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Nuclide identification algorithm for Polyvinyl Toluene scintillation detector based on Deep Neural Network

Cao Van Hiep[1,*], Dinh Tien Hung[1], Cao Dang Luu[2], Le Manh Duc[3], Pham Dinh Khang[4]

[1]*Military Institute of Chemical and Environmental Engineering*
[2] *Center 81, Chemical Corps, Ministry of National Defense*
[3]*Department of radiotherapy and radiosurgery, 108 Military Central Hospital*
[4]*Hanoi University of Science and Technology*
**E-mail: caovanhiep123@gmail.com*

Abstract: Radiation portal monitors (RPMs) are now stationed at strategic areas (airports, ports, etc.) to identify the illegal transportation of radioactive sources and nuclear items. RPMs are typically fitted with plastic scintillations detectors with high detection efficiency. Radioisotope identification from the gamma spectrum acquired on this detector usually is not regarded due to the low resolution. This research describes an artificial neural network-based isotope identification algorithm applied to the gamma spectrum collected from the RPM's PVT detector. Measured and simulated gamma spectra of 5 radionuclides are used for training and validating the proposed model, namely 241Am, ^{133}Ba, ^{137}Cs, ^{60}Co, and ^{152}Eu. The recognition accuracy of the proposed model with these radionuclides are 99.0%, 98.0%, 99.0%, 97.5%, and 98.5%, respectively. The model still recognizes the training isotopes with the lowest accuracy of 89.0% for spectra with the displacement in the range of 20%.

Keywords: Artificial neural network, plastic scintillation detector, nuclide identification algorithms.

I. INTRODUCTION

Radiation portal monitors (RPMs) are high-sensitivity radiation detection devices, often deployed at airports, seaports, and borders to detect individuals and vehicles illegally transporting sources and special nuclear materials (SNMs). RPMs are usually equipped with large-volume gamma and neutron detectors that increase the ability to detect radiation sources with low activity. Due to their advantages, including fast rise and decay time, high optical transmission, ease of manufacturing, low cost, large available size, and high durability, plastic scintillation detectors are now the most widely utilized gamma detector on RPMs, but due to the low Z-number components of the material and low density, the poor energy resolution shows the broad energy spectrum without clear photo-peak identifying the radionuclides. With limited spectral information, most RPMs utilize these detectors utilized conventional gross-count (GC) or energy windowing (EW) algorithms to detect radioactive sources. By comparing the gross counting rate to the background counting rate, the GC algorithm may identify the existence of radioactive sources, but it cannot discriminate between different types of radioactive sources. Based on their emission energy, the EW algorithm evaluates the counting rate ratios between two or more regions in the spectra to determine whether radioactive sources are natural or artificial. The capacity to identify radioisotopes from plastic-based spectrometers, on the other hand, is severely limited.

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Machine learning and deep learning algorithms have advanced significantly in the last decade and are now used in a variety of sectors, including nuclear physics and engineering. Examples of such applications include (i) nuclear power plant health and management (ii) radiation protection (ii) robotic and control system optimization. Model selection is based on the complexity of the training dataset, the required output of the problem, and the performance of the computational hardware. For radioisotope identification tasks, many learning-based methods have been presented in the literature [1]–[11]. The authors of [1] introduces a simple neural network for identifying automatically radiation spectra obtained in NaI(Tl) scintillation detector and Ge(Li) semiconductor detector. This approach can determine a given isotope presented in a mixture of elements as well as the relative proportion of each element by using Linear Associative Memory model trained by combination of a set of well-known spectra. However, this artificial neural network (ANN) is only applicable to high-resolution spectroscopy system (HPGe or equivalent), needing high-precision standard radiation sources in terms of activity to ensure recognition accuracy, also high requirements for energy calibration, detection efficiency, and measurement geometry during model training and validation. The authors of [2] has introduced a fast isotope recognition algorithm applied on the RPMs based on the combination of the Bayesian algorithm and spiking neural network. However, the model in this study is applicable to well-type NaI(Tl) detector, which are difficult to apply on the low-resolution detector, i.e., the plastic scintillation detector. The authors of [7] investigated the first application of the ANN model in the analysis for spectra from polyvinyl toluene (PVT) scintillation detector in RPMs, but this study still faces many difficulties in distinguishing NORMs isotopes from illegally transported radioactive materials. Recently, the authors of [11] built a radioisotope warning algorithm applied on PVT-based RPMs. There are two steps in the alarm algorithm: (i) Generate an alarm based on the conventional GC algorithm (ii) Process the spectrum in step 1 by using an ANN to identify radioisotope groups and reduce the probability of a nuisance alarm. The model was validated on eight radioisotopes, although they were only split into three separate groups based on their feature vectors. The approach requires extensive pre-processing of the input data; however, as the number of isotopes grows, particularly those with similar feature vectors, the problem gets complex, and grouping the isotopes becomes challenging. On a 2”×2” EJ-200 detector, the authors of [7] constructed a 3-layer fully connected ANN for simultaneous identification of multiple radioisotopes from PVT scintillation detector. Although this ANN achieves good accuracy, however, labeling too many classes (16 labeled for the 4 radioisotopes in training data set) might be challenging as the number of isotopes increases.

