A methodology for alpha/beta particles identification in Liquid Scintillation using a three-channel Convolutional Neural Network

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Abstract

To mitigate dark pulses and achieve low-background-level measurements, liquid scintillation counters (LSCs) are generally equipped with two or three photomultiplier tubes (PMTs) for coincidence measurements. However, traditional identification methods in LSC only utilize the anode pulse from a single PMT to identify / particles, which limits their ability to identify particles. We developed a three-channel convolutional neural network (TCNN) model, which integrates pulses from three PMT anodes to identify particle categories. Anode pulses are organized into a shape of (3,512) and subsequently fed into the TCNN for / pulse discrimination. To train and validate the TCNN, we prepared two samples: a 241Am sample as an alpha emitter and a 90Sr/90Y sample as a beta emitter. In the validation set, TCNN performed significantly better than traditional convolutional neural networks (CNN) in identifying / pulses, achieving accuracy, recall, and F1 score of 99.44%, 99.23%, and 99.34%, respectively. We also prepared a mixed-emitter sample exhibiting a beta activity of approximately 172 Bq and an alpha activity of 98 Bq to evaluate the impact of TCNN on spectral performance in practical applications. First, the category of pulses from the sample is identified by the TCNN, and then their amplitude is recorded in an -MCA spectrum or -MCA spectrum according to the identified category. The alpha particle peak in the -MCA spectrum is used to evaluate spectral performance. The optimal detection limit for the alpha particle peak is 0.3337 cps, which shows a sensitivity increase of 31.16% compared to the CNN method. This indicates that TCNN can effectively utilize three-channel pulses to enhance the ability to distinguish between alpha and beta particles when analyzing both simultaneously, thereby significantly improving the sensitivity of the detector.

Full Text

A Methodology for Alpha/Beta Particle Identification in Liquid Scintillation Using a Three-Channel Convolutional Neural Network

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Abstract

Liquid scintillation counters (LSC) are generally equipped with two or three photomultiplier tubes (PMT) for coincidence measurements to mitigate dark pulses and achieve low-background-level measurements. However, traditional identification methods in LSC only utilize the anode pulse from a single PMT to identify alpha/beta particles, which limits their discrimination capability. We developed a three-channel Convolutional Neural Network (TCNN) model that integrates pulses from three PMT anodes to identify particle categories. Anode pulses are organized into a shape of (3,512) and subsequently fed into the TCNN for alpha/beta pulse discrimination. To train and validate the TCNN, we prepared two samples: a $^{2\ 1}$Am sample as an alpha emitter and a Sr/ Y sample as a beta emitter. In the validation set, the TCNN performed significantly better than traditional convolutional neural networks (CNN) in identifying alpha/beta pulses, achieving accuracy, recall, and F1 scores of 99.44%, 99.23%, and 99.34%, respectively. We also prepared a mixed-emitter sample exhibiting a beta activity of approximately 172 Bq and an alpha activity of 98 Bq to evaluate the impact of the TCNN on spectral performance in practical applications. First, the category of each pulse from the sample is identified by the TCNN, and then its height is recorded in an alpha-MCA spectrum or beta-MCA spectrum according to the identified category. The alpha particle peak in the alpha-MCA spectrum is used to evaluate spectral performance. The optimal detection limit for the alpha particle peak is 0.3337 cps, which shows a sensitivity increase of 31.16% compared to the CNN method. This indicates that the TCNN can effectively utilize the three-channel pulses to distinguish between alpha and beta particles when analyzing both simultaneously, thereby significantly improving the sensitivity of the detector.

Keywords: Liquid Scintillation Counter · Convolutional Neural Network ·

Particle Identification · Simultaneous Alpha/Beta Analysis

1 Introduction

The measurement of radionuclides is critically important for various fields, including geochemistry, environmental monitoring, nuclear power generation, marine environment studies, and health physics. For example, assessing the gross activity of alpha and beta particles in water is essential to ascertain its suitability for commercial utilization [1]. Detecting the gross activity of alpha and beta particles in urine samples from populations suspected of internal contamination is highly valuable for treatment and prevention [2]. Radionuclides from uranium decay chains have been widely used as tracers for various processes in the marine environment [3]. Determination of $^{2\ 1}Pu$ in materials from nuclear facilities and environmental samples is an essential issue for radiation protection of workers and members of the public [4]. These applications all involve quantifying the activity of alpha or beta particles.

Liquid scintillation counting (LSC), characterized by excellent counting efficiency and minimal self-absorption, is appropriate for quantifying beta emitters and short-range alpha particles [5-6]. The decay energy of beta particles varies from 0 to 3 MeV, whereas the decay energy of alpha particles spans from 4 MeV to 7 MeV. The efficiency of fluorescence emission from scintillation liquid excited by alpha particles is approximately one-tenth of that excited by beta particles. Consequently, the pulse height spectrum produced by alpha particles may overlap with that produced by beta particles, presenting significant challenges for the simultaneous analysis of both particle types. Fortunately, Pulse Shape Discrimination (PSD) technology has been proposed to solve this problem.

