Measurements of Isomeric Yield Ratios for ^{133m,g}Ce and ^{137m,g}Ce at 55-, 60-, and 65-MeV Bremsstrahlung

H. J. Kim[ab], M. W. Lee[a], D. H. Jeong[a], J. K. Kim[a], K. M. Yang[a], Y. R. Kang[a*], T. I. Ro[b], S. G. Shin [c], Y. Kye [c], G. N. Kim[d]
[a]*Research Center, Dongnam Inst. of Radiological & Medical Sciences, Busan 619-953, Korea*
[b]*Department of Physics, Dong-A University, Busan 604-714, Korea*
[c] *Department of Advanced Nuclear Engineering , POSTECH, Pohang 790-784, Korea*
[d] *Kyungpook National University, Daegu 702-701, Korea*
[*]*Corresponding author:yeongrok@dirams.re.kr*

1. Introduction

Activation by gamma irradiation can be used for different practical purposes like activation analysis, production of isotopes for medical use, material science investigations, and biological studies, etc. Nuclear reactions with the emission of different particles become an important source of information on the structure and properties of the excited states of the investigated nuclei. The isomeric yield ratios has become an important approach for studying the angular momentum effects in nuclear reactions and spin dependence of the nuclear level density, refinements in gamma transition and testing of nuclear models [1-4]. Nuclei with a metastable (isomeric)- and an unstable ground-state can be used to measure the relative population of these two states in nuclear reactions, the so-called isomeric cross-section ratio. In case of the bremsstrahlung photon irradiation, due to the continuity of the energy spectrum, the isomeric ratio can also be presented through the yields of the two states instead of the two cross-sections, namely, IR = $Y_{high\text{-}spin}/Y_{low\text{-}spin}$ [5].

The energy dependent isomeric yield ratios for the ^{133m,g}Ce and the ^{137m,g}Ce were performed using the activation method at Pohang Accelerator Laboratory (PAL). The aim of the present work is to measure the iso-meric yield ratios of the ^{133m,g}Ce and the ^{137m,g}Ce with bremsstrahlung energy of 55- to 65-MeV with a step of ΔE = 5 MeV from natural Cerium.

2. Experimental procedure

The experiment was performed by using the end-point bremsstrahlung energies of 55-, 60-, and 65-MeV at the electron linear accelerator (linac) which was designed for 100 MeV and can be operated from 40-, to 70-MeV [6]. The bremsstrahlung was produced when a pulsed electron hits a thin W-target with a size of 100 × 100 mm and a thickness of 0.1 mm . The W-target is located at 18 cm from the beam exit window. The natural cerium foil was placed in air at 12 cm from the W-target and they were positioned at zero degree with the direction of the electron beam.

High-purity natural Cerium foils in square shape, made by the Alfa Aesar Inc. (USA) were exposed to uncollimated bremsstrahlung beams from the electron linac. The characteristics of the Ce foils and beam conditions are given in Table 1. Thin Ce foils were used for the irradiation which led to the strongly reduced or

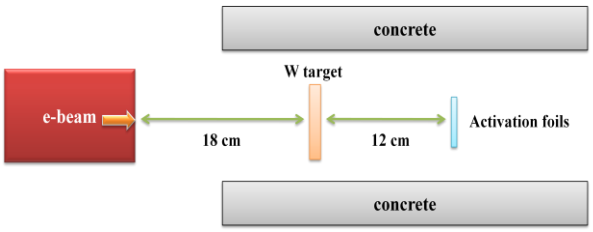

Fig. 1 Experimental set-up for sample irradiation

the negligible effect for a self-absorption of the measured γ-line. After an irradiation and an appropriate waiting time, the activated foils were take off, and then the induced gamma activities of the irradiated foils were measured by using γ-spectrometer without any chemical purification. The γ-spectrometer used for the measurements was a p-type coaxcial ORTEC high-purity germanium (HPGe) detector. The HPGe detector was connected to a PC-based multi-channel analyzer card system, which determined the photo-peak-area of the γ-ray spectra by using the GAMMA VISION (Ortec) data acquisition software. The energy resolution of the detector was 1.8 keV full width at half maximum (FWHM) at the 1332.5 keV peak of ^{60}Co. The standard sources of ^{133}Ba, ^{137}Cs, and ^{152}Eu were used for the energy and efficiency calibration of the detector system. In order to optimize the dead time and the coincidence summing effect we have also chosen the appropriate distance between the sample and the detector for each measurement.

