Chapter 1
Nuclear Transmutation of Long-Lived Nuclides with Laser Compton Scattering: Quantitative Analysis by Theoretical Approach

Shizuka Takai and Kouichi Hagino

Abstract A photo-neutron (γ, n) reaction with laser Compton scattering γ-rays has been suggested to be effective for the nuclear transmutations of fission products. The photo-neutron reaction occurs via a giant dipole resonance, which has a large cross section and whose properties are smooth functions of mass number. The laser Compton scattering can generate effectively and selectively high-energy photons with a desired energy range. In this chapter, we investigate quantitatively the effectiveness of the transmutation with laser Compton scattering based on the Hauser–Feshbach theory using the TALYS code. We carry out simulations for high-decay heating nuclide ^{137}Cs, in which the cross sections for ^{137}Cs (γ, γ), (γ, n), and $(\gamma, 2n)$ reactions, and the total photonuclear reaction cross sections versus incident photon energy, are calculated. The incident photon energy obtained by laser Compton scattering is also optimized. It is shown that the transmutation with medium-energy photon with a flux of more than 10^{18}/s effectively reduces the radioactivity of the target ^{137}Cs.

Keywords ^{137}Cs • Giant dipole resonance • Laser Compton scattering • Photo-neutron reaction • Radioactive wastes • Transmutation

1.1 Introduction

One of the major problems of the nuclear fuel cycle is the disposal of high-level radioactive waste that contains long-lived nuclides such as ^{129}I and high-decay heating nuclides such as ^{137}Cs. After the severe accident at the Fukushima Daiichi

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Nuclear Power Plant, there is also a problem of ^{137}Cs having been concentrated by treatment of contaminated water. Transmuting such nuclides into short-lived or stable nuclides is one possible way to resolve this problem. Neutron capture reactions have been proposed for transmutations of such fission products. However, the neutron capture cross sections differ significantly from nuclide to nuclide, and this transmutation method is not effective for nuclides with small neutron capture cross sections such as ^{137}Cs.

Recently, photo-neutron (γ, n) reactions with laser Compton scattering γ-rays have been suggested as an alternative method for nuclear transmutations [1, 2]. Figure 1.1 shows a schematic illustration of this transmutation. This transmutation uses γ-rays generated by laser photons backscattered off GeV electrons and photo-nuclear reactions via electric giant dipole resonance (GDR) [3], which has a large cross section for most nuclides. The GDR is a collective excitation of a nucleus that decays mainly by the emission of neutrons, and its total cross section is a smooth function of mass number. Therefore, this method is expected to be effective for transmuting fission products regardless of isotopes.

So far, transmutation with laser Compton scattering for some nuclides has been evaluated only in a simple manner. In this chapter, we investigate more quantitatively the effectiveness of the transmutation with laser Compton scattering, especially for ^{137}Cs.

1.2 Calculation Method

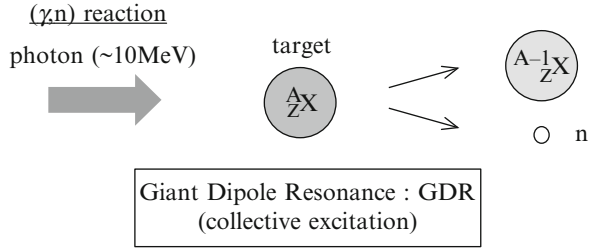### 1.2.1 Reaction via Giant Dipole Resonance

Nuclear transmutation with laser Compton scattering uses photonuclear reactions via GDR because the cross section of GDR is quite large and the total cross section is a smooth function of mass number. GDR is a collective excitation of a nucleus involving almost all nucleons, which is interpreted classically as a macroscopic oscillation of a bulk of protons against that of neutrons. The total cross section $\sigma_{\text{GDR}}^{\text{tot}}$, the resonance energy E_R, and the width Γ_R are given by [4]

$$
\begin{aligned}
\sigma_{\text{GDR}}^{\text{tot}} &= 60(1+\kappa)\frac{NZ}{A} \text{ mb} \cdot \text{MeV}, \\
E_R &= 31.2\,A^{-1/3} + 20.6\,A^{-1/6}\,\text{MeV}, \\
\Gamma_R &= 0.0026 E_R^{1.91}\,\text{MeV},
\end{aligned}
\tag{1.1}
$$

where N and Z are the neutron and proton numbers, $A = N + Z$ is the mass number, and κ, which is roughly equal to 0.2 for medium nuclides, is a correction coefficient for the pion exchange.