In organic scintillators, alpha particles possess greater stopping power compared to beta particles and produce higher quantities of delayed fluorescence light. Therefore, the alpha pulse output from the photomultiplier tube (PMT) exhibits a greater density of slow components. This property is essential for employing Pulse Shape Discrimination (PSD) technology to differentiate between pulses. In the early days, the practical PSD methods were Charge-Integration (CI) [7-8] and Zero-Crossing (ZC) [9-11]. Although these methods were originally proposed for neutron/gamma-ray discrimination, they are also applicable to alpha/beta particle discrimination. The tail-to-total charge ratio, which serves as the separation parameter for CI, is used to interpret the pulse category. The advantage of a simple algorithm structure allows it to be deployed in Field Programmable Gate Array (FPGA) for real-time measurement. In ZC, the zero-crossing time of shaped pulses from differential circuits is utilized to categorize the pulses. This technology is relatively easy to implement in analog circuits, but its resolution is slightly inferior to CI.

With the rapid development of digital and communication technologies, pulses are digitized and stored on personal computers. To achieve higher measurement accuracy, many intelligent algorithms have been proposed to handle offline digital pulses. Some frequency domain methods, which are less sensitive to high-frequency noise, have been proposed to identify neutron/gamma-ray events in high counting rate environments. These mainly include frequency gradient analysis [12], the Fourier Transform Method [13], and the wavelet transform method [14].

The K-means clustering algorithm [15], which does not require manual labeling, has been used to identify pulses. This method consumes less memory but has poor accuracy. Artificial neural network methods, such as Multi-Layer Perceptron (MLP) [16-17] and CNN [18], exhibit higher discrimination ability than traditional methods, especially thanks to their considerable advantages in extracting image textures and details, including local connection, parameter sharing, and adaptive learning. These characteristics enable them to demonstrate exceptional accuracy in particle identification. Furthermore, Recurrent Neural Network (RNN) [19] and Residual Network (ResNet) [20] have been introduced to differentiate between neutron, gamma-ray, and piled-up pulses.

Many scholars have applied some of these methods to identify alpha/beta particles in liquid scintillators [21-23]. Recently, an ANN algorithm designed to minimize computational resources has been deployed to ARM microprocessors for real-time identification of alpha and beta pulses in LSC [24].

The structural and operational methods of LSC used for alpha/beta measurements differ significantly from detectors used for neutron/gamma ray measurements. In LSC, the sample added to the scintillation liquid reduces the fluorescence yield through chemical quenching or delayed quenching [25]. When employing intelligent algorithms to identify alpha/beta pulses, it is necessary to fully evaluate whether the trained model maintains advanced performance in practical applications. In structural design, three PMTs are symmetrically arranged around the optical chamber of the LSC. The LSC can employ coincidence measurement techniques to eliminate dark pulses and achieve low background level measurements. Simultaneously, the LSC can execute the Triple to Double Coincidence Ratio (TDCR) method for activity standardization [26-27], which is used by national metrology institutions across numerous countries. These PMTs simultaneously measure fluorescence emitted from the scintillator stimulated by a radiation event. Except for fluorescence absorbed during transmission, the remainder randomly enters the three PMTs and strikes their cathode coatings to generate photoelectrons. Subsequently, the photoelectrons are amplified by the PMT, generating detectable pulses at their anodes. Therefore, the shape of the anode pulse exhibits greater complexity, including time misalignment, pulse disappearance, and significant changes in the proportion of post-pulses. Conventional techniques utilize only one anode pulse for identification, simplifying device architecture and mitigating the complexity of implementing the identification algorithm in hardware. However, the absence of the other two anode pulses results in incomplete waveforms that markedly reduce the performance of conventional identification algorithms. This paper proposes a three-channel convolutional neural network model (TCNN) that can integrate the waveforms of the three anode pulses from three PMTs to identify alpha and beta particles.

The remainder of this paper is organized as follows. Section 2 describes the LSC system, sample preparation, pulse shape characteristics, and evaluation methods for particle identification algorithms. Section 3 delineates the architecture of the TCNN model, the processes of data acquisition and preprocessing, as well as the training of the model. In Section 4, we employ the TCNN model for the simultaneous analysis of alpha/beta particles and evaluate its practical value. Finally, Section 5 summarizes the conclusions of this study.

