is zero-mean multivariate Gaussian distribution.

With zero-mean, Gaussian (instrument) measurement noise, the energy measurement can be modeled as

$$\xi_\epsilon(n) = \epsilon_m(n) + \vartheta_\epsilon(n) \tag{8}$$

for $\epsilon_m(n)$ is an expected energy of nth γ-ray of mth feature energy, and $\vartheta_\epsilon \sim N(0, R_{\vartheta_\epsilon \vartheta_\epsilon})$ is the noise produced by the instrument.

Finally, the energy measurement model of mth energy interval γ-ray sequences is given by a Gauss-Markov process as

$$\begin{aligned} &\text{Expected: } \epsilon_m(n) = \epsilon_m(n-1|n-1) + \omega_{\epsilon_m}(n-1) \\ &\text{Measured: } \xi_\epsilon(n) = \epsilon_m(n) + \vartheta_\epsilon(n) \end{aligned} \tag{9}$$

and the optimal estimate algorithm is the Kalman filtering is given by

$$\begin{aligned} \hat{\epsilon}_m(n|n) &= \hat{\epsilon}_m(n|n-1) + K_{\epsilon_m}(n) i_m(n) \\ i_m(n) &= \xi_\epsilon(n) - \hat{\epsilon}_m(n|n-1) \\ K_{\epsilon_m}(n) &= \tilde{\sigma}^2_{\epsilon_m}(n|n) / \sigma^2_{i_m}(n) \end{aligned} \tag{10}$$

where $\hat{\epsilon}_m(n|n)$ is the estimate of ϵ_m in the nth γ-ray of mth energy interval based on total n γ-rays; $\tilde{\sigma}^2_{\epsilon_m}(n|n)$ is the estimated error covariance; $i_m(n)$ is the innovations sequences; $\sigma^2_{i_m}(n)$ is the measured error covariance and K_{ϵ_m} is the gain ratio [5]. Obviously, $\hat{\epsilon}_m(n|n)$ is the Minimum Mean Square Error (MMSE) estimate of ϵ_m with conditional mean $\hat{\epsilon}_m(n|n) \coloneqq E\{\epsilon(n)|\Xi_n\}$ and posterior distribution given by $\Pr(\epsilon_m(n)|\Xi_n) \sim N(\hat{\epsilon}_m(n|n), \tilde{\sigma}^2_{\epsilon_m}(n|n))$

B. Radiation Branch Ratio Estimate

Radionuclide radiation branch ratio of different energy γ-ray is an important inherent feature that can be found from the energy decay schemes of different radionuclides. Practically, we use detection rate λ_m to represent the branch ratio just like $\lambda_m = \sigma M \alpha_m \eta_m S$. The detection rate λ_m of the mth monoenergetic source is determined by the absolute activity per unit mass σ, the radioactive material mass M, the radionuclide branch ratio α_m, the detection efficiency η_m at energy ϵ_m, and the geometric parameter S that typically depend on the solid angle and the distance between detector and radioactive material. Assuming there is a radionuclide with 3 monoenergetic sources as shown in Fig.1, considering photoelectric events only (by introducing the peak ratio p_m), the radiation branch ratio can be estimated by using detection rate of interested energy interval by the following approach:

$$\widehat{\alpha_m}(n) = \frac{N_{\epsilon_m}(n)}{\sigma MS \times t \times \eta_m \times p_m} \tag{11}$$

where $N_{\epsilon_m}(n)$ is the counts for the mth monoenergetic source ϵ_m in energy interval $\left[\epsilon_m + k \times \sigma_{\epsilon_m}, \epsilon_m - k \times \sigma_{\epsilon_m}\right]$ when *nth* gamma-ray was detected. Because the detection efficiency η_m and peak ratio p_m are fix parameters depends on the detector, then a one-to-one mapping relationship can be established between $\widehat{\alpha_m}$ and α_m after a calibration of measurement system that we could processing $\eta_m \times p_m$ as a constant "1" ,a simple algorithm is given like

