Nuclear Engineering and Technology 55 (2023) 3913–3923

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Application of peak based-Bayesian statistical method for isotope identification and categorization of depleted, natural and low enriched uranium measured by $LaBr_3$:Ce scintillation detector

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ARTICLE INFO

Article history:
Received 14 October 2022
Received in revised form
2 June 2023
Accepted 5 July 2023
Available online 26 July 2023

Keywords:
$LaBr_3$(Ce)
Scintillation detectors
Gamma– ray spectrum
Isotope identification
Bayesian statistical method
Detection algorithm
^{235}U
Nuclear security

ABSTRACT

Todays, medium energy resolution detectors are preferably used in radioisotope identification devices(RID) in nuclear and radioactive material categorization. However, there is still a need to develop or enhance « automated identifiers » for the useful RID algorithms. To decide whether any material is SNM or NORM, a key parameter is the better energy resolution of the detector. Although masking, shielding and gain shift/stabilization and other affecting parameters on site are also important for successful operations, the suitability of the RID algorithm is also a critical point to enhance the identification reliability while extracting the features from the spectral analysis. In this study, a RID algorithm based on Bayesian statistical method has been modified for medium energy resolution detectors and applied to the uranium gamma-ray spectra taken by a $LaBr_3$:Ce detector. The present Bayesian RID algorithm covers up to 2000 keV energy range. It uses the peak centroids, the peak areas from the measured gamma-ray spectra. The extraction features are derived from the peak-based Bayesian classifiers to estimate a posterior probability for each isotope in the ANSI library. The program operations were tested under a MATLAB platform.

The present peak based Bayesian RID algorithm was validated by using single isotopes(^{241}Am, ^{57}Co, ^{137}Cs, ^{54}Mn, ^{60}Co), and then applied to five standard nuclear materials(0.32–4.51% at.^{235}U), as well as natural U- and Th-ores. The ID performance of the RID algorithm was quantified in terms of *F-score* for each isotope. The posterior probability is calculated to be 54.5–74.4% for ^{238}U and 4.7–10.5% for ^{235}U in EC-NRM171 uranium materials. For the case of the more complex gamma-ray spectra from CRMs, the *total scoring* (S_T) method was preferred for its ID performance evaluation. It was shown that the present peak based Bayesian RID algorithm can be applied to identify ^{235}U and ^{238}U isotopes in LEU or natural U –Th samples if a medium energy resolution detector is was in the measurements.

1. Introduction

Todays, the concepts of nuclear security and nuclear safety have gained great importance due to emerging nuclear security threats. In particular, it is known that malicious use of dirty bombs or nuclear materials can directly threaten the health and security of public.

For nuclear security purposes, hand-held radioisotope identifiers (RIDs) are commonly used to characterize nuclear materials (NMs) including (SNM, RGPu, WGPu, ^{237}Np, etc.), NORMs/TENORMs, other radioactive materials (RMs) or medical and industrial radioisotopes. Many RIDs have already been deployed by International Atomic Energy Agency (IAEA) and other public authorities to facilitate the detection of threats, tracking radioactive material transportation by following primary inspections through radiation portal monitors (RPMs). Radioisotope Identification Devices (RID) can be used for searching, radioisotope categorization and localizing radioactive sources, and also simultaneously for making gamma dose rate measurements, and optionally indicating the neutron count/dose rate if it has neutron detector. The identification/categorization of radioactive or nuclear materials concealed in

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https://doi.org/10.1016/j.net.2023.07.006

cargoes is always a fundamental issue in nuclear security to prevent illicit trafficking of nuclear and radioactive materials. In this context, gamma-ray spectroscopy is a critical research and development priority to arrange of nuclear security missions, specifically the interdiction of special nuclear material involving the detection and identification of gamma-ray emitting sources [1]. To achieve these tasks, RIDs can be operated as gamma spectrometers to identify certain user defined radioisotopes. Basically, the gamma radiation spectra are compared with gamma lines or reference spectra of frequently observed radioisotopes and identified if statistically significant counting is achieved [2] [IAEA NSS6, 2007]. This implies that RIDs should be more effective and reliable in the field as a secondary inspection devices. Up to now, the majority of RIDs deployed in the field are based on the low energy resolution NaI(Tl) detector based devices and their performance for isotope identification has been well-documented. However, in practice, RIDS are mostly by the first responders or FLOs(Front-Line-Officiers) instead a trained spectroscopist who might capable of identifying complicated, multiple-line sources with even the poorest resolution detectors such as NaI(Tl) [3]. This is true that there are still some inabilities of the commercially available RID devices in view of correctly identifying isotopes. Todays, it is a fact that most of RIDs still use low energy resolution detectors since they have higher efficient and relatively lower cost NaI(Tl) scintillation detectors. Therefore, they do not also need to use sophisticated isotope identification algorithms [4]. This is a main drawback of NaI(Tl) detector due to worse energy resolution, Additionally, NaI(Tl) scintillation detectors also have gain shift/stability of temperature dependency in view of isotope identification performance. On the other hand, in recent years, with advancement of new detector technology, medium energy resolution detectors such as $LaBr_3$:Ce, CeBr and CdZnTe detectors allow new alternative isotope identification improvements due to their much better energy resolution and having a more linear energy response over a wide energy range of up to 2000 keV. Hence, especially medium energy resolution detectors are increasingly used for RIDs in nuclear material detection and radioisotope categorization. This implies that improving the energy resolution of any detector also needs to use for a more sophisticated RID algorithm rather than that of NaI(Tl) detectors. For instance, in the literature, some statistical methods, such as Bayesian Modelling Averaging and hierarchical and empirical Bayesian methods, might reduce the decision uncertainty on identification accuracy of any isotope in a nuclear/radioactive material [1]. If a medium energy resolution detector is used for isotope identification purpose in a conventional RID algorithms, there seems to be another problem that is related to be used “automated identifiers”. At this point, the literature survey implies that there is much room for improvement in the RID algorithms themselves even when automated algorithms fail [3]. Further, the performance of the current automated RID algorithms does not generally meet the requirements of the users nor the ANSI 42.34 standard [5–8]. Therefore, new algorithms are still ongoing in this field to enhance the capability of RIDs to allow more accurate isotope identification. Since RID algorithms provides the quantitative information from the spectral data, the extraction features on what isotopes are present and what type of radioactive material can be categorized from the inspected items can be provided through these RID algorithms in which either use all energy channels in the analysis region(ROI) or only energy channels in and near identified peaks [1]. However, in real situation, it is a fact that unknown shielding, isotope masking, low-count spectra, calibration drift, and other effects complicate the radioisotope identification process, often leading to an incorrect isotope identification (ID) or categorization.

