System Design of Photofission Reaction Ratio with High-energy Bremsstrahlung Photons for Nuclear Security

Kim Wei Chin[1], Hiroshi Sagara[1], Jun-ichi Hori[2], Yoshiyuki Takahashi[2], Chi Young Han[1]

[1] *Tokyo Institute of Technology*

[2] *Kyoto University*

Abstract

The applicability of the photofission reaction ratio (PFRR) method to identify high-enriched uranium is studied by switching the photon source from the Gaussian spectrum in extant studies to a bremsstrahlung spectrum. The principle of PFRR method is based on the difference of photofission cross section between ^{235}U and ^{238}U nuclides and it was validated using Gaussian photons in extant studies where PFRR increases linearly with uranium enrichment. A mock experiment was conducted using Californium-252 (Cf) neutron source as the reference target to estimate the minimum requirement of number of neutrons emission for detection by natural uranium target. The Cf-252 target used had the activity of 1.04×10^7 Bq and 1.21×10^6 neutrons/s. A 5” diameter x 5” thick organic liquid scintillator, BC-501A was used as the detector to measure neutron/gamma from Cf-252 source. To measure neutron in the mixed neutron/gamma field, electronic circuit with a PSD function was used. High voltage of 800V was supplied to the circuit and the signal is delayed using delay amplifier ORTEC 460 with 20x20 coarse gain and displayed on MCA coupled with rise time to height converter. A ROI of 21x85 on the contour plot of neutrons and gamma-rays distribution (512x512 arbitrary units) was defined as the region containing mostly neutrons. With 0.3% confidence, the minimum detectable activity for both natural background and background with beam operating at 18 MeV were 3.95 kBq and 10 kBq respectively. An accelerator experiment was conducted with natural uranium target with 18.0 MeV incident electron energy. The natural uranium target had a size of 4 x 4 x 0.6 cm, and the cross section of ^{238}U is estimated to be approximately 300 mb at peak around 14.0 MeV. As such, with the same ROI region defined, the natural uranium showed a fission rate larger than the minimum required reaction rate based on Cf-252 source for natural background.

Introduction

In the field of nuclear security and safeguards, many non-destructive assay (NDA) techniques have been developed where a few of them being successfully applied to mixed oxide powder, pellets, nitric solution of special nuclear materials (SNMs) in nuclear fuel cycles and uranium enrichment measurements [1]. The photofission reaction ratio (PFRR) method is an active NDA that can estimate uranium enrichment through the photofission reaction of fissionable nuclides using high-energy photons. It is based on the characteristic difference between the photofission cross-sections of different fissionable nuclides. The PFRR principle has been proposed and verified using an ideal Gaussian photon source simulation in extant studies [2, 3]. The feasibility of PFRR for detecting high-enriched uranium (HEU) using bremsstrahlung photons was validated since bremsstrahlung photon source can be realized on a practical scale and in portable size comparing to Gaussian source that requires costly laser Compton scattering instrument. And the ratio of the photofission reaction rate of the uranium target by a higher bremsstrahlung photon energy to that of a lower photon energy one (higher than the photofission threshold energy) exhibits a linearly increasing relationship with uranium enrichment [4-7]. One of the practical applications of this method is the detection of the presence of highly-enriched uranium in cargo containers crossing national borders, which can be achieved by injecting photon beams with two different energies into the shielded cargo containers, and the resulting ratio of photofission reaction is proportional to the uranium enrichment value. To examine the detectability of the neutrons emitted from photofission reaction by uranium target, an experiment was carried out at the linear accelerator facility of Kyoto University, KURRI using Cf-252 source as the reference target. In the present paper, the analysis of the minimum detectable activity based on statistical approach is presented.

PFRR Principle

The PFRR principle is heavily dependent on the photonuclear cross sections of different nuclides. The photonuclear cross sections against the incident photon energies for uranium and thorium isotopes from the ENDF/B-VII.1 nuclear data library is shown in Figure 1.

