Photo-transmutation of ^{100}Mo to ^{99}Mo with Laser-Compton Scattering Gamma-ray

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1. Introduction

Technetium-99m (^{99m}Tc) is an important medical radioisotope which is largely used in hospitals for diagnosing diseased organs. It is produced in the form of ^{99}Mo which decays to ^{99m}Tc with a half-life of 66 hours. There are about 40 million nuclear-medicine procedures performed worldwide per year and about 80-85% of all the procedures use Mo-99 [1].

Cyclotron proton accelerators are used worldwide to produce many short-living medical isotopes. However, few are capable of producing Mo-99 and none are suitable for producing more than a small fraction of the required amounts. More than 90% of the world's demand of ^{99}Mo is sourced from five nuclear reactors. Two of these reactors have already been decommissioned and the rest are more than 45 years old [2]. Relatively short half-life of the parent ^{99}Mo requires continuous re-supply to meet the requirements of medical industry. Therefore, there is an urgent need to produce the ^{99}Mo and ^{99m}Tc isotopes by alternative ways. One such alternative is giant dipole resonance (GDR) based photonuclear transmutation of ^{100}Mo to ^{99}Mo.

This paper presents a photonuclear transmutation method using laser Compton scattering (LCS) gamma-ray beam. Potential production rate (reaction rate) of ^{99}Mo using the photonuclear (γ,n) reaction is evaluated. Rigorous optimization of the LCS spectrum has also been performed to maximize production of the ^{99}Mo.

2. Laser Compton Scattering Photon

The laser Compton scattering is an elastic scattering of a low energy laser photon with a high energy electron, which results in an increase in energy (smaller wavelength) of the photon, as roughly depicted in Figure 1 [3,4].

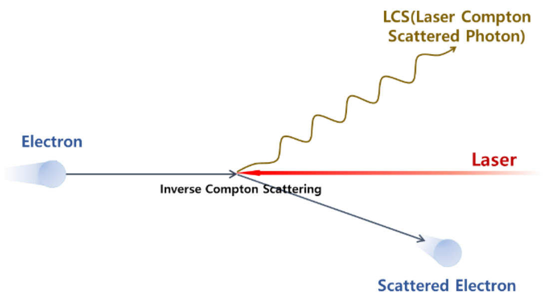

Fig. 1. The laser Compton scattering process

LCS photons can be used for photo-transmutation since they are energy-tunable and can be quasi-monochromatic. Intensity of the quasi-monochromatic LCS gamma-rays can be very high and it is strongly dependent on the electron current and the laser power.

3. The GDR Cross Section

^{99}Mo is primarily produced by the ^{100}Mo (γ,n) ^{99}Mo reaction over the GDR energies of 10-25 MeV. For the ^{99}Mo production from ^{100}Mo, it is assumed that the target is highly enriched (over 99.7%). In this work, 100 % ^{100}Mo target is assumed.

^{99}Mo production using (γ,n) largely depends on the GDR cross-section that follows the Lorentzian distribution as given below:

$$\sigma_{reac} = \sigma_{max}\left[4\left(\frac{E_{max}-E_{\gamma}}{\Gamma}\right)^2 + 1\right]^{-1}, \qquad (1)$$

where 'σ_{max}' is the maximum cross-section which appears at the maximum energy 'E_{max}' and 'Γ' is full width half maximum (FWHM) of energy range. Experimental data and thickness of ^{100}Mo target are summarized in Table 1, where 'd' is target thickness and E_{th} means the threshold energy for the GDR reaction.

Table 1. GDR cross-section data for ^{100}Mo

Nuclide	d (cm)	E_{th} (MeV)	Γ (MeV)	E_{max} (MeV)	σ_{max} (mb)
^{100}Mo	1	8.29	4	14.29	163.4

4. LCS Photon Spectrum Optimization

In order to produce ^{99m}Tc from ^{99}Mo, the effective photonuclear transmutation of ^{100}Mo to ^{99}Mo needs to be maximized by (γ,n) reaction. The aforementioned effective photonuclear transmutation does not only depend on the intensity of gamma-rays within the cone angle but also on further optimization of these gamma-rays spectrum. The following sections present rigorous optimization of the LCS gamma-ray spectrum. The section 4.1 shows the optimization using a fixed gamma-ray intensity values (i.e. 10^{10} and 10^{11} photons/sec) and the optimization of facility related parameters to maximize production rates is performed in section 4.2.