This paper presents a full-connected ANN to identify individual as well as mixture of radioisotopes in gamma spectra obtained in a large volume EJ-200 PVT scintillation detector. The isotopes used to train the model includes: ^{241}Am, ^{133}Ba, ^{137}Cs, ^{60}Co, ^{152}Eu. These isotopes were selected based on homeland security and emitted energies that range from 60 keV to 1400 keV. The input data of the ANN is a 1024 - channel normalized spectra. In addition, the model has ability to accurately identify isotopes in spectra with the relative peak positions shifted within ±10%.

II. MATERIAL AND METHODS

A. Artificial neural network

An artificial neural network is the piece of a computing science designed to simulate the

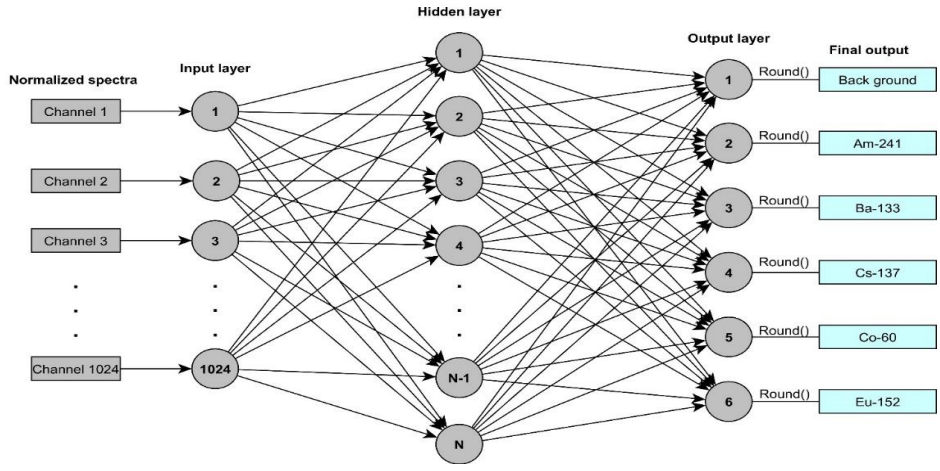

Fig. 1. The structure of the proposed ANN

way the human brain and processes information. It is the foundation of artificial intelligence (AI) and solves problems that would prove impossible or difficult by human or statistical standards. ANNs have self-learning capabilities that enable them to produce better results as more data becomes available. Mathematically, ANN is a model that attempts to map an arbitrary function from $\mathbb{R}^M$ to $\mathbb{R}^N$ where M and N are any integer. ANN accomplishes this by mimicking biological neurons. The structure of an ANN consists of an input layer, one or more hidden layers, and an output layer. Each neuron operates by summing the products of the previous layer values and each individual weight connection nodes. The value at one neuron before being passed to the next layer is transformed through a nonlinear function called the activation function, typically sigmoid, ReLU, Tanh, and etc. There is no method that can directly determine best-performing hyperparameters of the model. These parameters can be achieved by design an automated search to test different network configurations. Some popular search strategies include: random search, grid search, heuristic, exhaustive.