2 Detection System and Pulse Shape Analysis

2.1 3-PMTs Measurement System

The LSC used in this article was developed by Sichuan X-START Technology (M&C Co. Ltd). The instrument mainly comprises essential elements such as a lead shielding chamber (SC), a scintillation chamber (SCC), a signal conditioning circuit module (SCM), a data acquisition unit (DAU), and a data terminal. Figure 1 [Figure 1: see original paper] presents a partial structural diagram.

The SC, with a wall thickness of 10 cm and constructed from high-purity lead, was designed to reduce background radiation. The SCC is installed inside the SC. Signal and power cables are led out through through-holes on the side of the SC, each equipped with a specialized Pb plug to maintain the integrity of the shielding.

The SCC is made of pure copper material. Three PMTs are symmetrically installed on its side. A glass vial containing radionuclides and 15 ml of liquid scintillation cocktail is positioned at the geometric center to maximize photon collection efficiency. The geometric center can be accessed through the top through-hole in the SCC. Current signals from the PMTs are transmitted to the SCM through a 2 m coaxial cable.

These weak electric current signals are converted into amplified voltage pulses in the SCM via a preamplifier characterized by low noise and high bandwidth capabilities. Initially, these amplified pulses, also known as fast pulses, are digitized via a data acquisition (DAQ) board. Subsequently, particle type identification is conducted using digital PSD. Concurrently, the pulses are processed through summing circuits to ascertain the average amplitude, which is essential for measuring particle energy. Due to the short peaking time of the input pulses, the summed pulses suffer from significant ballistic deficits. To address this issue, an RC shaping circuit with a decay time constant of 750 nanoseconds is utilized to mitigate the effects of the ballistic deficit, although this results in a reduction in pulse amplitude. After shaping, the pulses are further amplified by another amplifier to ensure they fall within the amplitude range that can be measured by the ADC on the DAQ board before being routed to the DAQ board for further processing.

The DAU comprises four data acquisition (DAQ) boards. Each DAQ board is integrated with a Xilinx Zynq-7020 SoC, AD9434 ADC (500 Msps, 12 bits), and other components. Boards 1, 2, and 4 are used for digital fast pulses. Board 3 is used for digitizing summation pulses. Upon detecting a new pulse, DAQ board 3 immediately notifies the other DAQ boards to concurrently record the pulse. Simultaneously, the board produces a unique identification code and sends it to the other DAQ boards. This identification code is recorded in the last sequence of the pulse to identify the correlation of the pulses. In the experiment, the data depth was set to 2048 (4096 ns) and the delay depth was set to 450 (900 ns). This configuration fully preserves the pulse baseline and provides sufficient information for digital algorithms. Finally, all recorded pulse data is transmitted to the data processing terminal via Gigabit Ethernet for storage and analysis.

2.2 Sample Preparation

We conducted alpha/beta discrimination experiments based on two radioactive nuclides, $^{2\ 1}$Am and Sr/ Y. $^{2\ 1}$Am decays to ^{23} Np through alpha-particle emission, with a principal alpha energy of 5.486 MeV (84.5% branching ratio). This decay process has a half-life of 432.2 years. In the beta decay process, Sr disintegrates into Y, an antineutrino, and a beta particle, with a half-life of 28.79 years. The energy of the released beta particles is continuously distributed, ranging from 0 to 0.546 MeV. Y is an unstable intermediate with a half-life of 64.053 hours. It decays to stable Zr by emitting beta particles. The energy range of the emitted beta particles is from 0 to 2.28 MeV. The half-life of Y is much shorter than that of Sr, and the activity of these two radioactive nuclides in the prepared sample has reached equilibrium.

Other materials used in the experiment include Ultima Gold LLT scintillation cocktail, 20 mL glass vials, an electronic balance scale, rubber bulb pipettes, and a bottle-top dispenser. The Ultima Gold LLT is developed by PerkinElmer and has various advanced performance characteristics, including low background count, high light output, chemical stability, and efficient alpha/beta particle recognition.

We prepared three samples for the experiment: two single-emitter samples and one mixed-emitter sample. (1) An alpha-emitter sample consisting of 15 ml LLT scintillation cocktail and 229 Bq of $^{2\ 1}$Am. (2) A beta-emitter sample consisting of 15 ml LLT scintillation cocktail and approximately 335 Bq of Sr/ Y. (3) A mixed-emitter sample consisting of 15 ml LLT scintillation cocktail, 112 Bq of $^{2\ 1}$Am, and approximately 172 Bq of Sr/ Y.

It is critical to strictly control both the activity of radionuclides and the volume of scintillation cocktail during sample preparation to ensure accuracy and reproducibility. Each sample vial must contain 15.00 ± 0.05 milliliters of Ultima Gold LLT. Before measurement, the sample vials need at least 2 hours in a dark room to effectively reduce chemiluminescence interference.

