

## 使用激光康普顿散射伽马源的医用放射性同位素光核反应截面的可行性研究

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### Abstract

In recent years, the gap between the supply and demand of medical radioisotopes has widened, necessitating the development of novel production methods. Photonuclear reactions based on gamma sources offer unique advantages for producing high-specific-activity and innovative medical radioisotopes. However, the lack of experimental data on reaction cross sections for photonuclear reactions of medical radioisotopes of interest has severely constrained the development and production of photonuclear transmutation medical radioisotopes. In this study, the complete process of generation, decay, and measurement of medical radioisotopes was simulated via a combination of online gamma activation and offline gamma measurements using a shielded gamma-ray spectrometer. Utilizing a quasi-monochromatic gamma beam from the Shanghai Laser Electron Gamma Source (SLEGS), the feasibility of measuring production cross sections for targeted medical isotopes was investigated through simulations, and specific methodologies for measuring medical radioisotopes with low production cross sections were proposed. The viability of this approach for high-precision measurement of reaction cross sections for medical radioisotopes was demonstrated.

### Full Text

#### Preamble

#### Feasibility Study of Photonuclear Reaction Cross Sections for Medical Radioisotopes Using a Laser Compton Scattering Gamma Source

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In recent years, the gap between supply and demand for medical radioisotopes has widened, necessitating novel production methods. Photonuclear reactions based on gamma sources offer unique advantages for producing high-specific-activity and innovative medical radioisotopes. However, the lack of experimental cross-section data for photonuclear reactions involving medically relevant radioisotopes has severely constrained the development and production of these isotopes through photonuclear transmutation. This study simulates the complete process of generation, decay, and measurement of medical radioisotopes using online gamma activation combined with offline gamma measurements and a shielded gamma-ray spectrometer. Based on a quasi-monochromatic gamma beam from the Shanghai Laser Electron Gamma Source (SLEGS), we simulated the feasibility of measuring production cross sections for selected medical isotopes and propose specific solutions for isotopes with low production cross sections. The results demonstrate the viability of this method for high-precision measurements of photonuclear reaction cross sections for medical radioisotopes.

**Keywords:** Medical radioisotope; Photonuclear reaction; GEANT4; Shanghai Laser Electron Gamma Source (SLEGS); low-background gamma-ray spectrometer

## Introduction

The shortage of medical radioisotopes has become increasingly prominent in recent years [?, ?]. Currently, global production is concentrated in a few large research reactors that operate under a worldwide supply strategy, including the National Research Universal Reactor in Canada, the High Flux Reactor in the Netherlands, the High Flux Isotope Reactor in the United States, and the Missouri University Research Reactor. However, reactor-produced medical radioisotopes suffer from low specific activity, and many of these facilities—built in the 1950s and 1960s—are now being decommissioned. Consequently, new methods for producing medical radioisotopes with higher specific activity are urgently needed [?].

Photonuclear reactions have attracted considerable worldwide attention due to their excellent selectivity, relatively simple reaction products, and ability to pro-

duce high-specific-activity radioisotopes through photonuclear transmutation. The Nuclear Science Advisory Committee-Isotope Subcommittee concluded in its 2009 and 2015 reports that accelerator-based radioisotope production methods are essential, and that photonuclear transmutation using electron linear accelerators represents a unique source of radioisotopes [?, ?]. More recently, the committee identified this approach as one of the most compelling and influential opportunities for producing high-specific-activity medical radioisotopes.

Two primary methods exist for photonuclear transmutation. The first uses an electron accelerator to generate bremsstrahlung radiation that drives photonuclear reactions [?]. The second employs next-generation quasi-monochromatic gamma sources to directly drive photonuclear transmutation [?]. Regardless of the approach, the greatest uncertainty stems from the photonuclear reaction cross section. For most known medical radioisotopes that can be produced, as well as for innovative radioisotopes with significant potential applications, the scarcity of cross-section data severely limits research and development efforts, evaluations of photonuclear transmutation efficiency, and selection of optimal target reaction pathways. In electron-accelerator-driven schemes, the photonuclear reaction cross section determines the required maximum electron energy, which affects both the maximum yield of the target isotope and the specific activity of the product. Similarly, for quasi-monochromatic gamma sources, the cross section influences energy selection and product specific activity assessment. Therefore, measuring photonuclear reaction cross sections for medically relevant radioisotopes is of paramount importance.

High-precision cross-section measurements can be performed offline using shielded detection systems, which reduce interference from environmental gamma rays, neutrons, and cosmic rays while improving the signal-to-noise ratio—even when characteristic gamma-ray intensity branching ratios are small. In our simulation, a quasi-monochromatic gamma beam activates the target material, and an offline measurement device detects the characteristic gamma rays from photonuclear reaction products to identify the types and quantities of nuclides generated in the target. From this, the cross sections for  $(\gamma, n)$ ,  $(\gamma, p)$ , and  $(\gamma, \gamma)$  photonuclear reactions are obtained. A low-background gamma-ray spectrometer enables high-precision cross-section measurements. Using the GEANT4 toolkit [10–12], we simulated the entire process of medical isotope production, decay, and measurement based on the quasi-monochromatic gamma beam of the Shanghai Laser Electron Gamma Source (SLEGS) [?, ?]. Finally, we discuss the feasibility for several medically relevant radioisotopes. This study provides guidance for measuring reaction cross sections of medical radioisotopes produced via photonuclear reactions.

