JOINT INSTITUTE FOR NUCLEAR RESEARCH
Frank Laboratory of Neutron Physics (FLNP)

FINAL REPORT ON THE SUMMER STUDENT PROGRAM

Detection of explosives and shielded explosives based on 'TANGRA' setup: 'Romasha'

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Participation period:
July 02 – August 26

Dubna, 2017

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Abstract

In airports and borders, it is required to detect contrabands and illicit materials using a fast method. Due to development of detection methods using neutron based detection techniques, shielding of contrabands was developed to counter these new techniques. Based on the TANGRA setup 'Romasha', MCNP5 model was made and validated by comparison with the experimental results. This 'Romasha' configuration was used in MCNP5 to study the detection of explosives and shielded explosives using Prompt Gamma Neutron Activation Analysis (PGNAA). Detecting explosives was achieved distinguishing between different types of explosives. Shielded explosives were tested and detected as well as the shielding material. After making some modifications to 'Romasha' configuration, this method's validity in detection of illicit materials was proven successful and further usage of the method for more researches in this field can be done.

Introduction

TANGRA is an experimental setup based on the Tagged Neutron Gamma Rays. It was created in the Joint Institute for Nuclear Reactions (JINR) in Frank Laboratory of Neutron Physics (FLNP) to investigate the 14.1 MeV neutron induced nuclear reactions on nuclei of different materials using the tagged neutron method. The TANGRA setup consists of an ING-27 neutron generator with an incorporated 64-pixel α-particle detector, iron shielding collimator, a sample target for gamma ray emission, a multi-detector system for detecting gamma-rays and ADCM for data acquisition (a compact and universal Digital Pulse Processing system for nuclear physics experiments). *[1]*

Initially gamma rays were detected using 22 sodium iodide NaI(Tl) multi-detector system called "Romashka" which allowed the study the angular and energy distributions of gamma rays emitted from the investigated target. Now, the NaI detectors are replaced with BGO detectors which have higher efficiency due to the higher probability of interaction with high energy gamma rays. *[1]*

Due to the increase of terrorist attacks and trafficking, it became more and more required to have an efficient and fast way to detect illicit materials (explosives, drugs and contrabands) in airports and on borders. Conventional methods such as X-rays and sniffing dogs are not effective enough to detect these illicit materials. Instead, neutron based methods were introduced due to their high imaging capability and elemental identification.

As a result to this advancement in detection methods, much more terrorism and trafficking techniques were developed. Attempts to shield contrabands and explosives against neutrons were made to counter these neutron imaging and analysis techniques for detection.

The aim of this report is to detect explosives and the capability of detecting the shielded explosives using Monte Carlo computations based on the new BGO multi-detector configuration.

TANGRA Setup

The ING-27 neutron generator is a small-size generator of new generation based on sealed gas-filled neutron tube with a built-in multi-pixel alpha particles detector. New equipment for remote detection and identification of explosive and other hazardous materials using associated particle imaging method (API) is designed on the basis of ING-27. The generator is designed to operate in computer-based hardware systems, both stationary and mobile.

It consists of neutron emission unit with alpha detector, power supply & control unit, connection cables up to 15 m long.

Figure 1: ING-27 neutron generator

Figure 1, and table 1 show the ING-27 neutron generator and its technical characteristics respectively. [2]

Table 1: ING-27 neutron generator characteristics [1]

Neutron energy, MeV	14
Neutron flux, neutron/s	up to 10^8
Alpha detector emission mode	continuous
Number of alpha detector pixels	at least 9
Power supply, V	+200
Maximum power consumption, W	40
Operating lifetime, h (at neutron flux of 5 x 10^7 neutron/s)	1000
Dimensions, mm: - neutron emission unit - power supply and control unit	 220×130×179 279×193×94
Weight, kg: - neutron emission unit - power supply and control unit	 7.0 3.0

The neutron generator ING-27 produces a neutron beam resulting from a fusion reaction based on the following reaction:

$D_1^2 + T_1^3 \rightarrow \alpha_2^4 + n_0^1$ (Q=17.6 MeV)

Where the alpha particles are produced with energy = 3.5 MeV each and neutrons are emitted with energy = 14.1 MeV each. The two produced particle from this reaction are emitted back to back (nearly 180°). The multi-pixel α-particle detector measures the time the α-particle reaches the detector and the coordinates of the point where it hit the detector behind the iron shield collimator placed in front of the neutron beam as shown in figure 2. As a result, the position of the associated neutron and its direction can be specified, thus the neutron is "tagged" and counted. According to the tagged neutron detector pixels, the optimal position for placing the sample target is selected.

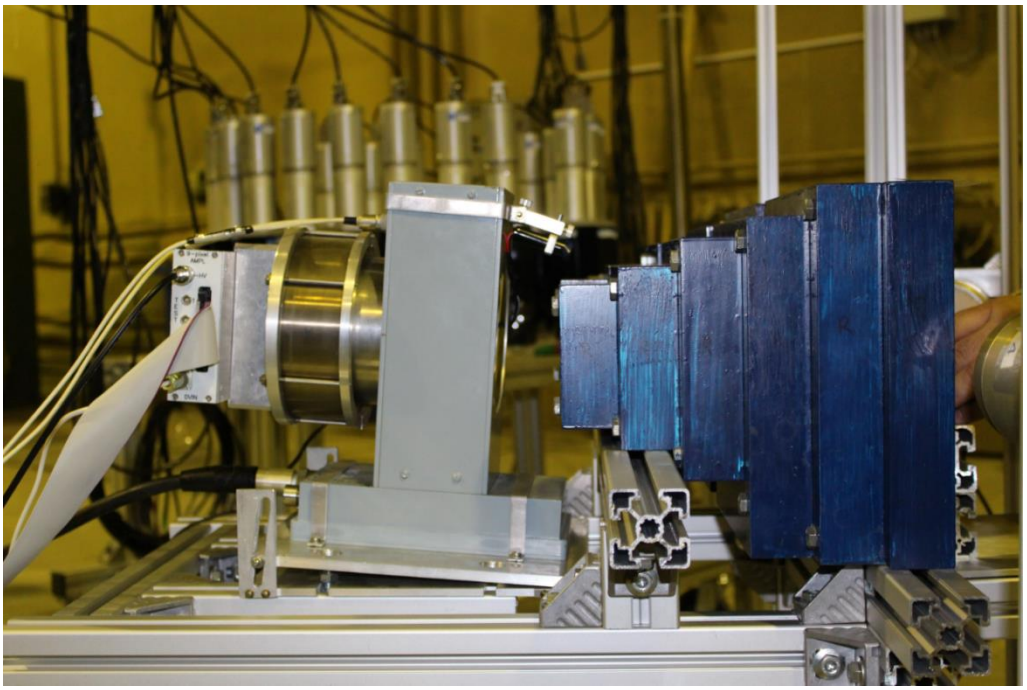


Figure 2: ING-27 neutron generator in 'Romasha'

The sample irradiated in 'Romasha' configuration of dimensions 10 x 10 x 5 is placed at a distance of 3.75 cm from the end of the iron collimator/shield. The 10 BGO gamma ray detectors were placed at a radius 30 cm from the center of the sample target in the form of a semi-circle to detect the gamma rays emitted from the sample target along 180° as shown in figure 3.

The calibration of the 10 BGO detectors was carried out using point gamma sources with known half-lives and activities. Cobalt-60 and Cesium-137 sources were placed at the target position in which they irradiate gamma rays of known energy to be detected by the array of BGO detectors. The 'Romasha' BGO detectors were calibrated according to the known peaks for each sample isotope.

