A Physics Study on Photoproduction of Tc-99m Using the NRF Phenomenon

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1. Introduction

Technetium-99m or Tc-99m, a metastable isomer of Tc-99, is most commonly used medical radioisotope. Tc-99m is used in over 40 million nuclear-medicine procedures performed throughout the world every year [1]. Its short half-life of ~6 hours linked with an easily detectable but relatively safe 143 keV gamma decay has made it an ideal radioisotope for medical applications. When coupled with suitable chemical compounds, it allows diagnosis and examination of specific physiological processes, making it essential for non-invasive medical procedures world-wide.

For Tc-99m, Mo-99 is first produced in nuclear reactors as a byproduct of fission reaction of U-235. Note that Mo-99 decays to Tc-99m with a half-life of ~66 hours. It is estimated that 30~40% of the produced Mo-99 is not utilized in hospitals and the unused Mo-99 nuclei all decay to Tc-99, which should be disposed of with a special care. Meanwhile, the ground state ^{99}Tc is produced in nuclear reactors and constitutes ~0.1% of total spent fuel inventory. It is a problematic radioisotope in terms of its disposal due to its long half-life (211,100 years) and high mobility in ground water.

In this study, authors investigate a novel way to photo-produce useful ^{99m}Tc by recycling hazardous ^{99}Tc. Feasibility of ^{99}Tc(γ,γ)^{99m}Tc reaction is investigated in view of the nuclear resonance fluorescence (NRF) reaction.

2. Methods and Results

2.1 Photoexcitation with NRF

The NRF reaction is the result of nuclear absorption and subsequent emission of high-energy photons. Figure 1 illustrates the essential concept of the NRF reaction. In Fig. 1, E_0 and E_γ represent energy of the ground state and the excited state of the nucleus, respectively. The ground state of ^{99}Tc nucleus can be excited to E_γ with a photon whose energy is very close to E_γ-E_0. Then the nucleus will stay in the excited state for very short time (usually ~ femtoseconds) and release a photon to decay to the original state.

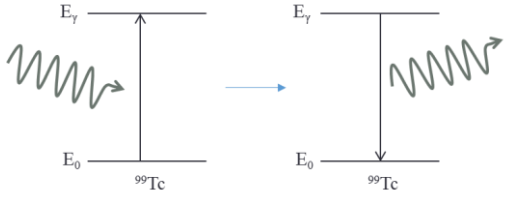

Fig. 1. The NRF reaction.

The NRF cross-section follows the Breit-Wigner formula and is given by Eq. (1):

$$\sigma(E) = \frac{1}{4\pi}\frac{2J+1}{2(J_0+1)}\left(\frac{hc}{E}\right)^2 \frac{\Gamma\Gamma_0}{(E-E_r)^2+\Gamma^2/4}, \quad (1)$$

where J is the spin in the excited state, J_0 is the spin in the ground state, Γ is the total decay width of all decay width at the excited energy level, Γ_0 is the partial decay width from the excited to ground state, E is the incident gamma-ray energy, and E_γ is the energy level of the excited state. In this research, nuclear data for ^{99}Tc is based on ENSDF and the NRF cross-section is calculated by the PHITS code [2, 3].

Figure 2 shows the NRF cross-section and BR_{total} at possible excited energy levels that can result in production of ^{99m}Tc. BR_{total} means the total branching ratio from the excited energy level to ^{99m}Tc. It should be noted that Fig. 2 shows cross-sections and BR_{total} for some of higher states for which the NRF cross-section and BR_{total} data can be evaluated. BR_{total} cannot be calculated if information is missing in ENSDF file about the photon branching ratios for each level deduced from isomer transition decay, even if it has a branching ratio to ^{99m}Tc. Some cross-sections may be erroneous if ENSDF does not have the transition path to ground state for respective excited energies.

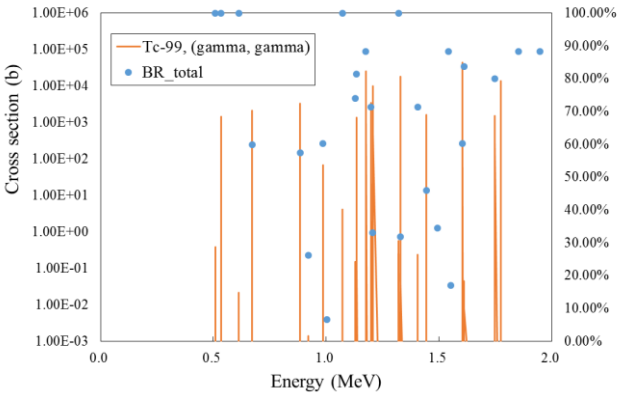

Fig. 2. NRF cross-section and BR_{total} for several higher states.