Table 1: Characteristics of the ^{nat}Ce activation foils

	Energy(MeV)	55	60	65
Beam Condition	Pulse width (μs)	1.6	1.6	1.6
	Repetition rate(Hz)	15	15	15
	Irradiation time (h)	3	3	3
	Waiting time (min)	31.7	30.7	32
	Counts time (h)	3600	3600	3600
Sample Condition	Purity (%)	99.9	99.9	99.9
	Size (mm^2)	1	1	1
	Weight (g)	0.082	0.078	0.081
	Thickness (mm)	0.1	0.1	0.1

3. Data analysis

The reaction products are identified based on γ-line energies and half-lives of the produced isotopes. The activities of the reaction product can be determined from the respective photo-peak area and the detection efficiency. As the pulsed nature of irradiation source is used, the relationship between the numbers of detected γ-rays, A_γ and the reaction cross-section σ can be expressed as follows:

$$A_\gamma = \frac{I_\gamma \varepsilon N_0 C}{\lambda(1-e^{-\lambda T})}\left(1-e^{-\lambda\tau}\right)\left(1-e^{-\lambda t_i}\right)e^{-\lambda t_w}\left(1-e^{-\lambda t_c}\right) \times \int_{E_{th}}^{E_{\gamma,max}} \emptyset(E)\sigma_k(E)dE \qquad (1)$$

where N_0 is the number of target nuclei, $\emptyset$ is the incident photon flux, ε is the detection efficiency for the measured γ-ray, C is the correction factor for coincidence summing and γ-ray self-absorption, I_γ is the intensity of the γ-ray, $\lambda$is the decay constant of the isotope of interest ($\lambda = \ln 2/T_{1/2}$), τ is the pulse width of electron beam, T is the cycle period, t_i is the irradiation time, t_w is the waiting time(the period between the end of irradiation and the start of counting), and t_c is the counting time. $\sigma_k(E)$ is the energy dependent reaction cross-section for k-state (where k represents the isomeric or ground state) and $\emptyset$(E) is the shape of the bremsstrahlung spectrum, $E_{\gamma,max}$ and E_{th} are the maximum bremsstrahlung energy and the reaction threshold, respectively.

In activation process, considering the production of nuclide of isomeric state and ground state at same time of irradiation and internal transfer, we can derive the isomeric ratio IR from the measured gamma activities as follows:

$$IR \equiv \frac{Y_m}{Y_g} = \left[\left(\frac{S_g}{S_m}\times\frac{\varepsilon_m I_{\gamma m}}{\varepsilon_g I_{\gamma g}}\times\frac{P\lambda_g}{\lambda_g-\lambda_m}\right)\times\frac{A_m B_m C_m D_m}{A_g B_g C_g D_g}+\frac{P\lambda_m}{\lambda_g-\lambda_m}\right]^{-1}$$

$$A_i = \frac{1-e^{-\lambda_i\tau}}{1-e^{-\lambda_i T}}e^{-\lambda_i(T-\tau)},\ B_i = \frac{1-e^{-\lambda_i t_i}}{\lambda_i}$$
$$C_i = e^{-\lambda_i t_w},\quad D_i = 1-e^{-\lambda_i t_c} \qquad (2)$$

where S_m and S_g are the photopeak areas for the detected γ-rays of the isomeric-state and the ground-state, ε_m and ε_g are the detection efficiencies for the γ-rays of interest, $I_{\gamma m}$ and $I_{\gamma g}$ are the γ-ray intensities, λ_m and λ_g are the decay constants of the two-states, and P is the branching ratio for the decay of isomeric-state to ground-state.

The photo-activation method was used to determine the isomeric yield ratios of the ^{nat}Ce(γ,xn)^{133m,g}Ce and the ^{nat}Ce(γ,xn)^{137m,g}Ce reactions. The produced nuclides in the irradiated foil together with reaction predecessors were identified based on the known spectroscopic data, such as energy and half-lives. The isomeric yield ratios were calculated from the measured activities of the high-spin state and the low-spin state of the produced radioisotope.

The ^{133m,g}Ce isomeric pair were identified based on their characteristic γ-ray energies and half-lives. The simplified level and the decay scheme of the ^{133m,g}Ce is given in Fig. 2. The ground state of ^{133}Ce (high-spin state, $1/2^+$) with a half-life of 97 m was decayed to the 174.1 keV state ($1/2^+$) and to the 97.3keV state ($3/2^+$) of ^{133}La by $EC + \beta^+$ processes with branching ratio 53% and 47%, respectively. The ground state ^{133g}Ce was identified when it was decayed to the 174.1 keV state of ^{133}La by $EC + \beta^+$ processes with a branching ratio of 53%, which was cascading to the 97.3 keV state ($3/2^+$) and again to the ground state ($5/2^+$) of ^{133}La by emitting 76.9- and 97.3-keV γ-ray, respectively. Meanwhile, the unstable isomeric state ^{133m}Ce (low-spin state, ($9/2^-$) with a half-life of 4.9 h decays to the various energy levels of ^{133}La by a both β^+ and/or electron-capture (EC) processes. By considering the γ-ray intensity and energy as well as the possible interferences shown in Fig. 2, the activity of the isomeric-state ^{133m}Ce was determined based on the 97.3 keV γ-line.

