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Original Article

Differential die-away technology applied to detect special nuclear materials

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ABSTRACT

Differential die-away analysis (DDAA) technology is a special nuclear material (SNM) active detection analysis technology. Be a nuclear material shielded or not, the technology can reveal the existence of nuclear materials by inducing fission from an external pulsed neutron source. In this paper, a detection model based on DDAA analysis technology was established by geant4 Monte Carlo simulation software, and the optimal sensitivity of the detection system is achieved by optimizing different configurations. After the geant4 simulation and optimization, a prototype was established, and experimental research was carried out. The result shows that the prototype can detect 200 g of ^{235}U in a steel cylinder shield that's of 1.5 cm in inner diameter, 10 cm in thickness and 280 kg in weight.

1. Introduction

According to the incidents and trafficking database (ITDB) of the International Atomic Energy Agency (IAEA), an annual average of 132 incidents of trafficking and malicious use occurred every year in the past ten years. With international terrorism going rampant, nuclear terrorist activities have become possible, and the global nuclear security risks are increasing [1]. Today, conventional radiative diffusion devices (RDD) based on cesium and cobalt radioisotopes are evolving into more complex transuranic devices [2]. In order to prevent the proliferation of nuclear materials and the danger of "dirty bombs", various countries have installed detection equipment at border crossings and ports where sees enormous volume, number and type of cargo containers. Given that the detection of SNM is the most time-consuming and expensive, its inspection system should be fast, efficient and with low probability of false positives [3–5].

Fissile materials contained in transuranic RDDs spontaneously emit neutrons and gamma rays with low intensity, weak energy and weak penetration. They can be effectively attenuated by shielding (lead and iron) of a few centimeters thick, which may lead to inaccurate passive detection or false alarm [6,7]. Passive detection technology can easily detect unshielded or lightly shielded nuclear materials, and doesn't work as well when they're heavily shielded. In such situations, active detection methods are used to induce fission by radiation sources for detection. DDAA [8–10] technology can be used for active detection and analysis of SNM. By adding a radiation source to induce fission from shield nuclear materials and detecting the time characteristics of fission neutron signals, detecting nuclear materials can be achieved. Compared with passive techniques, it has high sensitivity and wide application, and can detect smaller amount of fissile nuclear materials even under a high neutron gamma background [11]. Since the active detection devices based on the DDAA technology has better SNM detection performance, there are many practical application scenarios for such devices. For example, it can be applied to the detection of fissile material residues in vehicle contrabands and metal hulls (spent fuel transportation), the detection of border and customs cargo containers and the detection of SNM in samples or waste barrels in nuclear fuel reprocessing plants and waste management sites [12–14].

In this paper, the geant4 Monte Carlo simulation software was used to design the active detection device based on DDAA

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technology and study the detection performance on the same nuclear material under different geometric configurations. After simulating and optimizing the model, the prototype was established to carry out experimental research, and the application of DDAA technology in the field of SNM detection is evaluated. The results show that DDAA technology has a better performance in SNM detection.

2. Principle of differential die-away analysis technology

In DDAA technology, a pulsed neutron generator is used to generate microsecond wide pulses of neutrons that are directed to the cargo being examined. As each pulse passes through the cargo, the fast neutrons slow down sharply and disappear within a few microseconds. That is, the die-away time of fast neutrons is about a few microseconds. Whereas thermal neutrons produced by the source neutrons decay much more slowly (exponentially), on the order of hundreds of microseconds. If SNM exists in the cargo, it will be induced by fission to produce new fast neutrons. The decay of fast neutrons produced by such fission is much slower than that produced by a neutron generator. It decays with a decay time of the surrounding material. Therefore, generally the die-away time of the fission neutrons induced by SNM in the sample box is hundreds of microseconds to milliseconds [15].

Expected measured signal based on DDAA technology is as shown in Fig. 1. Fast interrogation neutrons have a certain die-away time as shown in area-0. In the absence of fissile nuclear material (solid curve), thermalized interrogation neutrons generate area-1 signal whose die-away time is orders of magnitude larger than those of fast neutrons, which is known as “background signal". In the presence of a nuclear material (dashed curve), area-1 signal is generated by thermal neutrons and prompt induced fission neutrons, both of which have approximately the same die-away time. As the mass of fissile nuclear material increases gradually, the difference between the two curves will increase gradually. The difference between the two curves would be even more pronounced if cadmium sheets of certain thickness are placed around the detector, since they would reduce the thermal neutron signal by several orders of magnitude.