The NRF cross-section for isomeric state is not depicted in Fig. 2 because it is evaluated to be negligibly small (order of 10^{-13} b). This is why Tc-99m production is explored with higher excited states. It is also very important to note that the NRF resonance is very sharp and the peak cross-section is quite high for many excited states. In the NRF resonances, the FWHM (full with at half maximum) is only about 2~3 eV.

Regarding the NRF cross-section in Fig. 2, it should be mentioned that the uncertainty of current cross-section estimates is considered to be very high at the moment, e.g., the actual cross-section can be between 1/100 of the evaluated value and 100 times the evaluated one [4].

2.2 Laser Compton Scattering Gamma-rays

The photonuclear excitation can be induced by using high-brightness gamma-rays generated from laser Compton scattering interactions. The laser Compton scattering (LCS) phenomenon is an elastic scattering of a low energy laser photon with a high energy electron to increase the energy of the photon (and reducing the wavelength), as roughly depicted in Fig. 3 [5, 6]. LCS gamma-rays can be used for photonuclear excitation since they are energy-tunable, quasi-monochromatic, and beam-like. The intensity of the LCS gamma-rays should be strong for efficient and high nuclear excitation rate.

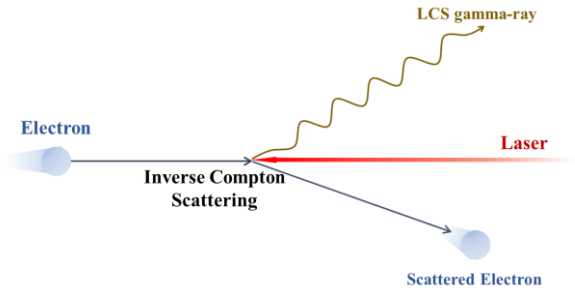

Fig. 3. Laser Compton scattering process.

In this research, an LCS facility with energy-recovery LINAC (ERL) system is considered as the gamma-ray source. T. Hayakawa et al. recently designed a high-flux LCS gamma-ray facility utilizing a 350 MeV ERL system [7], which is shown in Fig. 4. The facility was reported to yield a gamma-ray intensity in order of 10^{13} photons/second.

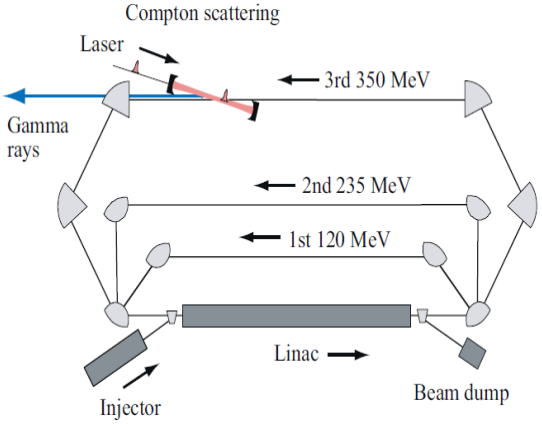

Fig. 4. Schematic of Hayakawa's ERL system.

In the above LCS facility, a 3-loop design is employed for cost reduction and compactness. An electron beam emitted from an injector is accelerated by a superconducting LINAC. After the three recirculation loops, the electron beam is re-injected into the LINAC with a deceleration phase, and the electron energy are fed back into radio-frequency [5] cavity of the superconducting LINAC. LCS gamma-rays are finally generated from the collisions of the electrons with the laser photons at the end of the loop. Important design parameters of the LCS facility are given in Table I.

Table I: Design parameters of Hayakawa's ERL system

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

3 Photoproduction of Tc-99m

The photonuclear reaction rate can be calculated using the NRF cross-section and the LCS spectrum as given by Eq. (2):

$$N_{reac} = n_{target} BR_{total} \int_{E_l}^{E_h} \sigma_{NRF}\, dE_\gamma \frac{dN_\gamma}{dE_\gamma} \int_0^d e^{-\Sigma_{NRF} x} dx, \quad (2)$$

where n_{target} is the number of atoms per cubic centimeter of the target material, d is the thickness of the target material, σ_{NRF} is the NRF-based resonance cross-section, E_l and E_h is the lowest and highest energy of LCS gamma-rays photon spectrum, respectively. In Eq. (2), Σ_{NRF} is the macroscopic NRF cross-section that is the product of σ_{NRF} and n_{target}, and dN_γ/dE_γ is the spectral density calculated using Eq. (3):