2.3 Analysis of Pulse Shape

A large category of practical organic scintillators can produce what is known as a -electron structure. The -electrons have two different energy level structures, namely singlet and triplet states. These structures enable excited organic molecules to return to their ground state through the emission of fluorescence, phosphorescence, or delayed fluorescence. The proportion of delayed fluorescence is related to the initial density of the triplet state into which the excited organic molecules fall. The higher the stopping power of charged particles, the higher the initial density of triplet states. Experimental results demonstrate that the delayed fluorescence produced by alpha particles exciting organic molecules is greater than that produced by beta particles. This is the fundamental principle of pulse discrimination.

In a PMT, photoelectrons are produced by the photoelectric effect when fluorescence is incident upon the cathode. Subsequently, the photoelectrons are amplified by the PMT, generating a detectable pulse at its anode. The pulse produced by beta particles is called a beta pulse, and the pulse produced by alpha particles is called an alpha pulse. A beta pulse and an alpha pulse are shown in Figure 2a [Figure 2: see original paper]. The width of a beta pulse is about 30 nanoseconds. The trailing edge of the pulse is smooth and has almost no post-pulse. Conversely, alpha pulses exhibit significantly longer durations, typically ranging from 200 ns to 400 ns. The trailing edge of the pulse is filled with pile-up post-pulses.

The waveforms of 100 alpha pulses are displayed in Figure 2b [Figure 2: see original paper]. The number of post-pulses fluctuates. Most pulses exhibit many post-pulses, while a few pulses exhibit very few or even no post-pulses. The waveforms of 100 beta pulses are displayed in Figure 2c [Figure 2: see original paper]. Only a small portion of pulses appear with a few post-pulses, while the rest have no post-pulses at all. The diversity of post-pulses limits the effectiveness of conventional PSD in practical applications.

The fast pulses from the three PMTs, produced by an identical event, are shown in Figure 2d [Figure 2: see original paper], labeled as CHA, CHB, and CHC, respectively. CHA, CHB, and CHC exhibit significant height variation, measuring 13 (a.u.), 198 (a.u.), and 86 (a.u.), respectively. CHA not only has a small pulse height but also a post-pulse. If we use the CI method to distinguish pulses, CHB and CHC will be identified as beta pulses, while CHA will be classified as an alpha pulse. This discrimination discrepancy can be explained by statistical fluctuations. The heights of the pulses fluctuate around a certain average value. The number of post-pulses also fluctuates randomly. These fluctuations are independent of each other and jointly influence the performance of PSD. Traditional methods only use pulses from one channel to identify particles. Abandoning the pulses from the other two channels limits the ability of these methods.

2.4 Evaluation Method

It is a binary classification problem to discriminate between alpha and beta pulses. Precision, recall, and F1-score are commonly used to evaluate classifier performance. The specific procedure for calculation is shown in equations (1)-(3).

Precision = Recall = 2×Precision×Recall F1Score = Precision+Recall

Where TP is True Positive, FN is False Negative, FP is False Positive, and TN is True Negative. In this paper, alpha pulse is defined as the positive class, while beta pulse is defined as the negative class. These classification indicators are used to evaluate the model in the validation set.

The pulses from liquid scintillation cocktails containing mixed-emitter samples cannot be manually labeled. For this situation, classification indicators cannot be used. The Figure of Merit (FoM) [28] can be used to evaluate classifier performance. The specific procedure for calculation is shown in equation (4).

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Where S is the distance between two Gaussian peaks, is the full width at half maximum (FWHM) of the alpha peak, and is the FWHM of the beta peak. The larger the FoM value, the better the classifier performance; otherwise, the worse.

The alpha particles produced by the decay of radioactive nuclides have characteristic energy, and alpha particle peaks are detectable in the energy spectrum. The detection limit can be used to evaluate instrument performance. The Currie criterion was used in this paper to calculate the Limit of Detection (LOD) [29] based on the statistical fluctuation of background counts.

where K = 1.645 is a normal distribution with 95% confidence, B is the total number of background counts, T is the measurement time, R is the chemical recovery fraction, is the efficiency fraction, and

3.1 Architecture of TCNN Model

The architecture of the Three-channel Convolutional Neural Network (TCNN) implemented in this article is shown in Figure 3 [Figure 3: see original paper]. The architecture consists of one input layer, five convolutional layers, four pooling layers, four activation functions, and one fully connected layer.

The shape of the input layer is (3,512), which consists of three pulses with a length of 512. In a convolutional layer, all neurons share a convolution kernel for computation. The size of the convolution kernel used by the TCNN is (Ci, Co, 1, 3), where Ci and Co represent the number of input channels and output channels of the convolutional layer, respectively. The pooling layer possesses no learnable parameters; it employs the principle of local image correlation to down-sample the feature map. This not only diminishes computational costs but also enhances the robustness of CNN to small positional changes. The pooling layer in the TCNN uses the maximum value for sampling. Consequently, the Rectified Linear Unit (ReLU) activation function is employed to nonlinearize the output of the pooling layer, thereby enhancing the model’ s expressive capacity.

