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ARTICLE

Reaction yield dependence of the (γ, γ’) reaction of ^{238}U on the target thickness

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Abstract

The dependence of the NRF yield on the target thickness was studied. To this end, an NRF experiment was performed on ^{238}U using a laser Compton backscattering (LCS) γ-ray beam at the HIγS facility at Duke University. Various thicknesses of depleted uranium (DU) targets were irradiated by an LCS γ-ray beam with an incident beam energy of ~2.475 MeV. The scattering NRF γ-rays were measured using an HPGe detector array positioned at scattering angles of 90° relative to the incident γ- beam. An analytical model for the NRF reaction yield (NRF RY model) is introduced to interpret the experimental data. Additionally, a Monte Carlo simulation using GEANT4 was performed to simulate the NRF interaction for a wide range of target thicknesses of the ^{238}U. The measured NRF yield shows the saturation behavior. The results of both of the simulation and the analytical model can reproduce the saturation curve of the scattering NRF yield of ^{238}U against the target thickness. In addition, we propose a method to deduce the precise integral cross-section of the NRF reaction by fitting the NRF yield dependency on the target thickness without any absolute measurements.

Keywords; NRF; SNMs; LCS; NRF RY Model; thickness dependence; saturation; ^{238}U; cross-section; Monte Carlo simulation; HPGe

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1. Introduction

The nuclear resonance fluorescence (NRF) technique has been introduced as a powerful tool to identify the isotopic composition of material based on the nuclear energy levels. NRF is a phenomenon in which a photon of an appropriate energy excites a nucleus to a higher state, which subsequently decays to the ground state or a lower-lying state by emitting a γ-ray with an energy equal to the energy difference between the resonance state and the lower state [1-2]. We have proposed an inspection system for hidden special nuclear materials (SNMs), such as ^{235}U and ^{239}Pu, based on the NRF technique [3], in which it is assumed that the target contains a certain amount of SNM with a high purity. Therefore, a study of the NRF yield dependence on the target thickness of SNMs is required to evaluate the performance of the inspection system.

Our study is based on the atomic and nuclear absorption of the incident and/or scattered NRF γ-rays. The prior attenuation and self-absorption measurements and the analytical approach are based on measurements of the NRF γ-rays from a scatter target that is placed downstream of the absorber material. In this method, the self-absorption has been measured by attenuation of the NRF γ-rays in the absorber prior to measuring it from the scatter material, as presented by F.R. Metzger [4] and S. Ofer, *et al*. [5]. Similarly, B.A. Ludewigt, et al. [6] and Q. Brian et al., [7] studied the relative attenuation of resonant and non-resonant γ-rays in NRF by a transmission technique in terms of two materials, the transmission material (with various thicknesses) and the scatter target, to determine the areal density of the transmission material relative to the scatter material. These previous studies used the absorption/attenuation technique to correct the calculations of the resonance parameters or took the relative values.

On the other hand, we studied the total attenuation factor of the resonant and non-resonant γ-rays energies on a direct NRF measurement (without an absorber target/material) of the resonant states of the isotope of interest (IOI) and will introduce a new method to extract the absolute values of the integrated NRF cross-section. The total attenuation factor can be deduced from a fitting function of the experimental saturation curve of the NRF yield against the target thickness. Thus, an NRF experiment that uses a 2.475-MeV LCS γ-ray beam and HPGe detector was conducted on the nuclear material ^{238}U. To extract the resonance attenuation factors of the excitation levels of the NRF, an analytical model for the NRF reaction yield (NRF RY model) was deduced. Calculations were performed to simulate the NRF experiment for ^{238}U by the Monte Carlo simulation toolkit GEANT4 [8-9] over a wide range of the target thicknesses (up to 20-mm-thick). The NRF cross-section of the induced excitation states of ^{238}U at approximately 2.5 MeV were deduced by the fitting of the saturation curves of the NRF yields in comparison with those from the common method.

2. Experimental

The NRF experiment was performed at the High Intensity γ-ray Source (HIγS) facility [10] at Duke University, using nearly mono-energetic γ-rays. The HIγS facility is based on the Compton backscattering of free-electron-laser (FEL) photons with electrons stored in the storage ring. Four NRF measurements were performed on eight disks of depleted uranium (DU), which has density of 19.1 g.cm^{-3}. Each disk of DU is 25.4 mm in diameter and approximately 0.6 mm in thickness with a mass of approximately 6.5 g, and each disk is encased within a thin plastic sealant. A target is assembled in four sets with two, four, six or eight disks stacked together; a summary of the runs is presented in Table 1. Each set of DU was placed within an evacuated acrylic tube to decrease the scattering of the incident γ-rays by air.