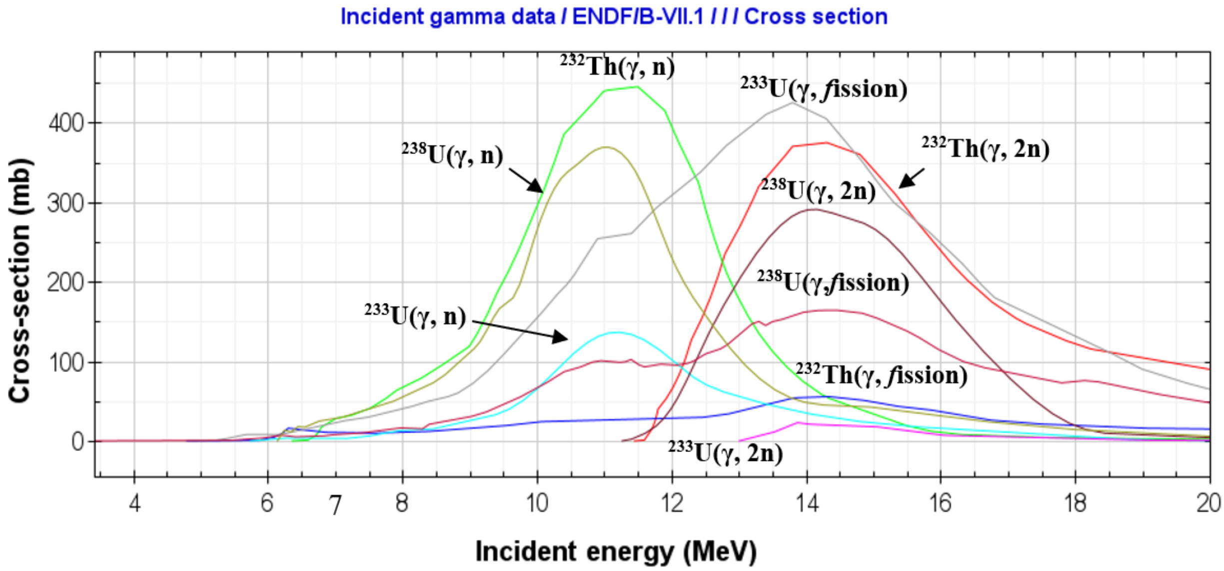


Figure 1: Photonuclear reaction cross sections against incident photon energies.

The photofission reaction occurred in a heavy metal target can be represented with the equation as follows:

$$R = N \int_{E_{thres}}^{E_{max}} \varphi(E)\sigma_f(E)\,dE \qquad \text{Eq. 1}$$

Where σ_f is the photofission cross section, φ is the photon flux and N is the atomic number density. The threshold energy for photofission reaction of both ^{235}U and ^{238}U isotopes are approximately 3.25 MeV while the maximum energy refers to the photon source energy. Based on a previous study, the combination of 6.0 and 11.0 MeV Gaussian photon energy was selected to measure uranium enrichment since the difference in photofission cross sections between ^{235}U and ^{238}U nuclides at these two energies vary significantly [2, 3]. The effort to reproduce the similar photofission reaction amount to that of Gaussian source by bremsstrahlung photons led to parameter survey of appropriate incident electron energies was carried out too [4-7]. And as a result, 7.0 and 13.5 MeV bremsstrahlung electron energies were selected after considering several factors including the measurement noise contributed from multiple neutrons reaction, (γ, 2n) [4].

Methodology

1. Bremsstrahlung photon source creation

The radiative stopping power of braking radiation, or bremsstrahlung is directly proportional to the particle's energy and square of the atomic number (Z) of the absorber, whereas the ionization stopping power decreases with particle energy and increases only with the first power of Z. In other words, bremsstrahlung is high for high-energy particles of small mass incident on high-Z material. The fraction of the incident electron kinetic energy, which is emitted as a bremsstrahlung photon, is always a small fraction for realistic shielding situations. For instance, only about 4% of the energy of a 0.5 MeV electron bombarded to lead is converted into bremsstrahlung [3]. The bremsstrahlung angular distribution exhibits an anisotropic trend and varies with incident electron energy. Further, as the electron energy increases, there is much less photons produced especially in high-energy region. PFRR was pre-investigated prior to the experiment using bremsstrahlung photons with a higher-Z converter target with different Z such as gold and tantalum in [5, 6]. The converter target used in the experiment was platinum which has a Z between gold and tantalum. Platinum is selected for its high photonuclear cross section to produce low number of fast neutrons because a filter of polyboron blocks were used to absorb the slow neutrons so that only fast neutrons contributed from photofission reaction reached the detector. The setup of the electron accelerator for production of bremsstrahlung photons and the beam profile of 13.5 MeV and 18.0 MeV with varied beam current are shown in Figure 2 and 3 respectively. 18 MeV beam energy is focused on present research because with end-point bremsstrahlung energy of 18 MeV, the photofission cross section which is around 14 MeV is believed to bring forth the most neutron numbers for ease of detection.

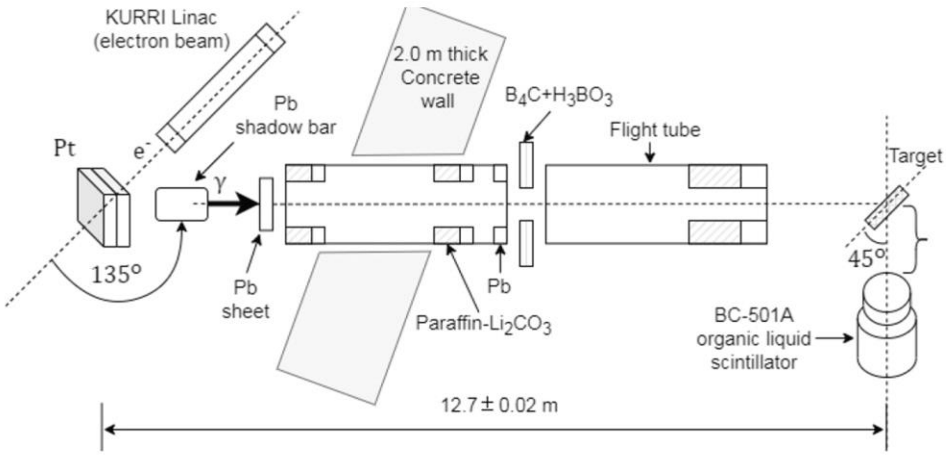


Figure 2: Experimental setup of KURRI Linac with BC-501A detector.

