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# A Study on Nuclear Specific Material Detection Technique Using Nuclear Resonance Reactions

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## Abstract

The non-destructive nuclear material detection technique is one of the novel methods under somewhat dangerous environments, for example, high level radiation or landmine areas. Specially, the detection of a landmine is a hot issue on the peaceful use of nuclear technology for human welfare. Generally, the explosives contain specific elements such as $^{14}$N or $^{35}$Cl. The photo-nuclear resonance gamma-rays are produced by nuclear reactions $^{13}$C(p , γ)$^{14}$N or $^{34}$S(p, γ)$^{35}$Cl in which target is bombarded by about 2MeV proton beam extracted from the proton accelerator. To avoid other neighboring resonant gamma-rays, we selected a higher resonant energy above 5MeV. The resonance gamma rays produced are absorbed or scattered when they react with $^{14}$N or $^{35}$Cl included in the mines and explosives. We can determine existence and position of mines or explosives by detecting the absorption and scattering gamma-ray signals.

## 1. Introduction

The non-destructive nuclear material detection technique is one of the novel methods under somewhat dangerous environments, such as high level radiation and landmine areas. Specially, the detection of landmine is a hot issue on the peaceful use of nuclear technology for human welfare.

Several methods of mine and explosive detection were suggested, for example, NQR(nuclear quadrupole resonance), metal survey meter and so on. However, while applied to an anti-personal mine which does not contain any metal components, a conventional method based on metal survey is not effective. The NQR method is deeply dependant on the large electric quadrupole moment compared with other stable elements underground, and it is known that NQR detection frequency varies with chemical composition and temperature.

In general, the explosives contain specific elements such as $^{14}$N or $^{35}$Cl. Our new landmine and explosive detection technique is to detect the nitrogen or chlorine nucleus directly by using the photo-nuclear resonant reactions $^{14}$N(γ, γ)$^{14}$N or $^{35}$Cl(γ, γ)$^{35}$Cl. The main goal of this work is to confirm our new detection mechanism in principle by performing a basic nuclear resonant reaction experiment. In this paper, the $^{14}$N detection method will be discussed in detail. The analogous method for the detection of $^{35}$Cl is also discussed.

## 2. The Proposed Detection Principle

The mine/explosive detection mechanism is in two steps.

Generation of $\gamma$ -ray

$$p + {}^{13}C({}^{34}S) \rightarrow {}^{14}N^*({}^{35}Cl^*) \rightarrow {}^{14}N({}^{35}Cl) + \gamma$$

Gamma resonance absorption and scattering

$$\gamma + {}^{14}N({}^{35}Cl) \rightarrow {}^{14}N^*({}^{35}Cl^*) \rightarrow {}^{14}N^*({}^{35}Cl^*) + \gamma \text{ or } {}^{13}C + p$$

The nuclear resonance gamma-rays are produced by nuclear reactions $^{13}$C(p, $\gamma$)$^{14}$N or $^{34}$S(p, $\gamma$)$^{35}$Cl in which the target is bombarded by about 2MeV proton beam extracted from the proton accelerator. To avoid other neighboring resonant gamma-rays, we selected a higher resonant energy above 5MeV. The resonance gamma rays produced are absorbed or scattered when they react with $^{14}$N or $^{35}$Cl nuclei included in the landmines and explosives. We can determine existence of landmines or explosives by measuring the absorption and scattering of gamma-rays.

In the study of explosives detection, we have surveyed the various present technologies and requirements. The explosive detector based on the gamma resonance reaction should be relatively superior to the present ones in accuracy. The gamma resonance absorption in $^{14}$N or $^{35}$Cl rich compounds was verified successfully by using a Tandem accelerator at low beam current. But for the verification of the field application, a higher beam current over 10mA is needed.