When a target nucleus is irradiated with a photon and excited to GDR, it often forms a compound nucleus with only a small contribution of a pre-equilibrium reaction [5]. The compound nucleus is an excited state in which the energy brought

Fig. 1.1 Schematic illustration of nuclear transmutation with laser Compton scattering

(γ,n) reaction

photon (~10MeV)   target $^{A-1}_{Z}$X

$^{A}_{Z}$X   ○ n

Giant Dipole Resonance : GDR
(collective excitation)

by the incident particle is shared among all degrees of freedom of the nucleus. The reaction cross section from an initial channel α to a final channel β proceeding through a compound nucleus state of spin J can be written by the Hauser–Feshbach formula as

$$\sigma^{cn}_{\alpha\beta} = \frac{\pi}{k^2_\alpha} \sum_J g_J \frac{T_\alpha \langle T_\beta \rangle}{\sum_\gamma \langle T_\gamma \rangle}, \qquad (1.2)$$

where k_α is the wave number in the initial channel, g_J is a statistical factor, T is a transmission coefficient, and $\langle T \rangle$ is the energy average of T. The statistical factor is

$$g_J = \frac{2J + 1}{(2i_\alpha + 1)(2I_\alpha + 1)}, \qquad (1.3)$$

where i_α and I_α are the projectile and target spins.

Calculations of reaction cross sections are performed using the nuclear model code TALYS (version 1.4) [6]. The neutron transmission coefficients are calculated via the global optical potential [7]. The gamma-ray transmission coefficients are calculated through the energy-dependent gamma-ray strength function according to Brink [8] and Axel [9]. We employed the level density given by Gilbert and Cameron [10].

Figure 1.2 shows the photonuclear reaction cross sections of ^{137}Cs calculated using the TALYS code. In the incident photon energy $B(n) \leq E_\gamma < B(2n)$, where B (n) and $B(2n)$ are the one- and two-neutron binding energies, respectively, we can see that the (γ, n) reaction mainly occurs. Because the resonance energy of GDR E_R is 15–18 MeV, which is roughly equal to $B(2n)$ for medium nuclides, about half the reactions via GDR are (γ, n) reactions, which occur at $B(n) \leq E_\gamma < B(2n)$.

1.2.2 High-Energy Photons Obtained by Laser Compton Scattering

Laser Compton scattering is a method to obtain high-energy photons by laser photons backscattered off energetic GeV electrons. In the case of head-on collision
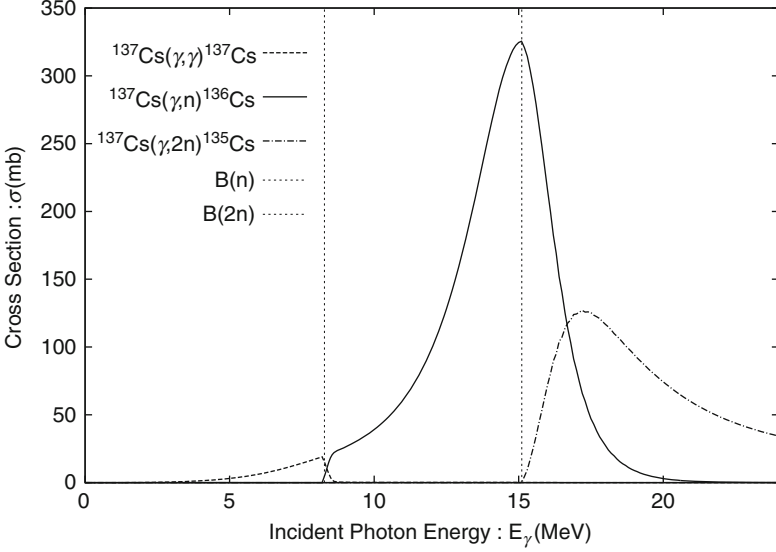
Fig. 1.2 Cross sections for ^{137}Cs (γ, γ), ^{137}Cs (*dashed line*), ^{137}Cs (γ, n), ^{136}Cs (*solid line*), and ^{137}Cs $(\gamma, 2n)$. ^{135}Cs (*dash-dot line*) reactions versus incident photon energy: *dotted lines* represent $B(n)$ and $B(2n)$ of ^{137}Cs

between relativistic electrons and laser photons, the energy of scattered photons is given by

$$E_\gamma = \frac{4\gamma^2 E_L}{1 + (\gamma\theta)^2 + 4E_L\gamma/m_e}, \tag{1.4}$$

where $\gamma = E_e/m_e$ is the Lorentz factor of the electron beam with energy E_e, m_e is the rest mass of the electron, E_L is the energy of the laser photon, and θ is the scattering angle. From Eq. (1.4), the energy of the scattered photon is maximum at $\theta = 0$, and it depends on the energy of incident electrons and photons. The minimum energy of the scattered photon can be fixed by controlling θ with collimators.