$$\widehat{\alpha_m}(n) = \frac{N_{\epsilon_m}(n)}{M_\epsilon(n)} \tag{12}$$

where $M_\epsilon(n)$ is the total count of all of the interested energy intervals at n. What needs to be emphasized is that $N_{\epsilon_m}(n)$ is increased by γ-ray detection in energy interval along with $M_\epsilon(n)$ so that

$$\lim_{n\to\infty} N_{\epsilon_m}(n+1) = \lim_{n\to\infty} N_{\epsilon_m}(n);$$

$$\lim_{n\to\infty} M_\epsilon(n+1) = \lim_{n\to\infty} M_\epsilon(n)$$

and

$$\lim_{n\to\infty} \widehat{\alpha_m}(n+1) = \lim_{n\to\infty} \widehat{\alpha_m}(n)$$

Thus, we have obtained two main characteristic parameters describing the sequence pattern of underlying radioactive material γ-ray events in interested energy intervals and introduced their estimation methods. This representation using less events and provide more detail information than traditional pulse height spectrum that was calculated by a number of measurement.

IV. IMPLEMENTATION AND PROOF-OF-CONCEPT EXPERIMENT

In the previous section, we introduce the event mode sequence and developed the basic estimate methods of the characteristic parameters. In this section, we discuss a more pragmatic approach to analysis the events pattern and test their validity.

A. Implementation

Based on the physics monoenergetic decomposition model, we structure a parallel/distribution analyzer which was composed of individual monoenergetic source processors in a sequential/photon-by-photon treatment method as depicted in Fig.2.

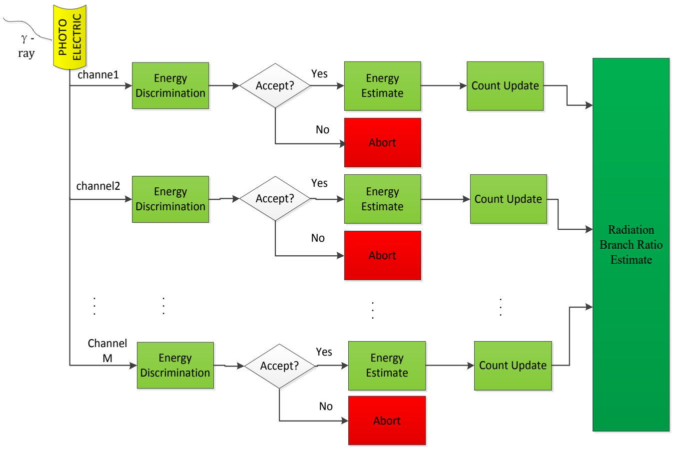


Fig. 2. Parallel/distributed radiation event sequences pattern analyzer: individual monoenergetic source (energy) processor for each interested energy interval

The implementation contains three phases with a beginning of acquiring the detector pulse and detection time measurement: (1) discrimination; (2) energy estimation; and (3) radiation branch ratio estimation as shown in Fig.2. The discrimination phase decides which channel will process the photon by an energy confidence interval test as

$$\left[\epsilon_{\mathrm{m}} + k \times \sigma_{\epsilon_m} < \xi_{\epsilon}(n) < \epsilon_{\mathrm{m}} - k \times \sigma_{\epsilon_m}\right] \tag{13}$$

where the confidence intervals are based on the underlying radioactive material with feature energy sets $\{\epsilon_{\mathrm{m}}\}$and detector energy resolution at interested energy ϵ_{m}. If the photon is accepted by channel m, we can begin the next step and estimate the characterize energy using a Kalman filtering was given in section Ⅲ. Subsequently, the radiation bran ratio is estimated photon-by-photon by counts updating in each channel. A individual monoenergetic source processor is depicted in Fig.3.

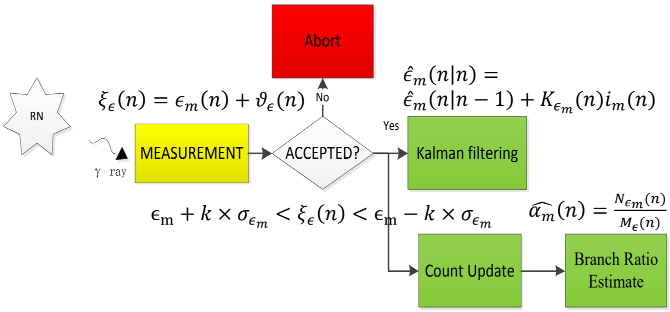


Fig. 3. Radiation event sequence monoenergetic source processor including: energy discrimination, energy estimation (LKF) and radiation branch ratio estimation.