1. Detection system

The detector used in the experiment, BC-501A is a type of organic liquid scintillator detector with excellent pulse-shape discrimination (PSD) properties and fast timing performance [8]. This detector was used for its exceptional distinguishability of neutron from gamma pulses especially high-energy neutrons without the use of moderator. Different from neutron counters such as He-3, BC-501A can record the neutron activities with energy variation. The difference in the intensity of slow neutron and gamma-ray light components serves as a basis for PSD methods used to identify and characterize neutron pulses [8]. The BC-501A that was used is 5 inches thick and has a diameter of 5 inches, as shown in Figure 4. The neutron source used for study is Cf-252 for its distinct neutron energy spectra, both in shape and average neutron energy.

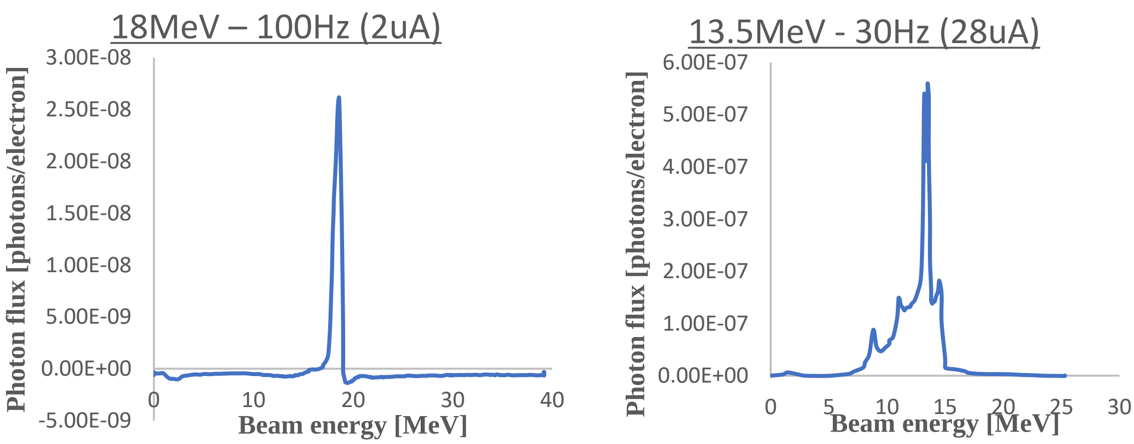


Figure 3: Beam profile of 18.0 and 13.5 MeV with different beam current.

The Californium source used has a radioactivity of 10.4 MBq while the uranium target for study is natural uranium. Cf-252 source was placed 1 cm from the detector. The measurement of Californium source was conducted without the accelerator beam operating, and the measurement time is approximately 5 minutes. Whereas the measurement of natural uranium target was conducted with various sets of varying conditions such as beam current (electron flux), incident electron energy, moderator material polyethylene boron blocks, etc. The natural uranium used are 12 pieces of which each has a size of 2 cm x 1 cm x 3 mm thick, and is placed 45º and 20 cm distance to the detector. KDS (ATL-35) laser was used for accurate positioning the target. To measure neutron in the mixed neutron/gamma field, electronic circuit with a PSD (W/Hz) function was used. The pulses were collected directly from an anode of a photomultiplier (PMT). The PMT had the same diameter as the scintillator. High voltage of 800V was supplied to the preamplifier of BC-501A and is connected to the ORTEC 460 Amplifier. The signal is delayed using delay amplifier with 20x20 coarse gain and together with rise time to height converter the signal by two different particles was displayed on the MCA. 18 MeV beam on uranium was operated with 138.2 kW linac power, 859 kV gun pulser voltage at 100 ns interval, 2 µA beam current and 0.27 µs long pulse width. The pulse height resolution, *dL/L* could be obtained as:

$$\frac{dL}{L} = \sqrt{A^2 + \frac{B^2}{L} + \frac{C^2}{L^2}} \qquad \text{Eq. 2}$$

Where L is light output or the energy of particles detected. A, B and C were determined to be approximately 8.7, 7.1 and 0.5 respectively for a 5” diameter x 2” thick BC-501A detector [9].