The LCS γ-beam was generated with almost 100% circularly polarized photons with an energy of 2.475 MeV and an average flux of approximately 3×10^{5} γ/s/keV. The γ-beam is collimated with a circular lead collimator 19.05 mm in diameter, which is located at approximately 60 m downstream from the collision point, where the electrons collide with the FEL photons, and it confines the beam spot to a certain size of photons per second to produce an energy spread (FWHM) of approximately 5% on the target.

Figure 1 shows the experimental setup of the NRF measurements. The scattering NRF γ-ray spectra were collected with an HPGe detector array with a total hours on-beam of LCS γ-ray source of ~32.5 h. Three HPGe detectors, each with 60% relative efficiency and energy resolution approximately 0.19% (~2.52 keV, FWHM) at 1332 keV, were placed downstream from the collimator and positioned at a scattering angle of 90° relative to the incident beam, apart from the ^{238}U target, two of them in a vertical position and the other in a horizontal position. The detectors were positioned at 10 cm from the center of the target. Two absorber sheets of copper (3.2 mm) and lead (4.0 mm) are set in the front of each detector to attenuate the low-energy background.

The incident beam energy was measured by the large volume of the HPGe detector with a 123% relative efficiency, which was placed in the beam axis prior to measuring the scattering NRF γ-rays. After measuring the beam energy, the HPGe (123%) detector (flux monitor) was moved out of the beam axis and positioned at an angle of 8.6° relative to the beam axis in the horizontal direction. The absolute beam flux on the target was measured during the data acquisition of each run using the measurement of Compton scattered γ-rays from a scatterer placed downstream in the beam path. A copper plate of 1.1 mm in thickness (with the size around 20×20 cm^2) was placed directly in the beam path, approximately 100 cm downstream from the detector setup and approximately 165 cm upstream from the flux monitor. The flux calculations were corrected to the attenuation through the DU targets and the scatter Cu-plate.

The energy calibration of all of the measured spectra was performed by the natural room background lines, which exist in each energy spectrum, namely the ^{40}K γ-ray line

(1460.8 keV) and the ^{208}Tl γ-ray line (2614.5 keV). The detection efficiency of all of the detectors was evaluated by placing a standard γ-ray source (^{56}Co) at the target position with γ-ray energies of 846.8 keV, and 1037.8 keV through 3253.5 keV. The efficiency of the flux-monitor detector was determined by positioning a ^{56}Co source at the copper plate.

3. Integrated NRF cross-section

The interaction cross-section for a state that undergoes NRF of a γ-ray as the nucleus transitions from an excited state to the ground state followed by de-excitation with emission of one or more photons is given by the Breit-Wigner equation [1]:

$$\sigma_{NRF}(E) = \frac{\pi g}{2}\frac{(\hbar c)^2}{E^2}\frac{\Gamma\Gamma_0}{(E-E_r)^2+(\Gamma/2)^2}, \tag{1}$$

where E denotes the incident-photon energy, and E_r represents the resonant energy. $\hbar$ is Planck's constant. Γ indicates the sum of the partial width of the various de-excitation states, *i.e.*, Γ is the total width of the NRF transitions, which is related to the state's mean lifetime, τ, by $\Gamma = \hbar/\tau$. In contrast, Γ_0 is the partial width of the state for decay by γ-ray emission to the ground state. g is a statistical factor that depends on the total angular momentum numbers of the ground state, J_0, and the excited-state, J_i, by the following expression:

$$g = \frac{2J_i+1}{2J_0+1}. \tag{2}$$

In addition, the ground state width is correlated to the total partial width of the NRF transitions by the following expression:

$$\frac{\Gamma_0^2}{\Gamma} = \frac{I_s}{g}\left(\frac{E_r}{\pi\hbar c}\right). \tag{3}$$

where I_s denote to the integral NRF cross-section. Because Γ is the sum of the partial-width transitions of the NRF γ-rays, the most probable NRF transitions of ^{238}U follow the spin sequence of $0^+ \rightarrow 1^{+/-} \rightarrow (0^+)$or ($2^+$) (the M1/E1 dipole transition) and $0^+ \rightarrow 2^+ \rightarrow 0^+$, (the quadrupole transition, E2). Thus, the total transition width of ^{238}U could be $\Gamma = \Gamma_0$ or $\Gamma = \Gamma_0 + \Gamma_1$.