The scattering cross section of laser Compton scattering is given by the Klein–Nishina formula:

$$\frac{d\sigma}{dE_\gamma} = \frac{\pi r_0^2}{2} m_e^2 E_L E_e^2 \left\{ \frac{m_e^4}{4E_L^2 E_e^2} \left(\frac{E_e}{E_\gamma - E_e} \right)^2 - \frac{m_e^2}{4E_L E_e} \left(\frac{E_e}{E_\gamma - E_e} \right) + \frac{E_e}{E_\gamma - E_e} + \frac{E_\gamma - E_e}{E_e} \right\},$$

$$r_0 = e^2/4\pi m_e,$$

$$N_\gamma = \int dE_\gamma \frac{dN_\gamma}{dE_\gamma} = \int dE_\gamma \frac{d\sigma}{dE_\gamma} \cdot \text{const.}$$
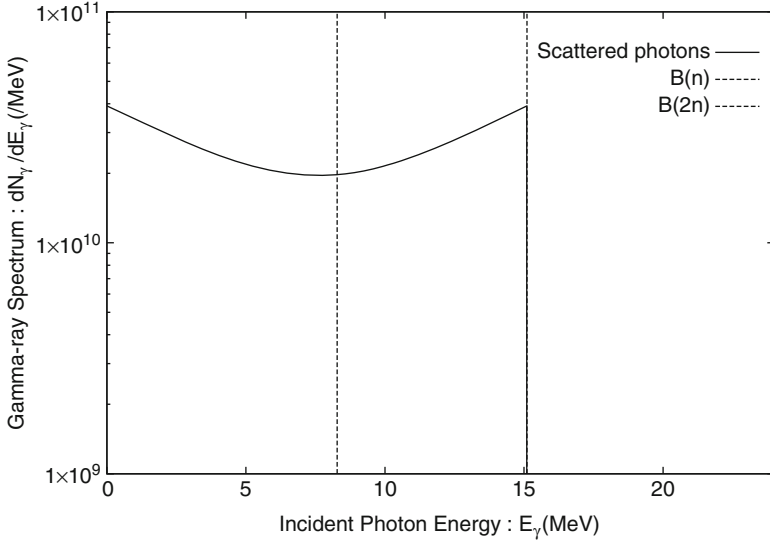
$$\tag{1.5}$$

Fig. 1.3 Calculated gamma (γ)-ray spectrum (*solid line*) generated by laser Compton scattering. The maximum energy, 15 MeV, was chosen to be equal to the binding energy $B(2n)$ of ^{137}Cs. The binding energies of $B(n)$ and $B(2n)$ for ^{137}Cs are indicated by *dashed lines*

To have a situation in which (γ, n) reactions occur, the photon beam with energy at $B(n) \leq E_\gamma < B(2n)$ is desired. In case of the free electron laser, we may assume/expect to get the total photon flux $N_\gamma \approx 2 \times 10^{12}$/s/500 mA for $E_e = 1.2$ GeV [1]. From Eq. (1.4), with $E_e = 1.2$ GeV and $E_L = 0.7$ eV, we obtain the maximum photon energy of $E_\gamma = 15$ MeV at $\theta = 0$, which is equal to $B(2n)$ for ^{137}Cs. Figure 1.3 shows the calculated γ-ray spectrum generated by laser Compton scattering using Eq. (1.5), where the total photon flux with energy from 0 to $B(2n)$ is $N_\gamma \approx 2 \times 10^{12}$/s.

From Fig. 1.3, we can see that about half the total scattered photons are in $B(n) \leq E_\gamma < B(2n)$ and contribute to generate the (γ, n) reactions for ^{137}Cs. In contrast, for the Bremsstrahlung that is usually used to generate high-energy photons, the photon intensity decreases rapidly as the photon energy increases, and only a small part of the high-energy tail is available for (γ, n) reactions [11].

