

Measurement of yields for the $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ reactions induced by 2.5 GeV bremsstrahlung

Nguyen Thi Hien ^a, Guinyun Kim ^{a*}, Kwangsoo Kim ^a, Muhammad Shahid ^a, Muhammad Zaman ^a, Muhammad Nadeem ^a, Pham Duc Khue ^b, Kim Tien Thanh ^b, Nguyen Van Do ^b

^aDepartment of Physics, Kyungpook National University, Daegu 702-701, Republic of Korea

^bInstitute of Physics, Vietnam Academy of Science and Technology, 10 Dao Tan, Hanoi, Vietnam

*Corresponding author: gnkim@knu.ac.kr

1. Introduction

The photonuclear reactions with high energy bremsstrahlung photons generated from the electron linear accelerators (Linac) have been the subject of many studies. One method of studying these reactions is to measure the yields of the radioactive residual nuclei [1]. The reaction yield is one of the basic data for use in both basic and applied nuclear physics research [2,3]. In practice, the reaction yield is computed from the production rate of a nuclide due to a certain nuclear reaction.

Most of the reaction yields reported so far have been measured at low energies [4-7]. At low photon energies only simple reactions such as (γ, n) , (γ, p) or similar processes are available. However, at high photon energies the photonuclear reactions with the emission of multinucleons are the most probable. Therefore, studies of high-energy nuclear reactions are of great important. It may help in deeper understanding of the reaction mechanisms and in extending of various fields of applications such as astrophysics, radiation physics, intense neutron source production and nuclear waste transmutation.

The aim of the present work is to investigate the multineutron photonuclear on ^{197}Au bombarded by 2.5 GeV bremsstrahlung. Most of the photodisintegration products of gold with half-lives sufficient for the activity measurement. In literature we have found some reaction yields for the $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ photonuclear reactions measured with low bremsstrahlung energies, namely from the reaction threshold up to just above the Giant Dipole Resonance (GDR) region. The neutron numbers ejected have been identified to be $x = 1-6$ [8-10]. In this work by using the 2.5 GeV bremsstrahlung we can obtain eight $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ reaction products. The residual nuclei $^{197-x}\text{Au}$ were identified from a knowledge of the target nuclide, the γ -ray

spectra, decay data and current literature data. The reaction yields were converted from the measured γ -ray activities. The high energy resolution HPGe γ -ray spectrometer makes such measurements relatively easy.

2. Experimental

2.1 Sample irradiation

The sample irradiation was carried out at the 2.5 GeV electron linac of the Pohang Accelerator Laboratory (PAL), POSTECH, Pohang, Korea. The bremsstrahlung photons were produced when a pulsed electron beam hit a thin W target with a size of 50 mm \times 50 mm and a thickness of 0.2 mm. The W target is located at 38.5 cm from the electron beam exit window. In this work, the natural gold foil in disc shape with diameter of 1.27 cm, thickness of 0.03 mm and purity of 99.95% was placed 24 cm from the W target. The irradiation geometry is shown in Fig. 1. The duration of irradiation was 170 min with total beam current of 7.26×10^{13} electrons. During irradiation the electron linac was operated with a repetition rate of 10 Hz, a pulse width of 1 ns, and electron energy of 2.5 GeV. The radioactive residual nuclei formed via the multiple photoneutron reactions $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ and their main decay data are given in Table 1 [11]. Beside the $^{197-x}\text{Au}$ radioactive nuclides a large number of radioactive nuclides are also formed simultaneously via the $^{197}\text{Au}(\gamma, xnkp)$ spallation reactions (with $x, k \geq 1$), and some of them pose a serious interfering problem in the activity measurements. The possible interfering reactions and their main decay data are given in Table 2.

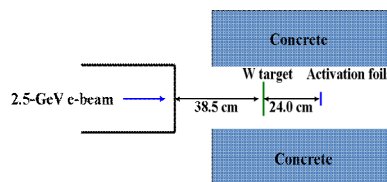


Fig.1. Experimental arrangement for the irradiation of activation foil.

Table 1: Nuclear reactions investigated and main decay data of the reaction products[1]:

Nuclear reaction	E_m (MeV)	Half-life	Main γ -ray energy (keV) and intensity (%)
$^{197}\text{Au}(\gamma, n)^{196}\text{Au}$	8.07	6.183 d	332.98 (22.9); 355.68(87); 426.0(7.0)
$^{197}\text{Au}(\gamma, 2n)^{195}\text{Au}$	14.71	186.09 d	98.85(10.9); 129.70(0.82)
$^{197}\text{Au}(\gamma, 3n)^{194}\text{Au}$	23.08	38.02 h	293.55(10.4); 328.45(61.0); 645.16(2.14)
$^{197}\text{Au}(\gamma, 4n)^{193}\text{Au}$	30.46	17.65 h	186.17(9.4); 255.57(6.2); 268.22(3.6)
$^{197}\text{Au}(\gamma, 5n)^{192}\text{Au}$	38.73	4.94 h	295.96(22.3); 308.46(3.45); 316.51(58); 612.46(4.34)
$^{197}\text{Au}(\gamma, 6n)^{191}\text{Au}$	45.73	3.18 h	166.5(3.32); 194.12 (2.74); 253.94(2.53); 277.88(7.2); 283.91(6.7); 399.84(4.7);
$^{197}\text{Au}(\gamma, 7n)^{190}\text{Au}$	54.77	42.8 min	295.78(71); 301.82(23.4); 597.67(9.4)
$^{197}\text{Au}(\gamma, 8n)^{189}\text{Au}$	62.14	28.7 min	447.65(55); 713.17(100); 812.68(63)

Table 2. Interfering reactions and main decay data [11].

The γ -rays of interest	The interference reactions; γ -ray energy, intensity and half-life of the reaction products
$E_\gamma = 426.0 \text{ keV}$ (7.0%) (^{196}Au)	$^{197}\text{Au}(\gamma, 15n5p)^{177}\text{W}$: $E_\gamma = 426.98 \text{ keV}$ (13.2%); $T_{1/2} = 135 \text{ min}$ $^{197}\text{Au}(\gamma, 8n2p)^{187}\text{Ir}$: $E_\gamma = 427.12 \text{ keV}$ (4.12%); $T_{1/2} = 10.5 \text{ h}$
$E_\gamma = 98.85 \text{ keV}$ (10.9%) (^{195}Au)	$^{197}\text{Au}(\gamma, 5n1p)^{191}\text{Pt}$: $E_\gamma = 96.52 \text{ keV}$ (3.28%); $T_{1/2} = 2.802 \text{ d}$ $^{97}\text{Au}(\gamma, 10n2p)^{182}\text{Ir}$: $E_\gamma = 97.4 \text{ keV}$ (4.2%); $T_{1/2} = 14.4 \text{ h}$
$E_\gamma = 129.7 \text{ keV}$ (0.82%) (^{195}Au)	$^{197}\text{Au}(\gamma, 5n1p)^{191}\text{Pt}$: $E_\gamma = 129.42 \text{ keV}$ (3.2 %); $T_{1/2} = 2.802 \text{ d}$
$E_\gamma = 328.45 \text{ keV}$ (61.0%) (^{194}Au)	$^{197}\text{Au}(\gamma, 18n5p)^{174}\text{W}$: $E_\gamma = 328.68 \text{ keV}$ (9.5%); $T_{1/2} = 31 \text{ min}$
$E_\gamma = 645.16 \text{ keV}$ (2.14%) (^{194}Au)	$^{197}\text{Au}(\gamma, 9n3p)^{185}\text{Os}$: $E_\gamma = 646.12 \text{ keV}$ (78.0%); $T_{1/2} = 93.6 \text{ d}$
$E_\gamma = 186.17 \text{ keV}$ (9.4%) (^{193}Au)	$^{197}\text{Au}(\gamma, 15n5p)^{177}\text{W}$: $E_\gamma = 186.42 \text{ keV}$ (7.8%); $T_{1/2} = 135 \text{ min}$
$E_\gamma = 255.57 \text{ keV}$ (6.2%) (^{193}Au)	$^{197}\text{Au}(\gamma, 10n2p)^{182}\text{Ir}$: $E_\gamma = 254.4 \text{ keV}$ (13.3%); $T_{1/2} = 14.4 \text{ h}$
$E_\gamma = 268.22 \text{ keV}$ (3.6%) (^{193}Au)	$^{197}\text{Au}(\gamma, 16n6p)^{175}\text{Ta}$: $E_\gamma = 266.9 \text{ keV}$ (10.8%); $T_{1/2} = 10.5 \text{ h}$
$E_\gamma = 308.46 \text{ keV}$ (3.45%) (^{192}Au)	$^{197}\text{Au}(\gamma, 3n2p)^{192}\text{Ir}$: $E_\gamma = 308.46 \text{ keV}$ (30%); $T_{1/2} = 73.831 \text{ d}$
$E_\gamma = 316.51 \text{ keV}$ (58%) (^{192}Au)	$^{197}\text{Au}(\gamma, 3n2p)^{192}\text{Ir}$: $E_\gamma = 316.51 \text{ keV}$ (82.81%); $T_{1/2} = 73.831 \text{ d}$
$E_\gamma = 295.96 \text{ keV}$ (22.3%) (^{192}Au)	$^{197}\text{Au}(\gamma, 3n2p)^{192}\text{Ir}$: $E_\gamma = 295.96 \text{ keV}$ (28.67%); $T_{1/2} = 73.831 \text{ d}$
$E_\gamma = 166.5 \text{ keV}$ (3.32%) (^{191}Au)	$^{197}\text{Au}(\gamma, 20n7p)^{170}\text{Hf}$: $E_\gamma = 164.71 \text{ keV}$ (26%); $T_{1/2} = 16.1 \text{ h}$
$E_\gamma = 194.12 \text{ keV}$ (2.74%) (^{191}Au)	$^{197}\text{Au}(\gamma, 8n1p)^{183}\text{Pt}$: $E_\gamma = 187.59 \text{ keV}$ (19.4%); $T_{1/2} = 10.2 \text{ d}$
$E_\gamma = 253.94 \text{ keV}$ (2.53%) (^{191}Au)	$^{197}\text{Au}(\gamma, 10n2p)^{183}\text{Ir}$: $E_\gamma = 254.4 \text{ keV}$ (13.3%); $T_{1/2} = 14.4 \text{ h}$
$E_\gamma = 283.91 \text{ keV}$ (6.7%) (^{191}Au)	$^{197}\text{Au}(\gamma, 12n2p)^{183}\text{Ir}$: $E_\gamma = 282.39 \text{ keV}$ (4.9%); $T_{1/2} = 58 \text{ min}$
$E_\gamma = 295.78 \text{ keV}$ (71.0%) (^{190}Au)	$^{197}\text{Au}(\gamma, 5n)^{192}\text{Au}$: $E_\gamma = 295.33 \text{ keV}$ (22.3%); $T_{1/2} = 4.94 \text{ h}$