Moreover, the scattering NRF γ-ray cross-section is Doppler broadened by the nuclear intrinsic width induced by the thermal motion of the nuclei. Consequently, the NRF cross-section can be written with the Doppler effect as follows [1]:

$$\sigma_{NRF}(E) = \pi^{3/2} \lambda\!\!\!^{-2} g \frac{\Gamma_0^2}{\Gamma} \frac{1}{\Delta} \exp\left[-\left(\frac{E - E_r}{\Delta} \right)^2 \right], \tag{4}$$

This form is valid only, when the Doppler width is larger than the intrinsic width. Where $\lambda\!\!\!^{-}$ is incident wave length of photon energy *E*. Δ is the Doppler width, which can be expressed in terms of

the atomic mass, *M*, and the absolute or effective temperature, *T*, as follows:

$$\Delta = E_r \sqrt{\frac{k_B T}{Mc^2}}, \tag{5}$$

where k_B is Boltzmann’s constant, and *c* is the speed of light in space. In the case of the ^{238}U target under the present experiment condition, the Doppler width for the emission line of 2.468 MeV is 1.99 eV at the absolute temperature of 300 K, where the intrinsic width is approximately 20.12 meV [7].

Another important term is the recoil energy of the nuclei after emission of the NRF γ-rays. In general, this is considerable and larger than the energy level (intrinsic or Doppler) width, and thus the scattered NRF γ-rays cannot be absorbed again by the mother isotope state. The recoil energy can be expressed by:

$$E_{recoil} = \frac{E_r^2}{Mc^2}. \tag{6}$$

For the emission line of 2.468 MeV of the ^{238}U nucleus, the recoil energy is deduced to be 13.74 eV, which is approximately seven times larger than the Doppler width for this transition.

4. Analytical Model for the NRF Reaction Yield

To estimate the reaction yield of the NRF interaction with the IOI, we will assume the slab geometry that is presented in Figure 2. The reaction yield of the NRF interaction caused by the incident γ-ray flux Φ_i can be described as follows.

i. The incident beam flux Φ_i will be attenuated prior to releasing the NRF γ-rays at a depth *x* by an attenuation factor of $\left\{\exp\left[-\mu_i(E_i)\cdot r_i\right]\right\}$, where $\mu_i(E_i)$ is the linear attenuation coefficient for the electronic $\left\{\mu_e(E_i)\right\}$ and nuclear $\left\{\mu_n(E_i)\right\}$ interactions. E_i is the incident γ-ray energy, and r_i represents the incident pass length.

ii. If the incident energy (E_i) or a part of it is within the nuclear level width (Γ) of the IOI, $\left\{(E_r-\Gamma/2)\leq E_i\leq(E_r+\Gamma/2)\right\}$, the NRF interaction will occur with the probability of the absorption NRF cross-section, σ. The released NRF γ-rays will have the angular distribution $W(\theta)$, described by [2,11]. These NRF γ-rays will undergo self-absorption by the electronic interaction in the target material prior to leaving it, by an attenuation factor of $\left\{\exp\left[-\mu_s(E_r)\cdot r_s\right]\right\}$, where $\mu_s(E_r)$ is the linear attenuation coefficient for the electronic interaction $\left\{\mu_e(E_r)\right\}$, E_r is the resonant energy of the NRF γ-rays for a particular level of IOI, and r_s is the scattering pass length. Additionally, the intensity of the NRF γ-rays depends on the number of nuclei (*N*) per cm^3 of the IOI in the target.

iii. The measured scattering yield of the NRF γ-rays released from the target is dependent on the detection efficiency of the detector, $\varepsilon(E_r)$, and the fraction of the solid angle subtended by the detector, $(\Omega/4\pi)$.

Consequently, the NRF yield can be given by following expression:

$$d^2Y = \left[\Phi_i \cdot e^{-\mu_i(E_i)r_i}\right] \cdot \left[N \cdot dx \cdot \sigma(E_i) \cdot dE \cdot W(\theta) \cdot e^{-\mu_s(E_r)r_s}\right] \cdot \left[\varepsilon(E_r) \cdot \frac{\Omega}{4\pi}\right]. \tag{7}$$

By integrating the reaction yield over the target thickness, x, and the incident γ-ray energy, E_i, we obtain:

$$Y = \left[\Phi_i \cdot N \cdot W(\theta) \cdot \varepsilon(E_r) \cdot \frac{\Omega}{4\pi}\right] \cdot \left[\int \sigma(E_i) \cdot dE\right] \cdot \left[\int e^{-\mu_i(E_i)r_i} \cdot e^{-\mu_s(E_r)r_s} \cdot dx\right]. \tag{8}$$

The first term of this equation represents the absolute values of the incident flux, the detection efficiency and the effective angular distribution. The second term represents the integration over the incident energy within the nuclear-level width of the IOI. The third term represents the integration over the target thickness in terms of the attenuation and the self-absorption of γ-rays through the target material.