B. *Proof-of-Concept experiment*

In order to assess the valid of the parallel/distributed analyzer, a proof-of-concept experiment was developed in a semi-physical simulation method. The experiment is divided into two stages: stage Ⅰ is numerical simulation; and stage Ⅱ is semi-physical simulation analysis.

In numerical simulation stage, a set of pairs is used to imitate a virtual radionuclide source with 3 feature energy. Each pair is treated as an event that containing a Gaussian distribution energy which expected value is a random variable from the 3 feature energy and an exponential distribution interval time, the distributed parameter listed in Table Ⅰ.

TABLE I. THE BRANCH RAIO, ENERGY, NOISE VARIANCE OF VIRTUAL RADIONUCLIDE MONOENERGETIC SOURCES

monoenergetic source No.	*branch ratio*	*energy /keV*	*measurement noise variance*
1	0.5	661.62	420.61
2	0.3	900.23	778.70
3	0.2	1332.50	1706.10

a. The detector energy resolution is calculated as 7.3% at all energy

Note that the difference of detection efficiency η_m at different energy is involved in the branch ratio $\widehat{\alpha_m}$, and the detector energy resolution diversity at different energy is ignored. The final pulse height spectrum (PHS) and the event mode sequence (EMS) consist of 2000 events are shown in Fig.4. There are 3 feature energy lines can be clearly observed in EMS of Fig.4a. In pules height spectrum of Fig.4b, 3 full-energy peaks correlation with feature energy can be found.

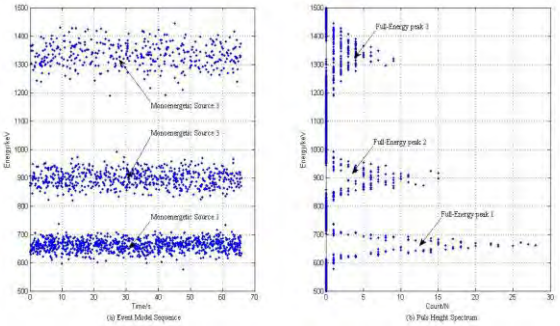


Fig. 4. Virtual radionuclide and 2000 photoelectricity measurement. (a) event mod sequence. (b) pulse-height spectrum with 3 full-energy peak

When a γ-ray is detected (photoelectricity only), the energy and time is measured and discriminated. If accepted, then the energy is used to estimate the monoenergetic source parameter using LKF processor and the right channel count is updated followed the branch ratio estimate sequentially. Fig.5 shows the branch ratio estimate of 3 monoenergetic sources photon-by-photon. The estimate error at the beginning (in the red rectangular zone) is mainly caused by the uncertainly of the radionuclide decay. By the accumulation of events, the estimated ranch ratios gradually become stabilized (0.5, 0.3, and 0.2).