In this paper, an ANN with the most basic structure in the form of multi-layer perception (MLP) is introduced. The structure of this ANN is illustrated in

Fig. **1** with 1024 neurons in the input layer, N neurons in the hidden layer and 6 neurons in the output layer corresponding to 6 radioisotope classes. The output of the hidden layer or the output layer layer is computed as in Eq.1:

$$a_i^{(l)} = f\left(\sum_{j=1}^{m} w_{ji}^l a_j^{(l-1)} + b_i^l\right) \qquad (1)$$

$a_i^{(l)}$ is the output values of the hidden layer or output layer; w_{ji}^l are the weights of the hidden/output layer; b_i^l is bias of input layer or output layer; m is the number of nodes in the hidden layer or the output layer. In this study, the ANN model choose *ReLU* and *sigmoid* as activation functions for the first and the second layer, respectively. The *ReLU* and *sigmoid* activation functions are described as follows:

$$ReLU(x) = \max(0, x) \qquad (2)$$

$$sigmoid(x) = \sigma(x) = \frac{1}{1+e^{-x}} \qquad (3)$$

The result at the output layer is rounded to two binary values of 0 and 1 corresponding to the presence or absence of that isotope on the input spectrum.

B. Data training set creation

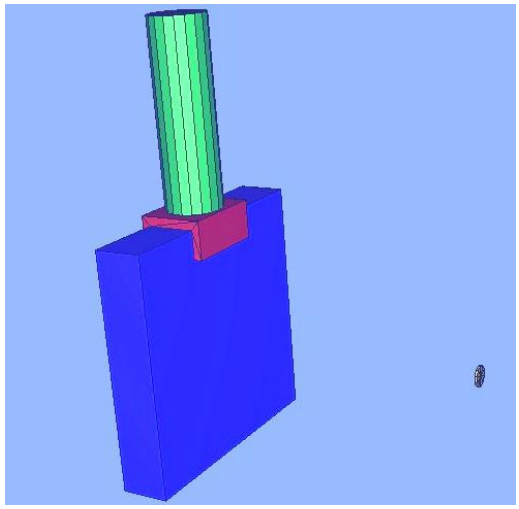

Fig. 2. The simulation model of plastic scintillation detector and radiation source

The training data set for this proposed model was generated in two ways: experimental measurement and MCNP simulation. The radioisotopes used for training model are include ^{241}Am, ^{133}Ba, ^{137}Cs, ^{60}Co, and ^{152}Eu. The simulation model of the PVT detector is based on the dimensions given by the manufacturer. The source-to-detector distance was fixed to 30 cm; the FT8 GEB (Gaussian broadening) card is investigated and applied to minimize the discrepancy between the simulated and the measured gamma spectra. The number of simulated particles is randomly selected in the range of $10^4 \div 10^6$ particles. Particularly, the background spectrum used in the model training process were measured by the EJ-200 detector with a random interval between 1 ÷ 30s.

The compton scattering region from the simulated spectrum will be substantially lower than the actual measured spectrum since the MCNP simulation model does not include materials surrounding the detector. To minimize the disparity, 30 mm thick aluminum plates are put around the detector to imitate dispersion from the environment. In Figure 3, the experimental

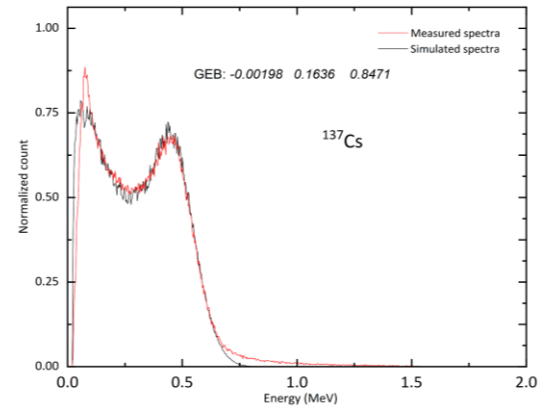

Fig. 3. Measured and simulated gamma spectra of ^{137}Cs radiation source with corresponding Gaussian broadening parameters

measurement spectrum and the simulated spectrum using ^{137}Cs radiation source are displayed. As shown in the Figure 3, the peak

position and Gaussian expansion of the calculated MCNP spectrum and the measured spectrum quite identical. After placing aluminum plates around the detector, the low-energy scattering region is also matched. The simulated spectrum does not contain count in the area with energy greater than the maximum emitted energy, however the count can still be seen in this region in the experimental spectrum due to the pulse pile-up. The contribution of this energy region to the overall spectrum, however, is negligible.