1.2.3 Setup of the Calculation for ^{137}Cs

When a target nucleus X is irradiated with a photon beam with energy E_γ, it forms a compound nucleus, which releases one neutron and becomes its isotope X′. The reaction rate of X(γ, n)X′ at time t is given by

$$r_{X \to X'}(t) = \int\limits_{B(n)}^{B(2n)} dE_\gamma \frac{dN_\gamma}{dE_\gamma} \sigma_{X \to X'}(E_\gamma) n_X(t) a, \qquad (1.6)$$

where $\sigma_{X \to X'}(E_\gamma)$ is the reaction cross section of $X(\gamma, n)X'$, $n_X(t)$ is the number of target nucleus per unit area at time t, and a is the attenuation factor of incident photons through a thick target. dN_γ/dE_γ is expressed with Eq. (1.5) and $\sigma_{X \to X'}(E_\gamma)$ is calculated from Eq. (1.2) using the TALYS code.

Figure 1.4 shows a calculation in which the photon beam is generated by the laser Compton scattering of 1.2 GeV electrons and 0.7 eV laser beams. We assume that the cylindrical target of ^{137}Cs of 1 g is irradiated with a photon beam with energy $B(n) \leq E_\gamma < B(2n)$ within a radius $r \approx 0.8$ mm at 2 m from the interaction point. When a target of ^{137}Cs is irradiated with photons and is excited to GDR, the $(\gamma, \gamma), (\gamma, n)$ and $(\gamma, 2n)$ reactions mainly occur. We consider ^{137}Cs, ^{136}Cs, ^{135}Cs, and ^{134}Cs as the isotopes generated by the transmutation. The numbers of these isotopes are expressed as

$$n_{137}(t + \Delta t) = n_{137}(t)e^{-\lambda_{137}\Delta t} - r_{137 \to 136}(t) - r_{137 \to 135}(t), \qquad (1.7)$$

$$n_{136}(t + \Delta t) = n_{136}(t)e^{-\lambda_{136}\Delta t} + r_{137 \to 136}(t) - r_{136 \to 135}(t) - r_{136 \to 134}(t), \qquad (1.8)$$

$$n_{135}(t + \Delta t) = n_{135}(t)e^{-\lambda_{135}\Delta t} + r_{137 \to 135}(t) + r_{136 \to 135}(t) - r_{135 \to 134}(t), \qquad (1.9)$$

$$n_{134}(t + \Delta t) = n_{134}(t)e^{-\lambda_{134}\Delta t} + r_{136 \to 134}(t) + r_{135 \to 134}(t). \qquad (1.10)$$

One can calculate the number of each isotopes by solving these equations with the Runge–Kutta method.

1.3   Results and Discussion

1.3.1   Nuclear Transmutation of ^{137}Cs with Laser Compton Scattering

Figure 1.5 shows the dependence of the reduction of 1 g ^{137}Cs on the photon flux $N_\gamma = 10^{12}, 10^{18}, 10^{19}, 10^{20}$/s, which is calculated with this setup (Fig. 1.4). The number of ^{137}Cs is effectively reduced with photon flux over 10^{18}/s, that is, the number of ^{137}Cs is reduced by 10 % for 24 h irradiation. Figure 1.6 shows the number of Cs isotopes when 1 g ^{137}Cs is irradiated with photon flux 2×10^{12}/s with the same setup. From this figure, we can see that the reduction rate of ^{137}Cs by the transmutation, which is nearly equal to the generation rate of ^{136}Cs, is two orders of magnitude smaller than the natural decay rate of 1 g ^{137}Cs. Thus, the transmutation of ^{137}Cs is not effective with photon flux 2×10^{12}/s, which is maximum with present accelerator systems.

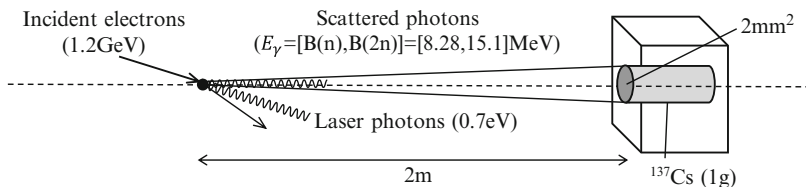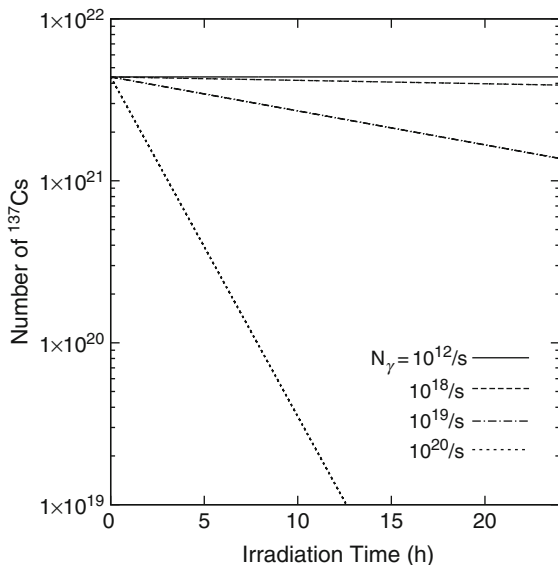**Fig. 1.4** Setup for calculation of transmutation of ^{137}Cs