The analytical model is introduced to study the attenuation and self-absorption of the incident γ-rays and scattering NRF γ-rays. We assumed the incident flux, Φ_i, with an angle θ_i relative to the perpendicular axis on the target surface, as shown in Figure 2, and an incident pass length of $r_i = x / \cos(\theta_i)$. Supposing an incident energy within the nuclear level width (Γ), the NRF γ-rays will be scattered in all possible directions with an angular distribution of $W(\theta)$. We assume that the backward scattering has an angle θ_b, and that the scattering length is $r_b = x / \cos(\theta_b)$. Thus, the attenuation term for the backward scattering will be as follows:

$$\int e^{-\mu_i(E_r)r_i} e^{-\mu_s(E_r)r_b}\, dx = \int e^{-\mu_i(E_r)\frac{x}{\cos(\theta_i)}} e^{-\mu_s(E_r)\frac{x}{\cos(\theta_b)}}\, dx. \tag{9}$$

By integration over the target thickness l, we obtain:

$$\int_0^l e^{-(\mu_i(E_r)\sec(\theta_i)+\mu_s(E_r)\sec(\theta_b))x} dx \tag{10}$$

$$= \frac{1 - e^{-(\mu_i(E_r)\sec(\theta_i) + \mu_s(E_r)\sec(\theta_b))l}}{\mu_i(E_r)\cdot \sec(\theta_i) + \mu_s(E_r)\cdot \sec(\theta_b)},$$

$$= \frac{1 - e^{-\mu(E_r)l}}{\mu(E_r)}$$

where $\mu_i(E_r) = \mu_e(E_r) + \mu_{NRF}(E_r)$ (11)

is the total attenuation, $\mu_i(E_r)$ is the attenuation for the incident γ-rays of both the electronic $\{\mu_e(E_r)\}$ and nuclear absorption by NRF $\{\mu_{NRF}(E_r)\}$; *i.e.*, $\mu_i(E_r) = \mu_e(E_r) + \mu_{NRF}(E_r)$, and $\mu_s(E_r)$ is the attenuation for the scattering NRF γ-rays for the electronic interaction only $\{\mu_s(E_r) = \mu_e(E_r)\}$. $\mu_e(E_r)$ and $\mu_{NRF}(E_r)$ can be expressed by:

$$\mu_e(E_r) = \frac{N_a}{M}\sum_n \sigma_n^a(E_r), \tag{12}$$

where M is the effective mass number of the target material, N_a is Avogadro's number, and σ_n^a is the atomic cross-section of the interaction type n, *e.g.*, n is the Compton scattering, photoelectric effect, pair production, *etc.*, and:

$$\mu_{NRF}(E_r) = \frac{N_a}{M_{IOI}}\sigma_{NRF}^{ij}, \tag{13}$$

where σ_{NRF}^{ij} represents the cross-section of the NRF transition line j of the i^{th} isotope, and M_{IOI} is the atomic mass of the IOI.

In the case of forward scattering, the total attenuation is given by replacing the scattering angle θ_b with θ_f, (see Figure 2), and therefore:

$$\mu(E_r) = \mu_i(E_r)\cdot \sec(\theta_i) + \mu_s(E_r)\cdot \sec(\theta_f). \tag{14}$$

The total attenuation term, which is given by Equation (10), represents the effective thickness for the NRF interaction through the target thickness. When the incident γ-rays are parallel to the x-axis $(\theta_i = 0)$, the total attenuation factor will be in the form:

$$\begin{aligned}\mu(E_r) &= \mu_i(E_r) + \mu_s(E_r)\cdot \sec(\theta) \\ &= \mu_e(E_r) + \mu_{NRF}(E_r) + \mu_e(E_r)\cdot \sec(\theta) \\ &= \mu_e(E_r)\left[1+\sec(\theta)\right] + \mu_{NRF}(E_r).\end{aligned} \tag{15}$$

where θ represents θ_b or θ_f. For a scattering angle of 90° and an incident angle θ_i equal zero, we assume a fixed shape. Let us assume a cylindrical shape with thickness l and radius r. The attenuation term will be in the form:

$$\frac{1-e^{-(\mu_{NRF}(E_r)+2\mu_e(E_r))l}}{\mu_{NRF}(E_r)+2\mu_e(E_r)} e^{-\mu_e(E_r)r}. \tag{16}$$