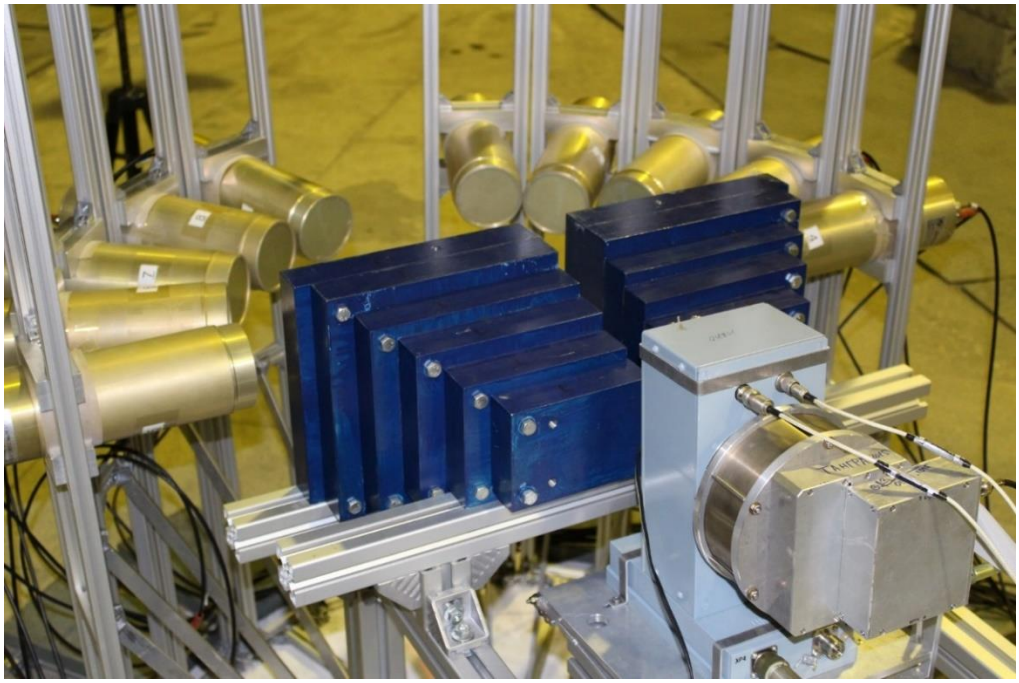


Figure 3: 'Romasha': ING-27, iron shielding - collimator assembly, 10 BGO detectors.

The signal processing and data collecting with 'Romasha' was done by a computerized 32-channel digitizer, using two ADCM16 (100MHz) ADC board (shown in figure 4).

The signals from all the detectors are registered in a digitized form and are saved in a file of extension '.root'.

Figure 4:32-channel digitizer used for DAQ

Literature review

Neutron Interaction with matter

Neutrons carry no charge and therefore cannot interact with matter by means of the Coulomb force, which dominates the energy loss mechanisms for charged particles and electrons. When a neutron does undergo interaction, it is with a nucleus of the target material. As a result of the interaction, the neutron may either totally disappear and be replaced by one or more secondary radiations, or else the energy or direction of the neutron is changed significantly. The relative probabilities of the various types of neutron interactions change dramatically with neutron energy according to the cross-section of nucleus and for each neutron energy. *[3]*

If the energy of the fast neutron is sufficiently high, inelastic scattering with nuclei can take place in which the recoil nucleus is elevated to one of its excited states during the collision. The recoil nucleus quickly de-excites, emitting a gamma ray, and the neutron loses a greater fraction of its energy than it would in an equivalent elastic collision. The produced gamma rays can determine the composition of the inspected material as each isotope has its own characteristic de-excitation gamma ray energy spectrum. *[3]*

For slow neutron interactions, the radiative capture reaction [or (n, ɣ) reaction] is the most probable. Because the incoming neutron energy is so low, all such reactions must have a positive Q-value to be energetically possible. *[3]*

Detection of explosives

Illicit materials and explosives are mainly composed of hydrogen, carbon, oxygen and nitrogen. In addition, most explosives have densities in the range 1.2–2.0 g/cm3 which is generally larger than most everyday H-C-N-O substances. Explosives are distinguished by relatively high proportions of nitrogen and oxygen and relatively low proportions of carbon and hydrogen. On the other hand, illicit drugs are generally rich in hydrogen and carbon and poor in nitrogen and oxygen. *[4]*

Most of the nuclides show difference in their scattering cross sections which enhances the probability of certain neutron interactions occurring at specific incident neutron energies and specific scattering angles. In particular, the main elemental constituents of narcotic and substances (H, C, N and O) differ strongly from one-another in their interactions with neutrons and can thus be characterized and distinguished via these differences. Neutron interrogation techniques generally depend on bombarding the nuclei of the interrogated object with neutrons of particular energy or energies, causing them to emit characteristic gamma rays or alter the energy of the interrogating neutrons. The attributes of the exploring neutrons and the energy and spatial distributions of the detected radiation (neutrons or de-excitation ɣ rays) may be used to determine the types, amounts and positions of the specific interacting nuclides, hence characterizing the specific materials making up the interrogated object. *[4]*

Thermal neutron analysis (TNA)

Fast neutrons produced by a radioisotopic source such as Cf_{98}^{252}, or a sealed tube neutron generator, are moderated to low energies (0.025 eV) within the object interrogation. A fraction of these thermal neutrons react with the nuclides in the object and the prompt capture gamma rays are detected. The application of TNA in the discovery of explosives is primarily based on the identification of nitrogen and hydrogen via the detection of the 2.22 and 10.83 MeV capture gamma rays from H_1^1 from N_7^{14}, respectively. The capture gamma ray spectra from arrays of detectors may be tomographically analyzed to provide the density distribution of nitrogen and hydrogen within the package. *[4]*

Fast neutron analysis (FNA)

A collimated beam of continuous fast neutrons bombards the object under inspection. The de-excitation gamma rays released from nuclei activated in fast neutron inelastic scattering events are detected by an array of detectors surrounding the object and shielded from the direct exposure to source neutrons. Specific elements present in each volume element or ‘‘voxel’’ of the object are identified through the de-convolution of the gamma ray spectra measured by each detector. The main signatures used are derived from detecting the 4.43 MeV gamma ray from C_6^{12}, the 1.64, 2.31 and 5.11 MeV gamma rays from N_7^{14}, and the 6.13 MeV gamma ray from O_8^{16}. Although the attenuation of fast neutrons within an object is much less than that of thermal

neutrons, FNA imaging is also limited to smaller objects due to the lack of geometrical definition when there are large distances between inner voxels and the detectors. *[4]*

Pulsed fast neuron analysis (PFNA)

The concept behind PFNA is similar to that of FNA except that a pulsed beam of neutrons is utilized. A focused collimated beam is passed through the object and the gamma rays emitted at certain energies are analyzed. The method uses penetrating neutrons at lower energies than FNA. The gamma rays are detected by scintillators that provide gamma ray energy information. The neutron beam provides position information required to determine the spatial distribution. The main advantage of using PFNA is that it unambiguously determines the elemental composition of explosives and the spatial location of these elemental concentrations. The main challenges in developing PFNA into a practical system is the tedious process of constructing a practical, collimated, pulsed energetic neutron beam that makes the operation safe and operationally acceptable and cost effective. *[4]*

Pulsed fast-thermal neutron analysis (PFTNA)

This technique provides a bulk analysis of the chemical present. Compared to PFNA which uses 2 ns pulsing, PFTNA uses 10 ms pulsing and has the main advantage of being portable. PFTNA allows the neutrons to be moderated and it can provide thermal neutron information like TNA but in addition it also measures fast neutron reactions for C, O and N and its advantage over PFNA is that it can measure thermal neutron capture gamma rays. *[4]*

Security screening technologies are used to either inspect passengers or their luggage. Screening of people walking through portals requires sophisticated technology. A screening solution must not only be accurate, but also publicly acceptable *[6].* In terms of what should be reported by a screening system and its performance characteristics, certain guidelines have been set by aviation authority *[7].* In particular, the screening equipment should report the following *[8]:*

(i) Type of illicit material.

(ii) The minimum quantity (mass).

(iii) The object shape (bulk, sheet, thickness, etc.).

(iv) The location of the illicit material in the bag. *[5]*

For different explosives: TNT, cyclotol, dynamite, PETN, TATP: carbon, nitrogen, oxygen and hydrogen compositions are shown in table 2.