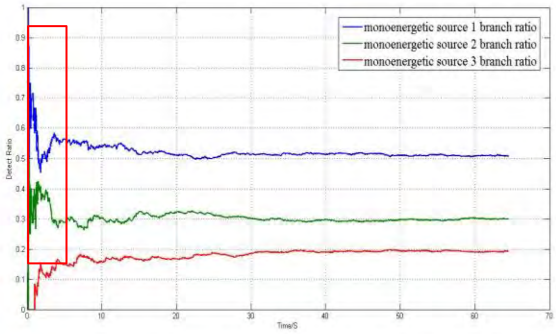


Fig. 5. Monoenergetic sources branch ratio estimate

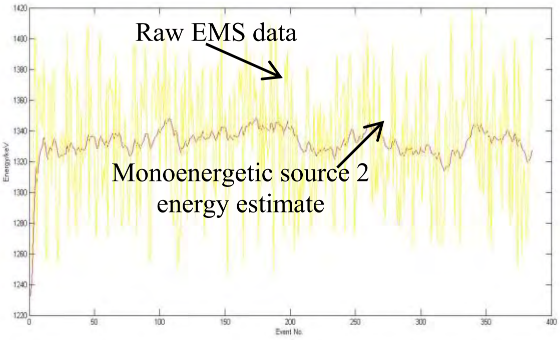


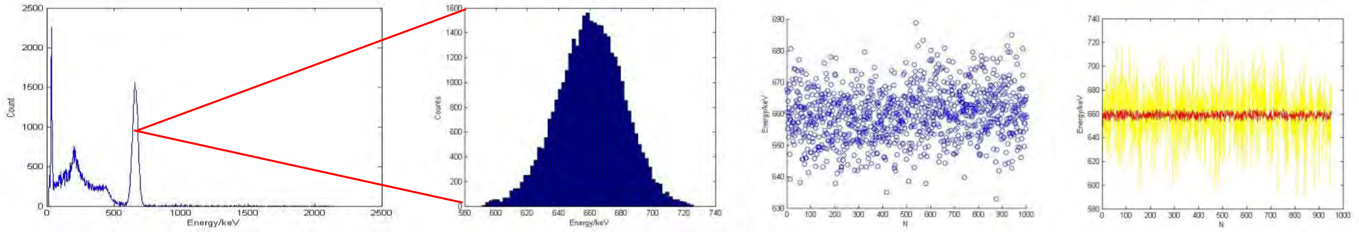
Fig. 6. Monoenergetic source energy parameter estimate using LKF

A typical energy estimate result of the processor targeted monoenergetic source 2 for an complete run is shown in Fig.6. The yellow line is the raw measurement energy sequence and the red line is the estimation monoenergetic source energy (1130keV). This clearly shows that the measurement noise is well suppressed as the fluctuation of the estimated energy curve (red) is obviously narrow than the original data curve (yellow).

In semi-physical simulation analysis stage, a cesium (^{137}Cs) source instead the virtual radionuclide, and the measurement instruments including a NaI(TI) detector, a Digital Multichannel analyzer and recording instrument are listed in Table Ⅱ. An environment background spectrum and a ^{137}Cs spectrum along with background and the potassium for 600 sec are measured. By deducting the contribution of background, we obtain a “pure” ^{137}Cs spectrum as shown in Fig.7a. Using this spectrum, we intercept the full-energy peak (at 661keV) zone as a probability distribution function (PDF) and then sample event sequence following the PDF while photoelectrical event is considered only. The imitate event sequence and energy analysis result is specified in Fig.7(c) (d). In Fig.7 (d), the yellow curve is the raw energy, and red curve is the estimated energy that completely coincidence with ^{137}Cs feature energy at 661keV. The result suggests that the structure is feasible to analysis underlying monoenergetic source level in right interval.

TABLE II. EXPERIMENT INSTRUMENT AND SOURCE INFORMATION

type	*activey*	*manuf ID*
^{137}Cs	0.635 μCi	
NaI(Ti) detector		independent developed by using HHP201NaI(Ti)
spectroscopy AMP		ORTEC 672
DMA		ORTEC972
recording software		ORTEC Maestro 7.01



(a) (b) (c) (d)

Fig. 7. Experiment Data. (a) ^{137}Cs pulse height spectrum. (b) full-energy peak at 661keV. (c) event mode sequence for 1000 times sample according energy PDF. (d) energy parameter estiamte (at 661keV).

V. CONCLUSIONS

In this paper, we introduced the event mode sequence (EMS) of radionuclide based on monoenergetic source decomposition, and developed a unified framework to define the radionuclide mathematically. Using this framework and certain assumed distribution, we explained how to analysis the radioactive pattern in interested energy interval that related with underlying radioactive material. A parallel/distribution structure was developed and used to proof-of-concept experiment containing numerical simulation and semi-physical simulation to demonstrate its feasibility. The analysis results of monoenergetic source energy and branch ratio are consistent with target radionuclides characterizing parameter.

Future work involves the research of a method to indicate underlying target radionuclide by pattern recognition photo-by-ph0ton, and furthermore analysis pattern feature from the measurement that contain Compton-scattering event in a high background noise environment.

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