After the measurement, the gamma spectrum is chosen at random and merged into

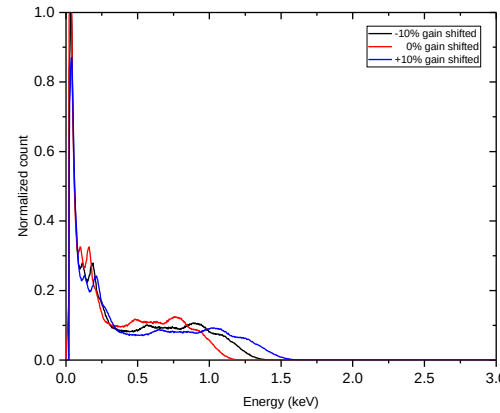

Fig. 4. Gamma spectra of ^{60}Co and ^{152}Eu source with different gain shift

classes of 2, 3, 4, and 5 isotopes. The source-to-background count rate ratio are adjusted at 1:1 and 1:2, respectively. The spectrum is normalized by dividing count rate in each channel to the maximum count rate. In addition, 2000 spectra with gain shifts in the 10% range were created for training ANN model. The gain adjusted spectra are shown in the Figure 4.

C. Validation and test data set creation

To create the validation and test sets, the gamma spectra were measured all combination of radioisotopes. For the test set, the radioactive check sources were placed 1 cm, 5 cm and 10 cm away from the detector (referred to as 1 cm away, 5 cm away and 10 cm away, respectively) and measured from 1s to 10s with intervals of 1s. This procedure was repeated 3 times. Therefore, there were 3000 spectra with 1024 channels in the test set. For the validation set, the spectra were generated for only the 10-cm-away case, and a spectrum of 1024 channels was extracted in a manner identical to that described in Section 2.2. The number of spectra in the validation set was 1440. The experimental setup to create training dataset is depicted in Figure 5. The radioactive source and detector are mounted to holders with a fixed source-to-detector distance. The signal from the detector is amplified and analyzed on the self-developed digital multichannel analyzer (DMCA)[12]. The radiation spectrum from the DMCA is transmitted to the PC for analysis and storage.

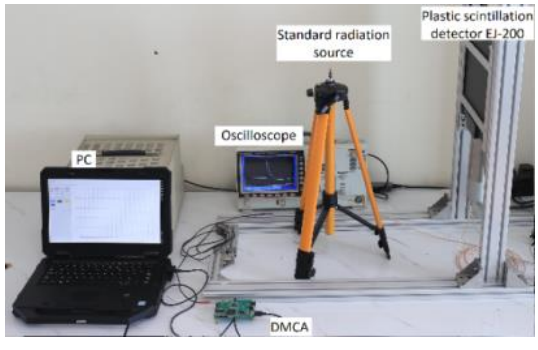

Fig. 5. Experimental set-up for the gamma spectra measurements

D. ANN model evaluation

In machine learning, Optimizers are algorithms or methods used to change the attributes of the neural network such as weights and learning rate in order to reduce the losses. The optimizer functions are selected based on the accuracy of the model after the first 1000 iterations. The optimizers ADAgrad, ADAdelta, and FTLR have very poor convergence during training, as seen in Figure 6. SGD and Adammax optimizers have average convergence, whereas RMSProp, Nadam, and Adam optimizers have the best convergence. In which RMSProp optimizer converges well for iterations under 500, while Nadam and Adam provide higher accuracy as the number of iterations grows. The model in this study used Adam to update all model parameters.