Table 2: C, N, O, H composition in different types of explosives

Explosive	Carbon (%)	Nitrogen (%)	Oxygen (%)	Hydrogen (%)
TNT	33	14	29	24
Cyclotol	24	21	29	26
Dynamite	15	15	45	25
PETN	17	14	41	28
TATP	25	25	25	25

Methodology

MCNP5 Model description

Monte-Carlo stochastic method was used through the MCNP5 code to simulate the 'Romasha' setup and calculate gamma fluxes emitted from explosives and shielded explosives. To use this model, it has to be validated first. For the validation purpose, carbon-12 sample was simulated and gamma fluxes were computed after irradiating it by a beam of fast neutrons. The used model is shown in figure 5.

As shown before in the 'Romasha' set up, figure 5 shows the MCNP5 model. The ING-27 neutron generator modeled as a neutron point source of energy 14.1 MeV at a distance 9.5 cm from the iron collimator/shield.

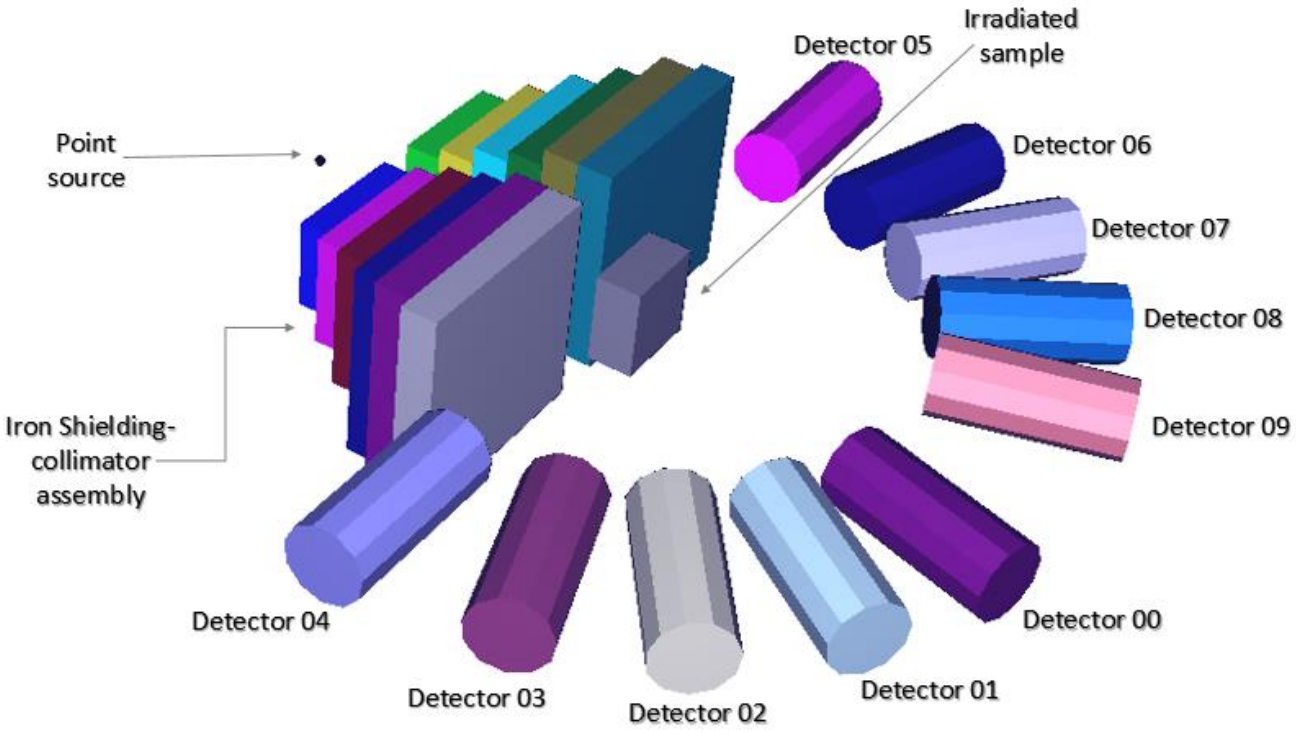
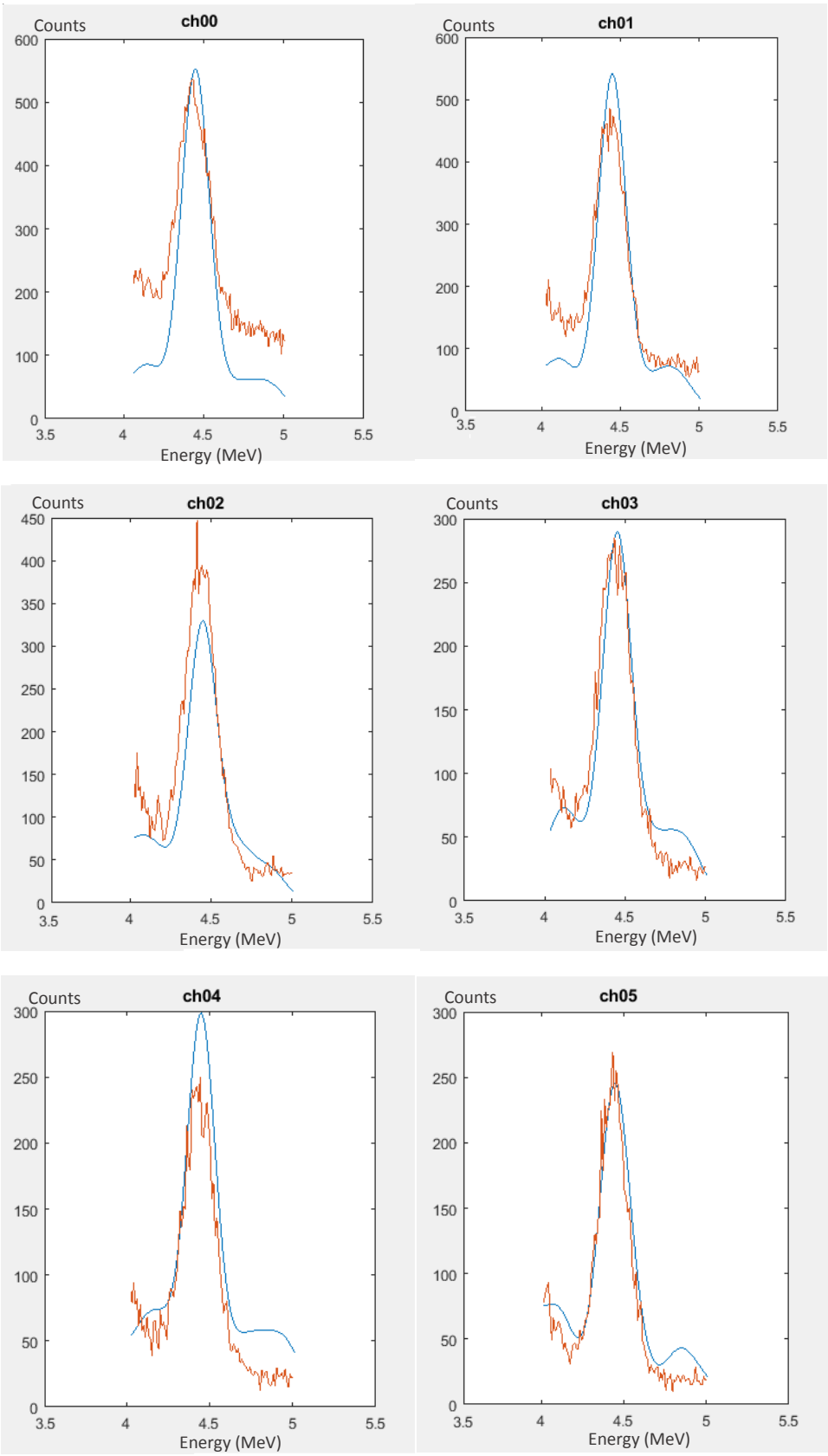