Because of the resonance that occurs within a very narrow energy window, the range of the level width Γ, the integration of the cross-section over the incident energy will be changed in the range of $(E_r - \Gamma_D/2)$ to $(E_r + \Gamma_D/2)$, considering the thermal motion and Doppler width. Thus the integration will contain the NRF cross-section, $\sigma(E_i)$, that is considerable a constant value within a small variation of energy width of the resonant level therefore:

$$\int_{(E_r-\Gamma_D/2)}^{(E_r+\Gamma_D/2)} \sigma(E_i)\cdot dE \simeq \Gamma_D \cdot \sigma_{NRF}(E_r), \tag{17}$$

where Γ_D is the Doppler width of a broadened resonance level [1]. Substituting Equation (10) or (16) and Equation (17) into Equation (8), we obtain the NRF reaction yield, Y, in the form:

$$Y = \Phi_i \cdot \Gamma_D \cdot N \cdot \sigma_{NRF}(E_r)\cdot W(\theta)\cdot \varepsilon(E_r)\cdot \frac{\Omega}{4\pi}\cdot \frac{1-e^{-\mu(E_r)l}}{\mu(E_r)}, \tag{18}$$

where the last term of the attenuation represents the effective thickness for the NRF interaction within the target thickness as follows:

$$x_{effective} = \frac{1 - e^{-\mu(E_r)l}}{\mu(E_r)} \quad . \tag{19}$$

If the thickness $l \geq x^{Max}$, where x^{Max} denote to the target thickness that approach the function of $\left\{\exp\left[-\mu(E_r)\cdot x^{Max}\right]\right\}$ to zero, then the maximum effective thickness of the NRF interaction becomes:

$$x_{effctive}^{Max} \approx \frac{1}{\mu(E_r)} = \chi_0, \text{ where } x_{effctive}^{Max} \leq x^{Max} \tag{20}$$

where χ_0 is the total attenuation length for the γ-rays. This means that the maximum effective length is equal to the total attenuation length. By this expression, we can estimate the maximum effective length for the NRF interaction in the interrogation sample by knowing the electronic attenuation in the material sample and the resonance cross-section of the transition level. Consequently, the maximum value for the reaction yield of the NRF interaction can be approximated as:

$$Y^{Max} \approx \Phi_i \cdot \Gamma_D \cdot N \cdot \sigma_{NRF}(E_r) \cdot W(\theta) \cdot \varepsilon(E_r) \cdot \frac{\Omega}{4\pi} \cdot \chi_0 . \tag{21}$$

Consequently, the yield of the NRF reaction has a maximum value that depends on the attenuation length through the target due to the atomic and nuclear absorption.

5. Data Analysis

Figure 3 shows an example of the Compton-scattering spectrum of the incident-beam flux for the incident energy of 2.475 MeV. Figure 4 presents an example of the summed energy spectra of the NRF γ-rays for three HPGe detectors at a scattering angle of 90° for four DU foils with a total thickness of approximately 2.7 mm. All of the measured spectra of the NRF are calibrated and scaled to a common energy bin and then subtracted from the nonlinear–background estimated by the Statistics-sensitive Non-linear Iterative Peak-clipping (SNIP) algorithm [12,14], as shown in Figure 4. This figure contains 12 peaks: one arises from the target–background of the ^{238}U that originating from the ^{214}Bi γ-line at 2.447 MeV, and the 11 NRF peaks are not observed in the room and target–background spectrum in the interested energy region. Thus, we consider that all of them arise from the ^{238}U target. The 11 NRF peaks include three pairs with an energy difference of 45 keV, which can be attributed to the three transitions of (2410, 2468, and 2499 keV) to the ground state with the spin sequence $(0 \rightarrow 1 \rightarrow 0)$, and the other three of (2365, 2423, and 2454 keV) to the first exited state at 45 keV with a spin sequence of $(0 \rightarrow 1 \rightarrow 2)$.

5.1. NRF yield vs. the target thickness

Figure 5 presents the results of the NRF yield versus the target thickness for the four different thicknesses (Table 1) of the DU target. In this study, we chose the NRF state of 2.468 MeV, which was measured in previous works [7,15,16]. The NRF yield increases with the target thickness initially (for a small thicknesses) and could be linear. Gradually, the NRF yield achieves saturation as the target thickness increases (thick target), as presented by Equation (18). The Monte Carlo simulations GEANT4 [17] which is modified to include all physical process of NRF were performed for various thicknesses of 0.5, 2, 4, 6, 8, 10, 12, 16 and 20 mm with a diameter of 25.4 mm to enable the measurement of the wider energy range behavior of the NRF yield. As shown in Figure 5, the simulation result agrees with the experimental results. It should be noted that we used the NRF cross-section obtained from the fitting results of the experimental data, which will be described in this subsection.

