

EXPERIMENT ON THE PERFORMANCE OF THE NEUTRON-BASED EXPLOSIVES DETECTION SYSTEM USING ^{252}Cf AND $^{241}\text{Am} / ^9\text{Be}$

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Abstract

This experiment used ^{252}Cf and $^{241}\text{Am} / ^9\text{Be}$ with activities of $2.215 \times 10^7 \text{ n/s}$ to test the performance of a neutron-based Explosives Detections System (EDS). The EDS is made up of a box-shaped paraffin-moderator, a thallium-doped sodium iodide gamma-ray detector, and a radioactive neutron source. Various parameters resulted from the inspection of TNT are measured and compared with the corresponding results from Monte Carlo calculations based on Monte Carlo N-Particle (MCNP) computer code. The following results are obtained: the thermal neutron fluxes at the center of the inspection cavity, from measurement and calculation, are $68.1 \pm 4.9 \text{ n/cm}^2\text{-s}$ and $58.7 \pm 7.6 \text{ n/cm}^2\text{-s}$, respectively; the EDS's detection sensitivities based on the detection of the 10.8-MeV gamma rays, from measurement and calculation, are $0.53 \pm 0.06 \text{ cps/kg}$ and $0.62 \pm 0.07 \text{ cps/kg}$, respectively; with a collimation of the gamma-ray detector in place, the measured intensity of the 2.2-MeV gamma rays resulted from interactions of thermal neutrons with H-chemical elements of TNT reduced by 40%; when $^{241}\text{Am} / ^9\text{Be}$ is used as the neutron source, the 4.4-MeV gamma rays from C can be observed along with the 2.2- and 10.8-MeV gamma rays from H and N, respectively; the measured fractional ratios of $N_{\text{H}}/N_{\text{C}}$, $N_{\text{H}}/N_{\text{N}}$ and $N_{\text{C}}/N_{\text{N}}$ are 1.44 ± 0.16 , 58.06 ± 8.17 and 40.37 ± 4.89 , respectively, while their corresponding theoretical values are 1.18 ± 0.12 , 50.13 ± 3.51 and 42.42 ± 4.74 , respectively; the maximum total doses at 1 m away from the EDS's surfaces, from the measurement and calculation, are 9.43 ± 0.36 and $10.51 \pm 0.53 \mu\text{Sv/h}$, respectively.

Keywords: Explosives detection, radioactive source, thermal neutrons analysis, fast neutron analysis, Monte Carlo calculations

Introduction

During the last several decades many efforts have been made to utilize neutrons in various kinds of material inspections including explosives detection. Three principal advantages of using neutrons in

the EDS involve their specificity, high penetrability, and non-destructivity. Many neutron-based nuclear techniques, such as Thermal Neutron Analysis (TNA), Fast Neutron

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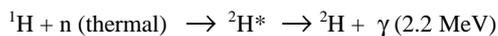
Analysis (FNA), and Pulse-Fast Neutron Analysis (PFNA) were proposed for explosives detection (Gozani, 1996; Buffler, 2003). In these techniques, neutrons are used as probes to penetrate the inspected object and create nuclear reactions in its constituents' chemical elements. For TNA, radioactive sources can be used to provide such neutrons, but for FNA and PFNA, neutron generators are required. Two advantages of using neutron generators to provide fast neutrons for the EDS are: 1) they can provide fast neutrons with available energies energetically possible to create inelastic nuclear reactions in chemical elements of explosives, and 2) they enable the deduction of timing information from inelastic nuclear reactions, which can be used as an additional indicator in the EDS. However, the neutron generator-based EDS also has disadvantages. It normally has a high price, heavy weight, and is not easy to operate. To avoid such disadvantages, radioactive sources such as, ^{252}Cf and $^{241}\text{Am}/^9\text{Be}$ should be used to provide neutrons for creations of the above mentioned nuclear reactions. After all, ^{252}Cf and $^{241}\text{Am}/^9\text{Be}$ also produce fast neutrons with energies that may be energetically possible to initiate inelastic nuclear reactions in some chemical elements of explosives. In fact, ^{252}Cf and $^{241}\text{Am}/^9\text{Be}$ produce neutrons with the highest energies of about 8 and 10 MeV, respectively (Hussein and Waller, 2000; Knoll, 2000). The objectives of this experiment are: 1) to investigate whether neutrons from ^{252}Cf or $^{241}\text{Am}/^9\text{Be}$ can create inelastic nuclear reactions in chemical elements of explosives or not, and 2) to deduce intensities of the gamma rays resulted from nuclear reactions between neutrons and the explosive's chemical elements (H, C, N, and O). These intensities will enable the estimations of the fractional ratios of the explosive's chemical elements which can be used as additional indicators for obtaining an effective EDS. Since this experiment is a pilot research project, the small and economy scale EDS is designed for testing its inspection performance in this experiment. The inspection cavity of the EDS is rather small, suitable for inspection of small objects. The larger and more practical EDS can