The main aim of this study is to modify a RID algorithm based on Bayesian statistics approach, first time developed by Stinnet [12,23] which can also be used for identification of uranium isotopes(^{235}U, ^{238}U) for SNM categorization from natural, depleted and low enriched uranium materials if a medium energy resolution detector is employed. The present RID algorithm uses both peak positions (centroids) and peak areas (ROI-Region of Interest) from the measured gamma-ray spectrum. To identify any radioisotope, the proposed Bayesian peak based RID algorithm was validated by means of single isotopes-gamma-ray spectra acquired with a $LaBr_3$:Ce detector in which we firstly optimized an energy tolerance of $E_D = \pm 5$ keV which is the difference between the measured peak energy and the peak energy specified in the isotope library. The modified Bayesian peak based-RID algorithm was tested up to 2000 keV energy range. To achieve this, the nergy and shape calibrations were already performed at a given shielding condition. This new RID algorithm was first time applied to the more complex gamma-ray spectra obtained from five EC-NRM171 samples containing depleted uranium(DU), natural uranium(NU) and low enriched uranium(LEU) and other some Certified Reference Materials(CRMs) containing U and Th decay products. The measured gamma-ray spectra from the above mentioned nuclear materials were used to establish « automated identifiers» in the proposed RID algorithm to decide whether the material are radioactive/nuclear characteristic or SNM in view of radioisotope categorization.

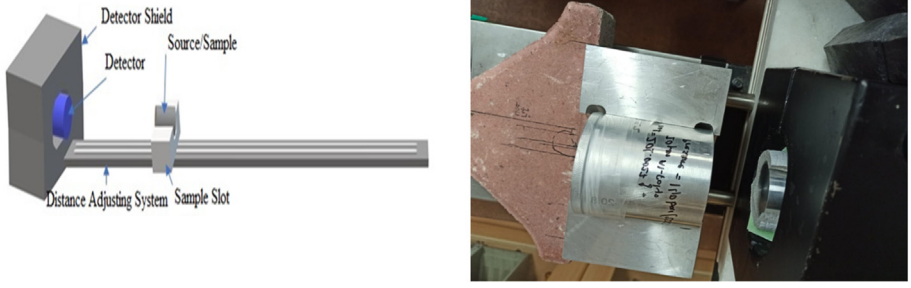
2. Measurements and isotope identification method

2.1. Experimental

In this study, a 38.1 mm × 38.1 mm LaBr3:Ce scintillation detector (BrilLanCe 380) crystal purchased from Saint Gobain Crystals(SGC) was used. SGC BrilLanCe 380 (LaBr3:Ce) crystal coupled directly to a specially selected photomultiplier tube (PMT) in sealed housing.The technical specifications of the detector as well as its energy resolution of 17.1 keV (2.59%) at 662 keV, and the relative efficiency value is 15.4% at 25 cm source-to-detector distance, relative to 3 inch × 3 inch NaI for 1332.5 keV (60Co). For data acquisition, a 2048 spectrum channels was used through Canberra Multiport II 16K ADC/MCA supported by [11] Genie 2000 Gamma Software.

In the measurement setup, the detector side was shielded with a 5 cm thick lead to reduce ambient background and then it was adjusted gain to cover up to 2000 keV energy range. The detector was calibrated in terms of energy and peak shape (in FWHM) using single isotope energies. As shown in Fig. 1, the measurements were carried out at a 5 cm, 15 cm and 25 cm distance from $LaBr_3$:Ce scintillation detector to observe the effect of counting statistics on the posterior probability for the isotopes such as ^{235}U. The measurement periods were chosen from 30s to 900s.

The certified uranium samples was pressed in an Al-can, back plugged with an sealed Al-cylinder as shown in Fig. 2(a). In order to prepare additional samples, a similar sample filling method was also employed to obtain the same apparent density at the sample height ($H_s = 2.11$ cm) and inside diameter ($D_s = 6.6$ cm) and $d = 1$ mm thin Al-window. Each aluminium can was fabricated in a CNC machine from grade 6061 aluminium-alloy complying with EN 573-3 (which is equivalent to ASTM 6061-T6 aluminium alloy), as shown in Fig. 2(b). The powder forms were pressed in each of cylindrical aluminium cans for filling CRMs minerals, purchased from

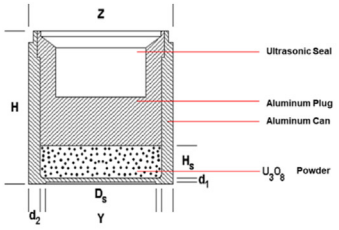


a) Source-detector distance adjustable radiometric bench

b) A fixed cylindrical source-to-detector measuring setup

Fig. 1. A gamma-ray measuring system with $LaBr_3$:Ce scintillation detector setup in a test rig.

a) CBNM Certified uranium cylinders (EC NRM 171)



b) Sample filling geometry in aluminium can

Fig. 2. EC NRM 171 Nuclear reference uranium samples and sample filling in aluminium cans.

Natural Resources Canada (NRCan) containing U-and Th-ores. For instance, CUP-2 certified uranium ore concentrate contains 75.42 ± 0.17% wt. U, BL-5 low-grade concentrate contains 7.09 ± 0.03 %wt. U, and OKA-2 Rare-Earth − Thorium Ore contains 2.893 ± 0.058 wt%Th and 218.6 ± 8.2 μg/g U [12]. ^{235}U isotopic abundances in EC NRM 171 standard uranium materials are given in Table 1.

2.2. Radioisotope identification algorithm

For automated radio isotope identification(RID), the RID algorithms can broadly be classified into several groups:

- Library comparisons,
- Region of interest (ROI) methods,
- Template matching,
- Expert interaction [14,15]

ID algorithms used in commercial detector devices are generally proprietary knowledge, only a few of these are practical for use on a RID device. Many algorithms for ID must first employ a peak finding method to quantify the centroid energy and possibly the area of all photopeaks in a spectrum. Bayesian statistics based algorithm has also developed for low energy resolution NaI detector in which it utilizes peak energies and peak areas to compute the posterior probabilities for each isotope given in adapted library that includes the isotopes of ANSI 42.34, and additional ones such as ^{226}Ra, ^{232}Th day products [17]. In the present developed algorithm, several steps were conducted as follows:

— Creating a library data base (SNM, Medical, Industrial, NORM, etc.), where based on ANSI library
— Energy calibration of detector, such as a quadratic function: $E(keV) = a + b \bullet X + c \bullet X^2$, where X is channel

Table 1
^{235}Uranium isotopic abundances in EC NRM 171-standard nuclear materials [13].