4.1 Spectrum Optimization using fixed intensity values

Figure 2 depicts LCS gamma-ray spectrum for the cone angle of 1.5 mrad. Incident laser energy in this case is 3.32 eV and electron energy is 550 MeV. GDR cross-section of ^{100}Mo is also shown in the figure. It is clear that that the maximum gamma-ray energy in this case is 15 MeV (energy where peak cross-section of GDR appears) and minimum energy is 4.327 MeV. On the other hand, the LCS spectrum is clearly not yet optimized. The LCS spectrum can be optimized in such a way that:

1. Spectrum overlapped with GDR region should be maximized.
2. Spectrum must have more number of photons where the GDR cross-section is maximum.

These conditions can possibly be satisfied in a number of different ways. However, in this section, focus is only on the laser incident energy-based optimization, which is based on various combinations of the optimized spectrum as shown in Figure 3. Further results of transmutation reaction rates are summarized in the Table 2. Incident laser energy for the optimized spectrum is set at 3.78 MeV.

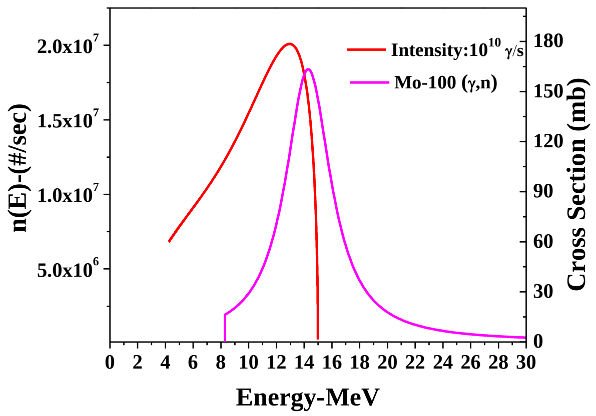

Figure 2. LCS spectrum and ^{100}Mo GDR cross-section

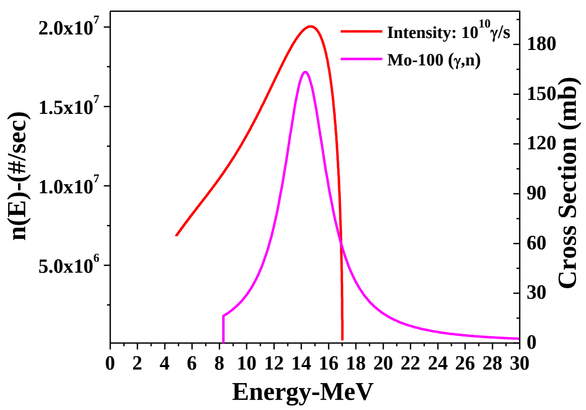

Figure 3. Optimized LCS spectrum and ^{100}Mo GDR cross-section.

Table 2 shows that the incident laser energy of 3.78 eV can yield the maximum reaction rate. Further increase in laser energy can increase the maximum energy of the gamma rays emitted but cannot increase the reaction rate, as the spectrum hardens and shifts towards the right of GDR cross-section peak where the cross-section is lower.

Table 2. Transmutation reaction rate calculations.

Incident Laser Energy (eV)	**Max. Gamma Energy (MeV)**	**Min. Gamma Energy (MeV)**	**Reaction Rate for 10^{11}(γ/s) intensity (#/sec)**
3.16	14.29	4.03	3.1×10^{8}
3.32	15.00	4.33	3.7×10^{8}
3.55	16.00	4.52	4.1×10^{8}
3.78	17.00	4.81	4.3×10^{8}
4.02	18.08	5.12	3.9×10^{8}

Activity according to the aforementioned reaction rate can be calculated by equation (2).

$$A = N_{reac}(1-e^{-\ln 2/T_{1/2}\times t}), \qquad (2)$$

where 'N_{reac}' is the number of reactions, '$T_{1/2}$' is the half-life of the product isotope and 't' is irradiation time.