be designed later if this experiment shows positive results. The theoretical backgrounds of TNA, FNA, and the brief MCNP Description are given in Section 2. Section 3 describes the experimental set-up, while results and discussion are given in Section 4. Section 5 comprises the conclusions.

Theoretical Background

TNA

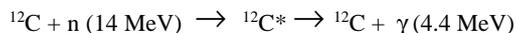
TNA is based on the capture of thermal neutrons by nuclei of the inspected object, in which the characteristic high energy gamma rays of specific nuclei are produced. Thermal neutrons are created by slowing down fast neutrons from radioactive sources in the moderator and in the inspected object itself. Since all commercial explosives contain H, C, N, and O nuclei (Buffler, 2003), the characteristic gamma rays of these nuclei are expected to be produced during the inspection. The following reactions are the results of the capture of thermal neutrons by H and N with cross sections of 330 and 11 mb, respectively (Hussein, 1992; Gozani, 1996):



Since the nuclear capture cross sections of C and O are very small, the gamma ray spectra produced during the inspection will not contain characteristic gamma rays from C and O.

FNA

FNA is based on the inelastic neutron scatterings of fast neutrons with the nuclei of the inspected object, in which the characteristic high energy gamma rays of specific nuclei are produced. When the explosive is inspected, gamma rays from the inelastic neutron scatterings with all the explosive's chemical elements (except H) are expected to be produced. The following reactions are the results of the inelastic neutron scatterings with C, N, and O with cross sections of 200 - 400, 430, and 474 mb, respectively (Hussein, 1992; Gozani, 1996):



MCNP Description

In this work, Monte Carlo calculation by using Monte Carlo N-Particles (MCNP) computer code is performed to compare its results with experimental values. Therefore, a brief description of MCNP is given below.

MCNP is a general-purpose particle transport code, where N represents neutron, photon, electron or combination of them. It can be used to simulate the transport of neutrons of energy from 10^{-11} - 20 MeV, and photons and electrons of energy from 1 keV- 10^3 MeV. In solving a problem, MCNP uses the statistical process to simulate the transport of individual particles and record some aspects of their average behavior which can be inferred to be the average behavior of the particle in the physical system. For the problem of interaction of nuclear particles with matters, the individual probabilistic events that comprise a process are simulated sequentially and the probability distribution governing these events is statistically sampled to describe the total phenomenon. In using MCNP for

calculation, the user must create an input file which contains information about the geometry specification, the source definition, the material description, the selection of cross section evaluations, and the type of answer and tally desired. MCNP-5 (X-5 Monte Carlo Team, 2003) is used for simulation in this work, in which the Evaluated Nuclear Data File B-VI is selected as the cross sectional data for nuclear interactions between neutrons and photons with matters. A point source of ^{252}Cf is used as the neutron source with the Watt fission energy spectrum. For most simulation cases, the numbers of simulated particles are at least one million. All of the simulation results have statistical uncertainties of less than 5%.

Experimental Set-up

This experiment was performed at China Institute of Atomic Energy, Beijing, China. The experimental set-up has the following components: a box-shaped neutron moderator, a neutron source, a gamma-ray detector and a data acquisition system. The experimental set-up is as shown in Figure 1.

