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STUDY OF MULTIPLE PHOTONEUTRON REACTIONS ON ^{197}Au INDUCED BY 2.5 GeV BREMSSTRAHLUNG

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Abstract. *We identified eight radionuclides ^{196}Au, ^{195}Au, ^{194}Au, ^{193}Au, ^{192}Au, ^{191}Au, ^{190}Au, ^{189}Au formed via the multiple photoneutron reactions $^{197}Au(\gamma, kn)^{197-k}Au$ with 2.5 GeV bremsstrahlung. The yields of radionuclides that decay by emitting γ-ray were measured using high purity germanium (HPGe) detector coupled to a PC-based multichannel analyzer. In order to improve the accuracy of the experimental results the necessary corrections were made. The obtained results are compared with reference data and the variations of the $^{197}Au(\gamma,kn)^{197-k}Au$ reaction yields according to incident bremsstrahlung energy and neutron multiplicity are also discussed.*

I. INTRODUCTION

When high-energy photons pass through matter multiparticle photonuclear reactions are induced. 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]. The measurements with high energy projectiles, especially with high energy photons are still very scarce. At low photon energies only simple reactions such as (γ,n), (γ,p) or similar processes are available. These reactions represent only a part of the possible interactions between the photons and complex nuclei. 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 measure the yields for the ^{197}Au(γ,kn)$^{197-k}$Au nuclear reactions induced by 2.5 GeV bremsstrahlung. We have chosen the gold target nuclei for the investigation because in many cases it has been used as a monitor reaction. 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}Au(γ,kn)$^{197-k}$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 k = 1- 6 [8-10]. In this work by using the 2.5 GeV bremsstrahlung we obtain eight ^{197}Au(γ,kn)$^{197-k}$Au reaction products. The radioactive residual nuclides $^{197-k}$Au were identified based on the knowledge and the information of the target nuclide, the γ-ray spectra and the decay data. The reaction yields were converted from the measured γ-ray activities. The high energy resolution γ-ray spectrometer based on a HPGe detector makes such measurements relatively easy. In order to improve the accuracy of the experimental results the corrections for the γ-ray interference and the coincidence summing effect were made. The obtained results are compared with those previously obtained by other authors, and the energy dependence of the reaction yields is discussed.

II. EXPERIMENTAL PROCEDURE

II.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 by bombarding a pulsed electron beam into a thin W target with a size of 50 mm × 50 mm and a thickness of 0.2 mm. It is located 38.5 cm from the electron beam exit window. The details of the 2.5 GeV electron linac and its applications were described elsewhere [11-12]. For irradiation, 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. At a given experimental arrangement the irradiation angle is small (about 3 degree), so that the total area of the sample is covered by the bremsstrahlung beam with high intensity and relatively high homogeneity.

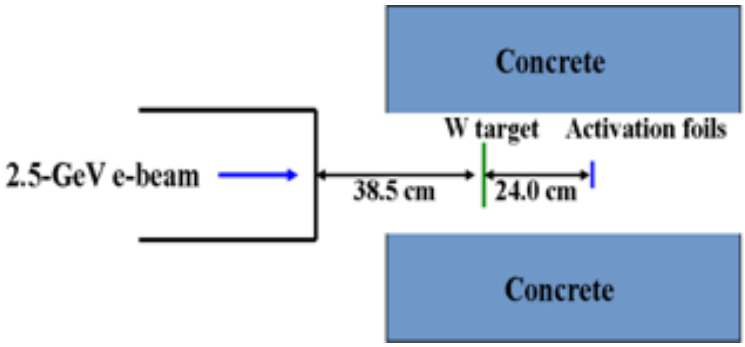

Fig. 1. Experimental arrangement for the irradiation of activation foil.

The gold foil was irradiated for 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}Au(γ,kn)$^{197-k}$Au and their main decay

data are given in Table 1 [13]. Beside the $^{197-k}$Au radioactive nuclides a large number of radioactive nuclides are also formed simultaneously via the photospallation reactions ^{197}Au(γ,knlp) with k, l $\geq$ 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.