The sliding step size is set to 1 in the last two convolutional layers and to 2 in the other layers. As the network depth increases, the number of channels in the feature maps progressively expands from 8 to 32. The feature map of the last convolutional layer is flattened into a 128-dimensional vector and further transformed into a two-dimensional vector through a fully connected layer to achieve binary classification.

The mathematical model of the TCNN is simplified to equation (5).

Where x is input data consisting of three fast pulses. Tcnn is the mapping function of the TCNN model. The z = (z0, z1) represents the predicted result and is also referred to as Logits. z0 and z1 are scalars that range from

During model training, z is converted into a probability distribution using the Softmax Function, as illustrated in equation (6).

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Where represents the probability of the input pulse being predicted as class m. m is 0 or 1. We train as predictive probabilities for alpha and beta pulses.

The Cross-Entropy Loss Function quantifies the model’ s prediction risk. The calculation formula is shown in equation (7).

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Where N is the number of input pulses, represents the true label of the i-th input pulse, and represents the probability of predicting the i-th input pulse as the m-th class.

The Adam optimizer is utilized to update the weight parameters. The learning rate (lr) is set to 0.2. L2 regularization is employed to prevent the weight parameters from becoming abnormally enormous. The regularization coefficient is set to 10 . During model training, excellent parameters are saved for further validation.

The trained model can be employed for identifying unlabeled alpha/beta pulses. First, we input pulses into equation (5) to obtain z = (z0, z1). The z0 represents the confidence score of alpha particles, and z1 represents the confidence score of beta particles. We can employ two distinct methods for classification.

The first method is the Score Comparison Method. This method directly compares the magnitude of z0 and z1. When z0 > z1, the pulse is interpreted as an alpha pulse. Otherwise, it is attributed to a beta pulse. This method is relatively simple and is suitable for evaluating performance in validation sets.

In practical applications, the Threshold Comparison Approach is advised for pulse discrimination. This method only uses Logits z0 or z1 for classification. For example, when z1 is used as a segmentation parameter, the larger the z1, the higher the probability that the pulse belongs to a beta pulse. Conversely, the higher the probability of it being interpreted as a non-beta pulse. In this article, non-beta pulses denote alpha pulses. The discrimination threshold is set to zT and has been previously provided. When z1 > zT, the pulse is interpreted as a beta pulse. Conversely, it belongs to an alpha pulse. The appropriate zT can be set according to specific needs. For example, the larger the zT is set, the higher the detection efficiency of alpha pulses. The smaller the zT is set, the higher the accuracy of the alpha pulse. In addition, we can also evaluate the performance of the model in identifying alpha/beta pulses by analyzing the distribution of z1.

Input → Conv1d stride=2 → Pool+Re → stride=2 → Conv1d stride=2 → Pool+Re → stride=2 → Conv1d stride=2 → Pool+Re → stride=2 → Conv1d stride=1 → Pool+Re → stride=2 → Conv1d stride=1

shape: (3,512) → (8,256) → (8,128) → (8,64) → (8,32) → (16,16) → (16,8) → (16,8) → (16,4) → (32,4) → (128)

Figure 3. The architecture of the Three-channel Convolutional Neural Network

3.2 Data Acquisition

The three samples prepared in Section 2.2 are measured by the LSC. The data terminal accepts pulses from the DAU and saves them to a file. A single-emitter sample collects 65,536 pulses. The pulses are equally split into a training set and a validation set. These two sets are employed for model training and validation, respectively.

In practical applications, alpha-emitters and beta-emitters are simultaneously added to liquid scintillation cocktails. The complex composition exposes the scintillator to more dangerous quenching effects. The increased quenching effect reduces the difference between alpha and beta pulses. The model' s capacity to correctly identify pulses produced by mixed-emitter samples is our primary concern. Consequently, we utilize pulses from the mixed-emitter sample as test pulses. It took a total of 115.14 seconds to collect 32,768 pulses.

The count rate of the alpha-emitter recorded by the instrument is nearly equivalent to its activity, given that its efficiency nears 100%. The activity of the alpha-emitter is 98 Bq. The total number of alpha pulses calculated in the test set is Na = 12,895. This value can be used to calculate the detection efficiency of alpha-emitters.

Preprocessing can mitigate the danger of gradient explosion or vanishing during training while enhancing model accuracy and generalization capability. The specific processing procedure is as follows.