The FOM is defined to represent the sensitivity of the DDAA nuclear material detection system. And it could be calculated by the following formula [16]:

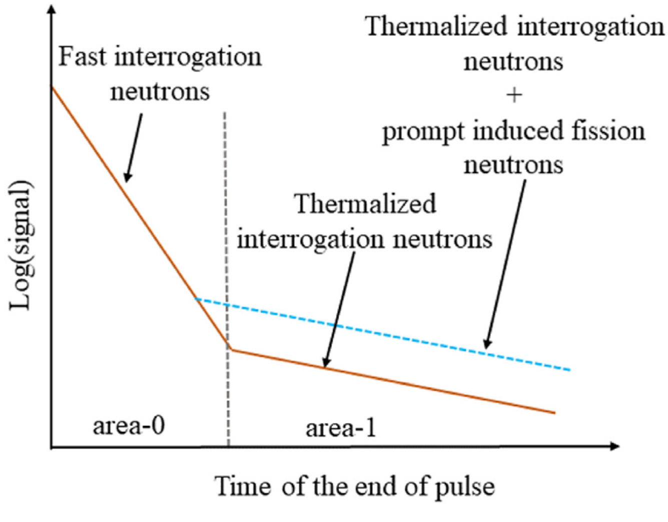

Fig. 1. Expected measured signal based on DDAA technology [6].

$$\mathrm{FOM} = \frac{1}{\sqrt{nps}} \times \frac{T - T_{bg}}{\sqrt{S + 2T_{bg}}} = \frac{S}{\sqrt{nps\left(T + T_{bg}\right)}} \tag{1}$$

where *nps* is a factor used as normalization when simulating different numbers of particles or measuring at different durations. Moreover, time integrated signal T (measured when there is fissile material) minus time integrated signal T_{bg} (without fissile material) at the same measurement time represents the net signal S.

The purpose of DDAA system is to find the maximum FOM value under different setups so that the detection system can achieve optimal sensitivity. Therefore, in section 3, geant4 is used to calculate under different configurations to work out various FOM results. At the same time, FOM can also be improved by increasing measurement time, source intensity, or detection efficiency of the ^{3}He detector array.

3. Geant4 simulation optimization for prototype

For optimizing the nuclear material detection device based on DDAA technology, simulations were carried out by the geant4 Monte Carlo toolkit [17]. Referring to the research of R. Remetti et al. [6], we designed and optimized the SNM detection system to meet our actual needs. As shown in Fig. 2, the detection device consists of a neutron source, a moderator and a neutron detector array. The neutron source emits pulsed neutrons of 14 MeV. Two pieces of cadmium sheets of 1 mm thickness are attached inside and outside the moderator made of high-density polyethylene. The neutron detector array consists of 8 ^{3}He detectors at a pressure of 2 Pa, 5.08 cm in diameter and 100 cm long each. And each detector is embedded in a polyethylene module. The cube sample box, length of 4 cm, is placed in the center of a cuboid polyethylene moderator (14 cm × 10 cm × 50 cm) or in a cylinder 22.3 cm in radius and 50 cm in height (to ensure the total mass of the sample moderator remains the same). Then 200 g ^{235}U was put in the sample box. The moderator around the sample is an important variable, if too heavy, rendering the device too bulky, too light, resulting insufficient fast neutron moderation, which can lead to possible failure of the DDAA technique. The moderator material is made of ordinary A4 paper with a density of 0.65 g/cm^3 and at a hydrogen atomic percentage of 47.6% [18]. In this paper, a polyethylene with a higher density, 0.95 g/cm^3 to be exact and at a hydrogen atomic percentage of 85.6% was used to slow down fast neutrons. Based on the above settings, while the total mass of the moderator remains constant, geant4 was used to simulate and optimize the sizes and shapes of moderators, as well as the placements of the detector array.

3.1. Simulation optimization results analysis

When the total mass of the sample and the moderator of the ^{3}He array detector stays constant (about 100 kg), relative position of each component between the detection equipment will affect detection result. Fig. 2 shows the optimization scheme under different placements of the detection equipment simulated by geant4.