Table 3. ^{99}Mo activity for high intensity of 10^{15}/sec.

Irradiation time	24 hr (Bq)	168 hr (Bq)	330 hr (Bq)
10^{11}/sec	9.58×10^{7}	3.56×10^{8}	4.17×10^{8}
10^{15}/sec	9.58×10^{11}	3.56×10^{12}	4.17×10^{12}

Table 3 shows the activity about the indicated irradiation time. First row shows the activity when the intensity is 10^{11}/sec, while the second row lists results at 10^{15}/sec, which is the intensity used by R. Hajima et al. to be produced by Compton back-scattering γ-beam using an energy recovery LINAC and super conducting "cold" cavities [4].

After five half-lives of the product isotopes, activity reaches the secular equilibrium state. For ^{99}Mo, the time needed to be in secular equilibrium state is 13.75 days (330 hrs). After 13.75 days irradiation, the activity of ^{99}Mo reaches to 4 TBq. For the 1 week production with the 10^{15}/sec gamma-ray intensity, the produced activity is 3.5 TBq which is 85% of the activity in the secular equilibrium state. Since 80 kCi (3000 TBq) of ^{99}Mo has to be produced around the world in a week [5], it is expected that ^{99}Mo production using the LCS beam can be a local source in a big city or region.

Table 4. Activity of ^{99}Mo vs. irradiation time (10^{15} #/sec)

Irradiation time	1 hr	6 hr	24 hr	168 hr	330 hr
Activity (TBq)	0.045	0.263	0.958	3.563	4.166
Weekly activity (TBq)	7.5	7.4	6.7	3.6	--

It is important to note that total weekly production can be significantly increased by reducing the irradiation time. For example, the total weekly activity of Mo-99 can be as high as 7 TBq if the irradiation time is less than 6 hours, which is almost twice higher than that of the continuous week-long irradiation.

4.2 Spectrum Optimization using existing facility

In section 4.1, we calculate the reaction rate between LCS gamma-ray and the target ^{100}Mo and estimate the activity of ^{99}Mo. We maximize the reaction rate by placing the LCS photon spectrum at the GDR cross-section of ^{100}Mo. Authors adjust the laser energy to make the spectrum have more number of photons where the GDR reaction rate is maximum. However, calculating the transmutation reaction rate with fixed intensities is not quite realistic. It is because the intensity of LCS gamma-ray beam is also the function of various parameters, especially laser energy and electron energy. Therefore authors calculate the transmutation reaction rate using the existing facilities in this section.

There are many operating LCS facilities around the world with high gamma-ray intensities. Among them, a three-loop Energy Recovery LINAC (ERL) proposed by Hayakawa et al. is introduced for the optimizing calculation. Some of important basic design parameters for the ERL facility are given in Table 5. The laser power, incident laser energy, and electron current are kept fixed to the original parameters of the Compton scattering facility. Only electron energy is used for the explicit optimization of the LCS spectrum.

Table 5. ERL facility design parameters. [6]

Design Parameter	**Value**
Electron energy	350 MeV
Laser wavelength	1064 nm
Electron beam current	100 mA
Average laser power	~100 W
Electron bunch charge	1 nC
Pulse energy	1.80 μJ
Amplification factor (Laser super cavity)	3000

The Designed electron energy is 350 MeV. However, the designed maximum LCS gamma-ray energy is 2.2 MeV which is not even close to the GDR cross-section. Authors increase the electron energy from 350 MeV to 970 MeV in this work. The corresponding intensity is 2.6×10^{13} γ/s. By using this intensity of LCS gamma-ray, authors optimize the LCS photon spectrum as shown in Figure 4. The transmutation reaction rate is 6.08×10^{10} /sec. The corresponding activity for the secular equilibrium state is 58.9 GBq.