The major difficulty is that the energy of the absorbed and re-emitted gamma-rays from the $^{14}$N/$^{35}$Cl($\gamma$, $\gamma$)$^{14}$N/$^{35}$Cl reaction is almost the same as that of the target gamma-ray from the $^{13}$C/$^{34}$S(p, $\gamma$)$^{14}$N/$^{35}$Cl reaction. An over 30 cm thick Pb shield needed to overcome the $^{13}$C/$^{34}$S target gamma-ray yield, and it is big burden for the system designer as a movable machine. The gamma-rays for $^{14}$N detection are produced via nuclear reactions when the $^{13}$C target is bombarded by 1.75MeV proton beam. The 9.17MeV gamma-rays produced are absorbed or scattered when they react with $^{14}$N nuclei included in the mines and explosives. In the case of $^{35}$Cl detection, it is more difficult than $^{14}$N, because target material has to be enriched to produce pure resonant gamma-rays and the 168 resonance states are near the 2MeV proton energy region. In any case, the re-emitted gamma-rays from the specific nuclei can give a position information of landmines or explosives.

## 3. Numerical Estimation Based on Nuclear Data

Before performing the experiment, we did an estimation calculation for our new detection principle. The properties of a 9.17MeV gamma-ray production $^{13}$C target by 1.75MeV proton are summarized as the natural abundance being 1.11%, $J^{\pi}$ being $1/2^{-}$, density being 2.253 g/cm$^3$, dE/dx = 35.69 MeV/mm, and range being 30.02μm. The range and stopping power is calculated by the TRIM code. The resonance width was reported as $\Gamma$=122eV and $\Gamma_{\gamma}$=6.3eV [1] , and the calculated thick target yield as $0.64 \times 10^{-8}$ [γ/p]. However, it is not consistent with the experimental data.

The 9.17MeV production yield as a unit of [γ/p] varied with respect to several experiments, so in the simulation we used $0.88(21) \times 10^{-8}$ [γ/p], which was obtained from the average of some previous experiments, $0.74 \times 10^{-8}$ [2], $1.01 \times 10^{-8}$ [3], $1.01 \times 10^{-8}$ [4] and $0.63 \times 10^{-8}$ [5]. The underground condition was assumed to be the attenuation coefficient μ=0.0289/cm, and the explosive part of the anti-personal mine dimension is 1.5 cm or 5 cm radius and 1cm or 5cm height with a density of $^{14}$N element 0.5 g/cm$^3$.

## 4. Experiments for the Proof of Principle

### A. $^{14}$N Detection

Experiments were performed several times by using KIGAM 1.7MV Tandem and SNU-AMS 3MV Tandetron accelerators. The main experiments were performed using SNU-AMS Tandetron. Firstly, we reviewed previous experiments by using the NaI(Tl) detector with anti cosmic ray shielding by thick plastic detectors. We used a BGO scintillator to increase detection yield rather than a NaI(Tl) scintillator. At least 30 cm Pb shield was required to distinguish the real signal of mines from observed signals.

The protons were accelerated to 1.75MeV, and then the excitation function, the angular distribution(Fig. 1), the photo-absorption(Fig. 2), and the scattering were measured. The proton beam current was over 1μA, and bombarded on an enriched $^{13}$C target foil(99%) with a 121 μg/cm$^2$ thickness. The detectors were 3"×3" NaI(Tl) and BGO scintillators and a 2"×3" HP-Ge detector with BGO Anti-Compton Shields. The mine/explosive emulator was 10cm and 20cm thick melamine or liquid nitrogen.

There is an energy difference between the Doppler shifted gamma-ray from the $^{13}$C(p, γ)$^{14}$N reaction and the resonant absorbed and scattered gamma-ray from the $^{14}$N(γ, γ)$^{14}$N reaction, especially for the 9.17MeV resonance state gamma-ray. The Doppler shift effect as a function of detection angle with respect to a proton beam is shown in Fig. 3. The solid line is the calculated result.

We also developed a new detection technique to detect the scattered gamma-ray from the nitrogen target by using the energy difference between the Doppler effected gamma-ray from the $^{13}$C(p, γ)$^{14}$N reaction, and the resonant absorbed and scattered gamma-ray from the $^{14}$N(γ, γ)$^{14}$N reaction, especially the 9.17MeV resonance state gamma-ray. In this experiment, we used liquid nitrogen 20*l* as a resonant absorption mine/explosive emulator. We used a HP-Ge detector with 30% efficiency and less 2keV energy resolution. Fig. 4 is the observed spectrum near the single escape peak region.