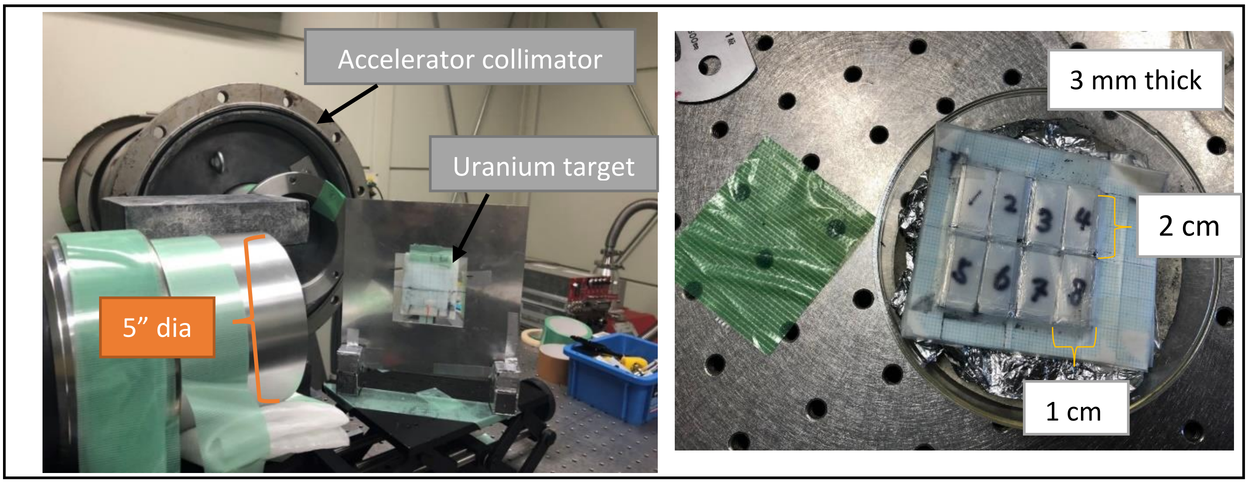


Figure 4 5” x 5” BC-501A and natural uranium target.

A digital PSD technique based on ratios of pulse integrals over different time periods was applied to discriminate neutrons from gamma rays. The equation of PSD can be represented as follows:

$$PSD = \frac{Q_{long} - Q_{short}}{Q_{long}} \qquad \text{Eq. 3}$$

Where Q_{short} and Q_{long} are the charge integrated during the short time-gate and long time-gate respectively. A depiction of the relationship between these two charges can be seen in Figure 5.

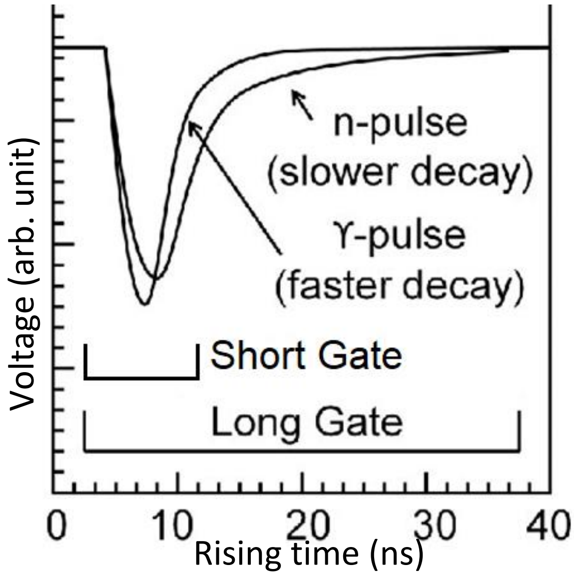


Figure 5: Depiction of the charge pulse difference for a neutron compared to a gamma.

Results and discussion

Figure of merit (FOM) is a statistical parameter often used to evaluate the performance of differentiability of neutrons from gamma rays. The FOM is proportional to the separation of the gamma and neutron peaks (△PEAK) and inversely proportional to their full-width at half-maximum ($FWHM_\gamma$ and $FWHM_n$ for the gamma-ray and neutron peaks respectively), defined as:

$$FOM = \frac{\triangle \text{ PEAK}}{FWHM_{\gamma} - FWHM_{n}} \qquad \text{Eq. 4}$$

FOM varies with the size and type of scintillator detector. For instance, typical FOM of a plastic scintillator EJ-270 for fast neutrons (taken as 450 keVee to 2.3 MeVee) ranges from 1.33 ± 0.01 to 1.77 ± 0.06. For liquid scintillator such as NE-213 (older version of BC-501A) and BC-501A, FOM measured at 1 MeV was 3.81 and 2.05 respectively [10]. The FOM measured for the Cf-252 source was 2.52236 ± 0.08785. For Cf-252 source, very good separation of neutrons from gamma rays can be observed from Figure 6.