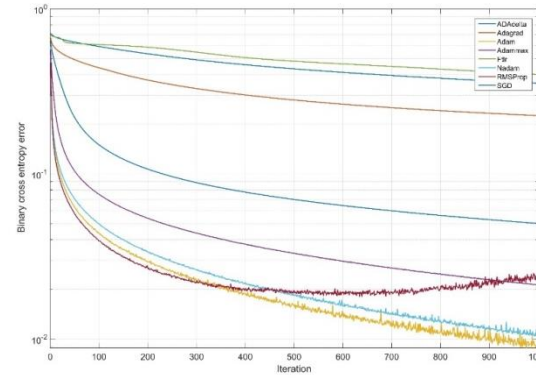

Fig. 6. Cross entropy loss comparison according to different optimizer

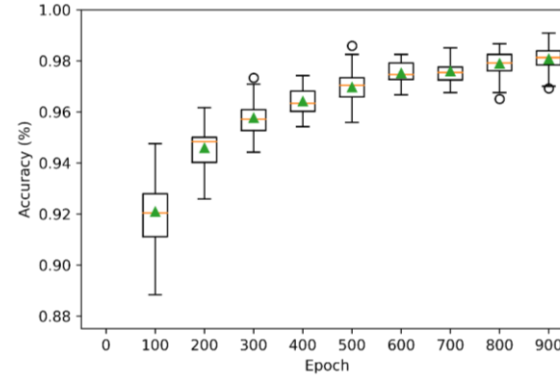

Fig. 7. Accuracy of the proposed ANN versus repeated k-fold cross validation (k=10, n_repeated = 5)

Repeated k-fold cross-validation provides a way to improve the estimated performance of a machine learning model. The k-fold cross-validation procedure divides the dataset into *k* non-overlapping folds. A total of k models are fit and evaluated on the *k* hold-out test set. The cross-validation procedure is repeated multiple times (referred to as *n_repeated* parameter) and the mean result across all folds from all runs is reported. This mean result is expected to be a more accurate estimate of the true unknown underlying mean performance of the model on the dataset, as calculated using the standard error.

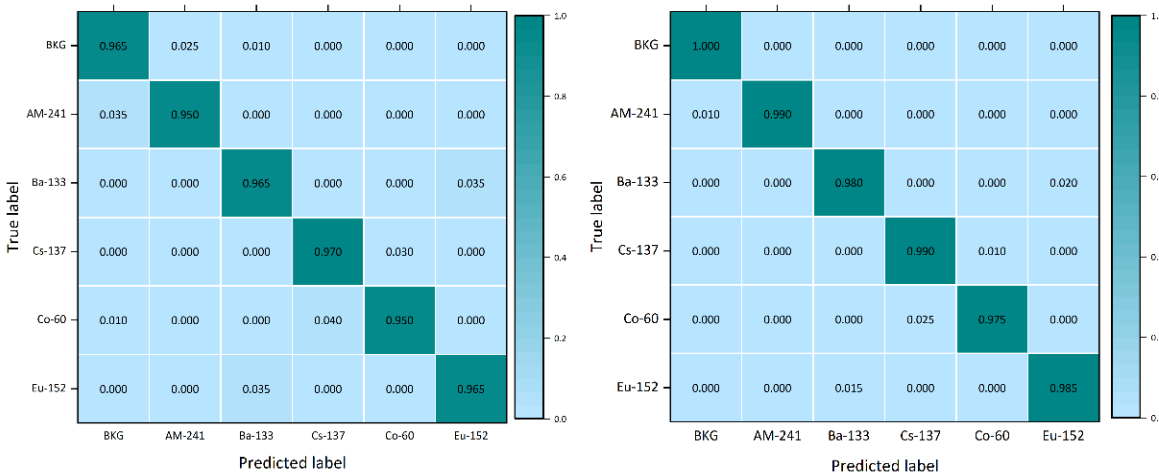

Fig. 8. Accuracy of the proposed ANN versus repeated k-fold cross validation (k=10, n_repeated = 5)