Figure 5: MCNP5 model for the 'Romasha' setup

Validation

Experimental data was obtained by data acquisition system for an hour of irradiation of the carbon-12 sample. Monte-Carlo results were calculated for the same duration. The model was validated on the experimental 4.44 MeV characteristic peak of C-12. The model results and the validation error for each channel are demonstrated in figure 6, and table 3 respectively.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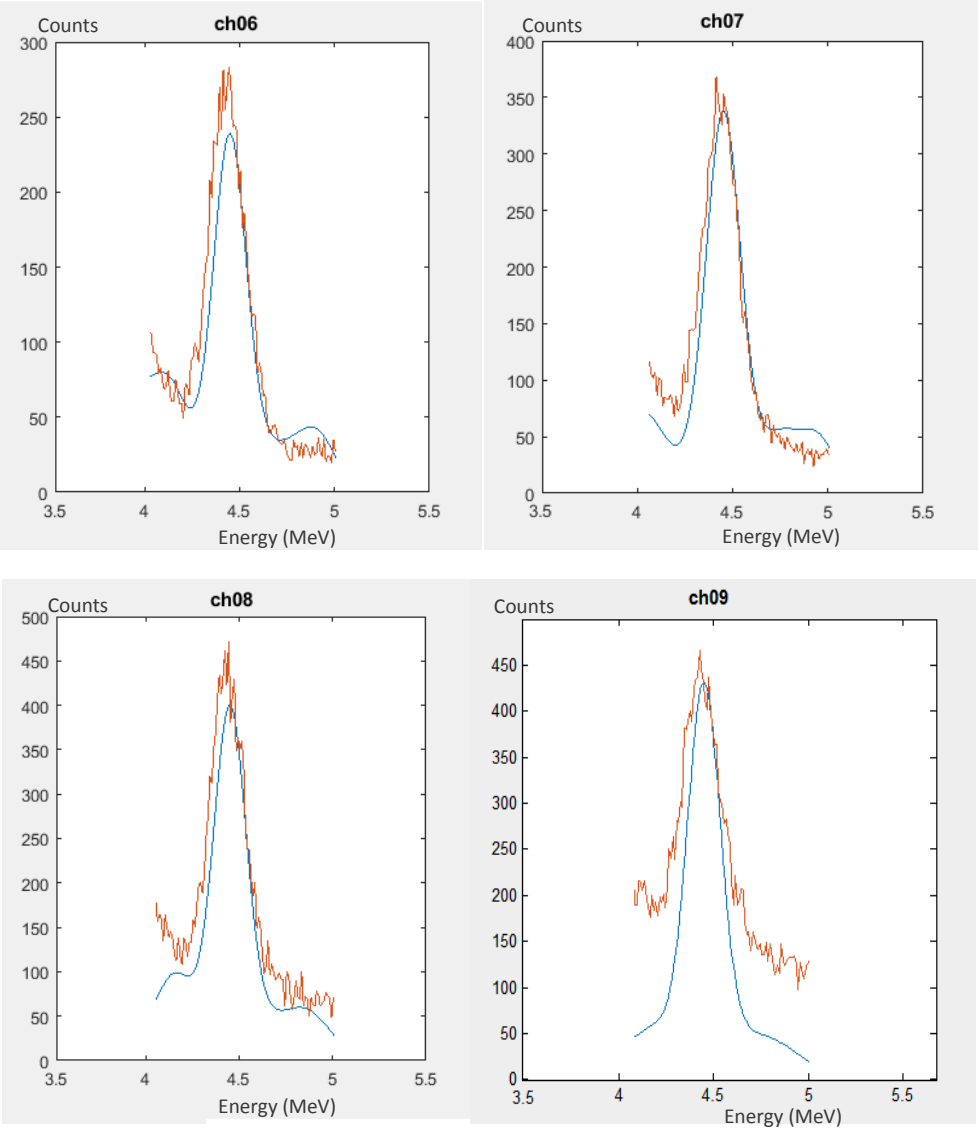

Figure 6: Experimental (orange) and MCNP5 (blue) results comparison (referring to detectors as channels)

Figure 6 shows the coincidence between the experimental results and the Monte-Carlo MCNP5 results. The MCNP5 results were fitted using Gaussian fitting. Table 3 shows the peak energy counts for both the experimental results and the MCNP5 results at 4.435 MeV emphasizing on the difference between them. The full width half maximum (FWHM) and resolution varies slightly from one detector to another as shown in table 3.

Table 3: Experimental and MCNP5 peak comparison for 10 BGO detectors, FWHM and resolution

Detector number	Peak counts of experimental results	Peak counts of MCNP5 results	ΔE (difference in exp. And mcnp peak counts)	FWHM (MeV)	Resolution (%)
0	537	548.21	11.21	0.29	6.54
1	486	527.32	41.32	0.24	5.42
2	387	329.1	57.91	0.25	5.64
3	285	287.77	2.77	0.24	5.3
4	250	298.69	48.69	0.25	5.64
5	232	245.24	13.24	0.2	4.51
6	270	237.23	32.77	0.25	5.53
7	338	334.69	3.31	0.23	5.19
8	423	404.19	18.82	0.25	5.53
9	431	420.66	10.34	0.34	7.56

It is observed from table 3 that the MCNP5 curves for detectors 03, 05, 07 have the highest accuracy validated on the experimental results with minimal difference between the peak energy of both. Both experimental and MCNP5 have approximately the same resolution with an average 5.86 % which is much better than the 8% resolution obtained from the NaI detectors in the previous 'Romashka' setup.

Detection of explosives

Based on the previously described 'Romasha' setup, MCNP5 code was used placing the explosives in the irradiated sample position for one minute of irradiation using neutron beam of from ING-27 neutron generator as shown in figure 5.

First, a TNT sample of volume 10 x 10 x 5 was tested. It was observed that only the carbon characteristic peak at 4.44 MeV and oxygen characteristic peak at 6.13 MeV were observed clearly. This is because these two peaks result from inelastic scattering with fast neutrons and since there is no moderator present between the neutron source and the tested sample, almost all neutrons reaching the sample are fast neutrons.

A modification was made in the used MCNP5 model by adding 5 cm of polyethylene in each side of the iron shielding/collimator assembly as shown in figure 7.

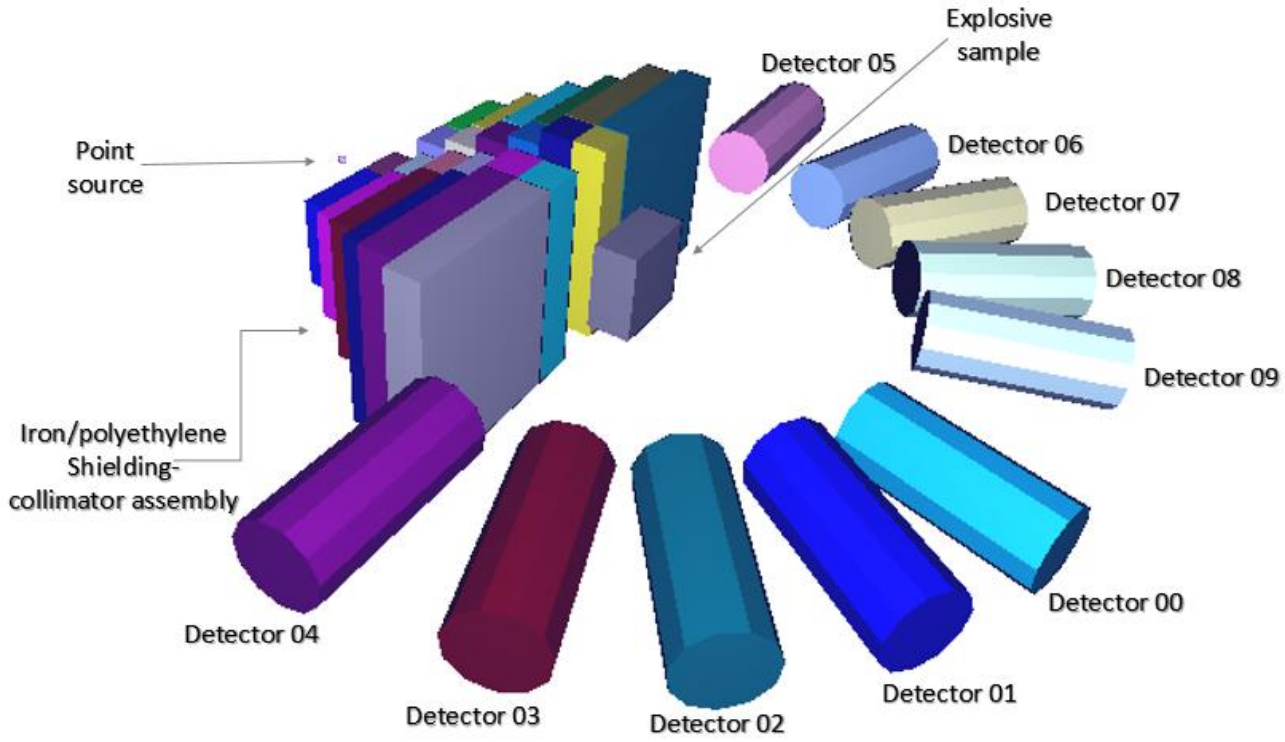

Figure 7: Modified 'Romasha' model with polyethylene in the iron shielding- collimator

The new composition for the shielding/collimator enhanced the results calculated. The effect of the iron-polyethylene shielding/collimator assembly was computed and eliminated from the results to calculate clearly the effect of the TNT sample. Other types of explosives were tested: cyclotol, PETN, TATP, dynamite.