Figures 1(a) and 1(b) are a photograph and a three-dimensional drawing of the experimental

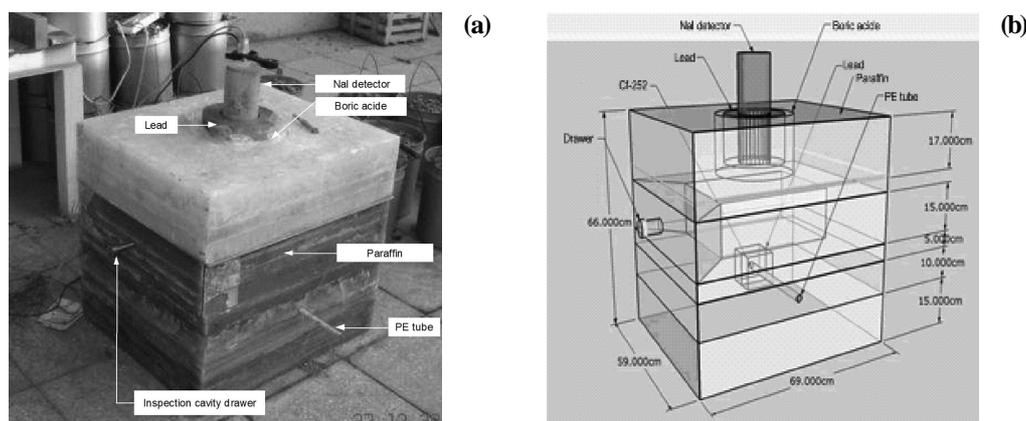


Figure 1. Experimental set-up: (a) photograph and (b) three dimensional drawing showing locations and dimensions of its components

set-up, respectively, in which the dimensions and locations of the experimental set-up's components are shown. The neutron moderator, made up of paraffin, is of 15-cm thickness with x, y, and z dimensions of $60 \times 70 \times 65 \text{ cm}^3$. Paraffin was chosen as the EDS's moderator instead of polyethylene because it is cheaper while giving about the same moderating power level. The paraffin moderator will slow down neutrons from the neutron sources before they interact with the nuclei of the sample to be inspected. The inspection cavity with x, y, and z dimensions of $30 \times 40 \times 15 \text{ cm}^3$ is located in the middle of the box in which the sample to be inspected can be inserted through a drawer located at about the middle of one side of the box. The source holder is made up of a lead cube of 5-cm thickness located at the lower part of the box under the inspection cavity. ^{252}Cf or $^{241}\text{Am}/^9\text{Be}$ with about the same activities ($2.215 \times 10^7 \text{ n/s}$) was inserted in the source holder through a hole using a polyethylene (PE) tube. A NaI (Tl) detector with dimensions of $7.62 \times 7.62 \text{ cm}^2$, located on top of the center of the inspection cavity, is used to measure the gamma ray flux resulted from inspection of the sample in the inspection cavity. The NaI (Tl) detector is shielded by a 3-cm thick boric-acid absorber to absorb most neutrons which would have otherwise been absorbed by its crystal. The boric acid is further shielded by 5-cm thick lead to prevent gamma rays coming from directions other than from the inspection cavity. A ^3He -detector with a volume of 76.18 cm^3 (2.5-cm diameter and 9.7-cm length) is used to measure the neutron flux in the inspection cavity. Gamma-ray and neutron dosimeters are used to measure gamma-ray and neutron doses around the box. The data acquisition system comprises an AFT-research amplifier (model 2025 from Canberra Inc.), a compact and fast module multi-channel analyzer (from Amptex Inc.), and a personal computer.

Results and Discussion

Various parameters resulted from interactions between thermal and fast neutrons and the constituents' chemical elements of the inspected

object are measured in this experiment. To compare these parameters to theoretical values, MCNP calculations of these parameters are made. The following sections discuss both of these sets of parameters.