Table 1. Nuclear reactions investigated and main decay data of the reaction products [13]

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

Table 2. Interfering reactions and main decay data [13]

The γ-rays of interest	The interference reactions; γ-ray energy, intensity and half-life of the reaction products
E_γ = 426.0keV (7.0%) (^{196}Au)	^{197}Au(γ,15n5p)^{177}W: E_γ=426.98 keV (13.2%); $T_{1/2}$=135 min
	^{197}Au(γ,8n2p)^{187}Ir: E_γ= 427.12 keV (4.12%); $T_{1/2}$ = 10.5 h
E_γ = 98.85keV (10.9%) (^{195}Au)	^{197}Au(γ,5n1p)^{191}Pt: E_γ = 96.52 keV (3.28%); $T_{1/2}$ = 2.802 d
	^{197}Au(γ,10n2p)^{185}Ir: E_γ=97.4 keV (4.2%); $T_{1/2}$=14.4 h
E_γ = 129.7keV (0.82%) (^{195}Au)	^{197}Au(γ,5n1p)^{191}Pt: E_γ=129.42 keV (3.2 %); $T_{1/2}$=2.802 d
E_γ = 328.45keV (61.0%) (^{194}Au)	^{197}Au(γ,18n5p)^{174}W: E_γ=328.68 keV(9.5%); $T_{1/2}$=31 min
E_γ = 645.16keV (2.14%) (^{194}Au)	^{197}Au(γ,9n3p)^{185}Os: E_γ= 646.12 keV (78.0%); $T_{1/2}$=93.6 d
E_γ = 186.17keV(9.4%) (^{193}Au)	^{197}Au(γ,15n5p)^{177}W: E_γ= 186.42 keV (7.8%); $T_{1/2}$=135 min
E_γ = 255.57keV (6.2%) (^{193}Au)	^{197}Au(γ,10n2p)^{185}Ir: E_γ=254.4 keV(13.3%); $T_{1/2}$=14.4 h
E_γ = 268.22keV(3.6%) (^{193}Au)	^{197}Au(γ,16n6p)^{175}Ta: E_γ=266.9 keV(10.8%); $T_{1/2}$=10.5 h
E_γ = 308.46keV(3.45%) (^{192}Au)	^{197}Au(γ,3n2p)^{192}Ir: E_γ= 308.46 keV (30%); $T_{1/2}$ = 73.831 d
E_γ = 316.51keV(58%) (^{192}Au)	^{197}Au(γ,3n2p)^{192}Ir: E_γ=316.51 keV (82.81%); $T_{1/2}$ = 73.831 d
E_γ = 295.96keV(22.3%) (^{192}Au)	^{197}Au(γ,3n2p)^{192}Ir:E_γ= 295.96 keV (28.67%); $T_{1/2}$ = 73.831 d
E_γ = 166.5keV(3.32%) (^{191}Au)	^{197}Au(γ,20n7p)^{170}Hf: E_γ=164.71 keV(26%); $T_{1/2}$ = 16.1 h
E_γ = 194.12keV(2.74%) (^{191}Au)	^{197}Au(γ,8n1p)^{188}Pt:E_γ= 187.59 keV(19.4%); $T_{1/2}$ = 10.2 d
E_γ = 253.94keV(2.53%) (^{191}Au)	^{197}Au(γ,10n2p)^{185}Ir: E_γ=254.4 keV(13.3%); $T_{1/2}$ = 14.4 h
E_γ = 283.91keV(6.7%) (^{191}Au)	^{197}Au(γ,12n2p)^{183}Ir: E_γ=282.39 keV(4.9%); $T_{1/2}$ = 58 min
E_γ = 295.78keV (71.0%) (^{190}Au)	^{197}Au(γ,5n)^{192}Au:E_γ=295.33 keV(22.3%); $T_{1/2}$ = 4.94 h

II.2. Activity measurement

The radioactivity measurements were made by taking the γ-ray spectra from the irradiated gold foil with a γ-ray spectrometer. It consists of a coaxial HPGe detector (Canberra) with the energy resolution of 1.8 keV and the relative efficiency is 20% at the 1332.5 keV γ-peak of ^{60}Co. The detector is coupled to a computer-based multichannel analyzer card system, which could determine the photopeak area of the γ-spectrum by using the software Genie 2000. The counting efficiency of the detector was determined using the standard γ-sources [14,15].

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.