To understand the behavior of neutron interaction within a compound material better, curves for each pure element (C, N, O, H) were obtained using the same configuration used before in figure 5.

Table 4 specifies each peak to a certain element to be able to understand the composition of the tested object and know the elements corresponding to the given peaks.

Table 2: Energy and the element it corresponds to

Energy (MeV)	Element
1.64	N
2.32	N
2.75	O
3.69	O
3.86	O
4.44	C
5.11	N
6.13	O
6.93	O
7.12	O

Taking TNT as an example, each curve of the four elements was multiplied by its proportion in a given explosive shown in table 2, then they were summed and demonstrated. This was done to show is the difference between the pure elements neutron interaction behavior and their behavior within a compound.

Detection of Shielded Explosives

After testing the behavior of explosives and their composition effects, shielded explosives were tested. A 10 x 10 x 5 TNT sample was used with a shielding of 1 cm surrounding it from all sides as shown in figure 8.

Three types of shields were used: natural boron, water, borated polyethylene with 3 % boron.

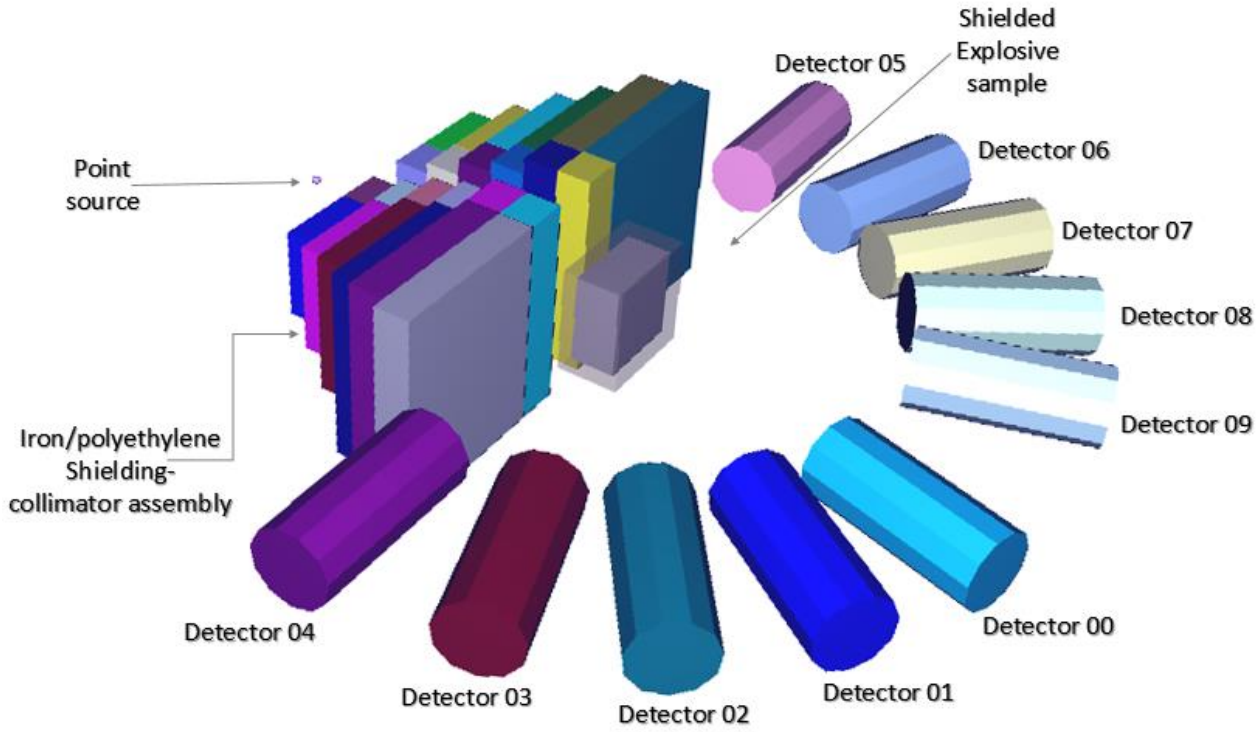

Figure 8: 'Romasha' setup irradiating a shielded sample

Results

After adding the polyethylene to the iron shielding-collimator, gamma peaks were obtained from MCNP5 model for different explosives (demonstrated in figures 9, 10, 11, 12, 13).

Carbon, oxygen peaks due to inelastic scattering with fast neutrons were visible and also oxygen and nitrogen radiative capture peaks were visible due to moderation of neutrons. Hydrogen 2.22 MeV peak was not clearly seen because of the much higher capture probability inside the polyethylene shield after moderation. The hydrogen peak was seen very clearly if there was no sample present; only due to the polyethylene present in the shield.