Simulating six possible situations shown in Fig. 2 and recording the time of the ^{3}He (n, p) reaction in the detector array, the curves corresponding to the results of each configuration can be obtained as shown in Fig. 2(ãf), in which the blue curve indicates the absence of nuclear material, and the red curve indicates the presence of fissile nuclear material. The FOM results under different setups are as in Fig. 3. Optimized calculation results show that configuration b is the worst, and the distinction efficiency between the two curves

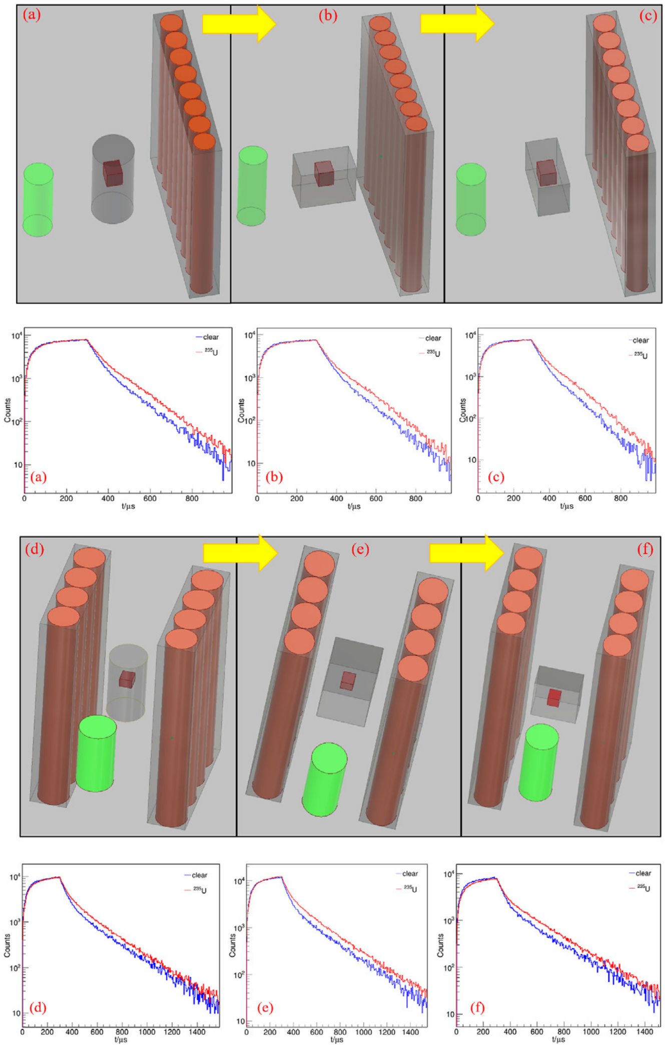

Fig. 2. The geant4 analog DDAA technique is used to probe SNM experiments with a fixed moderator mass. Green represents the neutron generator, transparent grey is the moderated structure around the sample to be tested (red), and transparent grey is the ^{3}He tube (orange) embedded in the polyethylene structure.

is the lowest. Besides, the results of configurations c, d, and f are relatively close, while configuration e is shown to be the best with the largest discrimination between the two curves.

3.2. Suggestions for detection system design

According to the optimized simulation results under different setups, we can draw the following conclusions under the detector prototype configuration.

1) Higher detection efficiency can be achieved when the volume of moderator between the detector and the sample is reduced appropriately (as shown in Fig. 3, setup c is better than b, and setup e is better than f);
2) Properly increase the volume of moderator between the neutron source and the sample to fully slow down fast neutrons and increase the probability of inducing fission. However, it should not be too thick, or it will affect the neutron flux (As shown in Fig. 4, with the side length of the moderator gradually increasing, the probability of nuclear material fission induced by neutrons first increases and then gradually decreases. And the probability of induced fission is the highest at the side length of 7 cm);