Initially, the baseline, which is the mean of the data sequence within the non-pulse region, is subtracted from the pulses. The continuous 512 sequence within the pulse region is cropped out as input data for the TCNN model. In the input data, the starting position of the pulse has been adjusted to the 50th sequence. The maximum normalization technique is employed to transform the data values to the interval of (-1, 1). The normalized data from three PMTs is organized into a shape of (3, 512) suitable for input into the TCNN model.

3.3 Training

The model can learn rich features if it is provided with substantial and high-quality pulses. An adequate quantity of training epochs is crucial for ensuring model accuracy and generalization capacity. The impact of these two factors will be studied in this section.

We planned 11 different numbers of training sets, ranging from 4 to 65,536 pulses. The model is trained for 100 epochs in each training set. Subsequently, it is assessed using a validation set consisting of 65,536 pulses. The results are shown in Figure 5a [Figure 5: see original paper]. The more pulses used to train the model, the more accurate the model' s ability to identify pulses. Upon training the model with 32 pulses, the precision, recall, and F1 score are 0.9262, 0.9624, and 0.9440, respectively. When the number of training pulses is increased to 65,536, the precision, recall, and F1 score improve to 0.9953, 0.9916, and 0.9935, respectively.

The experimental results indicate that, despite a limited number of training pulses, the model maintains comparatively high accuracy in pulse identification. When the number of training pulses is sufficient, the error rate in identifying pulses is reduced to below 1%. Finally, we use 65,536 pulses to train the model, which can reduce the time investment in data collection while maintaining accuracy.

Figure 4b [Figure 4: see original paper] depicts the relationship between model accuracy and epochs in the validation dataset containing 65,536 pulses. After the first epoch, measurement accuracy can reach a relatively high level, attaining precision, recall, and F1 scores of 0.9942, 0.9356, and 0.9640, respectively. The accuracy changes slightly with increasing training epochs, and the error rate is less than 1%. At 80 training epochs, measurement accuracy yields optimal results, with precision, recall, and F1 scores of 0.9958, 0.9915, and 0.9936, respectively. Finally, the optimal model is applied to research in subsequent chapters to guarantee reliability and consistency.

precision recall F1-score

precision recall F1-score

Number of training pulses

Epochs

Figure 4. (a) The impact of the number of training pulses on the accuracy of the validation set; (b) The impact of epoch on the accuracy of the validation set

3.4 Comparison

The CI and CNN are widely employed conventional techniques for pulse type discrimination. The structure of CI is simple, so it can be deployed to FPGA for real-time pulse discrimination. The CNN can achieve higher discrimination accuracy and is extensively employed to discriminate offline pulses. A comparative experiment is conducted on the validation set. The results are listed in Table 1 . The accuracy of CI is minimal, with precision, recall, and F1 scores of 0.8708, 0.9438, and 0.9058, respectively. The accuracy of CNN is higher than that of CI, with precision, recall, and F1 scores of 0.9989, 0.9395, and 0.9682, respectively. The accuracy of TCNN is highest, with precision, recall, and F1 scores of 0.9958, 0.9915, and 0.9936, respectively. The F1 score of TCNN surpasses that of CNN by 2.54 percentage points. The result indicates that the TCNN method can significantly improve the ability to distinguish alpha pulses from beta pulses.

Table 1. The accuracy of different methods in the validation set

Methods	precision	recall	F1 score
CI	0.8708	0.9438	0.9058
CNN [18]	0.9989	0.9395	0.9682
TCNN	0.9958	0.9915	0.9936

4.1 Analysis of Pulse Shape Discriminator

We use the test set pulses prepared in Section 3.2 to evaluate model performance in practical applications. These pulses cannot be manually labeled. Therefore, we first analyze the distribution of the segmentation parameter z1 and then use the threshold comparison method to identify the pulses.

We input these pulses into the TCNN model and calculate the segmentation parameter z1. Statistical analysis of z1 yields a bimodal distribution curve, as illustrated by the dotted line in Figure 5a [Figure 5: see original paper]. Similarly, the result produced by CNN also shows a bimodal distribution, as indicated by the dashed line in Figure 5a. The Charge-Ratio distribution produced by CI is shown by the solid line in Figure 5a. We have translated and scaled these curves to ensure that the bimodal distribution falls within the range of 1-200. The left peak of the curve is a short and wide alpha peak, while the right peak is a high and narrow beta peak. We use Levenberg-Marquardt nonlinear least squares to fit the bimodal curve and then calculate the FoM factor. The FoM values are 0.72 for CI, 0.81 for CNN, and 1.65 for TCNN. The FoM factor of TCNN is 1.04

times higher than that of CNN. The TCNN significantly improves the ability to distinguish between alpha and beta pulses in practical applications.