Certified Reference Material	Isotopic Abundance in atom %		Isotopic Abundance in weight %	
Code	$^{235}U/U$	$^{238}U/U$	$^{235}U/U$	$^{238}U/U$
EC-NRM171-031	0.3205 ± 0.0002	99.6627 ± 0.0004	0.3166 ± 0.0002	99.6668 ± 0.0004
EC-NRM171-071	0.7209 ± 0.0005	99.2738 ± 0.0002	0.7119 ± 0.0005	99.2828 ± 0.0002
EC-NRM171-194	1.9664 ± 0.0014	98.0159 ± 0.0009	1.9420 ± 0.0014	98.0406 ± 0.0009
EC-NRM171-295	2.9857 ± 0.0021	96.9826 ± 0.0015	2.9492 ± 0.0021	97.0196 ± 0.0015
EC-NRM171-446	4.5168 ± 0.0032	95.4398 ± 0.0016	4.4623 ± 0.0032	95.4950 ± 0.0016

*Data is provided by EC nuclear reference material EC NRM 171 certification report(No: EUR 10503 EN) -^{235}Uranium isotope abundance certified reference material for gamma spectrometry.

— Peak shape calibration of detector, such as root square of energy: $FWHM = \sqrt{a + b \bullet E}$, where FWHM is full width at half-maximum of the peak and E is the gamma-ray energy.
— Finding peak centroids from photopeaks in the acquired spectrum
— Compare the centroid with library and matching with the peak energy of isotope in the library
— Finding peak area in relevant ROI
— Calculation posterior probabilities by Bayesian Approach
— Scoring Iindex model, F-score(harmonic mean) or S_T-Total scoring model
— Identification of radioisotopes
— Reporting.

Since the presently developed RIID algorithm is based on both peak centroids and the peak areas of the relevant peaks for isotope identification, the determination of net peak area that under the photopeak in the measured gamma-ray spectrum is a very important step. This is because the net peak area provides the most quantitative information to estimate the posterior probability of the interested isotope. In order to determine net peak area in the region of interest (ROI), a simple but most practical method called-*Covell method* have been chosen and employed in this algorithm. The Compton continuity under the peak is calculated by using a linear background subtraction manner as follows:

$$A = \sum_{i=L}^{U} C_i - \frac{n\left[\sum_{i=L-m}^{L-1} C_i - \sum_{i=U+m}^{U+1} C_i\right]}{2m} \quad (1)$$

where, A is net peak area. C_i are counts in the i^{th} channel. L and U are left and right edge channel of peak. m is channel wide. n is the number of channels in the peak region [17], where m and n are adjustable parameters.

The net count of peaks must be above certain limits in order to be considered statistically significant. These limits are known as critical limit, upper limit, detection limit and decision limit. In order to calculate the critical limit, the confidence limit was chosen as 95%. The algorithm developed using these limits can identify significant peaks in the spectra for single and multiple isotopes, where those details are not given to save space because they are well described in text books[17] as well as other instruction manuals of the most common commercial softwares such as Genie 2000 and GammaVision.

2.3. Bayesian based isotope identification

The Bayesian statistics method based on Thomas Bayes' theorem is used in many applications and researches. This method is presented as an alternative to classical statistics approaches. For this reason, it has also been called "Inverse Probability" by some scientists [6]. In Bayesian statistics approach, the probability of an event can be determined by bringing together different sources of information within a certain methodology [18]. Bayesian statistical methods predict that a particular parameter M, such as an isotope responsible for a given data set, would be considered unknown but fixed by a frequentist, in a Bayesian view something is known about it [18]. Before taking any data, a prior distribution P(M) contains any a priori knowledge and assumptions about the tested parameter(M) and (D), data obtained related to parameter M. In the case of isotope identification, we adopt that a spectrum is produced by a radioactive isotope that must be in our spectral library. If the gamma-lines of the isotope do not match mostly to those of isotope in the library, then misidentification would be an inevitable result. Hence, the simple form of Bayes theorem on which Bayesian statistical approach is based is given by the following Eq. (2) [9,20].

$$P(M|D) = \frac{P(D|M) \cdot P(M)}{P(D)} \quad (2)$$

where P(M|D) is posterior probability, P(D|M) is likelihood function, P(M) is prior probability and P(D) is the normalization factor.

For the likelihood function P(D|M), there is a model based on four parameters [9]. This pattern depends on the position of the peak (f_{PP}), the percentage of peaks matching with the data in the library (f_{LPI}), the percentage of identification of the data peaks (f_{DPI}), and the peak area (f_{AR}):

$$P(D|M) = f_{LPI} \cdot f_{DPI} \cdot f_{PP} \cdot f_{AR} \quad (3)$$

where, f_{LPI} is the ratio of the sum of the gamma emission probabilities of the defined library peaks to the sum of the gamma emission probabilities of all the peaks of the relevant radioisotope in the library. f_{DPI} is estimated from the ratio of the peaks defined for a particular isotope to the number of all data peaks defined in the spectrum.

The f_{PP} parameter is a parameter that depends on the peak position. This parameter is determined by the difference between the energy of the peak obtained from the spectrum (E_D) and the energy that matches that energy in the library (E_L). This parameter is important for the correct identification of peaks in possible peak shifts. The f_{PP} parameter is expressed by a function dependent on the $\cos^2$ (x).

$$f_{PP}(E_D) = \begin{cases} b + (1-b)cos^2\left(\frac{\pi}{2} \cdot \frac{E_D - E_L}{w}\right) & E_D - E_L < w \\ b & E_D - E_L \geq w \end{cases} \quad (4)$$

where, b is a decreasing function with energy as follows:

$$b = 0.3 - \left[0.2 \cdot \frac{E_D}{2000}\right]$$

$$cos(x) = \frac{\pi}{2} \cdot \frac{E_D - E_L}{w}$$

The maximum energy that can be detected in scintillation detectors might be chosen more higher energies than 2000 keV, however, we tested that 2000 keV energy range is experimentally. This is a reasonable value for a 38.1mmx38.1 mm $LaBr_3$:Ce scintillation detector because the energy response is quite linear up to 2000 keV. Moreover, most of gamma lines below 2000 keV can easily be present in standard isotope libraries available in the commercial softwares for RIDs. Energy tolerance (w) is expressed in Eq. (5). In case of peak shifts, the energy of the peak of interest in the spectrum is considered valid in the range $E_D \pm w$. For a 38.1mm x 38.1 mm $LaBr_3$:Ce scintillation detector, we experimentally set the energy matching as a 5 keV between E_D and E_L and thus the energy tolerance width can be estimated by the following proposed equation:

$$w = 5 + \frac{E_D}{2000} x\, 5 \quad (5)$$

The last parameter in the Bayesian statistical model, f_{AR}, represents the effect of shielding conditions on the likelihood function.

This parameter is calculated by comparing the area between the E_1 and E_2 peaks in the data (r) and the reference area ratio (R) belonging to the library. The information about shielding is also included in the probability, depending on whether the value of r is greater or less than R[9].

2.4. Performance evaluation of peak based Bayesian RID algorithm

The reliability of radioisotope identification(RID) was quantified using two different performance approaches. The same scoring criteria were employed to objectively assess the performance of an isotope identification algorithm. For instance, a report of Domestic Nuclear Detection Office (DNDO) provides an overview of the equations, nuclide weighting factors, nuclide equivalencies, and configuration weighting factors used by the application for scoring RID algorithm performance models [14,19]. It is worth noting that, in the analyses made with Bayesian statistics, the deviation from the real value can be found by looking at *the precision* values instead of calculating *the variance* values as in classical statistics [23].