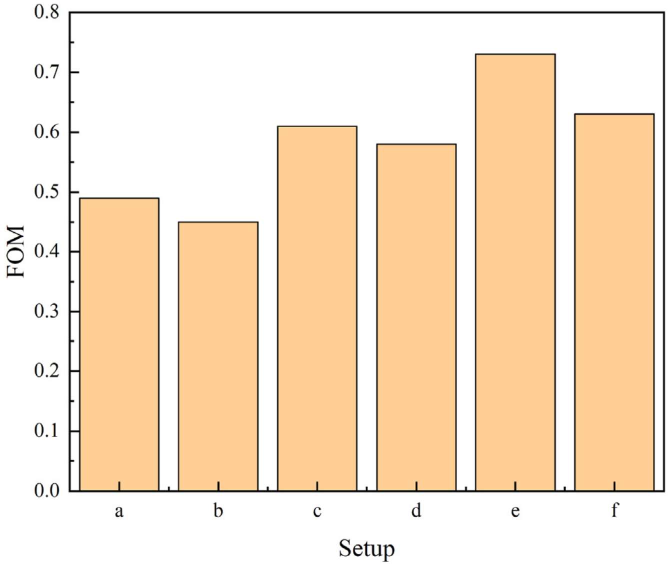

Fig. 3. FOM values calculated by six possible configurations of analog data show in Fig. 2

4. Experimental setup

Nuclear materials detection system based on DDAA analysis technology is as shown in Fig. 5, the controller of neutron generator was used to control it to generate pulsed neutrons of a certain frequency. When there was nuclear material in the sample box, pulse neutrons induced fission in nuclear material. The time and counts of neutrons can be detected by the ^{3}He array detector. Then collecting those data by the data acquisition (DAQ) system, the existence of nuclear material can be confirmed.

According to the optimized simulation results combined with the practical application requirements, a series of experiments were performed. The sample used in the experiments was 222 g of enriched uranium with the following composition: 10% of ^{238}U and 90% of ^{235}U, corresponding to almost 200 g of fissile ^{235}U. As shown in Fig. 6(a), 200 g, 400 g and 600 g of fissile ^{235}U were placed sequentially in a cube sample box with a side length of 4 cm. In order to study the application of DDAA technology for different masses of nuclear material detection under the configuration shown in Fig. 2(e), the sample box was put into the center of a cuboid polyethylene moderator with a size of 14 cm × 10 cm × 50 cm. A steel cylinder with an inner diameter of 1.5 cm, a wall thickness of 10 cm, and a weight of 280 kg was placed between the two groups of ^{3}He array detectors. 200 g fissile ^{235}U was placed in the center of the cylinder to simulate the application of DDAA technology with shielding. The experimental samples were placed between two sets of ^{3}He array detectors (^{3}He detector enclosed in high density polyethylene and covered with cadmium). The distance between the center of the sample and the detector array was 20 cm. ING-013 model (produced by All-Russian Research Institute of Automatics) pulsed neutron generator was used as the neutron source [19]. This system can generate up to 1 × 10^9 neutrons per second with an operation range of 1 Hz–50 Hz. The neutron generator emitted 14 MeV neutrons at a distance of 100 cm from the center of the sample. Stray neutrons were absorbed by boron filled polyethylene with a certain thickness around the non-incidence surface of the neutron generator. A pulsed D-T neutron

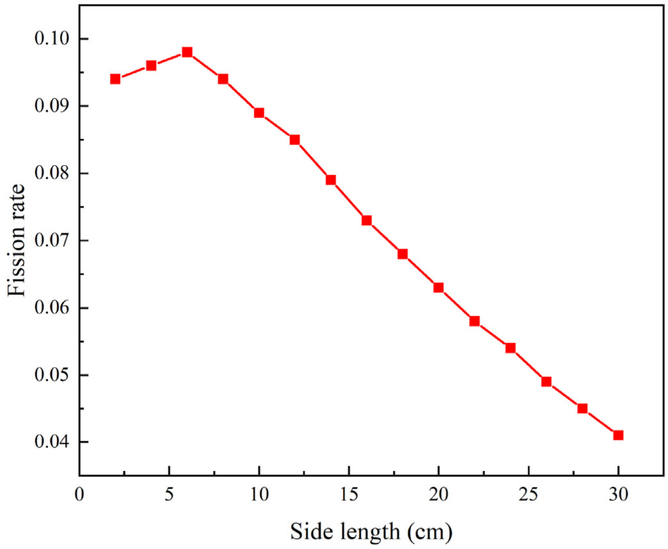

Fig. 4. Probability of induced fission neutrons while the thickness of moderator on the detection surface of array detector stays fixed and the thickness of moderator on the incident surface of neutron source changes.

generator was used as the induced fission neutron source. When its voltage was set to 4 kV, it could generate neutrons with a source intensity of 1 × 10^8 n/s. And the neutron pulsed was generated at a frequency of 50 Hz with of the synchronous pulse width of 5 μs The pre-amplifier signal from the ^{3}He detector array was processed through amplifier and discriminator. Then the total TTL signal was fed to DAQ system. The total acquisition time for each data set is 300 s.