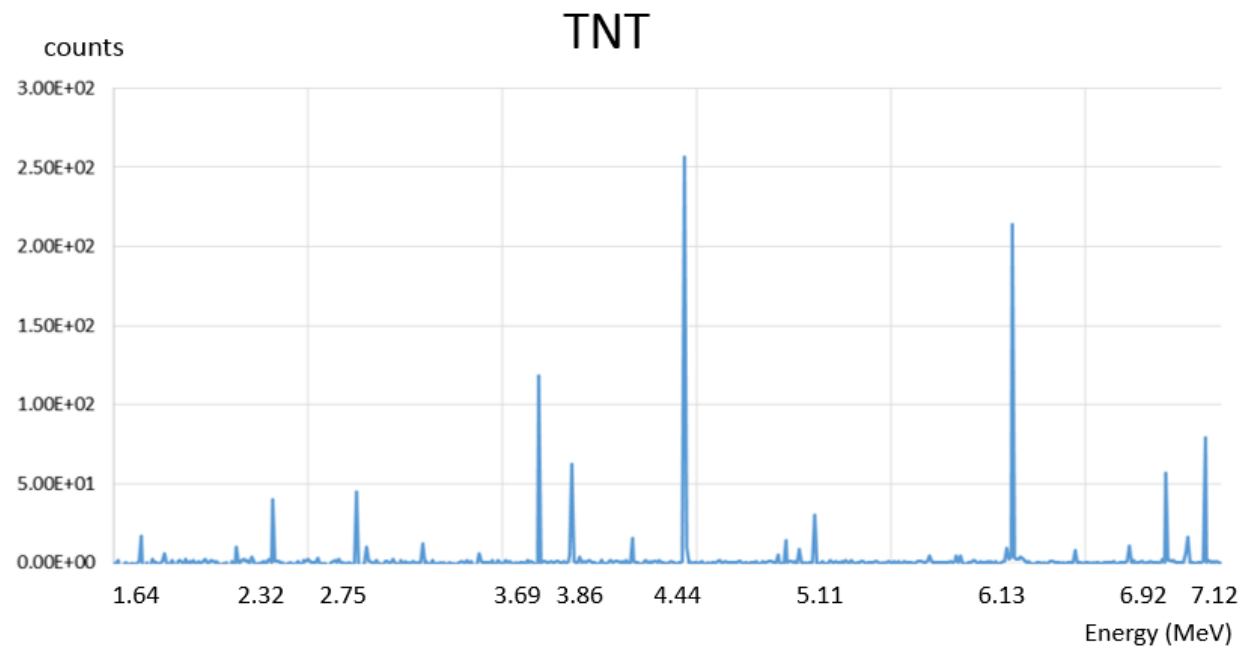

Figure 9: TNT gamma peaks

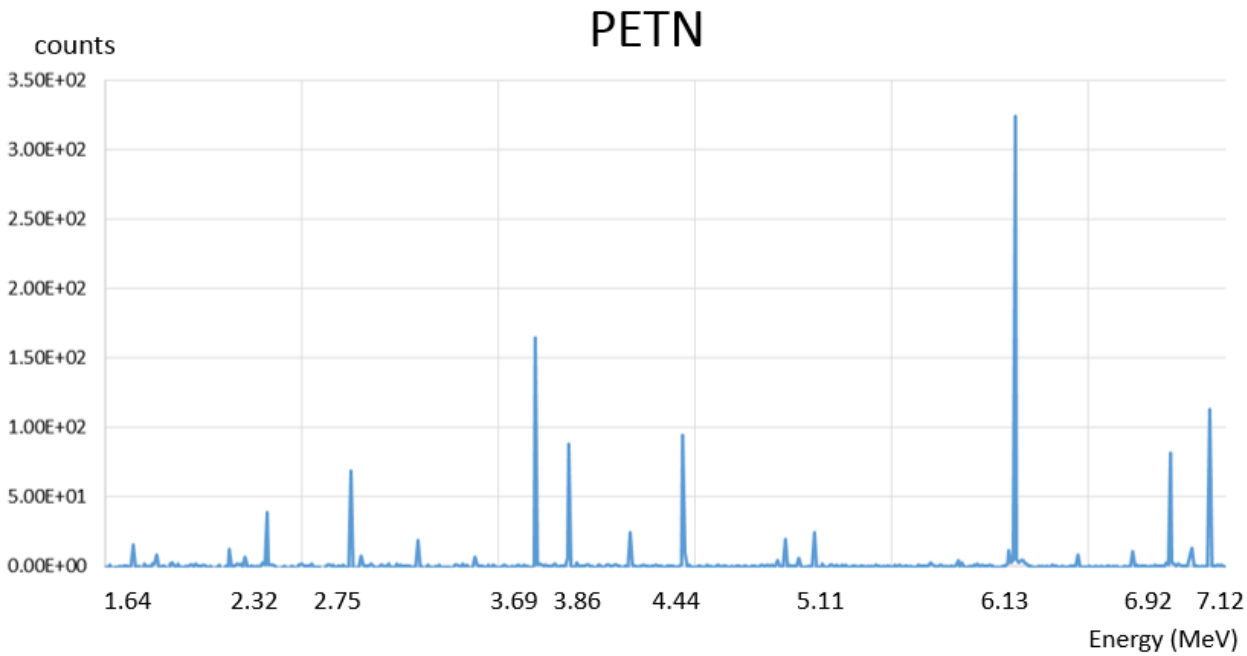

Figure 10: PETN gamma peaks

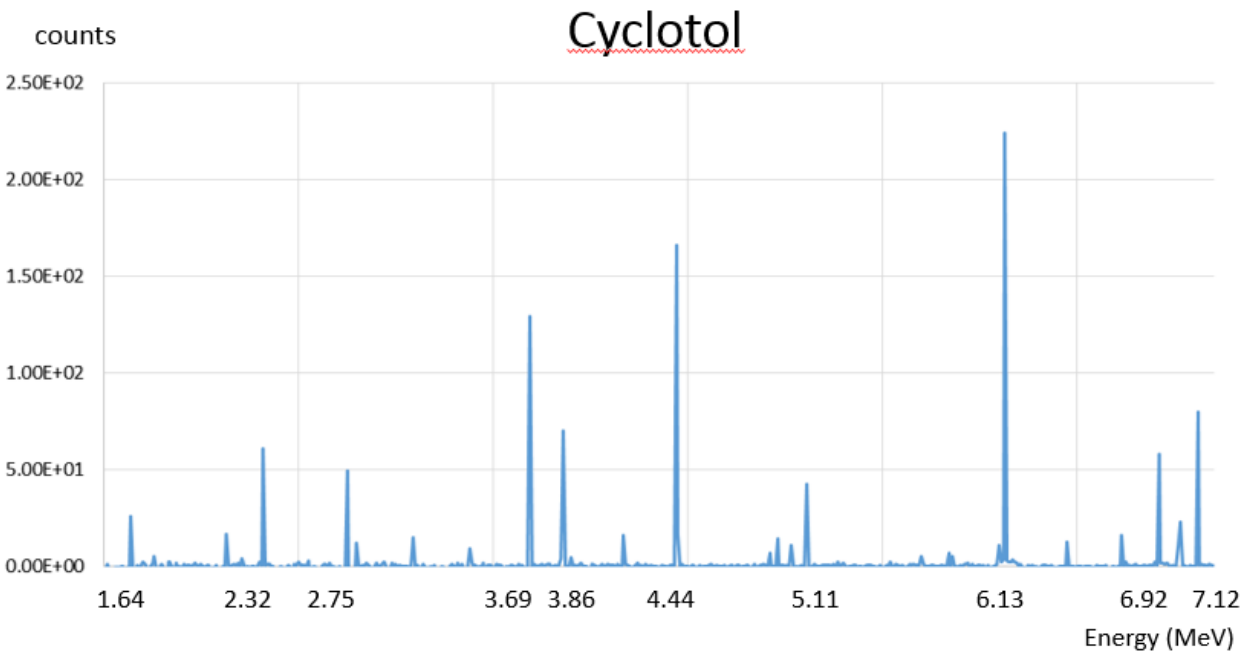

Figure 11: Cyclotol gamma peaks

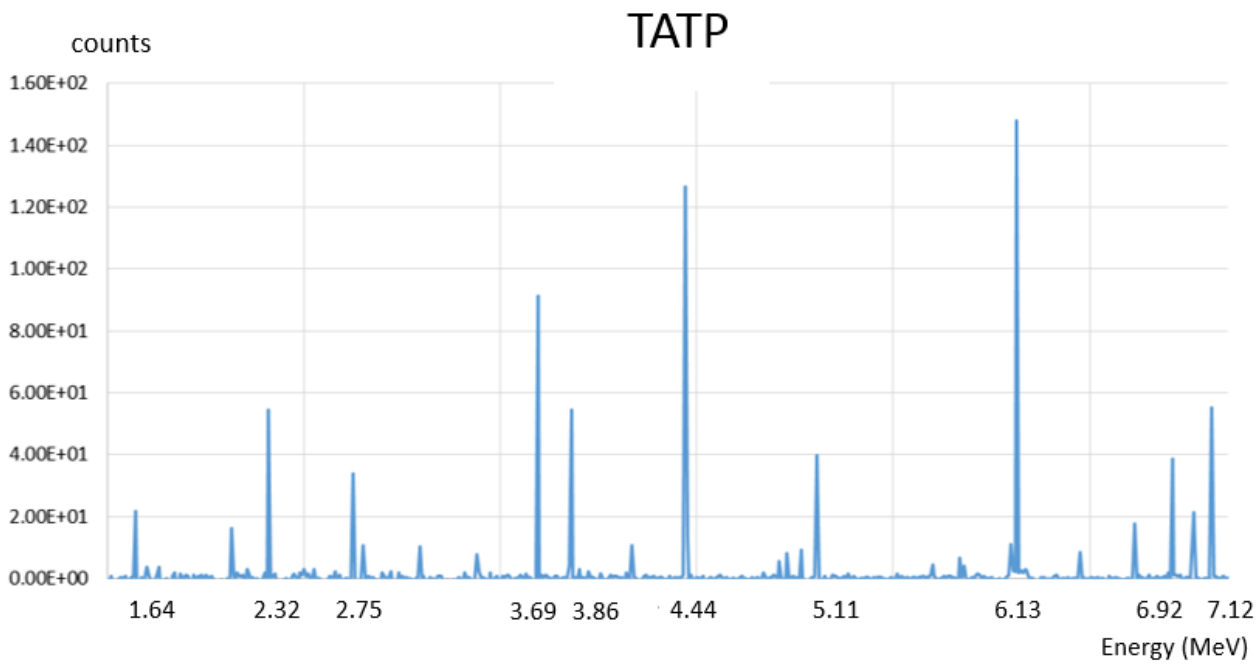

Figure 12: TATP gamma peaks

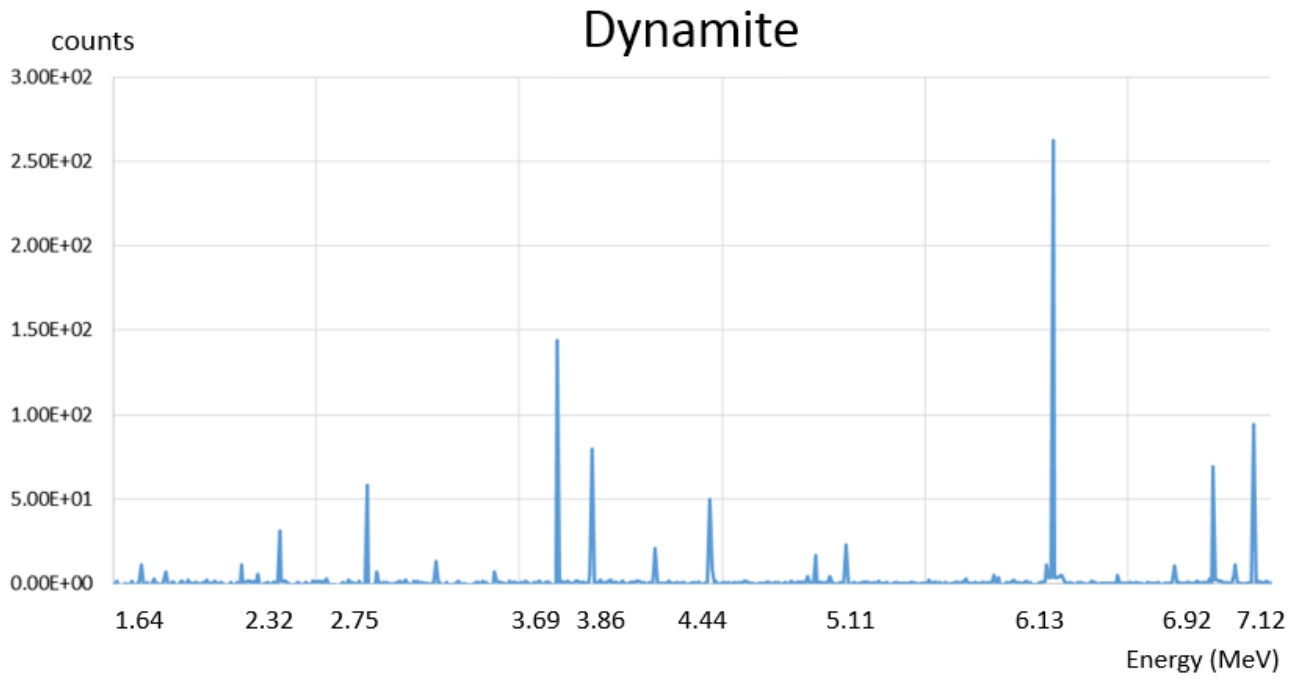

Figure 13: Dynamite gamma peaks

From figures 9, 10, 11, 12, 13, table 5 shows the ratios of the carbon characteristic peak at 4.44 MeV to oxygen peaks and nitrogen peaks. Knowing the approximate ratios can be an indication of the type of explosive being investigated.

Table 3: Carbon(4.44) to nitrogen and oxygen peaks ratios in different explosives

Explosive	**C : N (1.64)**	**C : N (2.23)**	**C : N (5.11)**	**C : O (2.75)**	**C : O (3.69)**	**C : O (3.86)**	**C : O (6.13)**	**C : O (6.92)**	**C : O (7.12)**
TNT	15.57	6.44	8.566	5.698	2.177	4.118	1.2	4.52	3.24
Cyclotol	6.434	2.734	3.9	3.37	1.286	2.37	0.74	2.87	2.077
Dynamite	4.5	1.618	2.183	0.86	0.347	0.63	0.19	0.72	0.533