The isomeric yield ratio of ^{nat}Ce (γ,xn)^{137m,g}Ce was measured with 55- and 65-MeV bremsstrahlung energies at the first time. The simplified level and decay scheme of the ^{137m,g}Ce is given in Fig. 2, it is seen that the isomeric state (low-spin state 11/2-), with a half life of 34.4 h decays to the unstable ground state ^{137g}Ce (high-spin state, 2/3+) by emitting 254.3 keV through an integral transition with a branching ratio 99.21%. Meanwhile, 0.79% of the isomeric state decays to the various energy levels of stable ^{137}La by a EC + β^+ processes. The unstable ground-state ^{137g}Ce with a half-life of 9.0 h decays to the 447.2 keV energy level ($5/2^+$) of ^{137}La by a $EC + \beta^+$ process with a branching ratio of 1.8%. As we can see from decay scheme in Fig. 2, the 254.3 keV photo-peak for ^{137m}Ce and the 447.2 keV photo-peak for ^{137g}Ce were used for the activity measurement.

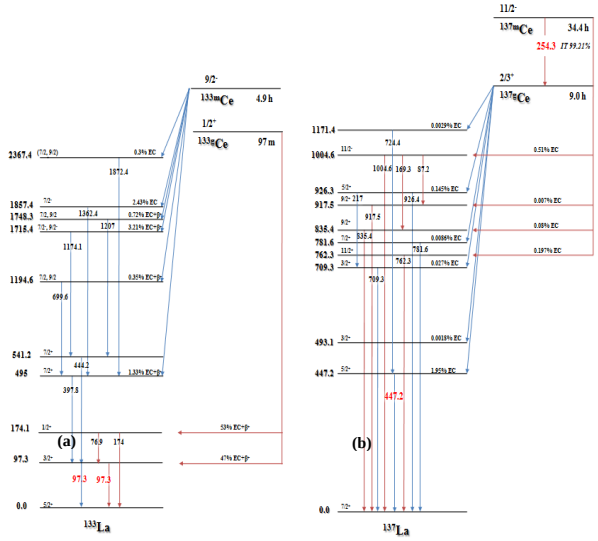

Fig. 2. Simplified representation of formation and decay scheme of isomeric pair; **(a)** ^{133m,g}Ce and **(b)** ^{137m,g}Ce. The nuclear level energies are in keV.

4. Results and discussion

The isomeric yield ratios for the $^{nat}Ce(\gamma,xn)^{133m,g}Ce$ reaction measured at 55-, 60-, and 65-MeV bremsstrahlung energies are 0.32 ± 0.03, 0.34 ± 0.03, and 0.29 ± 0.03, respectively. The uncertainties were calculated from Eq. 2 by using error propagation principle. The main sources of the uncertainties for the present results are due to statistical uncertainty, uncertainties in photo-peak efficiency calibration, nuclear data such as half-life, gamma intensity, IT, photo-peak area determination, coincidence summing, bremsstrahlung flux fluctuation, and others. There is no available data measured 55-, 60-, and 65-MeV bremsstrahlung energies. The present results are given in Table 2.

Natural Cerium consists of four stable isotopes with isotopic abundances as follows: ^{136}Ce (0.185%), ^{138}Ce (0.251%), ^{140}Ce (88.450%), and ^{142}Ce (11.114%). We measured the isomeric yield ratio for the $^{nat}Ce(\gamma,xn)^{137m,g}Ce$ reaction, where x denotes all the possible photonuclear reactions. When the natural Ce foils were irradiated with 55-, 60-, and 65-MeV bremsstrahlung, the $^{137m,g}Ce$ isomeric pairs can be formed $^{138}Ce(\gamma,n)^{137m,g}Ce$, ^{140}Ce $(\gamma,3n)$ $^{137m,g}Ce$, and $^{142}Ce(\gamma,5n)$ $^{137m,g}Ce$. The threshold energies corresponding to these reactions are 9.72-, 26.38-, and 38.98-MeV. The measured isomeric yield ratios of $^{137m,g}Ce$ are $0.21 \pm \mathbf{0.02}$, 0.22 ± 0.02, and 0.26 ± 0.03. The measured isomeric yield ratios and the uncertainties are summarized in Table 2.