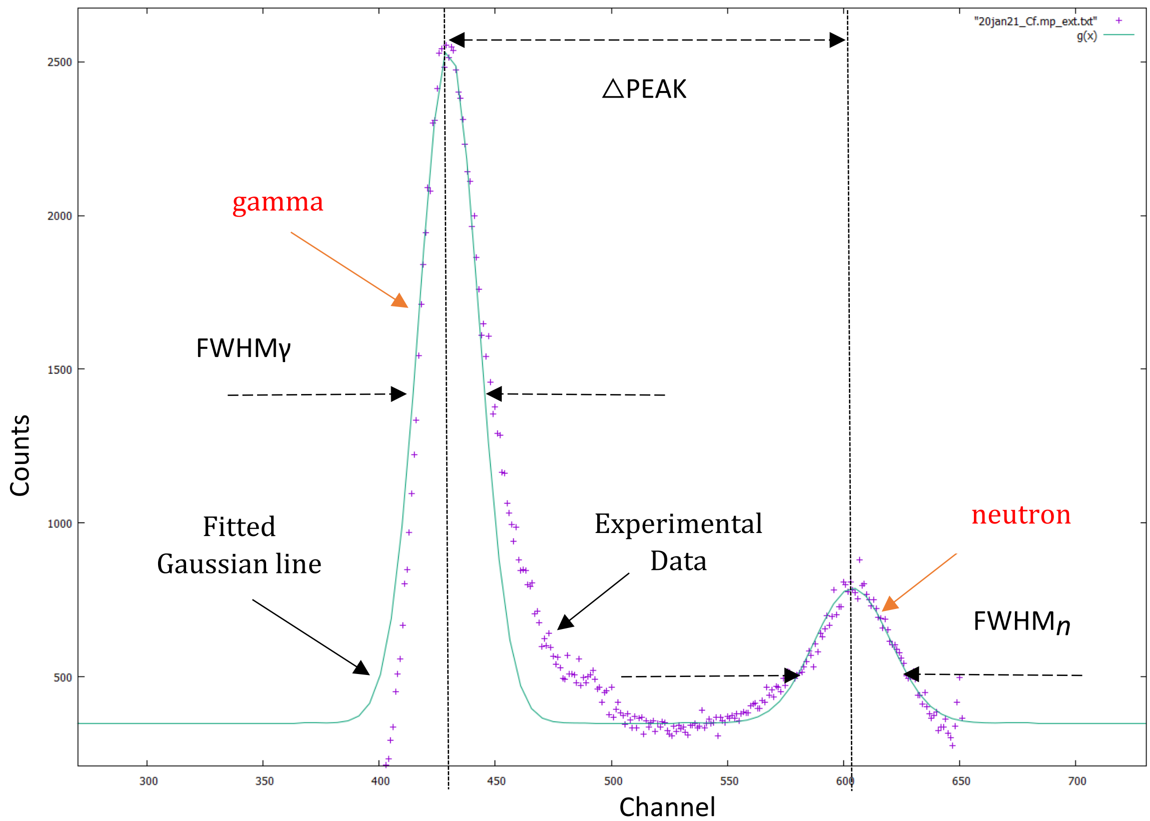


Figure 6: Neutron and gamma distribution Gaussian fits for 5” x 5” BC-501A scintillator.

Region of interest (ROI) of the experimental results were picked up to study focus on neutron counts based on Cf-252 observable neutron peak. Figure 7 shows the pulse height (corresponding to energy) against rising time (PSD) of natural background undergone 12.7 hours of measurement time and Cf-252 source measured within 5 minutes. It can be observed clearly that the neutron peak (on right) is separated from the gamma rays (on left). Since neutron is our particle of interest, a ROI was drawn around the neutron peak, 21 out of total 512 arbitrary units on x-axis and 85 out of total 512 units on y-axis. The light output counts from the ROI is tabulated in Table 1 where 4 cases are available: natural background (BG), Cf-252, background with 18 MeV beam ($BG_{beam\text{-}18MeV}$) and natural uranium (NU) with 18 MeV incident beam.

Table 1: The light output counts in the ROI defined.

ROI Cases	**Total count rate, cps**	**Total counts, N**	**Absolute error, $\sqrt[2]{N}$**	**Relative error, $\frac{\sqrt[2]{N}}{N}$**
Natural BG	0.021578	1025	32.01562	0.031235
$BG_{beam\text{-}18MeV}$ +PolyB	0.209391	754	27.45906	0.036418
Cf-252	75.87078	45706	213.7896	0.004677
NU + PolyB	0.775459	2793	52.84884	0.018922