The NRF RY model was applied to both the experimental and the simulation results. According to Equation (18), The NRF RY model can be expressed in the form of the fitting parameters C_0, C_1, and C_2 as follows and can then be used to fit the experimental or the simulation results, in the case of backward or forward scattering with a scattering angle of θ:

$$Y = C_0 \cdot \frac{1 - e^{-[C_1(1+\sec(\theta))+C2]x}}{C_1(1+\sec(\theta))+C2}. \tag{22}$$

In contrast, for scattering NRF γ-rays with an angle of 90°:

$$Y = C_0 \cdot \frac{1 - e^{-(2C_1+C_2)x}}{2C_1 + C_2} e^{-C_1 r}, \tag{23}$$

where

$$C_0 = \Phi_i \cdot \Gamma_D \cdot N \cdot \sigma_{NRF}(E_r) \cdot W(\theta) \cdot \varepsilon(E_r) \cdot \frac{\Omega}{4\pi} \cdot \prod_k \exp(-\mu_k x_k), \tag{24}$$

$C_1 = \mu_e(E_r)$, and $C_2 = \mu_{NRF}(E_r)$,

where $\prod_k \exp(-\mu_k x_k)$ is the multiplicand of the attenuations of the absorbers, *k*, that sit in front of the detector.

Using these equations, we can determine the total attenuation factor from the fitting parameters, as well as the NRF attenuation factor from the parameter C_2, and therefore, the NRF cross-section using C_2, and Equation (13) can be calculated.

5.2. Effective thickness and attenuation length

The attenuation length is equal to the maximum effective length of the NRF interaction within the target, as presented by Equation (20). For example, for the resonance level of 2.468 MeV, the attenuation factor of the atomic interaction at the γ-energy of 2.468 MeV is 0.852±0.008 cm^{-1} (where the atomic attenuation can be calculated from the XCOM [18] database or using GEANT4 that are derived from the evaluated photon data library (EPDL97), which states that photon cross sections should be accurate with less than 1% uncertainty [19], the second method is used), the total attenuation factor according to the experimental fitting is 2.91±0.38 cm^{-1}, and the attenuation factor for the NRF interaction is determined to be 1.21±0.16 cm^{-1}. Thus, the total attenuation length of the absorption and scattering γ-rays is 3.43 mm, whereas it is only 4.85 mm for the absorption component. On the other hand, the maximum effective thickness for the NRF interaction through the DU at any thickness ≥ (x^{Max} = 25 mm) is 3.43 mm according to Equations (19) and (20). Consequently, the total attenuation length through the DU target is equal to the maximum effective length for the NRF interaction at the maximum thickness of 25 mm for a resonance state of 2.468 MeV.

5.3. NRF cross-section calculations

We should emphasis that the NRF RY model can determine the NRF cross-section based on the attenuation factor of the NRF interaction that is determined from the fitting parameters of the experimental saturation curve. The attenuation factor for the NRF excitation level of 2.468 MeV of ^{238}U is 1.21 cm^{-1}, as discussed in the previous subsection. Therefore, the NRF cross-section for a state of 2.468 MeV of ^{238}U is 50.4±3.3 b.eV for an absolute temperature of 300 K (or 51.4±3.3 b.eV for an effective temperature of 312 K that is based on the Debye temperature of the metallic uranium of 207 K [20]). On the other hand, the NRF cross-section of this level is 38 b.eV [7], 61 b.eV [15] or 80±8 b.eV [16], as previously reported. The value determined by the NRF RY model is between these values and similar to the value reported in ENSDF [15].

Consequently, we propose a new technique to determine the NRF interaction cross-section based on the saturation-curve fitting by the NRF RY model to directly extract the NRF attenuation factor, and thereafter, the NRF cross-section. The merit of this method is that we can avoid many systematic errors that occur in the NRF experiments, such as the flux measurements and the detector detection efficiency. Additionally, the absolute values of the parameters of C_0, such as the incident flux, detector efficiency and so on are not required in this case.