Table 2 : Isomeric yield ratios of $^{133mg}Ce$ and ^{137mg} via photonuclear reactions with ^{nat}Ce

Nuclear reaction		Threshold energy (MeV)	Photon Energy (MeV)	IR($Y_{high\text{-}spin}$ / $Y_{low\text{-}spin}$)
	$^{136}Ce(\gamma,3n)^{133m,g}Ce$	28.30	55	0.32±0.03
$^{nat}Ce(\gamma,xn)^{133m,g}Ce$	$^{138}Ce(\gamma,5n)^{133m,g}Ce$	45.52	60	0.34±0.03
			65	0.29±0.03
	$^{138}Ce(\gamma,n)^{137m,g}Ce$	9.72	55	0.21±0.02
$^{nat}Ce(\gamma,xn)^{137m,g}Ce$	$^{140}Ce(\gamma,3n)^{137m,g}Ce$	26.38	60	0.22±0.02
	$^{142}Ce(\gamma,5n)^{137m,g}Ce$	38.98	65	0.26±0.03

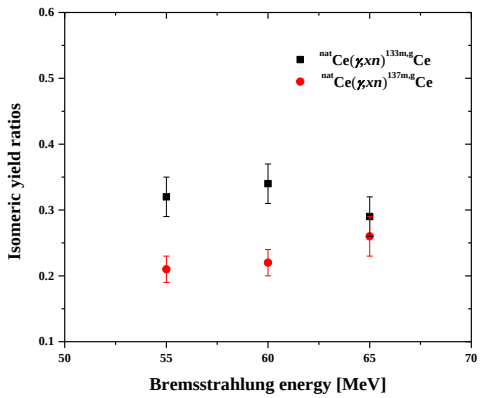

Fig. 3. Dependence of isomeric yield ratios of $^{133mg}Ce$ and $^{137mg}Ce$ on the incident bremsstrahlung energy via (γ,xn) re-actions.

5. Conclusion and future improvements

The isomeric yield retios for the $^{nat}Ce(\gamma,xn)^{133m,g}Ce$ and the $^{nat}Ce(\gamma,xn)^{137m,g}Ce$ reactions were measured with 55-, 60-, and 70-MeV end point bremsstrahlung energies by using the activation and the off-line γ-ray spectrometric technique in the electron linac Pohang Accelerator Laboratory. The reaction $^{nat}Ce(\gamma,xn)^{133m,g}Ce$ was studied for the first time which has no comparable literature data. Studies of $^{137m,g}Ce$ isomeric pair in the literature is based on $^{139}La(d,4n)^{137}Ce$ reactions from La_2O_3 powder in the low energy region [7]. The present experiment is based on (γ,n), $(\gamma,3n)$, and $(\gamma,5n)$ reactions which is the first time measurement at intermediate energy bremsst-rahlung 55-, 60-, 65-MeV from ^{nat}Ce target. From the reaction studied in present measurement, it is obser-vable that isomeric ratio is dependent on the spin of the target nuclei. The present results are the first measure-ments at these energy points. The detailed results of IRs for $^{133m,g}Ce$ and the $^{137m,g}Ce$ are available and will be given in a future publication.

Acknowledgment

This research was supported by National R&D Program through the Dongnam Inst. of Radiological & Medical Sciences funded by the Ministry of Science, ICT & Future Planning (50495-2014)

REFERENCES

[1] J. R. Huizenga and R. Vandenbosch, Phys. Rev. 120,1305 (1960).
[2] J. L. Need, Phys. Rev. 129, 1302 (1963).
[3] R. Vandenbosch and J. R. Huizenga, Phys. Rev. 120, 1313 (1960).
[4] N. D. Dudey and T. T. Sugihara, Phys. Rev. 139, B896 (1965).
[5] D. Kolev, E. Dobreva, N. Nenov and V. Todorov, Nucl. Instrum. Methods Phys. Res., Sect. A 356, 390 (1995).
[6] G. N. Kim, Y. S. Lee, V. Skoy, V. Kovalchuck, M. H. Cho, I. S. Ko, W. Namkung, D.W. Lee, H. D. Kim, T. I Ro, Y. G. Min, J. Korean Phys. Soc. 38, 14 (2001).
[7] H.Vignau, S. J. Nuclear Phys. 108, 112 (1961)