5. Experimental results

5.1. Fissile material of different masses

According to the simulation optimization calculation results, the configuration profile in Fig. 2(e) was used to carry out experimental research. A weight of 200 g, 400 g and 600 g^{235}U were placed in the sample box respectively. DDAA technology was used to analyze the relationship between neutron time and counts detected by ^{3}He detector array with and without nuclear material. As shown in Fig. 7, when there are ^{235}U nuclear materials of different masses, the existence of nuclear materials can be quickly distinguished by the neutron time spectrum. As nuclear materials increase in mass, the distinction becomes more and more obvious. When the ^{235}U is

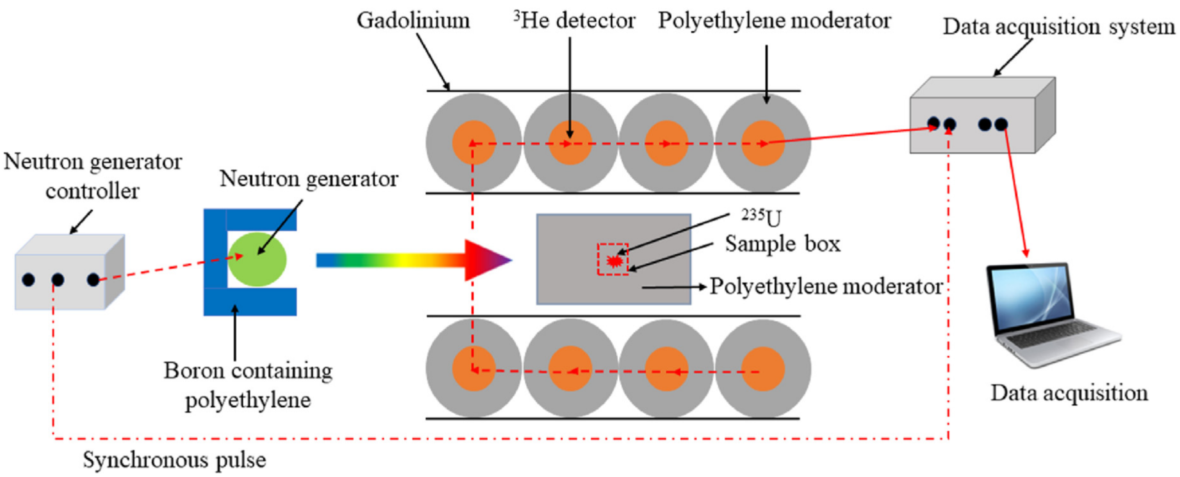

Fig. 5. Schematic diagram of detection system based on DDAA analysis technology.