Table 2 presents the NRF transitions for the γ-rays of ^{238}U at approximately 2.5 MeV, for which the incident beam energy is 2.48 MeV with an energy width (FWHM) of 5%. In addition, the attenuation factors of the atomic and the NRF interaction, which are extracted from the fitting parameters of the scattering yield of the NRF transitions with the target thickness, are presented in the third and fourth columns. The total attenuation length (or the maximum effective length) is determined from the total attenuation factor of the γ-rays due to the attenuation and the self-absorption component in the fifth column. The last two columns present the calculations of the NRF cross-section transitions by both methods determined from the absolute measurement (common method) and from the NRF saturation curve (proposed method).

The common method is based on a single measurement; thus, the absolute values presented in Equation (18) are necessary to evaluate the cross-section, including the incident flux per eV, the effective angular distribution of the scattering NRF γ-rays, the detection efficiency and so on. Therefore, the uncertainties in the common method are numerous due to all of the parameters presented in Equation (22) or (23) which have three constant parameters C_0 (Equation (24)), C_1 and C_2 as well the two variables of NRF yield (Y) and the target thickness (x). Thus, the uncertainty in NRF cross-section due to the absolute measurement can be expressed by:

$$\frac{\Delta(\sigma_{NRF})}{\sigma_{NRF}}=\left[\left(\frac{\Delta(Y)}{Y}\right)^2+\left(\frac{\Delta(x)}{x}\right)^2+\left(\frac{\Delta(\Phi_i)}{\Phi_i}\right)^2+\left(\frac{\Delta(N)}{N}\right)^2+\left(\frac{\Delta(W(\theta))}{W(\theta)}\right)^2\right.$$
$$\left.+\left(\frac{\Delta(\varepsilon(E_r))}{\varepsilon(E_r)}\right)^2+\left(\frac{\Delta(\mu_e(E_r))}{\mu_e(E_r)}\right)^2+\left(\frac{\Delta(\mu_{NRF}(E_r))}{\mu_{NRF}(E_r)}\right)^2\right]^{1/2} \tag{25}$$

In contrast, the uncertainty in the cross-section that is extracted from the saturation-curve fitting of the NRF yield, using the fitting function of Equations (22) or (23), is derived from the error in the attenuation factors of, $\sigma(\mu_e)$ and $\sigma(\mu_{NRF})$, that is determined by the uncertainty in the parameter C_1 and C_2 (directly determined from the fitting). Therefore, the uncertainty in NRF cross-section, $\sigma(\sigma_{NRF})$, can be addressed by:

$$\frac{\Delta(\sigma_{NRF})}{\sigma_{NRF}} = \sqrt{\left(\frac{\Delta(Y)}{Y}\right)^2 + \left(\frac{\Delta(x)}{x}\right)^2 + \left(\frac{\Delta(\mu_e(E_r))}{\mu_e(E_r)}\right)^2 + \left(\frac{\Delta(\mu_{NRF})}{\mu_{NRF}}\right)^2} \quad (26)$$

While as, the uncertainty in C_1 and C_2 which getting by the fitting, could include the uncertainty in the variables of NRF yield (*Y* – NRF peak area) and the target thickness that were included in the plotted data. Thus, the uncertainty in the NRF cross-section in this case simply giving by:

$$\frac{\Delta(\sigma_{NRF})}{\sigma_{NRF}} = \sqrt{\left(\frac{\Delta(\mu_e(E_r))}{\mu_e(E_r)}\right)^2 + \left(\frac{\Delta(\mu_{NRF})}{\mu_{NRF}}\right)^2} \quad (27)$$

The NRF cross-sections deduced by both methods are consistent with each other within the uncertainty. Moreover, the cross-sections calculated by the saturation method show slightly higher values than those from the other method. For example, the NRF cross-section of the state at 2.468 MeV is 47.4±4.3 b.eV, whereas the cross-section extracted from the fitting function for the multiple measurements is deduced to be 51.4±3.3 b.eV. The difference in the values estimated by the two methods arises from the precise correction for the attenuation and the self-absorption by the thickness dependency estimation of the incident γ-ray. Consequently, the scattering yield for the thickness-dependence measurements of the NRF with the RY model could introduce the precise estimation of the NRF cross-section without a need for absolute measurements. In addition, the uncertainty will be reduced.