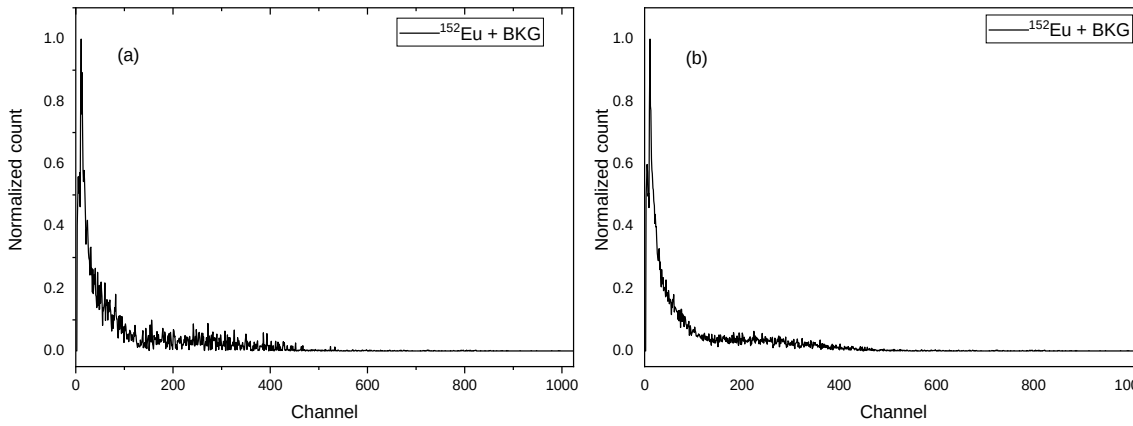

Fig. 9. ^{152}Eu gamma spectra with the source-background count rate ratio of (a) 1:2 and (b) 1:1

In the k-fold cross validation method, *k* is the most important parameter. In this study, the value of k was fixed at 10, and *n_repeated* was fixed at 5, commonly used values and proven to give small error, low variance. The accuracy of the mode on the data set are reported in the Figure 7, it can be seen that the model has high accuracy (~90% at 50 iterations, ~97% at 5000 iterations and 99% at 10000 iterations) with low variance.

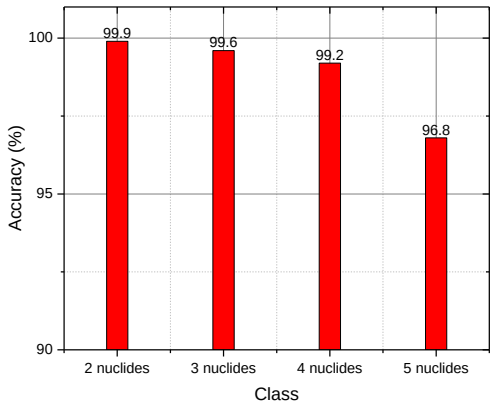

Fig. 10. Accuracy of the proposed ANN versus repeated k-fold cross validation (k=10, n_repeated = 5)

III. RESULTS

A. Single isotope identification

In order to evaluate the performance of the model in identifying single-radioisotope, two normalized confusion matrix with the source to background ratio equal to 1 and 0.5 are presented in the Figure 8.

ANN model is validated with 200 gamma spectra in each case. An example of the ^{152}Eu isotope spectrum with different count rate ratios is shown in Figure 9.

From the above 2 confusion matrices, it can be seen that: in the case of low count contribution from the source, the probability of isotope detection is lower than in the other case. With a count rate ratio of 1:2, the probability of detecting ^{241}Am isotope is the lowest with 95%, 5% false negative of ^{241}Am source falls into the cases of background prediction. This can be explained by the fact that the background spectrum and the ^{241}Am source spectrum are quite similar, especially when the count rate contribution from the ^{241}Am source is smaller than that of the background. The other sources in the training set have an accuracy rate of 96.5 ÷ 97.0%.