We also extracted the height of the summed pulse and combined it with z1 to obtain a two-dimensional distribution (z1, height). The statistical results are shown in Figure 5d [Figure 5: see original paper]. Similarly, the two-dimensional distribution (z1, height) produced by CNN is shown in Figure 5c, and the two-dimensional distribution (Charge Ratio, height) produced by CI is shown in Figure 5b. In the two-dimensional distribution, isolated Gaussian peaks are derived from alpha pulses, while the rest of the distribution comes from beta pulses. The beta pulse exhibits a continuous distribution in the height dimension and a Gaussian distribution in the segmentation parameter dimension.

In Figure 5b, the resolution of the charge ratio for low-energy beta pulses is poor, which is the main reason why it is difficult for the CI method to distinguish between alpha and beta pulses. In Figure 5c, the resolution of z1 has been improved for beta pulses but has decreased for alpha pulses. The boundary of the z1 distribution between alpha and beta pulses is ambiguous, limiting the ability of CNN to distinguish them. In Figure 5d, the distribution of z1 from beta pulses is independent of pulse height and has very high resolution. The boundary of the z1 distribution between alpha and beta pulses is easily identifiable, which is very advantageous for TCNN to identify them.

Figure 5. Distribution of segmentation parameters from composite sample; (a) One-dimensional distributions produced by CI, CNN, and TCNN; (b) Two-dimensional distribution (charge ratio, height) produced by CI; (c) Two-dimensional distribution (z1, height) produced by CNN; (d) Two-dimensional distribution (z1, height) produced by TCNN

4.2 Simultaneous Alpha/Beta Analysis

We performed statistical analysis on the heights of 32,768 summation pulses from the test set to construct the energy spectrum. The statistical results are shown in Figure 6a [Figure 6: see original paper]. The Gaussian peak in the energy spectrum is the Alpha Particle Peak produced by the decay of $^{2\ 1}$Am. The background of the Alpha Particle Peak and the rest of the energy spectrum are generated by beta particles. In the energy spectrum, the beta particles increase the background of the alpha particle peak, adversely affecting the detection limit and measurement accuracy of the instrument. The counting of alpha particles also interferes with measurement of beta particle activity.

Another method of obtaining pulse height spectra involves using a classifier to identify pulses. Two pulse height spectra, also known as alpha-multichannel analyzers (MCA) and beta-multichannel analyzers, are used to record alpha or beta pulse heights, respectively. If the pulse is recognized by the classifier as an alpha particle, the height of the summation pulse is recorded in the alpha-MCA. Otherwise, it is recorded in the beta-MCA. We use CI, CNN, or TCNN to identify pulse categories, and subsequently record their heights into alpha-

MCA or beta-MCA. The statistical results are shown in Figures 6b, 6c, and 6d, respectively. The alpha-MCA spectrum has an alpha particle peak and a small background count of beta particles. The beta-MCA spectrum no longer shows the alpha particle peak, mainly comprising the height spectrum of beta pulses.

Figure 6. The spectrum from the composite sample; (a) The unseparated spectrum; (b) The separated spectrum generated by CI; (c) The separated spectrum generated by CNN; (d) The separated spectrum generated by TCNN

The correlation between alpha-MCA and beta-MCA spectra is so strong that either of these two spectra can be employed to evaluate instrument performance. In the alpha-MCA spectrum, the Alpha Particle Peak is easily recognizable from the background. Therefore, we use the alpha-MCA spectrum to evaluate detector performance. The net count of the Alpha Particle Peak divided by the calculated total count Na yields the recall. The net peak count divided by the total count from the alpha-MCA spectrum yields the precision. The F1 score is computed by substituting recall and precision into equation (3).

We set various discriminative thresholds around the valleys of the bimodal distribution shown in Figure 7a [Figure 7: see original paper]. The alpha-MCA spectrum is recreated, and relevant calculations are performed to obtain different precision, recall, and F1 scores. The precision-recall curve is illustrated in Figure 7a. The F1-score-recall curve is depicted in Figure 7b.

As the recall produced by the CI method rises from 0.8562 to 0.9168, the precision declines from 0.9168 to 0.8562, while the F1 score fluctuates between 0.8513 and 0.8649. The optimal F1 score is 0.8513. As the recall in the CNN approach increases from 0.9142 to 0.9578, the precision decreases from 0.9730 to 0.9495, while the F1 score varies between 0.9427 and 0.9536. The optimal F1 score is 0.9536. As the recall in the TCNN method increases from 0.9224 to 0.9553, the precision decreases from 0.9835 to 0.9252, while the F1 score varies between 0.9520 and 0.9637. The optimal F1 score is 0.9637, which is higher than the optimal value of CNN by 1.01 percentage points. Meanwhile, the recall and precision are 0.9528 and 0.9748, respectively.

The indicators that directly reflect detector performance are detection limit and background. The total count subtracted by the net count of the alpha particle peak yields the background. The LOD-recall curve and background-recall curve are illustrated in Figures 7c and 7d, respectively.