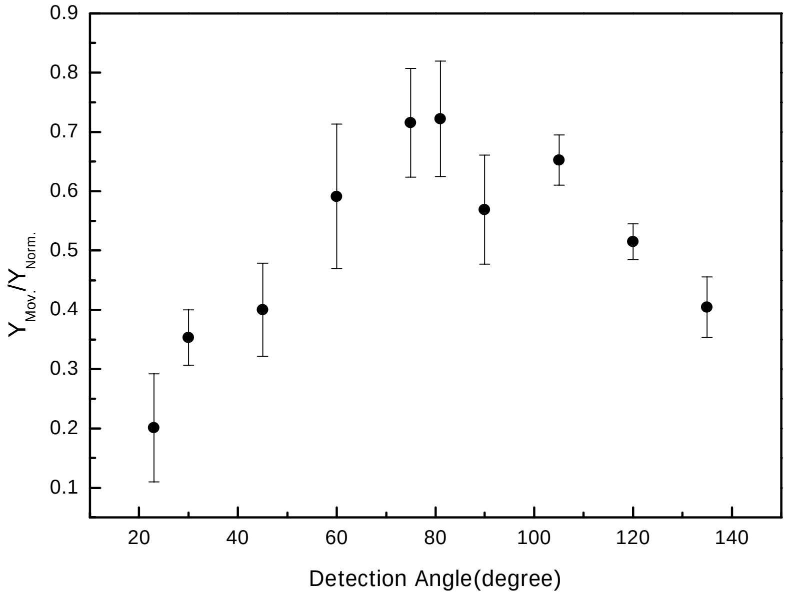

Fig. 1. The angular distribution of 9.17 MeV gamma-rays.

