Preliminary study of prompt gamma neutron activation analysis for Melamine

Muhamad Noor Izwan Ishak1, a), Susan Maria Sipaun1, Normanshah Dahing1, Haerie Hassan1, Nurliyana Abdullah1

1Plant Assessment Technology (PAT) Group, Industrial Technology Division, Malaysian Nuclear Agency, Bangi, 43000 Kajang, Selangor, Malaysia

a)noorizwan@nm.gov.my

Abstract. Prompt gamma neutron activation (PGNA) technique can be applied for detecting explosives in hidden places by using spontaneous fission source 252Cf source or neutron generator that emits thermal and cold neutrons. Some of emitted neutrons will be captured by nuclei and results in production of prompt gamma rays. Explosive materials can be identified by observing prompt gamma energy spectrum using high purity germanium (HPGe) detector. Gamma energy peaks indicate the presence of corresponding explosive nuclei in scanned materials. This paper describes the experiments conducted to investigate the possibility of explosive detection by using PGNA with 241Am/Be neutron source and limitations that come with it.

1. Introduction

The threat of explosive devices on military and civilian is one of the most important issues in many countries. The detection of explosives in hidden places has become an important aspect of the problem. Prompt gamma neutron activation analysis is capable of detecting elements in explosive item. It also offers the possibility of measuring the elemental density of most elements in inspecting materials. Melamine (C₃H₆N₆) contains 67% nitrogen by mass. Melamine is used as dummy explosives sample in this experiment due to its chemical formula. Nitrogen (N) in melamine is a key element of chemical explosives as shown in Table 1 below.

Table 1. Some Representative Common High Explosives and Their Compositions.

| Explosives                  | Formula     | wt % C | wt % H | wt % N | wt % O   |
|-----------------------------|-------------|--------|--------|--------|----------|
| Ammonium nitrate (AN)       | H₃N₆O₃     | 0      | 5.04   | 35.01  | 59.97    |
| Ammonium picrate (Expl D)   | C₆H₇N₆O₇   | 29.28  | 2.46   | 22.76  | 45.5     |
| Cyclonite (RDX)             | C₅H₁₀N₄O₆  | 16.22  | 2.72   | 37.84  | 43.22    |
| Pentaerythritol tetranitrate (PETN) | C₅H₁₀N₄O₁₂ | 19.00  | 2.55   | 17.72  | 60.73    |
| Trinitrotoluene (TNT)        | C₇H₅N₃O₆  | 37.02  | 2.22   | 18.5   | 42.26    |
Figure 1. The process of neutron capture by target nucleus followed by the emission of prompt gamma ray, delayed gamma and beta particle.

The basic principle of prompt gamma activation analysis techniques is illustrated in Figure 1 [1]. When neutron emitted by a neutron source (i.e. $^{252}$Cf) enter a medium of the target, depending on the type of material, neutrons can pass through with long distance before interacting with the nucleus. Neutrons can interact with the nuclei of materials in two ways to yield characteristic prompt gamma-ray emission. First, fast neutron (> 1 MeV) can scatter elastically from a nucleus, losing energy in the process and exciting the nucleus to a higher energy state. Immediate excitation of the nucleus produces a gamma-ray characteristic of the excited nucleus. Next, neutrons with energy around the thermal value (0.025 eV) can be captured by a nucleus. A compound nucleus is formed in an excited state and the prompt de-excitation of this capture nucleus causes the emission of a characteristic capture gamma-ray. The presence of a gamma-ray line with a particular energy is used for qualitative analysis. In addition, the intensity of the line is used for quantitative analysis. The range of gamma-ray in a typical bulk medium is approximately 20 to 25 cm.

2. Materials and method

The main components of PGNAA used in this research are High Purity Germanium (HPGe) semiconductor detector, neutron beam source (usually Cf-252 or Am Be) and multichannel analyzer system (MCA). MCA is calibrated using Co-60 and C-137 energy peaks as the reference energy. Sometimes, MCA is calibrated using hydrogen peaks especially on site. Prompt gamma energy peaks formation depends on: (1) The concentration of the elements near the detector, (2) Neutron absorption cross section (unit: barns) of elements. The high cross section of H nucleus (0.3326 barn) causes the energy peak 2223 eV to be detected easier than other energy peaks, (3) Thermal or cold neutron flux emitted from neutron source, and (3) Geometrical alignment of the 3 components and sample. PGNAA system is also simulated using Particle and Heavy Ion Transport System (PHITS) code by using pure Si, C, and N$_2$ sample in ideal environment.

The multi-element samples will produce multi-energy prompt gamma rays. This is a complex spectrum, which typically contains several hundreds of peak energies. Therefore, if the background is omitted, the most easily determined element is the one which has highest abundance, cross-sections and the intensity of gamma ray. Detection of prompt gamma with certain energy is used for qualitative analysis and the intensity of gamma rays is used for quantitative analysis. Among the advantages of this technique compared with conventional techniques is that there is no damage to the sample, preparation is simple, in-situ measurement, portable and uses only low radioactivity of radioactive sources in the range millicurie (mCi). The prompt gamma spectrum is obtained in neutron flux quantity and compared with experimental results.
3. Results and discussion

Figure 4 and 5 show results from the simulation of mono energetic 0.025 eV neutron beam bombarded in a direction toward melamine cube melamine sample with dimension 2x2x2 cm. It was observed that neutron scattered in all directions as a result of elastic collision with the nuclei contained in melamine. Figure 5 shows the production of prompt gamma rays as a result of neutron capture. The ratio of the number of prompt gamma produced to the number of neutrons collided with melamine is proportional to the neutron capture cross section of nuclei in melamine.