Fig. 1.5 Dependence of the reduction of 1 g ^{137}Cs on photon flux $N_\gamma = 10^{12}, 10^{18}, 10^{19}, 10^{20}$/s

1.3.2 Comparison with Other Nuclides

Table 1.1 summarizes the generation rate of $^{A-1}$X from of 1 g of a fission product ^{A}X with photon flux 2×10^{12}/s also for ^{129}I, ^{135}Cs, and ^{90}Sr in addition to ^{137}Cs. The reduction rate, which is nearly equal to the generation rate of $^{A-1}$X, does not depend significantly on nuclides because the properties of GDR are smooth functions of the mass number. From this table, we can see that the reduction rate for the transmutation of ^{137}Cs can be similar to that of other medium nuclides.

Fig. 1.6 Number of Cs
isotopes when 1 g ^{137}Cs is
irradiated with photon flux
2×10^{12}/s. *Dotted line*
shows the number of ^{137}Ba
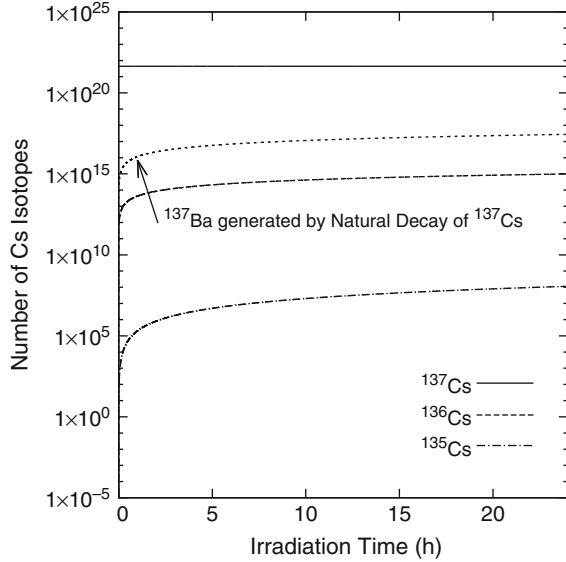that are generated by the
natural decay of 1 g ^{137}Cs

Table 1.1 Generation rate of $^{A-1}$X from 1 g of fission product ^{A}X with photon flux 2×10^{12}/s

Target (^{A}X)	[B (n), B (2n)] (MeV)	E_{GDR} (MeV)	$\sigma_{\text{GDR}}^{\text{tot}}$ (b·MeV)	N ($^{A-1}$X) (/s)
^{129}I	[8.83, 15.7]	15.3	2.25	1.24×10^{10}
^{135}Cs	[8.76, 15.7]	15.2	2.31	1.31×10^{10}
^{137}Cs	[8.28, 15.1]	15.1	2.37	1.19×10^{10}
^{90}Sr	[7.81, 14.2]	16.7	1.58	4.71×10^{9}

1.4   Conclusion

In this work, the effectiveness of transmutation with laser Compton scattering for
reducing fission products was quantitatively investigated. The transmutation of
^{137}Cs is effective with photon flux greater than 10^{18}/s, which results in 10 %
reduction for 24 h irradiation. However, transmutation with photon flux 2×10^{12}/s,
which is achievable with present maximum accelerator systems, is not effective,
and the reduction rate is approximately two orders of magnitude less than the
natural decay rate.

Nuclear transmutation with laser Compton scattering can transmute selectively a
medium mass nuclide ^{A}X into $^{A-1}$X, and its reduction rate is independent of
isotopes. Because the transmutation with laser Compton scattering can almost
exclusively generate desired nuclides, this method will be useful for the generation
of isotopes for medicine [1].

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