Thermal Neutron Flux at the Center of the Inspection Cavity

Since the intensity of the gamma rays resulted from TNA is proportional to the thermal neutron flux created in the inspection cavity, the thermal neutron flux at the center of the inspection cavity is estimated in this experiment. This is done by measuring the intensity of thermal neutrons at the center of the inspection cavity, using a ^3He -detector. Three measurements were taken with an average reading of 19,533 counts per second (cps). The thermal neutron flux at the center of the inspection cavity can be estimated by substituting the average value of thermal neutron intensity in the formula, $\phi_{\text{th}} = \text{cps}/N\sigma$, where N and σ are the number of atoms and neutron capture cross section of ^3He -atoms, respectively. With $N = 5.38 \times 10^{21}$ atoms and $\sigma = 5,327 \times 10^{-24} \text{ cm}^2$, the above equation gives a thermal neutron flux at the center of the inspection cavity of $68.1 \pm 4.9 \text{ n/cm}^2\text{-s}$. The corresponding MCNP calculation value is $58.7 \pm 7.6 \text{ n/cm}^2\text{-s}$. The discrepancy between experimental and calculation values may be due to two sources: 1) the large uncertainty involving using the low activity neutron source, and 2) the large uncertainty involving the estimation of number of ^3He -atoms in the ^3He -detector.

Gamma Ray Spectra Resulted from Inspection of TNT by ^{252}Cf

Figure 2 shows the gamma ray spectra resulted from inspection of 1,000, 520, and 200 g of TNT in the inspection cavity, using ^{252}Cf for 900 s counting time.

Included in Figure 2 are the corresponding background spectra resulted from inspection of air-samples for the same counting period. Since gamma-ray intensities from each TNT mass can not be distinguished graphically, the gamma-ray intensities from 520 and 1,000 g are multiplied by 10 and 100 respectively, before being plotted

in Figure 2 for better visualization. It is clearly shown that while the 2.2-MeV gamma-ray lines from H are well resolved, the 10.8-MeV lines from N are not. This is because the neutron capture cross section of N is much smaller than that of H. However, the appearance of the 10.8-MeV gamma-ray regions is similar to the results from an inspection of 300 g of C-4 explosive, using

$^{241}\text{Am}/^9\text{Be}$ of 10^9 n/s activity as shown in Figure 3 (Gozani, 1996). Nevertheless, the 10.8-MeV gamma-ray line in this figure is more pronounced than those shown in Figure 2 because C-4 has nitrogen density about twice higher than that of TNT and the activity of neutron source is about two orders higher. By taking the differences of photo peak areas of the 10.8-MeV gamma rays

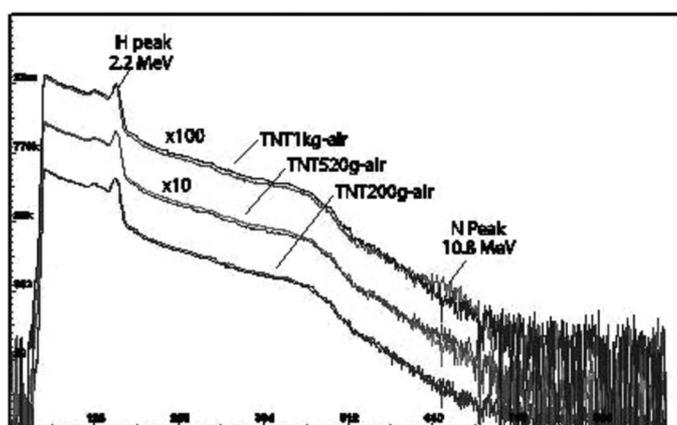


Figure 2. Gamma ray spectra resulting from inspection of 1,000, 520, and 200 g of TNT in the inspection cavity using ^{252}Cf for 900 s

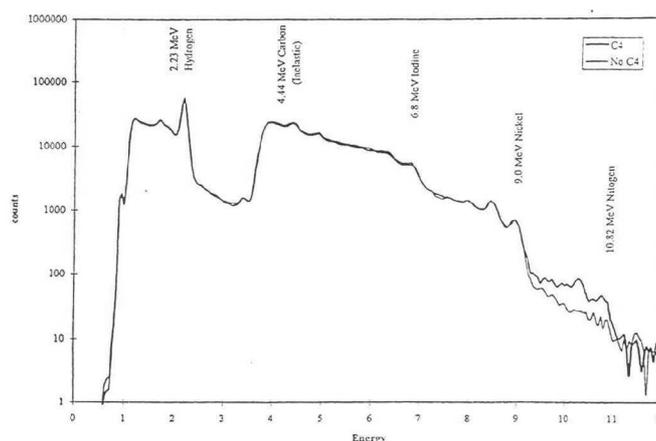


Figure 3. Gamma ray spectrum resulting from inspection of 300 g of C-4, using $^{241}\text{Am}/^9\text{Be}$ (Gozani, 1996)