6. Summary

The NRF reaction-yield dependence on the target thickness was studied. NRF measurements with four different thicknesses of ^{238}U, namely 1.287, 2.703, 4117 and, 5.563

mm, were performed in the HIγS facility. The NRF yield shows a saturation behavior for the NRF interaction with the target thickness. An NRF RY model was designed to interpret the saturation behavior of the NRF yield. Additionally, Monte Carlo simulation by GEANT4 was conducted for the NRF interaction to study a wide range of thicknesses. The simulation results were consistent with the experimental result for a transition level of 2.468 MeV. The analytical model of the NRF RY can reproduce both the experimental and simulation results. The most significant outcome of this study was the estimation of the NRF cross-section precisely without any absolute measurements of the photon flux, detection efficiency and so on. This method precisely analyzes the attenuation of the incident γ-rays due to atomic and nuclear absorption and the self-absorption of the emitted NRF γ-rays. The NRF cross-sections for 11 excitation levels were deduced from the absolute measurements and the saturation-curve fitting of the scattering yield of the NRF versus the target thickness. The NRF cross-sections deduced from the saturation-curve fitting show reasonable agreement with these from the absolute measurement with high accuracies.

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Table 1. Run summary of the NRF measurements for various thicknesses of the DU target. Eight foils from the DU target were used in these measurements. The total mass of each target is presented in units of g, and the total thickness is in units of mm. Each target had a cylindrical shape with a diameter of 25.4±0.5 mm. The uncertainties in DU masses and the thicknesses are approximately 1%.

#Run	#Foils	Mass (g)	Thickness (mm)
1	2	12.42	1.287
2	4	26.08	2.703
3	6	39.72	4.117
4	8	53.67	5.563

Table 2. The NRF excitation energies and their Doppler widths of ^{238}U at an effective temperature of 312 K are presented in the first and second columns. The third to fifth columns present the attenuation factors for the atomic and NRF interactions as well as the attenuation length or the maximum effective thickness for NRF γ-rays. The last two columns present the integrated NRF cross-sections calculated by the saturation-curve fitting and by the absolute measurements. The last column is calculated for a target thickness of 1.287 mm (run #1). The uncertainties in the atomic attenuation factors are less than 1% according to the EPDL97 [19].

E_γ (keV)	Γ_D (eV)	μ_e (cm^{-1})	μ_{NRF} (cm^{-1})	$x_{effective}^{max} \| \chi_0$ (mm)	σ_{NRF}(b.eV) Saturation curve	Absolute measurement
2365	1.939	0.8591	1.36±0.21	3.25	54.1±4.4	44.1±4.9
2410	1.976	0.8557	1.12±0.15	3.53	45.6±3.1	37.1±3.9
2423	1.986	0.8547	0.81±0.13	3.96	34.0±2.8	33.0±3.5
2442	2.002	0.8534	0.12±0.10	5.48	4.9±2.1	3.9±1.9
2454	2.012	0.8525	0.97±0.16	3.75	40.8±3.4	38.5±3.8
2468	2.023	0.8516	1.21±0.16	3.43	51.4±3.3	47.4±4.3
2472	2.027	0.8512	0.31±0.13	4.98	12.8±2.7	10.1±2.9
2492	2.043	0.8500	0.19±0.12	5.29	8.2±2.5	7.5±2.7
2499	2.049	0.8495	1.11±0.16	3.56	46.8±3.3	45.5±4.1
2511	2.059	0.8488	0.10±0.08	5.57	4.3±1.7	4.1±1.9
2529	2.073	0.8477	0.27±0.13	5.10	11.6±2.7	9.5±2.7

Figure captions List

Figure 1. Schematic side view of the experimental setup of the NRF measurements of the ^{238}U at HIγS facility at Duke University, including the γ-ray track simulated using the MC simulation, GEANT4.

Figure 2. Schematic view of the slab geometry for the NRF interaction point, as well as the scattering γ-rays in the backward and forward directions.

Figure 3. The Compton-scattered energy spectrum for the incident-beam energy of 2.475 MeV from a scatter Cu-plate (1.1-mm-thick) with an angle of 8.6°. The Compton-scattered full energy photo-peak obtained by a double Gaussian fit to the photo-peak of the source spectrum to isolate the photo-peak from the Compton edge, as shown by the black peak.

Figure 4. The summation of the experimental energy spectra of the scattering NRF γ-rays with an angle of 90°. The on-beam spectrum, the nonlinear–background estimation curve and the subtracted spectrum are presented. The statistical errors for both spectra are less than 5%.

Figure 5. The NRF yield dependence on the target thickness of the ^{238}U in the state of 2.468 MeV. The experimental results of the four measurements for the various thicknesses of the DU. The simulation results for a wide range of the various thicknesses, x = 0.5, 2, 4, 6, 8, 10, 12, 16 and 20 mm. The reaction-yield model (RYM) of the NRF interaction performed for both the experimental and the simulation results, as shown by the fitting curves in the figure.