2.2 Activity measurement

The measurements started soon after the end of the irradiation and continued for several weeks. The measuring times were varied from few tens of minutes to some hours depending on the statistics of the γ -ray peaks of interest. A typical gamma spectrum of the activated gold foil is given in Fig. 2.

The activity of each radioactive reaction product was determined from the measured γ -spectra based on the γ -peak with high intensity, well separated, and relatively low background. The photopeak area was corrected for the counting efficiency of the detector and the γ -ray branching ratio in the decay scheme. The resulting activity was corrected to the end of the irradiation time. For the photopeaks having

interferences the corrections were made by the same ways as mentioned in our previous work [12].

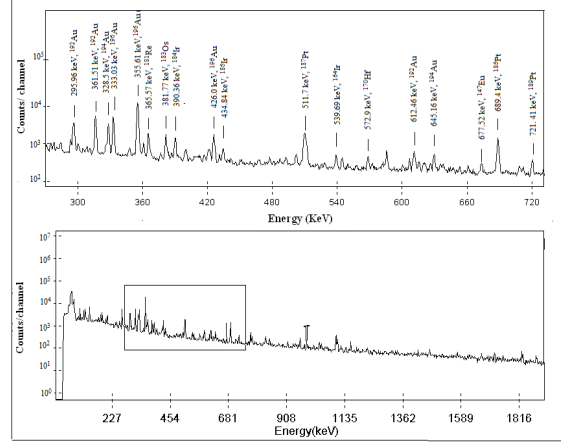


Fig.2. Typical gamma-ray spectrum of the gold foil irradiated with 2.5 GeV bremsstrahlung with irradiation time 240 min, waiting time 215 min and measuring time 5 min.

3. Data analysis

For any nuclear reaction, the yield can be expressed as follows:

$$Y(E^{\max}) = N \int_{E_{th}}^{E_{\max}} \sigma(E) \phi(E) dE \quad (1)$$

where $\sigma(E)$ is the energy dependent reaction cross-section, $\phi(E)$ is the flux of bremsstrahlung photon at the sample position, integral extends from the reaction threshold energy, E_{th} , up to the maximum bremsstrahlung energy, E_{\max} , respectively.

By considering the pulse nature of the bremsstrahlung beam, relation between the reaction yield and the photopeak area of the measured γ -ray, S , can be expressed as follows:

$$Y(E^{\max}) = \frac{S \lambda (1 - e^{-\lambda T})}{N_0 I \varepsilon (1 - e^{-\lambda \tau}) (1 - e^{-\lambda t_i}) e^{-\lambda t_w} (1 - e^{-\lambda t_c})} \quad (2)$$

where N_0 is the number of target nuclei, I is the intensity of the measured γ -ray, ε is the detection efficiency for the γ -ray of interest, λ is the decay constant of the radioactive isotope, τ is the pulse width, T is the cycle period, t_i is the irradiation time, t_w is the waiting time, and t_c is the counting time.