(in the region of 9.45 - 11.43 MeV) from TNT and air samples of each spectrum, the intensity of the 10.8-MeV gamma rays resulted from inspections of 1,000, 520, and 200 g of TNT are 481, 307, and 76, respectively. Dividing these numbers by the counting time of 900 s and the corresponding TNT masses, the EDS's detection sensitivities for the detection of 1,000, 520, and 200 g of TNT are 0.53, 0.65, and 0.4 cps/kg, respectively. The average value of these detection sensitivities is 0.53 ± 0.06 cps/kg, which is quite low. This rather low detection sensitivity may be due to the low activity of ^{252}Cf . Moreover, it is about 11% lower than the corresponding MCNP-calculation value of 0.62 ± 0.07 cps/kg. This discrepancy may be mainly due to the large uncertainties involving the deductions of the low intensities of the 10.8-MeV gamma-ray lines. Another source of discrepancy may be due to the effect of the two-step MCNP calculation which may neglect the thermal neutron self-absorption by TNT. Unfortunately, the method to estimate this effect is not well understood. Figure 2 clearly shows that gamma rays from inelastic nuclear reactions were not observed. The lack of this observation implies that, though ^{252}Cf produces some neutrons with available energies energetically possible for FNA in chemical elements of TNT, the yields of such

neutrons may be too low.

Effect of Collimation of the Gamma-ray Detector

It is well known that the 2.2-MeV gamma rays will not be useful for TNA-based EDS, unless proper collimation or shielding of the gamma-ray detector is in place to prevent the detection of the unwanted 2.2-MeV gamma rays resulted from the capture of thermal neutrons by the H-chemical elements of the moderator. A creative attempt is performed in this experiment to study the effect of detector collimation by moving NaI (Tl) detector 6 cm further away from its original position. This collimation will reduce the detector solid angle for the detection of the unwanted 2.2-MeV gamma rays resulted from the capture of thermal neutrons by the H-chemical elements of the paraffin-moderator. This attempt may enable the use of the 2.2-MeV gamma-ray intensity as an additional indicator for the EDS. Figure 4 shows the comparison between the gamma-ray spectra resulted from the inspection of 1,000 g of TNT using the collimated and un-collimated detectors. It was found that the intensity of the 2.2-MeV gamma-rays from the collimated detector is about 40% lower than that of the un-collimated counterpart. This result suggests that proper collimation of

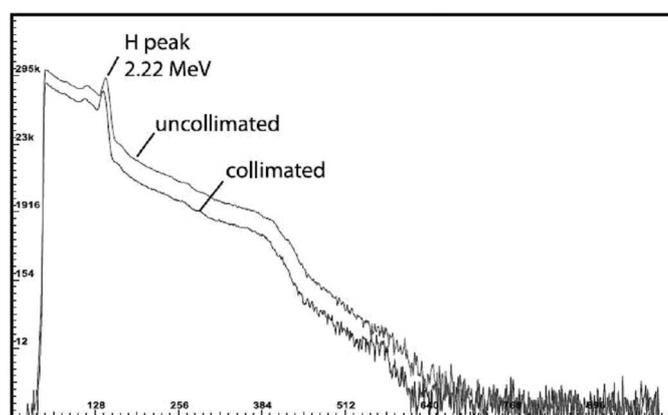


Figure 4. A comparison between gamma-ray spectra resulting from inspection of 1,000 g of TNT using the collimated and un-collimated gamma-ray detectors

the gamma-ray detector must be achieved if the 2.2-MeV gamma rays from H are to be useful for the TNA-based EDS.