The models such as F − score (i.e., harmonic mean) or total scoring(S_T) used in performance evaluation in radioisotope identification applications can be described as follows:

2.4.1. F − score model

The F − score is a criterion for the basic statistical model used in the RID algorithm performance evaluation [24]. The data obtained in this evaluation model are classified in four different ways as true positive (TP), true negative (TN), false positive (FP) and false negative (FN). With this classification obtained, precision (P), sensitivity (i.e., Recall, r), accuracy (A) and F - score (F - score) values can be calculated. While evaluating the performance of the algorithm applied here, calculating accuracy may not always give correct results. For this reason, the *F-score model* has been developed relating to both precision (P) and recall (r) parameters.

$$Precision\ (P) = \frac{TP}{TP + FP} \tag{6}$$

$$Recall\ (r) = \frac{TP}{TP + FN} \tag{7}$$

$$Accuracy(A) = \frac{TP + TN}{TP + FP + TN + FN} \tag{8}$$

Thus, F - score value is simply calculated the harmonic mean of the precision and recall(i.e.,sensitivity) values, as seen in Eq.10.

$$Score\left(F\right) = 2\frac{P \cdot r}{P + r} \tag{9}$$

The F - score is estimated as a decimal number between 0 and 1, and as this F - score approaches 1, that is to say 100% corresponds to the accuracy of the algorithm which is the best score.

2.4.2. Total scoring model, S_T

The total scoring model which is used to evaluate radioisotope identification applications, works within certain rules as seen in Table 2 and Table 3 instead of two different possibilities as true or false as in other models. Evaluation of the radioisotope identification results of spectra containing only one radioisotope is done by scoring with the rules in Table 2 to quantify total scoring number(S_T),. The scoring system in Table 3 is used to examine the performance of RID algorithms on more complex spectra of radioactive materials which might contain many radioisotopes than one single radioisotope such as enriched uranium, reactor fuel and U- or Th-ores and their daughters.

3. Results and discussion

In this study, a new technology $LaBr_3$:Ce scintillation detector having a better energy resolution than that of NaI(Tl) scintillation detectors, conventionally used in RIDs was considered to develop this new algorithm for ID purpose. Since a $LaBr_3$:Ce scintillation detector gave the well resolvable photopeaks in the spectrum, the developed algorithm have resulted in more precise and accurate radioisotope identification, thus also allowing SNM categorization. To this end, the point single- sources such as ^{241}Am, ^{57}Co, ^{54}Mn, ^{137}Cs and ^{60}Co were used to validate the proposed RID algorithm based on Bayesian statistical approach. In the present modified peak based Bayesian RID algorithm, the posterior probability values using Eq. (2) were obtained for single isotopes having one or two separate peaks such as ^{241}Am (59.6 keV) seen in Fig. 3, ^{57}Co (122.1 keV; 136 keV) seen in Fig. 4, ^{137}Cs (661.6 keV) seen in Fig. 5, ^{54}Mn (834.8 keV) seen in Fig. 6 and ^{60}Co (1173.2 keV; 1332.5 keV)

Table 2
Identification scoring for spectra containing a single radioisotope [21].

Score	Scoring for identification of a single radioisotope
1	Only one radioisotope identified and it is correct.
0.50	2+ radioisotopes identified: one radioisotope is correct radioisotope and it has the highest confidence
0.25	2+ radioisotopes identified: one radioisotope is correct, but an incorrect radioisotope has the highest confidence.
0	No radioisotope identified
−1	1+ radioisotopes identified but none is correct

Table 3
Identification scoring for spectra containing more than one radioisotope [21].

Score	Score radioisotope identification results
1	2 radioisotopes identified and both are correct.
0.75	3+ radioisotopes identified, including the two correct radioisotopes (and one of the correct radioisotopes has the highest confidence)
0.50	3+ radioisotope identified, including the two correct radioisotopes (but the highest confidence radioisotope is not correct), or 1 radioisotope identified and it is correct.
0.25	2+ radioisotopes identified but only one is correct
0	No radioisotope identified
−1	1+ radioisotope (s) identified but none is correct.

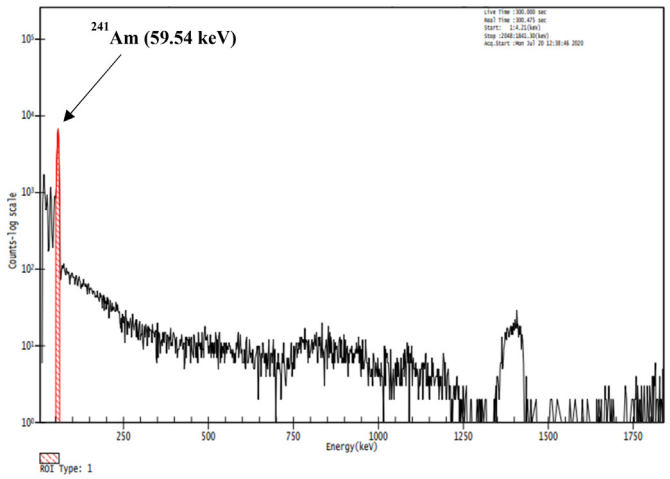


Fig. 3. Posterior probability values estimated from Bayesian statistical approach for ^{241}Am gamma-ray spectrum measured by a 38.1 mm × 38.1 mm $LaBr_3$:Ce scintillation detector.

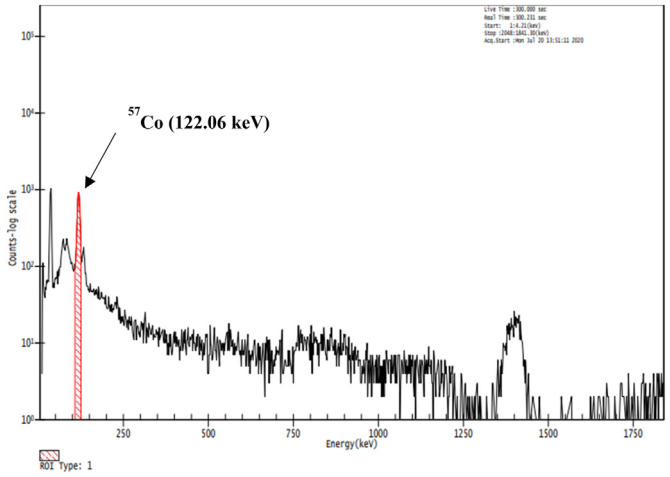


Fig. 4. Posterior probability values estimated from Bayesian statistical approach for ^{57}Co gamma-ray spectrum measured by a 38.1 mm × 38.1 mm $LaBr_3$:Ce scintillation detector.