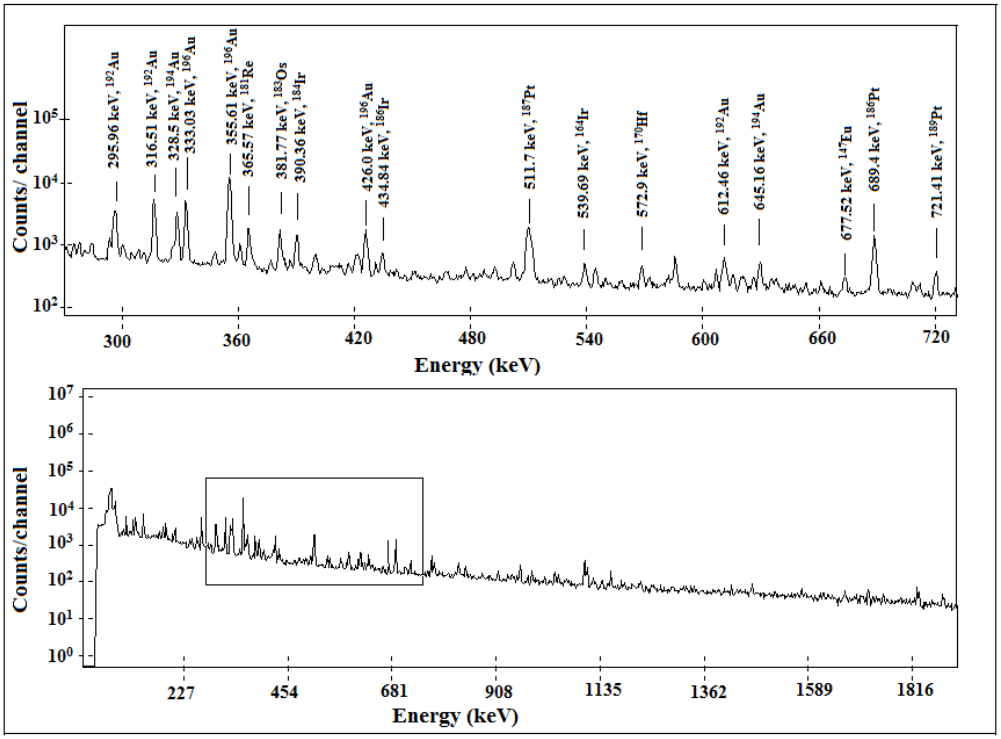

Fig. 2. Typical gamma-ray spectrum of the gold foil irradiated by 2.5 GeV bremsstrahlung with irradiation time 240 min, waiting time 215 min and measuring time 15 min.

II.3. Data analysis

Due to the continuity of the bremsstrahlung spectrum, the relationship between the yield of a photonuclear reaction, $Y(E^{\max})$, and the reaction cross section 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.

The activity of each radioactive reaction product was determined from the measured γ-spectra based on the γ-peaks with high intensity, well separated, and relatively low background. The photopeak area (or the number of counts) 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 [16].

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 correction factor for a γ-ray energy attenuation, F_{att} in the activation foil at a given γ-ray energy was approximated as follows:

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

where μ is the linear attenuation coefficient (cm^{-1}) and t is the thickness of the sample (cm). The measured activity can be corrected to zero attenuation by dividing with factor F_{att}.

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 [17,18]. The summing correction factors, C, corresponding to the sample-detector distances of 2.5- and 5-cm are given in Table 3.

III. RESULTS AND DISCUSSION

We identified and determined yields of eight radioactive nuclides formed via the multiple photoneutron reactions ^{197}Au(γ,kn)$^{197-k}$Au with 2.5 GeV bremsstrahlung. The

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

Radioactive isotopes	E_γ (keV) and I_γ(%)	Coincidence summing correction factor	Summing correction factor C at sample - detector 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

bremsstrahlung photon spectrum is approximated to be $1/E$ shape. Hence, if the integrated cross section for the (γ,1n) reaction is available, the integrated cross-section for any (γ,kn) 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}Au(γ,n)^{196}Au reaction. By this way we can avoid a number of systematic errors.

For comparison, we calculated the reaction yields for the ^{197}Au(γ,kn)$^{197-k}$Au reactions by using the following formula [19]:

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

The normalized yields obtained for the ^{197}Au(γ,kn)$^{197-k}$Au reactions together with the reference data measured at 65 MeV [9] and 67.7 MeV [10] bremsstrahlung beams 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.

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 beams.

IV. CONCLUSION

Relative yields of eight radioactive nuclides formed in the multiple photoneutron reactions ^{197}Au(γ,kn)$^{197-k}$Au with maximum end-point energies of 2.5 GeV bremsstrahlung have been measured by direct γ-ray counting of irradiated gold target nuclei. In order to improve the accuracy of the experimental results the necessary corrections were made.

Table 4. Yields of the ^{197}Au(γ,kn)$^{197-k}$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±0.0013	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]*

The obtained experimental results are in reasonable agreement with the calculated values. The yields for the ^{197}Au(γ,kn)$^{197-k}$Au reactions depend not only on the excitation energies but also on the number of neutrons ejected.

V. ACKNOWLEDGMENTS

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