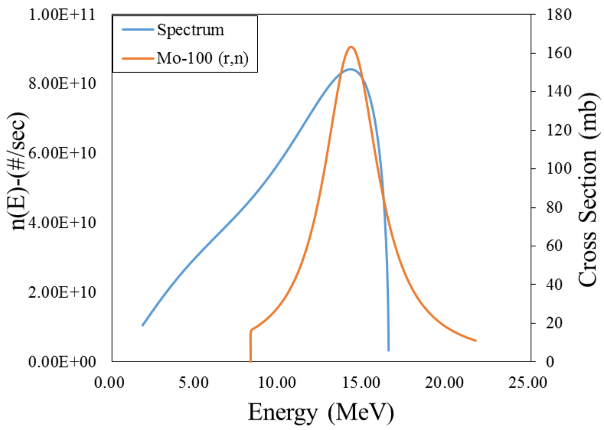

Figure 4. LCS spectrum and ^{100}Mo GDR cross-section for futuristic ERL in Japan.

Until now we perform the optimization by only changing laser energy or electron energy. Authors now compare the spectrums and study which way between increasing the electron energy or the laser energy would give us better transmutation reaction rate. For the ERL facility, we use the own laser energy of facility and change the electron energy for the optimization. This time we set the laser energy as 2.7 eV whose value means the increase in laser energy. The electron energy is then automatically chosen as 640 MeV, which is in contrast to decrease in electron energy. The maximum energy of LCS gamma-ray energy is then 16.5 MeV with 2.7 eV laser energy and 640 MeV electron energy. Comparison of spectrum is depicted in Figure 5. As we increase the laser energy, spectrum become compact, increasing minimum energy of LCS gamma-ray. However, since we decrease the electron energy, intensity of LCS gamma-ray decrease, resulting in decrease of transmutation reaction rate. Therefore, increasing the electron energy rather than laser energy for the sake of same maximum energy of LCS gamma-ray is better option for higher transmutation rate.

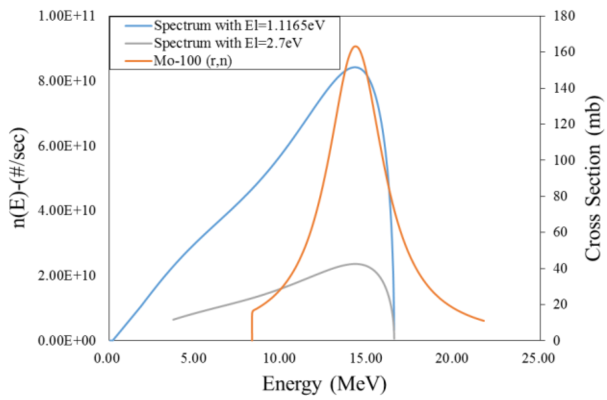

Figure 5. LCS spectrums for futuristic ERL facility with 1.165 and 2.7 eV laser energy.

Another conceptual way for higher gamma-ray intensity is to extract multiple LCS beams from a single electron accelerator source. Then the total gamma-ray intensities can be added up to give one order of magnitude higher flux [7]. It can be realized in a way that a single electron beam coming from the LINAC source is passed through multiple laser cavities and the resulting gamma-ray flux are targeted towards the target radionuclide.

5. Conclusions

For ^{99}Mo production with the LCS photons using GDR-based (γ,n) reaction, the gamma-ray energy should be around 15 MeV. This study indicates that optimization of LCS spectrum by varying the electron and laser energies within practical limits can enhance the transmutation of Mo-100 to M-99 quite significantly. It has been found that irradiation time should be rather short, e.g., less than 6 hours, to maximize the weekly production of Mo-99 in the GDR-based Mo-99 production facility using the LCS photons. The analysis shows that production of ^{99}Mo using a high-performance LCS facility offers a potentially-promising alternative for the production of ^{99m}Tc.

For future work, more detailed evaluation of the photonuclear transmutation efficiency considering interaction of gamma-rays with the target material other than GDR-based (γ,n) reaction will also be evaluated.

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