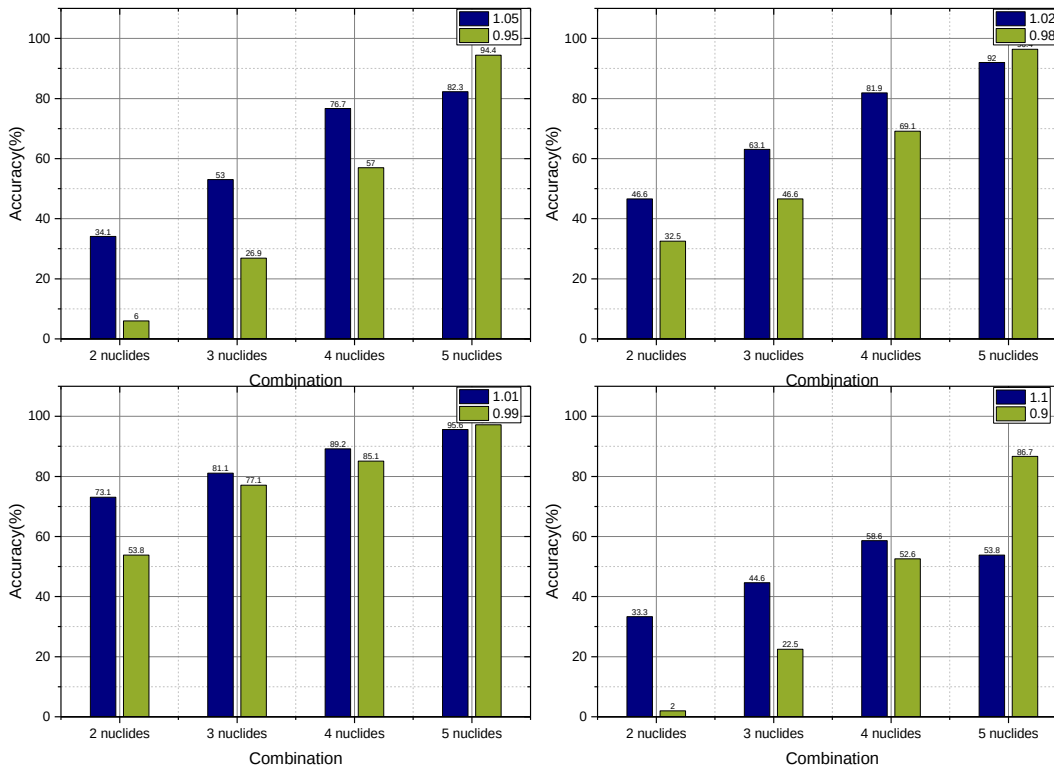

Fig. 11. Accuracy of the proposed ANN with excluding shifted spectra in the training data set

In this case that the count ratio of the source to the background is 1:2, the probability of detecting the correct isotope ranges from 98 ÷ 99%. For the background, the rate of correct identification is 100%. Pair of isotopes with probability of misidentification include ^{137}Cs and ^{60}Co, ^{152}Eu and ^{133}Ba. This occurs with isotopic pairs with similar emission energy leading to similar spectral shapes, especially when the count rate of the radiation source is low.

B. Mixture isotopes identification

In order to evaluate the effectiveness of identifying isotopes mixtures, the accuracy of the model when evaluated the mixture of radioisotopes are investigated. Each combination of 250 spectra has the contribution proportions of randomly selected isotopes. The accuracy of identification with these combinations is shown in Figure 10, Figure 11. From Figure 10,**Error! Reference source not found.** it can be seen that the accuracy of the model decreases with increasing number of isotopes present in the spectrum. When many isotopes are present at the same time, it becomes more difficult to accurately identify all the isotopes because the spectral features will be dominated by the isotopes with a large contribution ratio.

The gamma spectrum obtained on the scintillation detectors is strongly dependent on the ambient temperature, especially in outdoor measurement [12]. Therefore, the recognition model needs to be verified with gain-shift gamma spectra. In this study, the model is also verified with spectra that has a gain shift up to