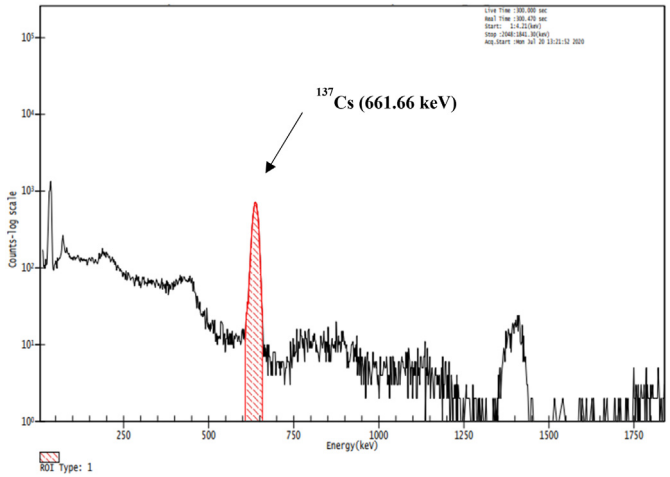


Fig. 5. Posterior probability value estimated from Bayesian statistical approach for ^{137}Cs gamma-ray spectrum measured by a 38.1 mm × 38.1 mm LaBr3:Ce scintillation detector.

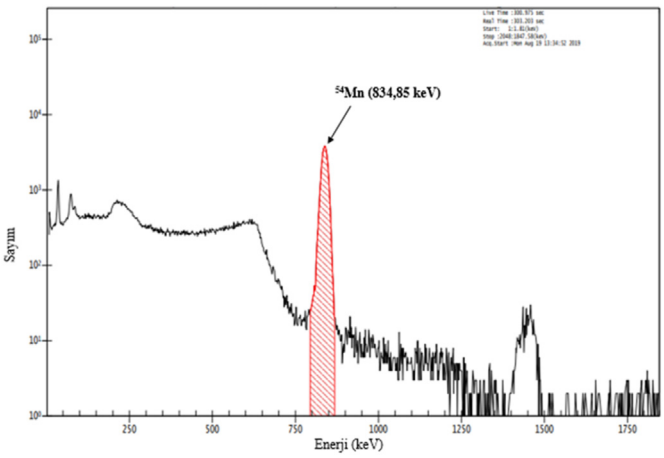


Fig. 6. Posterior probability value estimated from Bayesian statistical approach for ^{54}Mn gamma-ray spectrum measured by a 38.1 mm × 38.1 mm LaBr3:Ce scintillation detector.

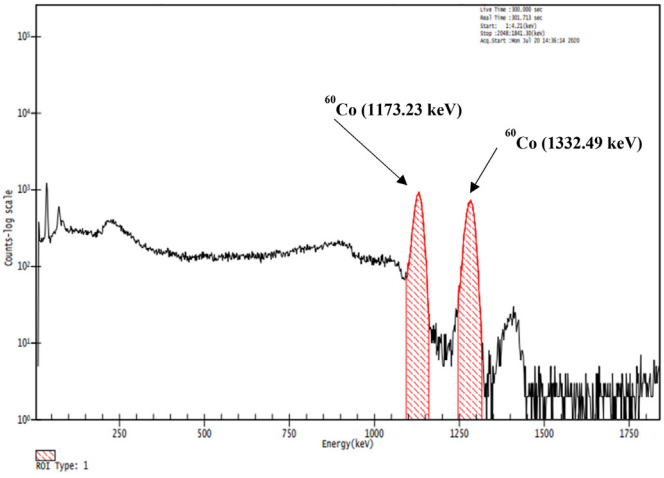


Fig. 7. Posterior probability value estimated from Bayesian statistical approach for ^{60}Co gamma-ray spectrum measured with a 38.1 mm × 38.1 mm LaBr3:Ce scintillation detector.

seen in Fig. 7. Each source was measured separately at a 5 cm distance from the detector, as shown in Fig. 1. These posterior probability results for single isotopes validate the present peak based Bayesian RID algorithm when the isotopes in ANSI library matching with the measured peak centroids in the spectra according to the rules considered in this statistical approach.

	Estimated from Fig. 3
Identified isotope	**Posterior probability (%)**
^{241}Am	62.6025
^{238}U	20.6347
^{239}Pu	16.7628

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

	Estimated from Fig. 4
Identified isotope	**Posterior probability (%)**
^{57}Co	76.3677
^{152}Eu	17.6987
^{239}Pu	5.9336

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

	Estimated from Fig. 5
Identified isotope	**Posterior probability (%)**
^{137}Cs	100.00

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

	Estimated from Figure 6
Identified isotope	Posterior probability (%)
^{54}Mn	100.00

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

	Estimated from Fig. 7
Identified isotope	Posterior probability (%)
^{60}Co	100.00

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

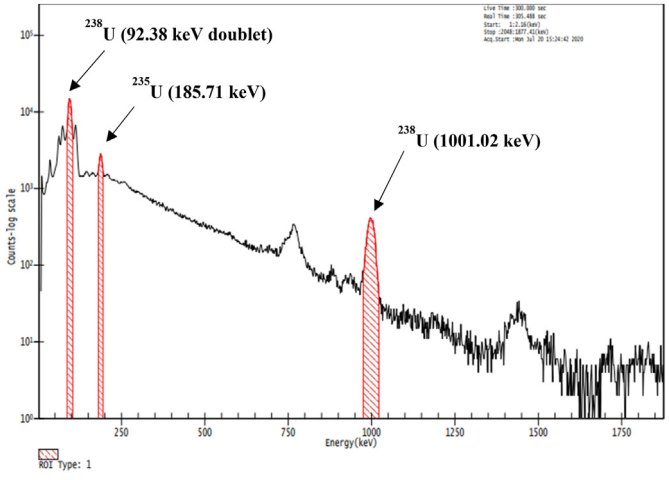


Fig. 8. Posterior probability values estimated from Bayesian statistical approach for uranium isotopes in NRM171-031 depleted uranium material measured with a 38.1 mm × 38.1 mm $LaBr_3$:Ce detector.

After the peak based Bayesian RID algorithm was validated by using single isotopes with one or two-emitting gamma-ray peaks, the posterior probabilities of the uranium isotopes were then estimated for the more complex spectra from NRM 171-031

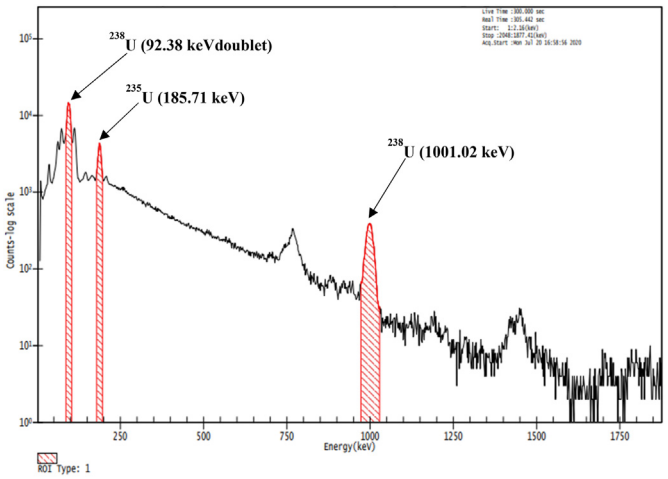


Fig. 9. Posterior probability values estimated from Bayesian statistical approach for uranium isotopes in EC NRM171-071 natural uranium material measured with a 38.1 mm × 38.1 mm $LaBr_3$:Ce detector.