$$\frac{dN_\gamma}{dE_\gamma} = \frac{N_\gamma}{\sigma_t} \int_{E_0-\delta_E}^{E_0+\delta_E} \frac{d\sigma}{dE_\gamma} \frac{1}{\sqrt{2\pi}\,\delta_E} exp\left[-\frac{(E_e - E_0)^2}{2\delta_E^2}\right] dE_e, \quad (3)$$

where N_γ is the total gamma-ray intensity of the facility in the units of #/sec, σ_t is the total Compton scattering cross-section in mb , $d\sigma/dE_\gamma$ is the differential Compton scattering cross-section with unit of mb/MeV, E_0 is the central electron beam energy in MeV, and δ_E is the dispersion of the electron beam from its central energy value.

It is worthwhile to note that gamma-ray attenuation in the target region is also considered in Eq. (2). This is because the σ_{NRF} cross-sections are large that they may have a non-negligible impact on the gamma-ray attenuation. It is assumed that the spectral density is constant within the energy range of E_l and E_h. This can be accepted because the energy range is less than 10 eV.

The designed electron energy is 350 MeV in the original ERL facility shown in Fig. 4. However, the electron energy is decreased from 350 MeV to 315 MeV in this study as the required LCS photon energy is reduced. The maximum LCS photon energy is adjusted to 1.77 MeV because the NRF cross-sections above this

energy are quite small and a lot of information is missing. The corresponding intensity of LCS photons is 2.1×10^{13} γ/s. By using this intensity of LCS gamma-ray, the LCS photon spectrum is optimized as shown in Fig. 5. The total photonuclear reaction rate is calculated to be 6.64×10^{10} /sec in this work. It is important to note that only 21 energy states are considered in the current study although higher energy state may additionally contribute to the photo-production of Tc-99m.

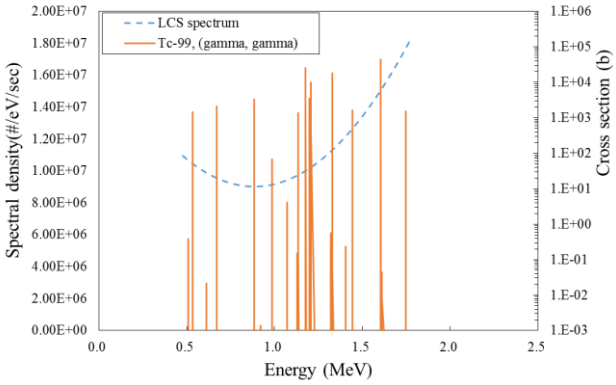

Fig. 5. LCS spectrum and ^{99}Tc NRF cross-section.

Activity according to the aforementioned reaction rates can be calculated using Eq. (4):

$$A = N_{reac}(1 - e^{-\ln 2/T_{1/2} \times t}), \quad (4)$$

where N_{reac} is the reaction rate, $T_{1/2}$ is the half-life of the product isotope, Tc-99m, which is ~6 hours, and t is the irradiation time. Table II displays the Tc-99m activity for several irradiation times. The time needed to achieve a secular equilibrium state is 30 hours for Tc-99m. Table II provides Tc-99m activity produced for 0.5-, 1-, 3-, 6-, 12- and 30-hour irradiation times.

Table II: Activity of ^{99m}Tc as a function of irradiation time

Irradiation time (hr)	0.5	1	3	6	12	30
Activity (mCi)	0.2	0.3	0.9	1.5	2.3	2.9

As shown in Table II, the available Tc-99m activity is rather small since the LCS beam intensity is relatively small. Taking into account futuristic LCS facilities, however, the LCS gamma-ray beam intensity is expected by several orders of magnitude and the resulting activity of Tc-99m can be increased drastically. It should also be considered that the compact LCS facility can be located in big cities near the hospitals and Tc-99m can be produced on demand and its loss can be also minimized due to proximity to the customers.

4. Conclusions

The proposed NRF-based photo-nuclear reaction is anticipated to innovate production of the important medical isotope ^{99m}Tc by recycling long-living radioactive isotope ^{99}Tc from nuclear waste. The results imply some possibility of ^{99}Tc recycling, although the LCS beam intensity should be increased a lot for a practical implementation of the proposed method. More detailed investigations will be performed in the future.

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