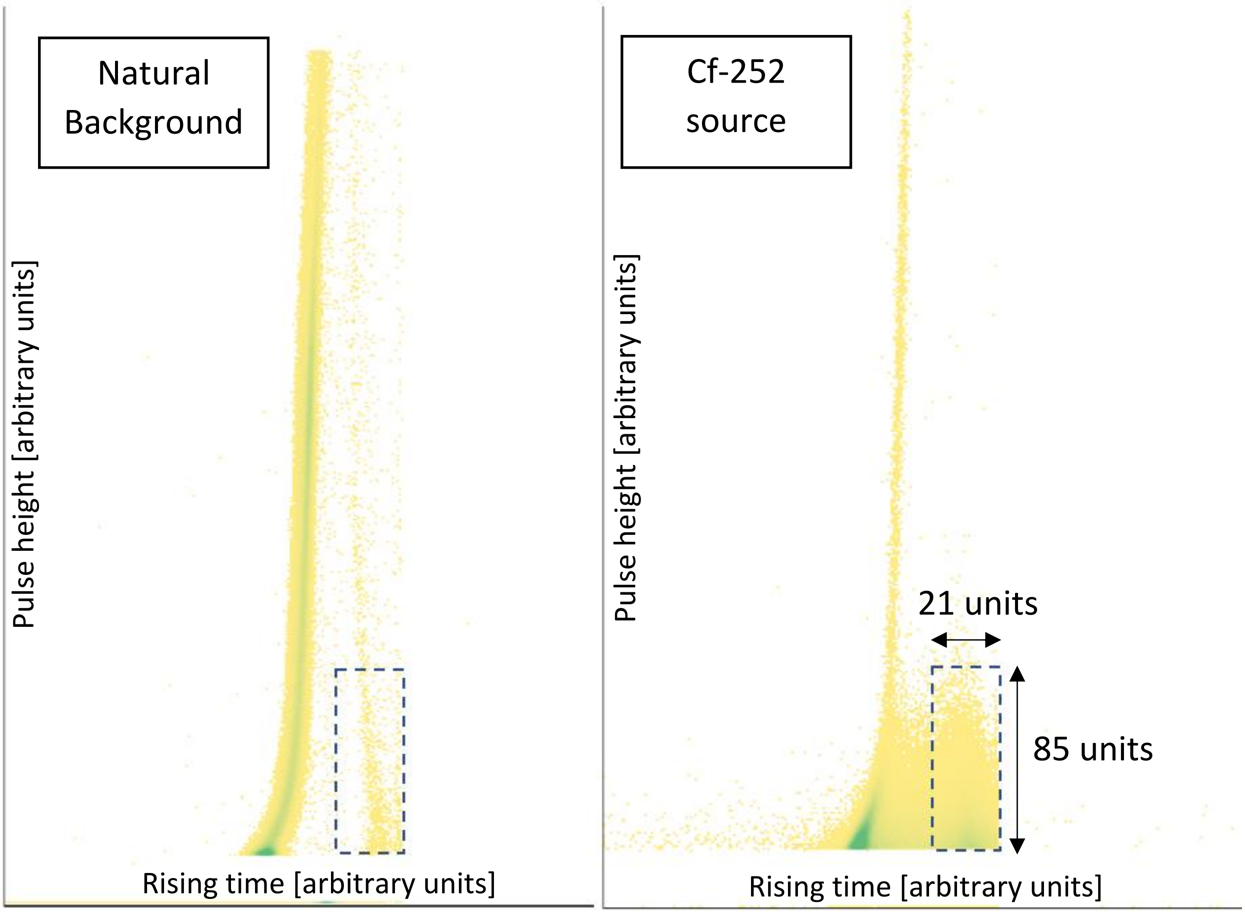
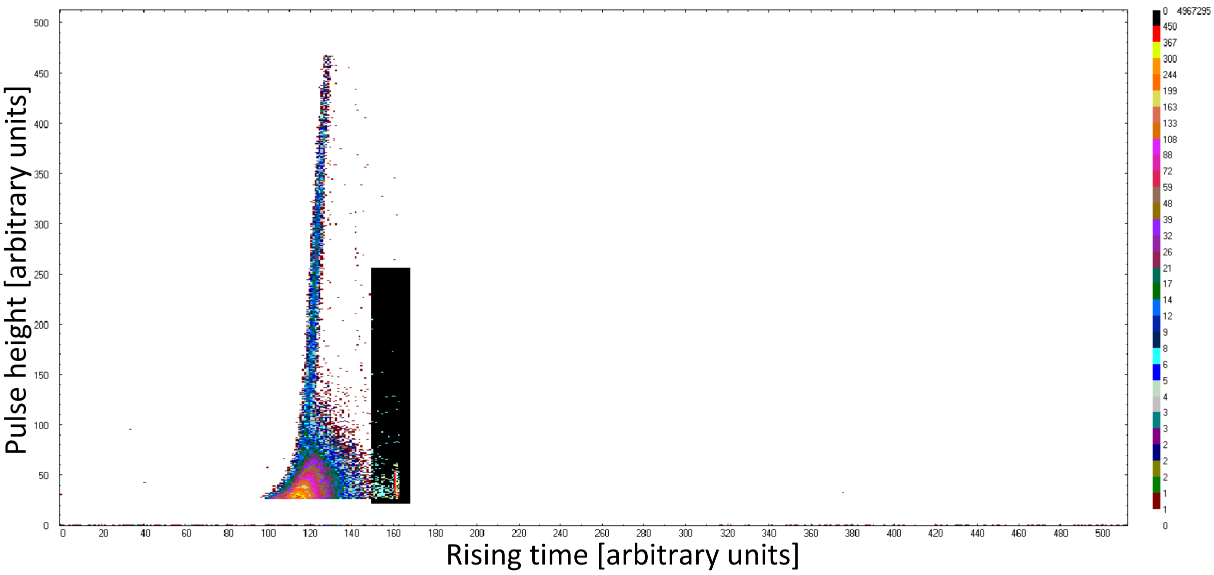
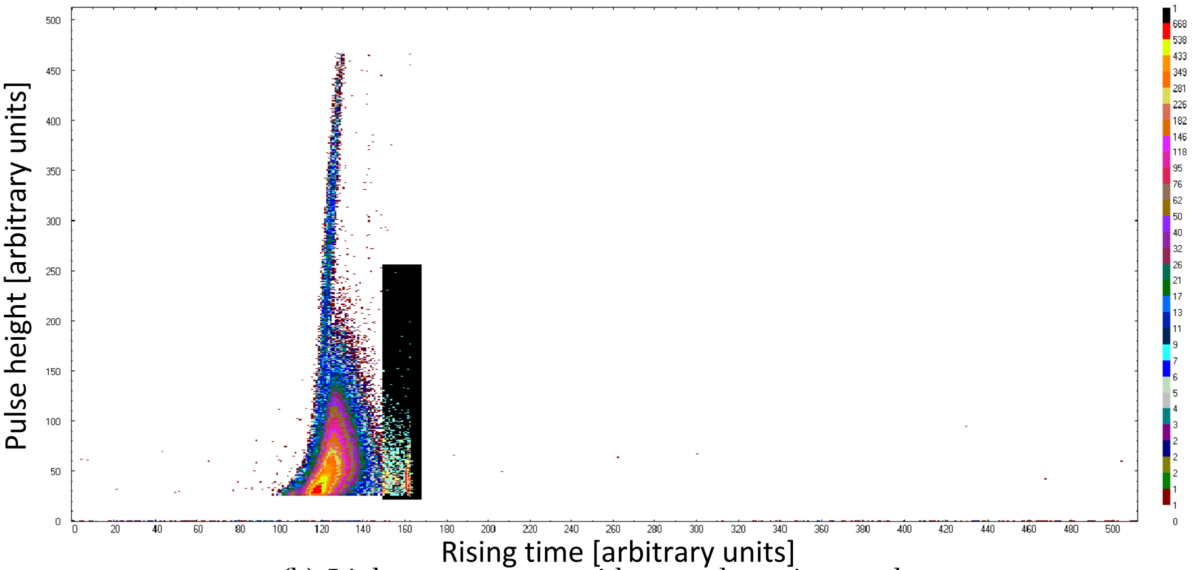