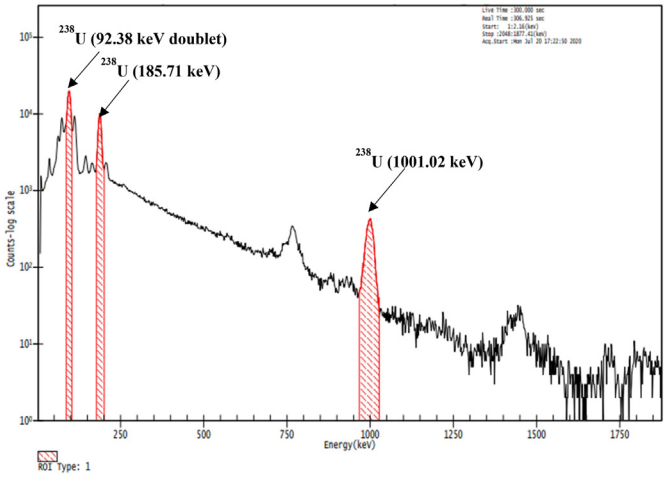


Fig. 10. Posterior probability values estimated from Bayesian statistical approach for uranium isotopes in EC NRM171-194 low enriched uranium material measured with a 38.1 mm × 38.1 mm $LaBr_3$:Ce detector.

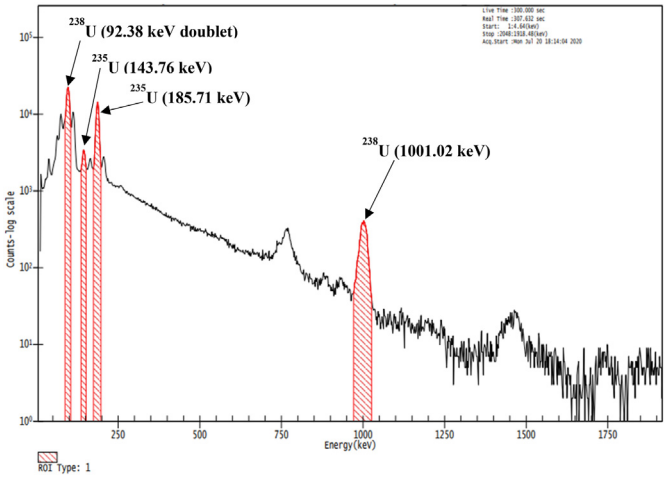


Fig. 11. Posterior probability values estimated from Bayesian statistical approach for uranium isotopes in EC NRM171-295 low enriched uranium material measured with a 38.1 mm × 38.1 mm $LaBr_3$:Ce detector.

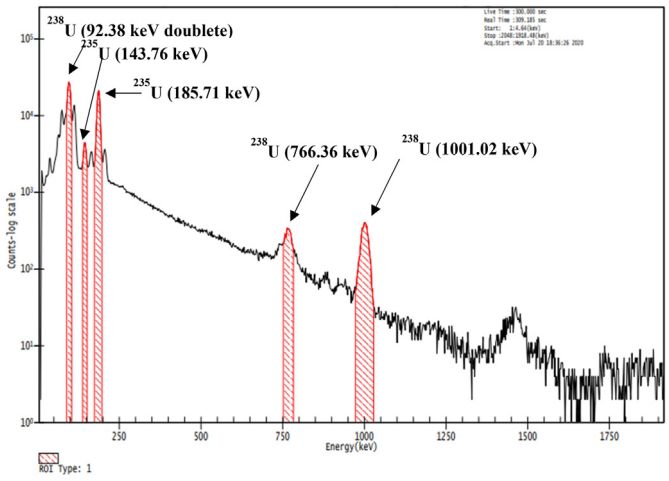


Fig. 12. Posterior probability values estimated from Bayesian statistical approach for uranium isotopes in EC NRM171-446 low enriched uranium material measured with a 38.1 mm × 38.1 mm $LaBr_3$:Ce detector.

depleted uranium (in Fig. 8), NRM171-071 natural uranium (in Fig. 9) and low enriched uranium materials NRM 171-194,-295,-446(in Fig. 10 through 12).

EC NRM171-031 depleted uranium material	Estimated from Fig. 8
Identified isotope	**Posterior probability (%)**
^{238}U	74.4058
^{67}Ga	16.5048
^{235}U	5.0032
^{233}U	3.5724
Others	0.5138

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

EC NRM171-071 natural uranium material	Estimated from Fig. 9
Identified isotope	Posterior probability (%)
^{238}U	68.2645
^{67}Ga	15.2629
^{133}Xe	6.2735
^{235}U	4.7041
Others	5.495

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

EC NRM171-194 low enriched uranium material	Estimated from Fig. 10
Identified isotope	**Posterior probability (%)**
^{238}U	67.7222
^{67}Ga	15.2465
^{133}Xe	6.4350
^{235}U	4.8253
Others	5.7710

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

EC NRM171-295 low enriched uranium material	Estimated from Fig. 11
Identified isotope	**Posterior probability (%)**
^{238}U	54.4962
^{235}U	16.2997
^{67}Ga	11.7540
^{133}Xe	6.0953
Others	11.3548

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

EC NRM171-446 low enriched uranium material	Estimated from Fig. 12
Identified isotope	Posterior probability (%)
^{238}U	70.9222
^{235}U	10.5085
^{67}Ga	8.6511
^{133}Xe	3.9297
Others	5.9885

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

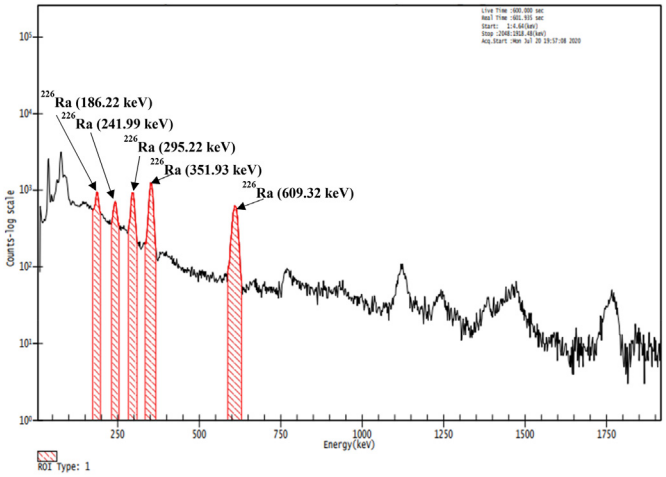


Fig. 13. Posterior probability values estimated from Bayesian statistical approach for radium-decay isotopes in BL-3 uranium ore material measured with a 38.1 mm × 38.1 mm $LaBr_3$:Ce detector.