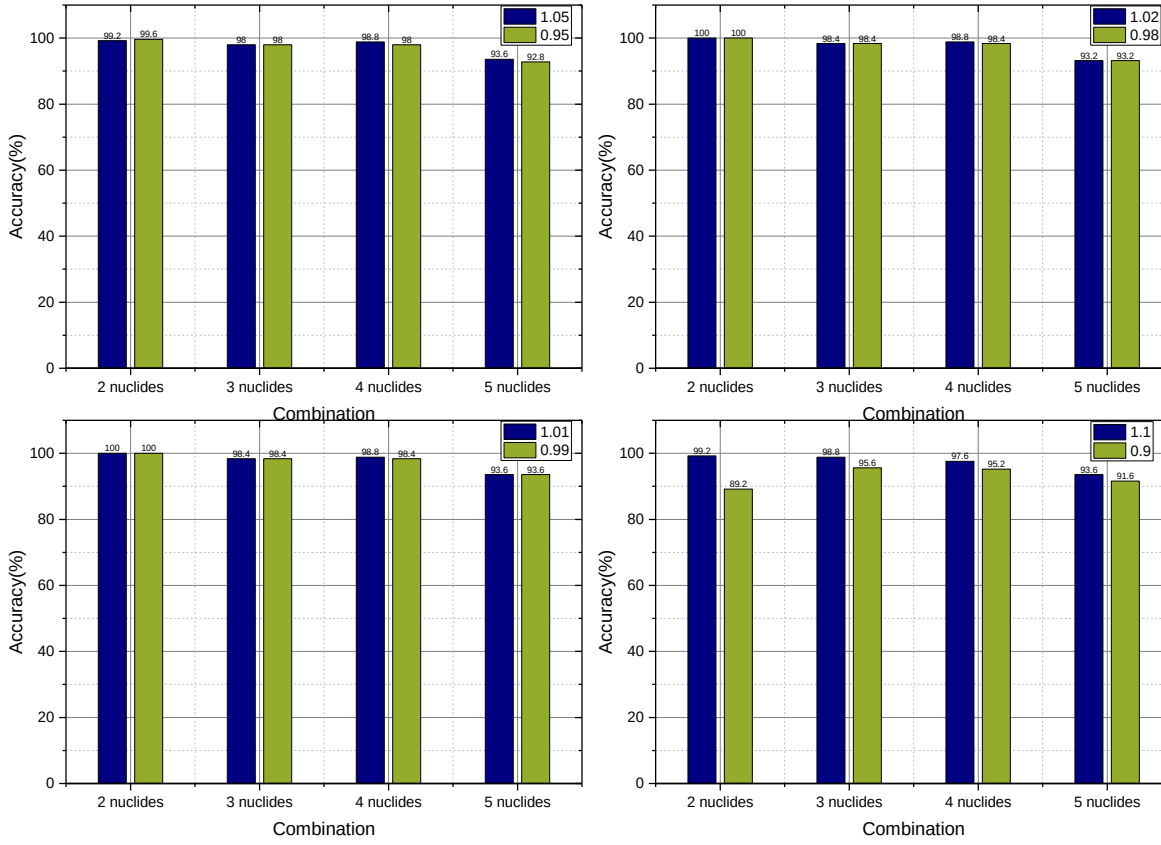

Fig.12. Accuracy of the proposed ANN with different gain shifted gamma spectra

±10%. Accuracy values were investigated at ±1%, ±2%, ±5%, ±10% gain drift and shown in the Figure 11. From Figure 11, it can be seen that the accuracy of the model decreases with the larger gain shift, and with the decrease in the number of isotopes appearing in the spectrum.

This can be explained that the ANN is trained only with the well-calibrated training data set. When changing the gain factor by +10%, the accuracy of radionuclide recognition with a mixture of 2 isotopes was only 33%, and 2% when the -10% gain shift is applied. The accuracy for the mixtures of 3, 4, and 5 isotopes are 44% to 86%. This accuracy reduction due to the ANN lacks training on shifted gamma spectra. To overcome the above limitation, 500 spectra with gain shift randomly selected in range of 10% are added to the training data set. The accuracy of the ANN after retraining are shown in Figure. 12. On Figure. 12, it can be seen that the accuracy with isotope combinations is increased to 93.2 ÷ 98.5% with the gain variation up to 10%.

This study proposed an ANN-based nuclides identification algorithm that applied on PVT scintillation detection. Different from work [8], this study presents a method that is well applicable to scintillation detectors with poor energy resolution. This simple approach can overcome the limitations that arise from the conventional gross-count algorithm or energy-weighted window algorithm.

III. CONCLUSIONS

This study presented an ANN model that successfully identifies single/mixtures radioisotopes on low-resolution PVT-based spectrometer. The hyperparameters are optimally selected for the training data set. The proposed model has high accuracy when identifying with a combination of one or more radioisotopes, even when the spectrum has a gain shift up to 10%. This study has confirmed the application of deep learning models in the practical problem of isotope identification on a low-resolution gamma spectrometer.

Some aspects of radionuclide recognition algorithm for plastic scintillation detector can be further explored based on this work. The number of input data dimensions is still high (1024 for this study), so dimension reduction methods need to be applied to reduce the model architecture and optimize training time. In addition, the number of considered isotopes also needs to be increased to improve the model's applicability.

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