A gamma-ray Spectrum Resulted from Inspection of TNT using $^{241}\text{Am}/^9\text{Be}$

To investigate whether neutrons from $^{241}\text{Am}/^9\text{Be}$ can initiate inelastic nuclear reactions in C- and O-chemical elements of explosives or not, the $^{241}\text{Am}/^9\text{Be}$ is used to inspect TNT of 1,000 g in the inspection cavity. For this inspection, creativity was done by putting an additional shielding in the inspection cavity to prevent the 2.2-MeV gamma rays from activation of H-chemical elements of paraffin from reaching the detector. Lead blocks of 5-cm thickness were put around the inspection cavity except at its ceiling, where the gamma ray detector's front face is located. With this shielding in place, the chances of the unwanted 2.2-MeV gamma rays due to activation of H-chemical elements of paraffin reaching the gamma ray detector should be greatly reduced. The reduction may enable the use of the 2.2-MeV gamma-ray intensity as an additional indicator for the identification of explosive. Figure 5 shows the gamma ray spectrum resulting from this inspection in which the 4.4-MeV gamma rays with their single and double escapes of 3.9 and 3.4 MeV energies,

respectively, are observed along with the 2.2- and 10.8-MeV gamma rays. It is obvious to see that intensity of the 2.2-MeV gamma-ray lines in this figure is very much less than those of Figures 2 and 3. This observation implies that additional shielding of the inspection cavity can prevent some of the unwanted 2.2-MeV gamma rays from reaching the detector. However, the 6.1- and 5.1-MeV gamma rays from inelastic nuclear reactions in O and N, respectively, do not appear in Figure 5.

These observations imply that, though some of the neutrons from $^{241}\text{Am}/^9\text{Be}$ have available energies energetically possible for such reactions, the yield of such neutrons is too low. The deductions of photo peak areas of gamma rays in Figure 5 give intensities of the 2.2-, 4.3-, and 10.8-MeV gamma rays of 14,816, 8,860, and 222, respectively. By using the ratios of these intensities (C_i/C_j), the measured fractional ratios of TNT's chemical elements (N_i/N_j) can be deduced by using the formula:

$$\frac{N_i}{N_j} = \frac{C_i / \varepsilon_i f_i}{C_j / \varepsilon_j f_j}$$

where ε_i and ε_j are the detecting efficiencies of gamma rays of energies E_i and E_j , and f_i and f_j are the correction factors due to attenuation of

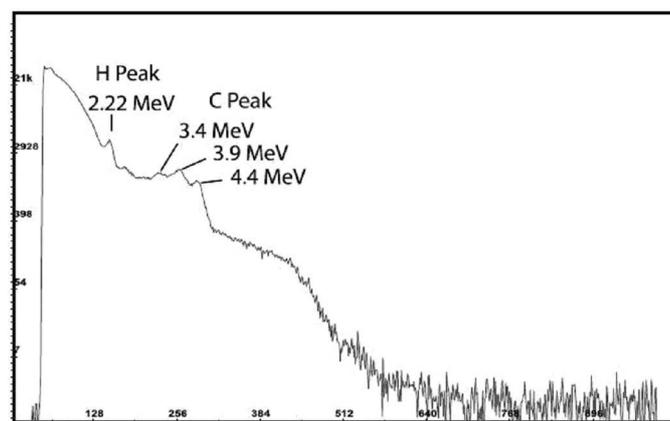


Figure 5. The gamma ray spectrum resulting from inspection of TNT of 1,000 g for 900 s, using $^{241}\text{Am}/^9\text{Be}$, in which the 4.4-MeV gamma rays with their single and double escapes of energies of 3.9 and 3.4 MeV, respectively, are observed in the spectrum

gamma rays of energies E_i and E_j , respectively. With $C_H=14,816$, $C_C=8,860$, $C_N=222$, $\epsilon_H=1.00$, $\epsilon_C=0.86$, $\epsilon_N=0.87$, $f_H=0.99$, $f_C=0.99$, and $f_N=0.99$, the above equation gives $N_H/N_C=1.44 \pm 0.16$, $N_H/N_N=58.06 \pm 8.17$ and $N_C/N_N=40.37 \pm 4.89$. The corresponding theoretical ratios can be calculated by using the following formula:

$$\frac{N_i}{N_j} = \frac{\sigma_i / A_i}{\sigma_j / A_j}$$

where σ_i and σ_j are the neutron cross sections for activations of TNT's chemical elements and A_i and A_j are the number of atoms of TNT's chemical elements. Since the chemical formula of TNT is $C_7H_5O_6N_3$, $A_H=5$, $A_C=7$, and $A_N=3$. By substituting these values and $\sigma_H=0.33b$, $\sigma_C=0.2b$, and $\sigma_N=0.011b$ in the above equation, we obtained the corresponding theoretical ratios as the followings: $N_H/N_C=1.18 \pm 0.12$, $N_H/N_N=50.13 \pm 3.51$ and $N_C/N_N=42.42 \pm 4.74$. Though the measured and theoretical values do not agree within their error limits, they are reasonably close. The discrepancies may be due to the low counting statistics due to low activity of neutron source. A study based on MCNP calculations suggested that efficient radioactive source-based explosives detection requires a source with activity in the order of 10^8 n/s (Uchai *et al.*, 2007). In the large ($4 \times 2.4 \times 1.8$ m³) TNA-based EDS, built by Science Application International Corporation, activity of the ²⁵²Cf-neutron source is even higher, being in the order of 10^9 n/s (Gozani *et al.*, 1992).

Radiation Doses Around the EDS

Since the safety aspect of the EDS is just as important as its performance aspect, radiation doses around the EDS were measured in this experiment. Two radiation dosimeters, one for neutrons and one for gamma rays, were used to measure the respective radiation doses at a distance of 1 m away from the EDS's surfaces in x- and y-directions. The measured total radiation doses in x- and y-directions are 9.43 ± 0.36 and 4.84 ± 0.31 μ Sv/h, respectively. The corresponding MCNP-calculation values are 10.51 ± 0.53 and 6.72 ± 0.34 μ Sv/h, respectively. Using

the measured values, we can estimate the dose rate that a control operator receives while working with the EDS in one year. Assuming that the control operator works with the EDS for 48 weeks/year, 5 days/week, and 8 h/day (assuming that he takes 4 weeks vacation), he will receive radiation of $(9.43 \times 10^{-6}$ Sv/h) \times (48 w/y \times 5 d/w \times 8 h/d) = 0.02 Sv/y. This value is about 60% below the whole body ICRP- Maximum Permissible Dose (MPD) of 0.05 Sv/y (ICRP 75, 1997).

Conclusions

The conclusions of this work can be summarized as follows. When ²⁵²Cf is used as the neutron source, it can create TNA-based nuclear reactions in H- and N-chemical elements of TNT but not the FNA-based nuclear reactions. However, the measured intensity of the 10.8-MeV gamma rays is quite low, making the detection sensitivity of the EDS low. When the gamma-ray detector is properly collimated, the intensity of the 2.2-MeV gamma rays due to activations of the H-chemical elements of paraffin is very much reduced, implying that an additional indicator for explosives detection can be obtained from H-information. When ²⁴¹Am/⁹Be is used as the neutron source, inelastic nuclear reactions in C-chemical elements of explosives are possible, enabling the deduction of fractional ratios of some explosive chemical elements. It was found that the measured and theoretical fractional ratios are reasonably close. The measured total radiation doses at 1 m distance from the EDS's surfaces are quite low, being well under the whole body ICRP-MPD value. However, some of the experimental results are quite different from the corresponding MCNP-calculation values. The discrepancies may be due to the large statistical uncertainty involving using the low activity radioactive sources, the low detection efficiency of gamma ray detector, and improper shielding of the gamma ray detector. To improve the performance of the TNA- and FNA-based EDS, ²⁴¹Am/⁹Be with higher activity and better shielding of the gamma ray detector should be in place. Moreover, by using a detector with higher

efficiency (such as the larger NaI (Tl) detector) or with higher resolution (such as bismuth germanate), the EDS's performance may be improved.

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