Figure 7: ROI selection based on Cf-252 source.

Figure 8 shows the contour plot with the same axes of background with 18 MeV beam and natural uranium with 18 MeV incident beam. Both of these two had the filter of polyethylene-boron (PolyB) blocks which were used to absorb thermal neutrons mostly coming from platinum converter target (which could be confused with the neutrons from photofission of uranium target). Though faintly, it can be seen that the counts in the ROI of natural uranium is more than that of background with 18 MeV beam, some of which were contributed from scattering due to the deflection angle of 45° of uranium target.



(a) Background count



(b) Light output count with natural uranium as the target

Figure 8: Contour plot of pulse height vs rising time with beam operating at 18 MeV and 1.7 uA with a polyethylene-boron block filter.

Minimum detectability

The minimum detectability section consists of two parts: the finding out of minimum detectable activity from 0.3% confidence statistical point of view using the propagation error of Cf-252 source and background; the feasibility of NU being detected considering photofission reaction point of view. The propagated error was calculated between both natural background and background with 18 MeV beam with gross counts of sample, say, Cf-252 source or natural uranium. The standard deviation, σ is calculated with √[(Rg/tg)+(Rb/tb)] where Rg is the gross count rate, Rb is the background count rate, tg and tb are measurement time for gross count and background count respectively. σ divided by net count rate, (Rg – Rb) obtains the fractional standard deviation or known as the coefficient of variation, COV, a statistical parameter to evaluate the need to increase the experimental measurement time for an increment of precision by a factor of two requires the increment of time by a factor of four. σ and COV obtained for Cf-252 vs natural background were 0.355 and 0.468% respectively. For Cf-252 vs $BG_{beam\text{-}18MeV}$, σ and COV obtained were 0.355 and 0.469% respectively. While for NU vs $BG_{beam\text{-}18MeV}$, σ and COV obtained were 0.0165 and 2.921%. If assuming the minimum net count rate, Rm that can be detected with 0.3% confidence is given by:

$$Rm = 3\sigma(Rg - Rb) \qquad \text{Eq. 5}$$

Solving this equation for Rg will give the minimum detectable gross counts Rm:

$$Rg = \frac{\left(2\text{Rb} + \frac{9}{\text{tg}}\right) + \sqrt{\frac{36Rb}{tg} + \frac{81}{tg^2} + \frac{36Rb}{tb}}}{2} \qquad \text{Eq. 6}$$