PETN	5.92	2.4	3.84	1.375	0.57	1.066	0.29	1.147	0.83
TATP	5.77	2.311	3.18	3.738	1.38	2.311	0.85	3.255	2.28

TNT mixture of proportions of pure elements curves was calculated and compared with the real TNT curve shown previously in figure 9. Figure 14 demonstrates the differences between the two curves.

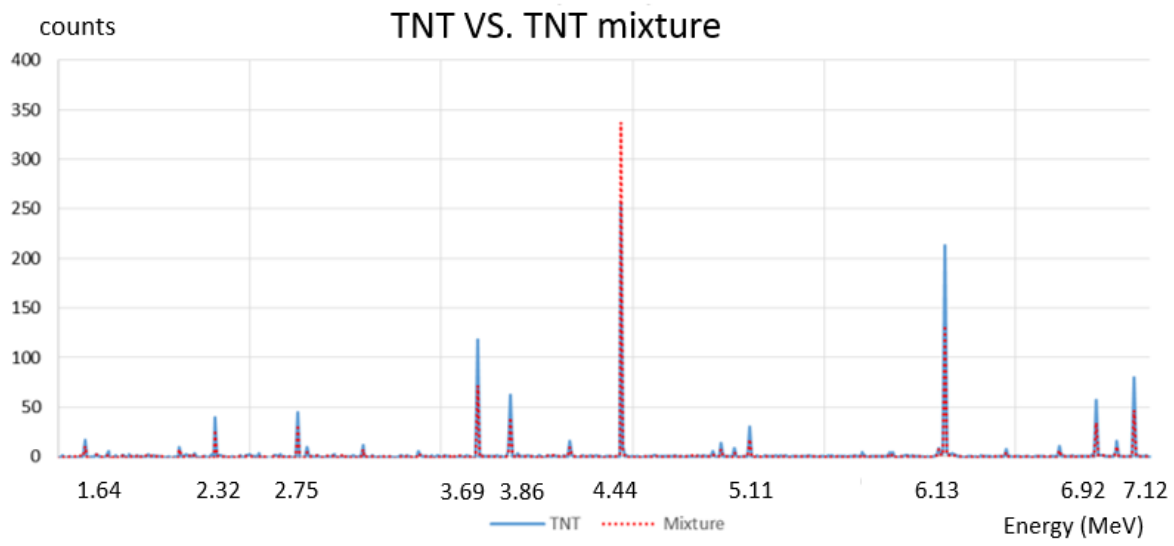

Figure 14: TNT vs TNT proportion mixture gamma curves

It is observed that all peaks are higher in case of the real TNT sample except for the carbon peak at 4.44 MeV. This is because in real TNT sample neutrons undergo some moderation through the light elements and through hydrogen mainly which gives rise to higher neutron capture probability, thus shows higher peaks. On the other hand, carbon 4.44 MeV peak results from inelastic scattering of high energy neutrons, therefore due to moderation its probability decreased.

After adding boron, water, borated polyethylene shields respectively to the TNT sample, results obtained are shown in figure 15, 16, 17.