In the present work, the energies of γ -rays to be measured are varied from about 100 keV to 800 keV. In addition the gold is a heavy element, therefore the counting loss due to the γ -ray attenuation was taken

into account. The self attenuation factor, F_{att} was approximated by the following expression:

$$F_{att} = \frac{\mu t}{1 - \mu t} \quad (3)$$

where μ is the linear attenuation coefficient and t is the thickness of the sample.

As can be seen in Table 1, most radioactive products emit two or more γ -rays in cascade, therefore in order to improve the accuracy of the activity measurement appropriate corrections for the summation of coincident γ -rays were made [13,14]. The summing correction factors, C , corresponding to the sample-detector distances of 2.5- and 5-cm are given in Table 3.

3. Results and discussion

Total eight radioactive nuclides $^{197-x}\text{Au}$ (with $x = 1-8$) formed in the $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ multiple photoneutron reactions with 2.5 GeV bremsstrahlung were identified based on their characteristic γ -ray energies and half-lives. The bremsstrahlung photon spectrum is approximated to be $1/E$ shape. Hence, if the integrated cross section for the $(\gamma, 1n)$ reaction is available, the integrated cross-section for any (γ, xn) process can also be calculated from the relative yield ratio. The method does not require a reasonably accurate evaluation of the value of E_{max} . Due to this convenient, in this work the obtained yields were normalized to that of the $^{197}\text{Au}(\gamma, n)^{196}\text{Au}$ reaction. By this way we can avoid a number of systematic errors.

For comparison, we calculated the reaction yields for the $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ reactions by using the following formula [15]:

$$Y(\gamma, xn) = 0.135A^{0.684} \exp[-46A^{-0.72}(x-1)^{0.85}] \quad (4)$$

The normalized yields obtained for the $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ reactions together with the reference data measured at 65 MeV [9], and 67.7 MeV [10] are given in Table 4. The total error for each reaction yield was calculated by adding in quadrature the statistical counting error, the error in the HPGe detector efficiency, the estimated error in the variation of the bremsstrahlung beam and the uncertainties of the decay data used in calculation.

From Table 4 we can see that the reaction yields vary with bremsstrahlung energies. They all show

similar decreasing trend with increasing number of ejected neutrons. At lower incident bremsstrahlung energies the decreasing trend is faster. In addition, we also recognize that there is a reasonable agreement between the reaction yields measured with 65 MeV [9] and 67.7 MeV [10] bremsstrahlung.

Table 3. Coincidence summing correction factors for the γ -rays used in yield determinations:

Radioactive isotopes	E_γ (keV)	Coincidence summing correction factor	Summing correction factor, distance d (cm)	
			d1 = 2.5	d2 = 5.0
^{196}Au	$\gamma_1 = 333.03$	C_1	1.21	1.09
^{196}Au	$\gamma_2 = 355.68$	C_2	1.05	1.02
^{194}Au	$\gamma_1 = 293.58$	C_1	1.23	1.09
^{194}Au	$\gamma_2 = 328.50$	C_2	1.03	1.01
^{192}Au	$\gamma_1 = 295.96$	C_1	1.23	1.09
^{192}Au	$\gamma_2 = 316.51$	C_2	1.08	1.03
^{190}Au	$\gamma_1 = 301.82$	C_1	1.25	1.10
^{190}Au	$\gamma_2 = 295.78$	C_2	1.07	1.03

Table 4: Yields of the $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ photonuclear reactions

Neutron number	Reaction yields (rel. unit)			
	2.5 GeV [this work]	65 MeV [9]*	67.7 MeV [10]	2.5 GeV [Cal.]
1	1.0000	1.0000	1.0000	1.0000
2	0.329 ± 0.021	0.197 ± 0.012	0.160 ± 0.01	0.2393
3	0.0828 ± 0.0056	0.0250 ± 0.0017	0.023 ± 0.002	0.1051
4	0.0480 ± 0.0037	0.0097 ± 0.0012	0.0074 ± 0.001	0.0491
5	0.0169 ± 0.0014	0.0035 ± 0.0004	0.0025 ± 0.0002	0.0239
6	0.0126 ± 0.0012	0.00034 ± 0.00005	0.00050 ± 0.00007	0.0119
7	0.0077 ± 0.0008	-	-	0.0061
8	0.0027 ± 0.0004	-	-	0.0031

* The reaction yields were taken from the original data from Fig.3 of ref. [9].

4. Conclusion

Multiple photoneutron reactions on ^{197}Au induced by 2.5 GeV bremsstrahlung have been investigated by using the activation method. We identified and determined yields of 8 radioactive residual nuclides formed via the multiple photoneutron reactions $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$. In this work, the necessary corrections were made in order to improve the accuracy of the experimental results. The obtained experimental results are in good agreement with the calculated values. The yields for the $^{197}\text{Au}(\gamma, xn)^{197-x}\text{Au}$ reactions depend not only on the excitation energies but also on the number of neutrons ejected.

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