Prompt gamma spectrum from both PHITS simulation and PGNAA experiment are shown in figure 6 and 7 for silicon sample. More energy peaks for silicon nuclei are observed in simulation compared to experiment due to low neutron flux and short scanning time (30 minutes). Other energy peaks in the spectrum are formed due to other elements in the surrounding and background radiation.

Results for simulation with carbon and nitrogen samples are shown in figure 8 and 9. The simulation with these two elements are implemented for comparison with melamine sample due to its chemical formula. It can observe that energy peaks for carbon are easier to obtain compared to nitrogen, which needs longer scanning time or higher neutron flux. Hydrogen energy peak (2223 eV) from melamine sample in figure 10 caused other peaks to be not apparent enough to be observed.
Figure 4. Monoenergetic thermal neutron of 0.025 eV bombarded toward pure melamine cube 103 sample in vacuum space 100. Neutrons scattered across all direction after collision with melamine.

Figure 5. Prompt gamma produced from melamine sample and hitting Ge detector 104 as a result of simulation setup in Figure 4.
Figure 6. Prompt Gamma Spectrum from Si sample from PHITS simulation.

Figure 7. Gamma spectrum of sand measured during prompt gamma neutron activation in the energy range from 0 to 3700 keV for 30 minutes.
Figure 8. Prompt Gamma Spectrum from C sample from PHITS simulation.

Figure 9. Prompt Gamma Spectrum from N$_2$ sample from PHITS simulation.
Figure 10. Prompt Gamma Spectrum from C\(_3\)H\(_6\)N\(_6\) (melamine) sample from PHITS simulation.

Figure 11. Gamma spectrum of melamine measured during prompt gamma neutron activation in the energy range from 0 to 3700 keV for 30 minutes.

Six energy peak spectrum for N are obtained in Figure 11 representing nuclear reaction: \(^{14}\text{N}(n,n').\) The gamma peak observed at \(E_\gamma = 2223\) keV results from the \(1\text{H}(n,\gamma)\) reaction. It is difficult to obtain precise estimation of concentration of individual elements in sample considering low thermal and cold neutron flux from \(^{241}\text{Am}/\text{Be}\) neutron source and much longer detection time is needed to obtain more prompt gamma energy peaks having smaller neutron
absorption cross section. Background gamma radiation and surrounding materials can produce prompt gamma from neutron capture forming additional energy peaks. For the further research, improvisation of various parameters such as detection time, distance between source, detector and sample and source activities need to be taken into account. Other techniques like CHNS analyzer and XRF spectrometer will be considered to obtain mass composition of elements in the melamine sample before investigating the capability of PGNAA make the same assessment.

4. Conclusion
From the preliminary result, it can be concluded that PGNAA technique can be utilized to detect the presence of specific elements in melamine sample. Based on the application of PGNAA in research and industries, PGNAA can also use to study the elemental composition of a sample. This technique is nondestructive, with simple sample preparation, in-situ measurement, portable and use very low activity of radioactive. PGNAA instruments are capable to detect the presence explosive materials once the composition of elements of scanned sample (ex, C/H or N/C ratios) are determined. Although this sample need to be bombarded with high thermal neutron flux. Neutron generator or laser induced neutron source can be utilized for this purpose.

5. References
[1] Dahing L 2014 Non-destructive multi elemental analysis using prompt gamma neutron activation analysis techniques: Preliminary results for concrete sample AIP Conf. Proc. 1614 p 20.
[2] Viestia G, Pesentea S, Nebbiaa G, Lunardona M, Sudacb D, Nadb K, Blagusb S, Valkovićb V 2015 Detection of hidden explosives by using tagged neutron beams: Status and perspectives Nucl. Instr. and Meth. in Phys. Res. B: Beam Interactions with Mater. and Atoms 241(1–4) p 748-752
[3] Seabury E H, Caffrey A J 2006 Explosives detection and identification by PGNAA (Idaho Falls, Idaho: Idaho National Laboratory) p 83415.
[4] Lee S, Lee K, Cha H 2014 Activation analysis of indium, KCl, and melamine by using a laser-induced neutron source J. of the Korean Phys. Soc. 64(7) p 982-986.
[5] International Atomic Energy Agency 2006 Database of prompt gamma rays from slow neutron capture for elemental analysis (Vienna: International Atomic Energy Agency).
[6] Molnar Gabor L 2004 Handbook of prompt gamma activation analysis with neutron beams (Kluwer Academic Publishers) ISBN 978-1-4757-0997-1
[7] Ithnin H 2016 Modeling and simulation of portable prompt gamma neutron activation analysis (PGNAA) system using MCNPX code Int. Nucl. Sci. Tech. and Eng. Conf. 2018 iNuSTEC 2018 (Universiti Teknologi Mara).