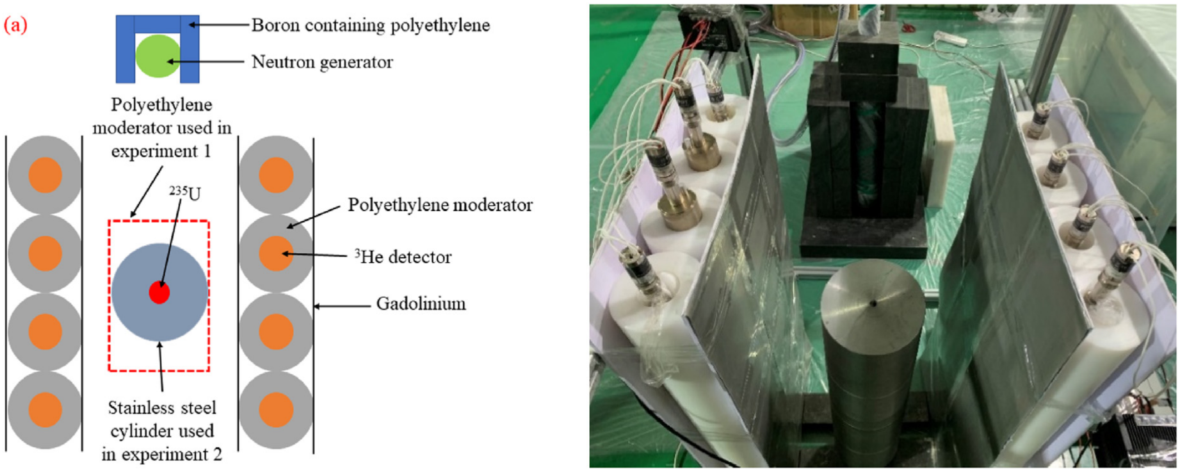

Fig. 6. (a) Schematic diagram of the experimental setup of the two groups (b) SNM experimental site map measured in a steel shield with an inner diameter of 1.5 cm, a wall thickness of 10 cm, and a weight of 280 kg.

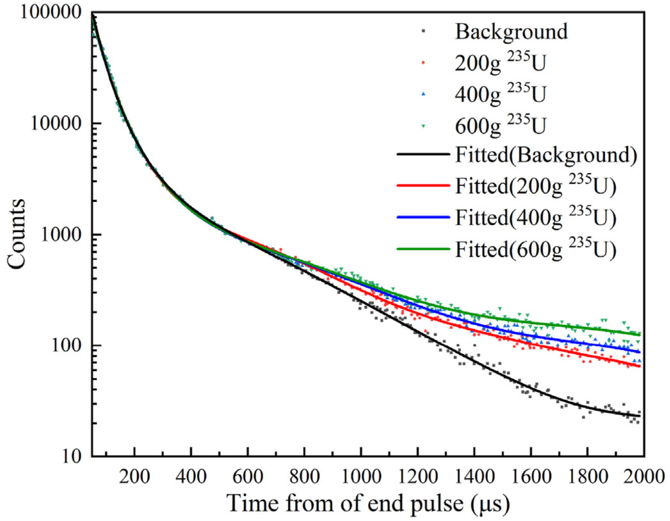

Fig. 7. Plot of neutron counts as a function of time for 200 g、400 g and 600 g^{235}U.

200 g, 400 g and 600 g in mass, its FOM values are calculated to be at 0.34, 0.47 and 0.55. The results show that 200 g nuclear material can be quickly detected by the detection system.

5.2. Effects of fissile material in the presence of shielding

To study the feasibility of DDAA technology in detecting shielded nuclear materials, 200 g^{235}U sample was placed in a steel cylinder. The steel cylinder has an inner diameter of 1.5 cm, a wall thickness of 10 cm and a weight of 280 kg. The experiment setup is shown in Fig. 6(b), and the relationship between neutron time and counts detected by the array detector with and without nuclear material was measured. As shown in Fig. 8, 200 g^{235}U in the steel cylinder shield can be quickly distinguished by DDAA technology. Combined with the real-time alarm function of the DAQ software, the alarm function within a few seconds can be realized. In other words, the experimental results indicate that the entire SNM active detection system can realize detection and alarm functions within a few seconds, which provides possibility for future practical application.

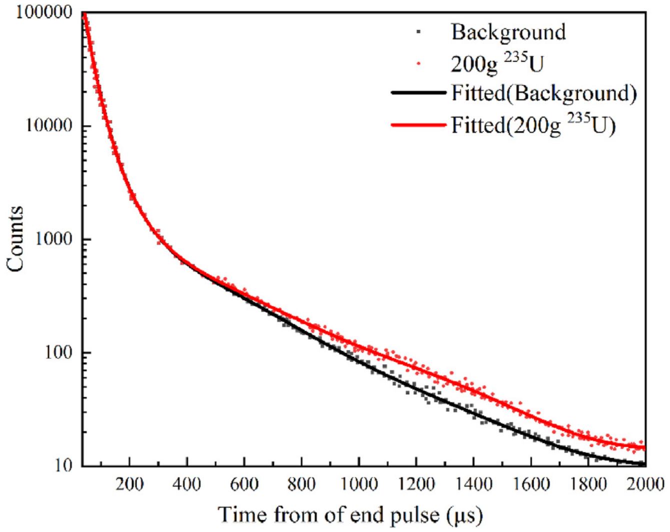

Fig. 8. Detected neutron counts versus time when 200 g of ^{235}U nuclear material was placed in a steel shield.