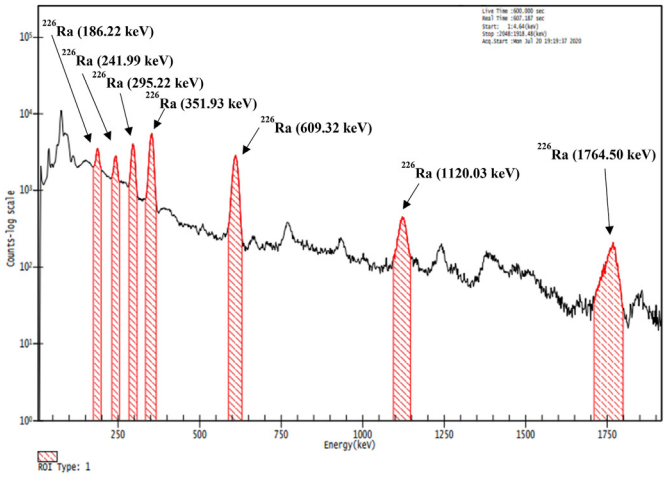


Fig. 14. Posterior probability values estimated from Bayesian statistical approach for radium-decay isotopes in BL-5 uranium ore material measured with a 38.1 mm × 38.1 mm $LaBr_3$:Ce detector.

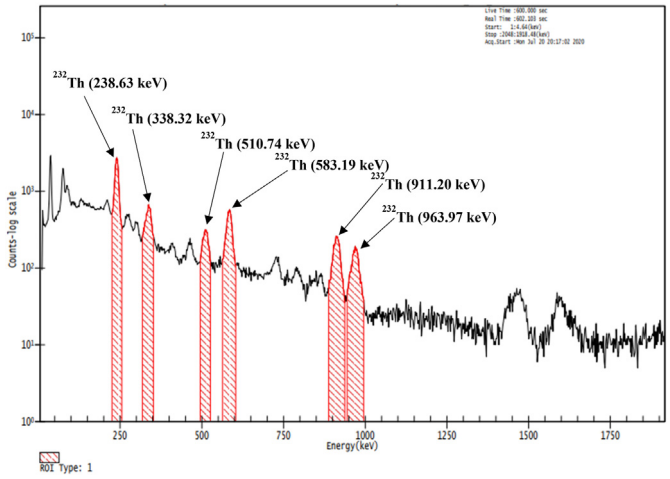


Fig. 15. Posterior probability values estimated from Bayesian statistical approach for thorium isotopes in OKA-2 rare-earth thorium ore material measured with a 38.1 mm × 38.1 mm LaBr3:Ce detector.

For the case of more complex gamma-ray spectra from natural U-ore(BL-3 in Fig. 13, BL-5 in Fig. 14) and OKA-2 Th-ore material in Fig. 15 were also acquired by a calibrated $LaBr_3$:Ce scintillation detector at different measurement periods and source-to-detector distances. In this paper, however, the exampled results are presented at d = 5 cm distance and 5 min measurement periods to evaluate the performance of the peak-based Bayesian approach in which a posterior probability is estimated for each isotope in ANSI library.

BL-3 certified natural uranium ore material	Estimated from Fig. 13
Identified isotope	**Posterior probability (%)**
^{226}Ra	70.7556
^{133}Ba	15.5356
^{67}Ga	4.5558
^{133}Xe	2.3812
Others	6.7718

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

BL-5 certified natural uranium ore material	Estimated from Fig. 14
Identified isotope	Posterior probability (%)
^{226}Ra	86.1230
^{133}Ba	7.3764
^{152}Eu	2.3656
^{235}U	0.7236
Others	3.4114

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

OKA-2 certified thorium ore material	Estimated from Fig. 15
Identified isotope	Posterior probability (%)
^{232}Th	89.8463
^{152}Eu	4.9187
^{22}Na	2.1027
^{133}Xe	1.9161
Others	1.2162

Source to detector distance, d = 5 cm
Measurement time, t = 300 s.

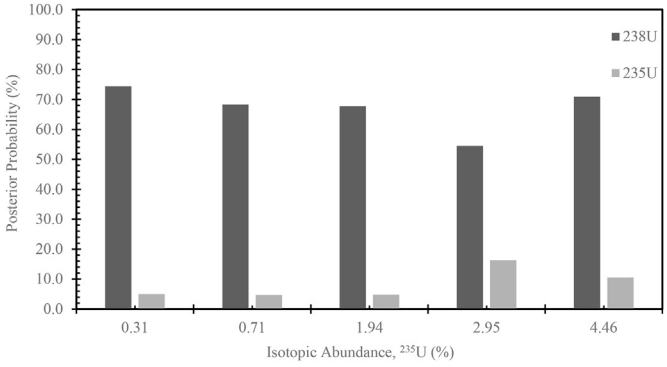


Fig. 16. The percentage posterior probability of ^{235}U and ^{238}U isotopes for the different low enriched uranium reference materials.

The posterior probabilities of most abundant isotope ^{238}U and fissile isotope ^{235}U and in low enriched uranium materials are shown in Fig. 16 for a measurement period of 300s at a 5 cm distance,. However, in short measurements, say 30–60s, ^{235}U isotope could not be identified in depleted uranium due to mainly very low counting statistics while the present peak-based Bayesian RID algorithm is employed.

As a histogram, Fig. 16 shows the percentage posterior probabilities for both ^{235}U and ^{238}U contained in depleted uranium(0.31% wt. ^{235}U), natural uranium, (0.71%wt. ^{235}U) and three low enriched uranium (1.94%wt. ^{235}U, 2.95%wt. ^{235}U and 4.46%wt. ^{235}U) standard materials(i.e., EC-NRM-171). ^{235}U isotope is identified in low enriched uranium but the posterior probability is less than 10%, as expected. On the other hand, ^{238}U isotope is identified more accurately with a posterior probability of 55–71% in the same samples. Although the posterior probability of ^{235}U isotope identification is much lower, these results indicate that the present peak-based Bayesian RID algorithm works well to identify quantitatively ^{235}U fissile isotope in uranium even if they are depleted or natural uranium forms.

The applicability of the presently developed peak based-Bayesian RID algorithm to a medium energy resolution $LaBr_3$:Ce scintillation detector is given in terms of ID performance index either total scoring (S_T) or F-score. The developed peak-based Bayesian algorithm was applied to not only single isotopes but also uranium isotopes. The ID performance was quantified in terms of F-score for each isotope (see Table 7, 8 and 9). In case of more

Table 4
Total scoring results for radioisotope identification performance of the algorithm in EC-NRM-171 nuclear standard materials for different measurement periods.