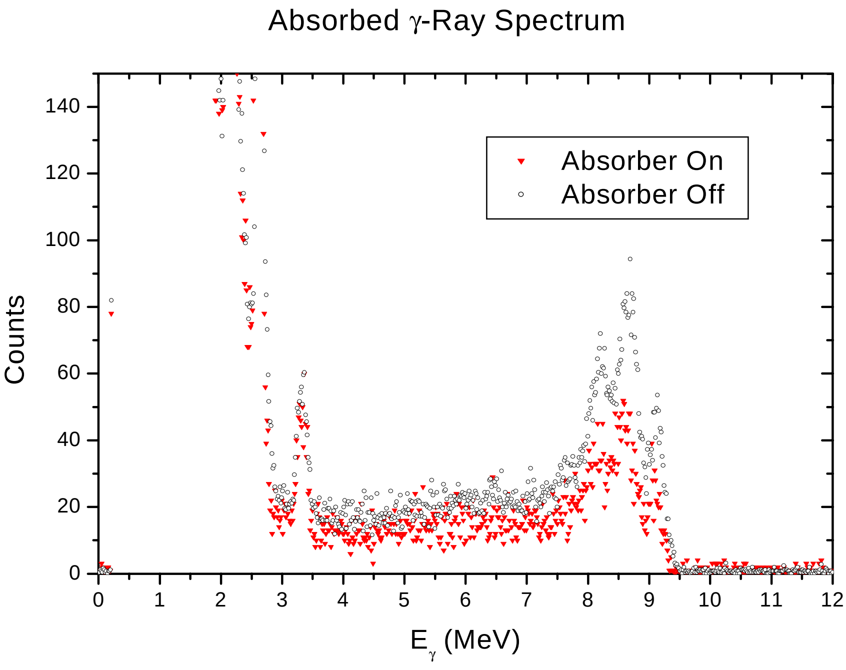

Fig. 2. Resonant absorption reaction spectrum for the melamine

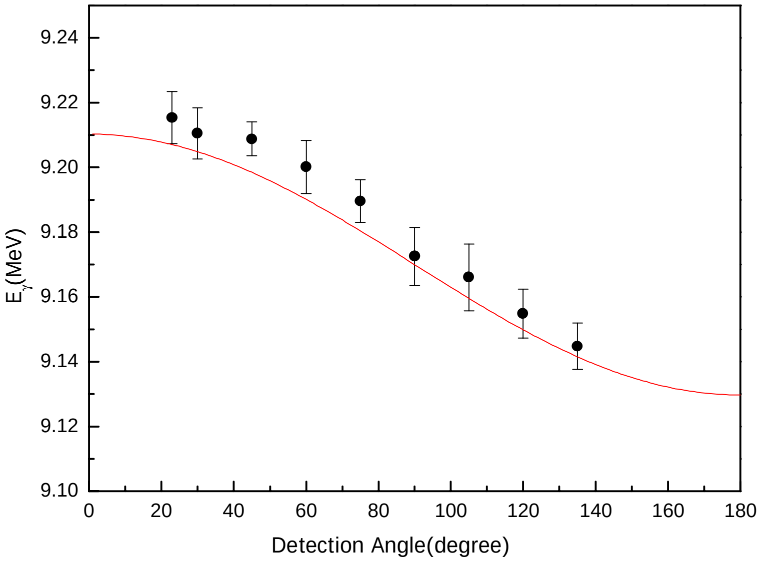

Fig. 3. Doppler effect due to the velocity of the compound nucleus. The solid line is a calculated result for the $^{13}C(p, \gamma)^{14}N$ reaction at 1.75 MeV proton energy.

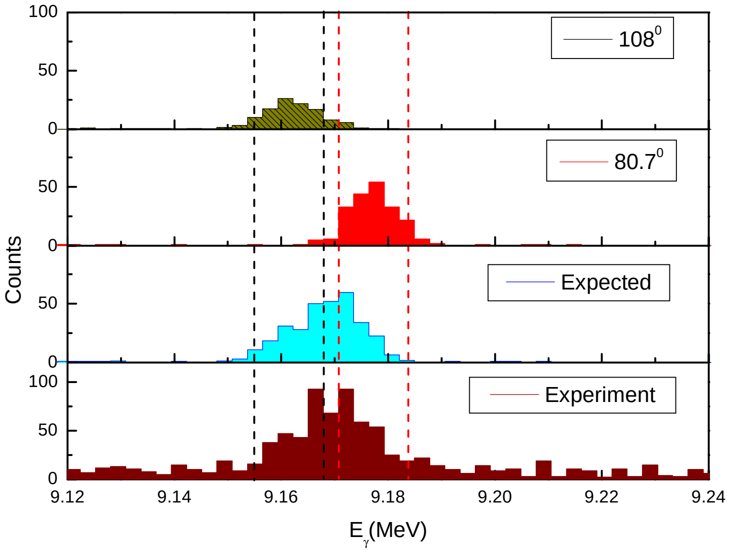

Fig. 4. The spectra measured for the photo-nucleus resonant reaction. Spectrum at 108° is measured at an off-resonance angle and spectrum at 80.7

B. $^{35}$Cl Detection

Almost similar experiment was performed for the $^{35}$Cl detection. The differences are the target material and the incident proton energy. The target was natural sulfur in which $^{34}$S is included as a 4.21% of sulfur. The incident proton energy was 2MeV, and the proton could populate lots of resonance states and resonant gamma-rays. The target was Zn$^{nat}$S with 3mm thickness. The liquid $CCl_4$ was used as a $^{35}$Cl absorber. The production yield was expected to be $7.6x10^{-10}$ $\gamma$/p for 100% enriched target[7, 8]. Figure 5 is the measured spectrum at the resonance angle of 83°.

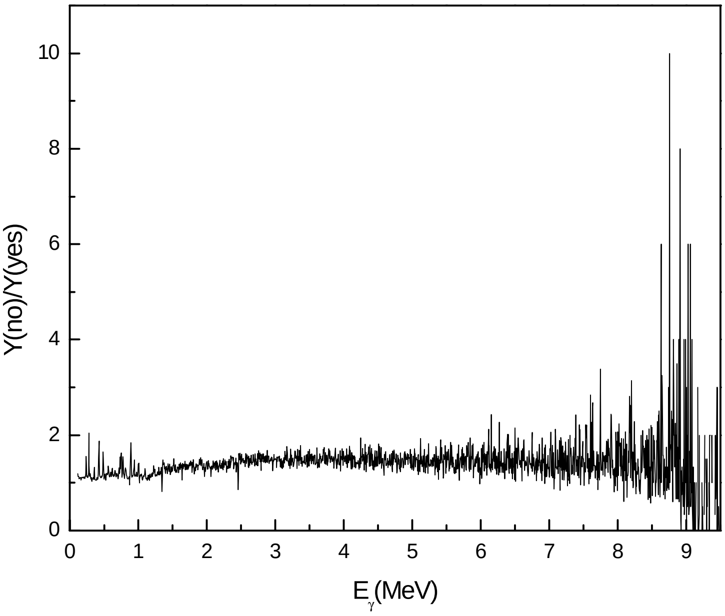

Fig. 5. The spectra measured for the photo-nucleus resonant reaction at an on-resonance angle.