6. Conclusion

In this paper, Monte Carlo simulation and experiments were studied on the detection of SNM based on DDAA technology. The detection device was optimized by geant4 to obtain the configuration scheme for the optimal sensitivity. After building the detector prototype, experiments were carried out. The results show that 200 g^{235}U in a steel shield can be detected within a few seconds and an alarm is triggered immediately. In future work, the experimental equipment will be further optimized to improve the detection capability of SNM based on DDAA technology, so as to improve the detection capability of nuclear security.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

[1] IAEA Incident and Trafficking Database (ITDB), Incidents of Nuclear and Other Radiocation Material Out of Regulator Control 2021 Fact Sheet [EB/OL], 2022, 07.15), https://www.iaea.org/sites/default/files/22/01/itdb-factsheet.pdf.
[2] Asaf Durakovic, Medical effects of a transuranic "dirty bomb", Mil. Med.: Off. J. AMSUS,Soc. Fed. Health. Agencies. 182 (3) (2017) e1591–e1595.
[3] U. Bravar, R.S. Woolf, P.J. Bruillard, et al., Calibration of the fast neutron imaging telescope (FNIT) prototype detector, IEEE Trans. Nucl. Sci. 56 (5) (2009) 2947–2954.
[4] J.L. Jones, W.Y. Yoon, D.R. Norman, et al., Photonuclear-based, nuclear material detection system for cargo containers, Nucl. Instrum. Methods Phys. Res. B 241 (1/4) (2005) 770–776.
[5] D. Cester, G. Nebbia, L. Stevanato, et al., Special nuclear material detection with a mobile multi-detector system, Nucl. Instrum. Methods Phys. Res. A 663 (1) (2012) 55–63.
[6] R. Remetti, G. Gandolfo, L. Lepore, et al., In field application of differential die-away time technique for detecting gram quantities of fissile materials, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. (870) (2017) 123–130.
[7] R. Rober t, C. David, T. Scott, Rattling nucleons: new developments in active interrogation of special nuclear material, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 663 (1) (2012) 75–79.
[8] A. Belian, H.O. Menlove, M. T, et al., New design of the differential die-away self-interrogation instrument for spent fuel assay, Nucl. Mater. Manag. 40 (3) (2012) 58–60.
[9] H.O. Menlove, S.H. Menlove, S.J. Tobin, Fissile and fertile nuclear material measurements using a new differential die-away self-interrogation technique, Nucl. Instrum. Methods Phys. Res. 602 (2) (2009) 588–593.
[10] A.C. Kaplan, V. Henzl, H.O. Menlove, M. T, et al., Determination of total plutonium content in spent nuclear fuel assemblies with the differential die-away self-interrogation instrument, Nucl. Instrum. Methods Phys. Res. 764 (2014) 347–351.
[11] K.A. Jordan, T. Gozani, Pulsed neutron differential die away analysis for detection of nuclear materials, Nucl. Instrum. Methods Phys. Res. B 261 (2007) 365–368.
[12] N. Cherubini, A. Dodaro, G. Gandolfo, et al., Field prototype of the ENEA neutron active interrogation device for the detection of dirty bombs, Challenges 7 (2016) 1–9.
[13] K.A. Jordan, T. Gozani, J. Vujic, Differential die-away analysis system response modeling and detector design, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 589 (2008) 436–444.
[14] Yogesh, Ashish Kashyap, et al., Differential Die-Away Analysis for detection of 235U in metallic matrix, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 806 (2016) 1–4.
[15] K.A. Jordana, T. Gozani, Detection of 235U in hydrogenous cargo with Differential Die-Away Analysis and optimized neutron detectors, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 579 (1) (2007) 388–390.

[16] K.A. Jordan, J. Vujic, E. Phillips, et al., Improving differential die-away analysis via the use of neutron poisons in detectors, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 579 (1) (2007) 404–406.
[17] S. Agostinelli, J. Allison, K. Amako, et al., Geant4—a simulation toolkit, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 506 (3) (2003) 250–303.
[18] K.A. Jordan, T. Gozani, J. Vujic, Differential die-away analysis system response modeling and detector design, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 598 (3) (2008) 436–444.
[19] V. Mikerov, Yu Barmakov, E. Bogolubov, et al., Portable neutron generators of VNIIA and their applications, Proc. Sci. 25 (2007) 1–7.