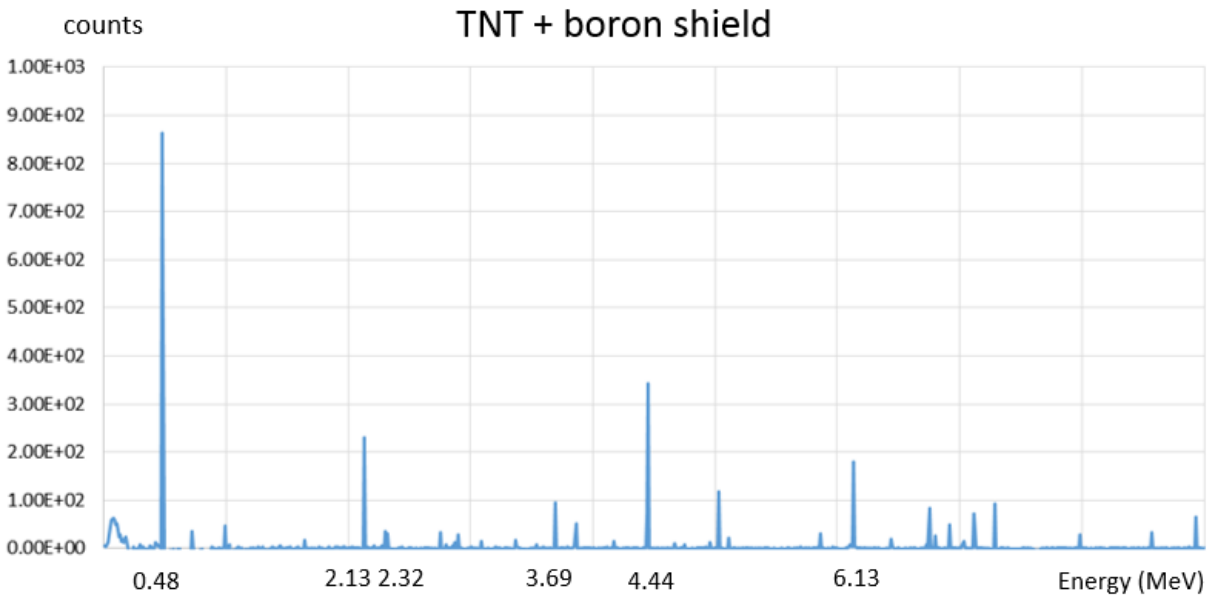

Figure 15: TNT + Boron shield gamma peaks

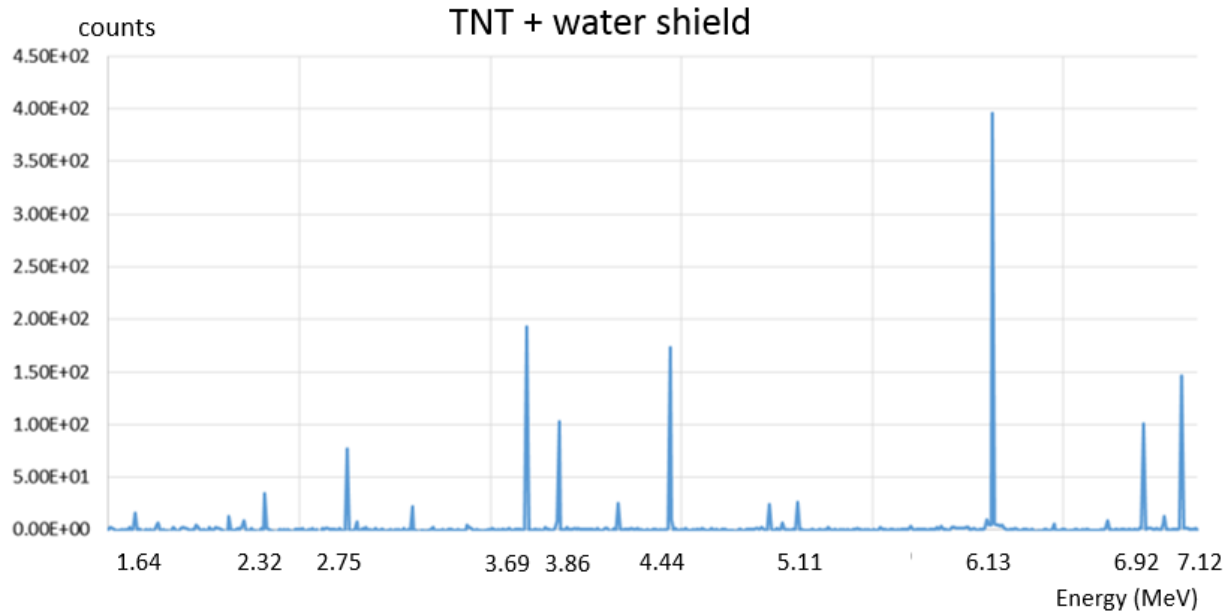

Figure 16: TNT + water shield gamma peaks

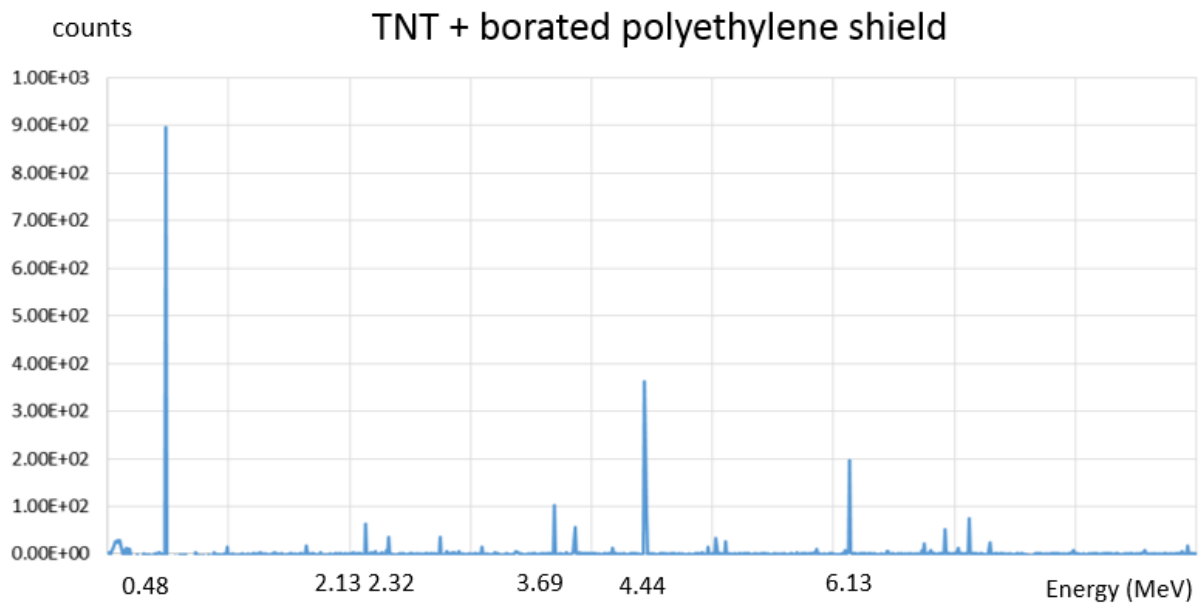

Figure 17: Borated polyethylene gamma peaks

In figure 15, TNT with boron shield shows carbon, oxygen peaks and traces of nitrogen peaks along with characteristic boron peak at 0.48 MeV and at 2.13 MeV. This indicates the presence of an explosive inside boron shield.

In figure 16, TNT with water shield shows carbon, oxygen and nitrogen peaks significantly, this is due to water composition of oxygen which heightens the oxygen peaks and hydrogen which moderates neutrons so they are captured more in nitrogen. It is not significant as a shield on its own but we can still detect the explosive elements.

In figure 17, TNT with borated polyethylene shield shows carbon, oxygen and nitrogen peaks as well as boron peaks. This way the presence of an explosive is detected along with a boronated shield.

Conclusion

In conclusion of the previously demonstrated results, it was proven that adding polyethylene to the shielding-collimator was essential for the detection of explosives and shielded explosives possible by having a wide range of neutron energy spectrum where each energy interacts with every isotope with different cross section. This modification allows the interaction of neutron with matter through inelastic scattering and radiative capture producing different characteristic gamma rays. Different types of explosives can be detected and distinguished from one another. Different interaction phenomena can be analyzed and understood through adding polyethylene to the iron shielding-collimator in the 'Romasha' setup.

Future research

In future, this methodology can to be used to:

1- Detect other illicit materials.
2- Determining the minimum detectable mass for illicit material.
3- Determining the thickness limit of the shield for detection of illicit materials.

Acknowledgement

I would like to thank my supervisor Dr. Vadim Skoy for his patience, support and guidance. Also, I would like to thank the technical assistant Dimitar Grozdanov.

I would also like to express gratitude to Dr. Dorota Chudoba, Ms. Julia Rybachuk and Ms. Elena Karpova for their support and help throughout my time in Dubna.

Also, I would like to thank the University Center of JINR and Frank Laboratory for neutron physics for giving me this summer student program opportunity and for their financial support of the summer program and my accommodation in Dubna.

Finally, I would like to give special thanks to Karim Hossny, my mentor in Alexandria for giving me the basics to start a research and for his never ending support.

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