And the minimum detectable activity, Am can be calculated as follows:

$$Am = \frac{Rm}{S} \qquad \text{Eq. 7}$$

Where S is the sensitivity of the detector expressed in count rate per Becquerel. Minimum count rate, Rm calculated for Cf-252 vs natural BG and Cf-252 vs $BG_{beam-18MeV}$ were 0.02877 and 0.0728 respectively. Then, using the count rate in the ROI from Table 1 and activity information of Cf-252 as 1.04×10^{7} Bq, the sensitivity for Cf-252 vs natural BG and Cf-252 vs $BG_{beam-18MeV}$ were 7.29×10^{-6} and 7.28×10^{-6} respectively. Consequently, Am obtained for Cf-252 vs natural BG and Cf-252 vs $BG_{beam-18MeV}$ were 3950 Bq and 10 kBq respectively. In other words, target with radioactivity higher than 3950 Bq is required to be detectable in natural background while for background with beam operating at 18 MeV the target require a radioactivity higher than 10 kBq.

On the other hand, the relationship between the source strength and detector efficiency could be used to evaluate the minimum detectability of NU target considering the induced fission reaction:

$$Source\ strength \times Detector\ Efficiency > BG \qquad \text{Eq. 8}$$

$$fission\ reaction\ rate\ of\ NU = \nu N\sigma\varphi V > fission\ reaction\ rate\ of\ Cf-252 \qquad \text{Eq. 9}$$

Where ν is the neutron multiplication factor = 3.34367, N is the number atomic density, σ is the photofission cross section, φ is the photon flux, V is the target volume. The detector efficiency was calculated with simply the ratio of net detected light output counts in the ROI of Cf-252 (assume all counts are neutrons) over the neutrons emitted from Cf-252 source (1.21×10^{6} n/s with 0.117 n/s/Bq): $75.8492/1.21 \times 10^{6} = 6.2685 \times 10^{-5}$. As a calculation results from Eq. 8, the source strength required for natural background $BG_{beam-18MeV}$ were 344.23 n/s and 3340.35 n/s, or in fission rate as 117 fission/s and 3120 fission/s respectively. Using eq. 9 we can calculate the fission reaction rate of NU where N = 19g/cc ÷ 238 (99.283% ^{238}U) x 6.02×10^{23} atom/cm^3; σ was given the maximum approximate value of photofission cross section at peak around 14 MeV (with end-point bremsstrahlung incident energy of 18 MeV) which is about 300 millibarn; φ is photon flux per area of target which will be discussed later; V is NU target volume of 4 cm x 4 cm x 0.6 cm thick (four pieces stacking on top of the eight pieces shown in Figure 4). Photon flux from the beam profile of 18 MeV and 2 µA gives a value of 3.22×10^{8} photons, however this value could not be used since it is the photons produced detected from near within the converter target platinum. Instead, dose rate was measured at the gap of collimator (where the filter of PolyB block was placed, from Figure 2) and through conversion factor from 1977 and 1991 ANSI/ANS gamma flux-to-dose conversion factor models [11], detected dose rate of around 30 mSv/h during the beam operation of 100 Hz, 13.5 MeV had the conversion factor of 1.5×10^{-1} µSv/h/photons/cm^{2}s yielded photon flux of 2×10^{5} photons/cm^{2}s. Since the photon flux of interest is near end-point energy, the proportion of photon flux in the region of 10 to 15 MeV is around 10%, so the flux of interest is expected to be only 10% of 2×10^{5} photons/cm^{2}s. Furthermore, the distance of NU target from the collimator gap is around 3 m, loss by inverse square law was considered too. The effective photon flux is around 2.22×10^{3} photons/cm^{2}s. Using this value of photon flux yields the fission rate of NU as 512 fission/s, or 1711.96 n/s as the source strength. Both fission rate and source strength of NU is between 117 and 3120 fission/s as well as 344.23 n/s and 3340.35 n/s of Cf-252 to the two different backgrounds, thus it can be deduced that the photoneutrons induced from NU is enough to be detectable in natural background but not adequately detectable in the background with beam operating at 18 MeV.

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Conclusion

FOM method shows the good separation of neutrons from gamma rays of Cf-252 source with 5” diameter x 5” thick BC-501A organic liquid scintillator. With 0.3% confidence, the minimum detectable activity for both natural background and background with beam operating at 18 MeV were 3.95 kBq and 10 kBq respectively. Using Cf-252 source strength as the requirement standard, natural uranium can be detected in natural background environment but not in the environment with beam operating at 18 MeV. To improve the visibility of photofission reaction, few factors could be considered in future such as uranium enrichment value (^{235}U has about two times higher photofission cross section than ^{238}U), uranium target size (larger is better but could be complicated with complexity of interactions within target), moderation of neutrons from converter target (using water tank, aluminium filter, etc. to capture slow neutrons), housing of lead around detector to reduce gamma flush from accelerator and others.

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