As the recall in the CI method rises from 0.8562 to 0.9168, the background increases from 17.38 cps to 26.12 cps, while the LOD fluctuates within the range of 0.4998 cps and 0.7807 cps. The optimal LOD is 0.4998 cps. As the recall in the CNN approach increases from 0.9142 to 0.9578, the background elevates from 2.84 cps to 3.12 cps, while the LOD varies between 0.4848 cps and 0.5937 cps. The optimal LOD is 0.4848 cps. As the recall in the TCNN method increases from 0.9224 to 0.9553, the background elevates from 1.72 cps to 8.64 cps, while the LOD varies between 0.3337 cps and 0.4738 cps. The optimal LOD is 0.3337 cps. Meanwhile, the recall and precision are 0.9591 and 0.9632, respectively. The detection sensitivity of $^{2\;1}$Am generated by the TCNN method is 31.16% higher than that generated by the CNN method. Moreover, at the same recall rate, the background level from the alpha-MCA spectra produced by the TCNN method is lower. This enables the detector to achieve higher efficiency and sensitivity in simultaneous alpha/beta analysis.

Figure 7. Analysis of alpha-MCA spectra from composite sample; (a) Precision-recall curve; (b) F1-score-recall curve; (c) LOD-recall curve; (d) Background-recall curve

5 Conclusion

This study aims to improve the accuracy and sensitivity of LSC in simultaneous alpha/beta analysis. A three-channel Convolutional Neural Network (TCNN) model is proposed. This technique identifies pulse types by integrating three pulses produced by LSC simultaneously.

We established a $^{2\;1}$Am sample, a Sr/ Y sample, and a composite sample of both $^{2\;1}$Am and Sr/ Y using Ultima Gold LLT scintillation cocktail, $^{2\;1}$Am source, and Sr/ Y source. Data acquisition was conducted on the LSC manufactured by X-STAR Technology Company. The pulses produced by alpha-emitter and beta-emitter samples are utilized to train and validate the model. During the training procedure, we performed experimental analysis on the epoch and pulse size. The experimental results indicate that the model trained with 32 pulses possesses significant discriminative ability. Utilizing 65,536 pulses to train the model over 80 epochs achieves optimal discriminating performance. We utilize the optimal model to identify pulse types in the validation set and compare it with conventional methods. The results demonstrate that TCNN possesses better capability to differentiate between alpha and beta particles relative to conventional techniques.

The TCNN model' s capability for simultaneous alpha/beta analysis was evaluated using the prepared composite sample. The segmentation parameter z1, produced by the model, is utilized to differentiate between alpha and beta pulses. Statistical analysis indicates that the segmentation parameters exhibit a bimodal distribution. The resolution of the alpha peak is lower than that of the beta peak. The FoM factor of the bimodal distribution was determined to be 1.62, which is 1.04 times superior to that of the CNN method. The results obtained from two-dimensional statistics (segmentation parameter, height) indicate that the pulse discrimination capability of the TCNN is not influenced by pulse height. Upon determining the segmentation threshold, we combine the classifier to extract the alpha-MCA spectrum and beta-MCA spectrum. The experimental results indicate that beta-MCA mainly comprises the distribution spectrum of beta particles. The alpha-MCA spectrum mainly includes alpha particle peaks and a minor beta spectrum. We assessed the influence of different classifiers on instrument performance utilizing the alpha-MCA spectrum. Under the same recall rate, the alpha-MCA spectra separated by the TCNN model have lower background level and smaller detection limits.

These results indicate that the TCNN model possesses strong ability to distinguish alpha pulses from beta pulses in practical applications. Simultaneously, it enhances the background rejection capability and sensitivity of LSC.

The TCNN exhibits outstanding performance in experiments; nevertheless, the complex calculations limit its application in real-time scenarios. Future research can further optimize the computational efficiency of the TCNN and explore its application in neutron/gamma-ray detectors, especially in scenarios with high real-time requirements.

In summary, this study not only provides an efficient and reliable method for identifying alpha and beta particles in LSC, but also offers a new idea for processing multi-channel radiation detectors. The TCNN method is expected to play an important role in fields such as nuclear physics experiments, environmental monitoring, and radioactive waste disposal.

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Author Contributions

All authors contributed to the study conception and design. Material preparation, data collection, analysis, and the first draft of the manuscript were written by Ming Wang. All authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Data Availability

The data that support the findings of this study are openly available in Science at https://doi.org/10.57760/sciencedb.26212 and https://cstr.cn/31253.11.sciencedb.26212.

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Declarations

Conflict of interest: The authors declare that they have no conflict of interest.

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Note: Figure translations are in progress. See original paper for figures.