Measurement Time (s)	Depleted, natural and low enriched uranium (LEU) standard materials					Total score, S_T (%)
	EC NRM171-031	EC NRM171-071	EC NRM171-194	EC NRM171-295	EC NRM171-446	
30	0.50	0.75	0.75	0.75	0.75	70.00
60	0.75	0.75	0.75	0.75	0.75	75.00
300	0.75	0.75	0.75	0.75	0.75	75.00
600	0.75	0.75	0.75	0.75	0.75	75.00
900	0.75	0.75	0.75	0.75	0.75	75.00

Source to detector distance, d = 5 cm

Table 5
Total scoring results for radioisotope identification performance of the algorithm in U and Th- ore certified reference material for different measurement periods.

Measurement Time (s)	U- and Th- ore certified materials				Total score, S_T (%)
	BL-2	BL-3	BL-5	OKA-2	
30	0.00	0.50	0.50	0.75	37.50
60	0.00	0.50	0.75	0.75	37.50
300	0.50	0.50	0.75	0.75	56.25
600	0.50	0.75	0.75	0.75	62.50
900	0.50	0.75	0.75	0.75	62.50

Source to detector distance, d = 5 cm

complex spectra such as from U–Th ore materials, the total scoring (S_T) method was employed for the performance evaluation(see Table 4, 5 and 6). These ID performances are given in Table 4 through Table 9 for different measurement periods and source-to-detector distances for both EC-NRM 171 uranium standards and CRMs containing natural U and Th (see Table 5) (see Tables 6 and 7).

At different measuremet times and source-to-detector distances, the presently developed peak-based Bayesian RID algorithm allows us to radioisotope categorization and SNM categorization. This shows that it works well for the case of $LaBr_3$:Ce medium resolution detector if one wants to identify SNM as well as other radioisotopes given the library.

4. Conclusion

The peak based Bayesian RID algorithm was modified from the well-known Bayesian statistical approach which was already used for the low energy resolution NaI:Tl detectors. It has been successfully validated for a medium energy resolution $LaBr_3$:Ce detector by using single isotopes of ^{241}Am, ^{57}Co, ^{137}Cs, ^{54}Mn and ^{60}Co. Then this RID algorithm was applied first time to five EC NRM171 nuclear uranium standard materials (containing 0.32–4.51% at.^{235}U), as well as natural U- and Th-ores such as BL-2, BL-3, BL-5 and OKA-2 ore materials. In order to get ID extraction features from the measured gamma ray spectrum in the peak based Bayesian approach, essentially two basic classifiers(experimentally determined peak centroids and peak areas) were used., Thus we estimated a posterior probability for each isotope if it matched with the isotope in the library. In this work, ANSI standard libraries are preferably considered to match peak centroids within energy tolerances for the case of a calibrated detector.

In conclusion, the ID performance of the developed peak based Bayesian RID algorithm was quantified in terms of F-score for each isotope in the acquired gamma-ray spectrum. The posterior probability is calculated to be 54.5–74.4% for major isotope ^{238}U and 4.7–10.5% for minor isotope ^{235}U in uranium standard materials. When this developed RID algorithm was applied to more complex gamma-ray spectra, for eaxample, obtained from CRMs containing U and Th, then, the total scoring index (S_T) for the ID performance evaluation of the present algorithm resulted in more reliable decisions to identify ^{226}Ra, ^{238}U and ^{232}Th and their decay products. Since total scoring (S_T) index considers many radioisotopes than one radioisotope present in the spectrum, it is a more useful ID performance evaluation method for the peak based Bayesian RID algorithm. In conclusion, it was shown that the developed Bayesian RID algorithm can be applied to identify ^{235}U and ^{238}U isotopes in depleted, natural and LEU or natural U–Th samples, and other medical, industrial isotopes if the measurements are carried out using a medium energy resolution scintillator detector such as $LaBr_3$:Ce.

Table 6
Total scoring results for radioisotope identification performance of algorithm in EC NRM 171 nuclear standard materials at different measuring distances.

Source to detector distance (cm)	Depleted, natural and low enriched uranium (LEU) standard materials					Total score, S_T (%)
	EC NRM171-031	EC NRM171-071	EC NRM171-194	EC NRM171-295	EC NRM171-446	
5	0.75	0.75	0.75	0.75	0.75	75.00
15	0.75	0.75	0.75	0.75	0.75	75.00
25	0	0.75	0.50	0.50	0.50	45.00

Measurement time, t = 300 s.

Table 7
F-score results for radioisotope identification performance of algorithm in EC NRM 171 nuclear standard materials for different measurement periods.

Measurement time (s)	True positive (TP)	False positive (FP)	False Negative (FN)	Precision (P)	Recall (r)	Performance Index (F –Score)
30	5.00	0	0	1.00	1.00	1.00
60	5.00	0	0	1.00	1.00	1.00
300	5.00	0	0	1.00	1.00	1.00
600	5.00	0	0	1.00	1.00	1.00
900	5.00	0	0	1.00	1.00	1.00

Source to detector distance, d = 5 cm

Table 8
F – score results for radioisotope identification performance of algorithm in U and Th ore certified reference materials for different measurement periods.

Measurement time (s)	True positive (TP)	False positive (FP)	False Negative (FN)	Precision (P)	Recall (r)	Performance Index (F –Score)
30	2.00	1.00	1.00	0.67	0.67	0.67
60	2.00	1.00	1.00	0.67	0.67	0.67
300	4.00	0	0	1.00	1.00	1.00
600	4.00	0	0	1.00	1.00	1.00
900	4.00	0	0	1.00	1.00	1.00

Source to detector distance, d = 5 cm

Table 9
F – score results for radioisotope identification performance of algorithm in EC-NRM 171 nuclear standard materials at different measuring distances.

Source to detector distance (cm)	True positive (TP)	False positive (FP)	False Negative (FN)	Precision (P)	Recall (r)	Performance Index (F –Score)
5	5.00	0	0	1.00	1.00	1.00
15	5.00	0	0	1.00	1.00	1.00
25	1.00	3.00	1.00	0.25	0.50	0.33

Measurement time, t = 300 s.

Declaration of conflict of interest

Authors have no conflict of interest to declare.

Acknowledgement

This work is fully supported by International Atomic Energy Agency (IAEA) J02012 coded Coordinated Research Project (2017-2022) on Advancing Radiation Detection Equipment for Detecting Nuclear and Other Radioactive Material out of Regulatory Control (MORC), which is under contract Nr.20908, entitled "*Method Development for High, Medium and Low Resolution Detector Based Gamma Spectroscopic Determination of ^{235}U Isotopic Abundance in Nuclear Material Detection and Characterization*".

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