## 5. Discussion

The resonant gamma-ray production yield of the thick target with 121μg/cm$^2$ is maximized near 1.745MeV, which agreed well with the previous result [1]. The derived resonant width from the experiment is 600eV of FWHM, and the compiled data is about 120eV [1]. However, the width data are not consistent with each other, and depend on the experimental method.

Even though the detection system based on the BGO scintillator has much more yield, about 100 times that of the HP-Ge detector, we wish to prove our principle by using the

Doppler shift effect. So we made a new system based on a HP-Ge detector, which guaranteed 2keV energy resolution at several MeV energies. This system did not need any shield in principle, but when performing the experiment a 10cm Pb brick was used between $^{13}$C target and HP-Ge detector. The major merit of this system is that we can separate the gamma-ray from $^{13}$C and mine/explosive emulator when we choose the detection angle of the mine gamma-ray properly. This method is deeply related to the Doppler effect, because the gamma-ray energies coming from the $^{13}$C target are a function of the detection angle, and the gamma-ray energy from the mine emulator is the resonant energy with a small recoil correction energy, about 6 keV in this $^{14}$N(γ, γ)$^{14}$N reaction. Such a gamma-ray energy deviation is maximized when the detection angle is far from the resonant angle.

The gamma-ray production and absorption process are inverse mechanisms, and the available angle of this mechanism is only 80.7°(0.7). This phenomenon results in the Doppler shift effect from the reaction kinematics. The difference from the Doppler shift effect between the forward and backward detection angle is about 130 keV. We also measured the angular distribution of the gamma-ray production by using the SNU-AMS tandetron accelerator. We used two HP-Ge detectors: one was used for the movable angular detector and the other was used for the normalization detector. Fortunately, the yield shows a maximum at the detection angle 90° near the resonant angle and the symmetric yield distribution.

The scattered gamma-ray yield from the $^{14}$N(γ, γ)$^{14}$N resonant reaction is somewhat less than the expected yield calculated by using previous data. The measured scattered yield is 1.8(0.4)/sec/10mA and the expected one is 4.2(2.5)/sec/10mA. When we calculated the yield, some assumptions were used that thick target yield is $1 \times 10^{-8}$ γ/p, HP-Ge detector absolute efficiency is 0.1% for the 59.2 mm(diameter) × 76.2mm(length) detector crystal dimension, and the attenuation is 30% due to liquid nitrogen Dewar[6].

The $^{35}$Cl case shows so many resonant re-emitted gamma-rays after passing absorber material above 8 MeV. This result shows that if target material is enriched and thickness is thin enough to select the each resonance state, the detection system can be utilized for the explosive detection. Now we are on further experimental study.

## 6. Conclusion

In the present work, we measured resonant energy and angular distribution, and photo-nucleus resonant gamma-rays from the $^{14}$N/$^{35}$Cl(γ, γ)$^{14}$N/$^{35}$Cl and $^{13}$C/$^{34}$S(p, γ)$^{14}$N/$^{35}$Cl reactions by using HP-Ge, BGO and NaI(Tl) Detectors. We proved a new detection principle and observed the scattered gamma-ray by using a new method based on energy difference originated from the Doppler shift effect. We have plans for further experiments at the SNU-AMS facility to confirm and collect meaningful physical nuclear data. The experimental setup will be a detector system shielded to suppress cosmic ray and gamma-ray from a target, including an angular positioning system. The proton beam current will be larger than 10μA. We will improve our data quality from a nuclear data point of view.

## Acknowledgements

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## References

[1] Table of Isotopes, 8th ed. (Wiley-Interscience City, 1996), Vol. I.
[2] S. S. Hanna and L. Meyer-Schutzmeister, Phys. Rev. 115(1959) 986
[3] J. D. Seagrave, Phys. Rev. 85 (1952) 197
[4] D. A. Close, NIM 113 (1973) 561
[5] LA-12777-MS, LANL Report(1994)
[6] Nuclear Reaction Analysis, J. B. Marion, F. C. Young(1967) 67
[7] Nuclear Physics A265 (1976) 416
[8] Nuclear Physics A265 (1976) 429