## II. Measurement Method

The parameters for candidate medical radioisotopes produced through  $(\gamma, n)$  and  $(\gamma, p)$  reactions are listed in Table 1 [13–15]. Theoretical cross-section data in the relevant energy region were obtained from the TENDL-2021 database.

Most photonuclear reaction cross sections for medically relevant radioisotopes in the giant dipole resonance energy region lack experimental data and require accurate measurement.

Two general methods exist for measuring cross sections of  $(\gamma, n)$ ,  $(\gamma, p)$ , and  $(\gamma, \gamma)$  reactions. The first method measures neutrons, protons, and  $\gamma$  products directly online, while the second performs offline measurements of characteristic gamma rays from target nuclei produced through online activation. However, natural targets typically contain multiple isotopes, making it impossible to determine which isotope produced the detected neutrons, protons, or gamma rays. Even with enriched isotopic targets, achieving 100% purity is difficult, and accurate cross-section measurements using high-purity isotopic targets are expensive. Furthermore, measuring small reaction cross sections is challenging due to background from bremsstrahlung radiation in the gamma beam and natural environmental background.

Compared to direct online measurements, offline measurements completely separate radionuclide activity measurement from the photonuclear excitation process, effectively avoiding interference from low-energy neutrons, X/ $\gamma$  rays of various energies, and positron pair spectra generated during online irradiation [16–18]. Moreover, medical radioisotopes typically have long half-lives and emit gamma rays during decay, making them well-suited for offline decay measurements. However, most medical radioisotopes have relatively small photonuclear reaction cross sections, and the characteristic peaks from photonuclear activation products in irradiated targets are often submerged in environmental background when measured offline. Thus, the detection limit depends critically on background levels.

Low-background gamma-ray spectroscopy using germanium (Ge) detectors has become a fundamental tool for identifying and measuring radionuclide activities, determining radioactive decay emission probabilities, and performing low-level counting [?]. Table 2 presents a survey of reported low-background systems, all achieving integrated backgrounds of  $\leq 0.12$  c/s/100cm<sup>3</sup>Ge [20–25]. With passive material shielding alone, integrated backgrounds can reach levels below 2 c/s under optimal conditions. Therefore, we propose using a low-background gamma-ray spectrometer for offline measurements of medical radioisotopes to reduce background, achieve high-precision cross-section measurements, decrease irradiation time, and conserve valuable beam time.

The target nuclei produced by online irradiation can be identified by their characteristic gamma energies and half-lives. Let  $N_0$  be the number of target nuclei obtained at the end of irradiation, and  $N$  be the integral count of the gamma full-energy peak:

$$N = N_0 I_{\gamma} \epsilon_{\gamma} C_{k\gamma} e^{-\lambda t_a} (1 - e^{-\lambda t_m}),$$

where  $I_{\gamma}$  is the gamma decay intensity from the NNDC database,  $\epsilon_{\gamma}$  is

the HPGe detection efficiency (calibratable using a gamma standard source of known intensity),  $C_{\gamma}$  is the penetration factor for gamma rays from target nuclear decay,  $\mu$  is the mass attenuation coefficient ( $\text{cm}^2/\text{g}$ ),  $x$  is the target mass thickness ( $\text{g}/\text{cm}^2$ ),  $\lambda$  is the decay constant of the target nucleus,  $t_d$  is the time interval between irradiation end and measurement start, and  $t_m$  is the offline measurement duration.

As a general activation measurement [?],  $N_0$  is given by:

$$N_0 = \sigma(E)N_tQC_{k\text{beam}} \int_0^{t_i} \phi(t)e^{-\lambda t} dt,$$

where  $\sigma(E)$  is the energy-dependent reaction cross section,  $N_t$  is the surface density of target nuclei ( $\text{atoms}/\text{cm}^2$ ),  $Q$  is the abundance of the target isotope,  $C_{k\text{beam}}$  is the penetration factor for the incident gamma beam in the target,  $\phi(t)$  is the gamma beam intensity as a function of time, and  $t_i$  is the irradiation duration. If  $\phi(t)$  is approximated as constant, then:

$$N_0 = \sigma(E)\phi N_tQC_{k\text{beam}} \frac{(1 - e^{-\lambda t_i})}{\lambda},$$

where  $\phi$  is the integrated gamma-ray flux. Since  $N_0$  can be determined from  $N$  using Eq. (1), the reaction cross section can be obtained from Eq. (2). The gamma flux from actual SLEGS measurements is stable due to the stable electron beam intensity of the SSRF storage ring and the laser beam used for Compton scattering. A specific form of beam intensity  $\phi(t)$  was required for simulation, and a constant beam-intensity approximation was employed to study the feasibility of cross-section measurements based on realistic beam-intensity measurements. After establishing the photonuclear reaction cross-section measurement method, we constructed a low-background gamma spectrometer using GEANT4 and assessed its feasibility by simulating the entire process of photonuclear reaction production and offline measurement of medical radioisotopes.

### III. Simulation Modeling

GEANT4 is a Monte Carlo simulation toolkit widely used in particle physics, astrophysics, nuclear physics, medical physics, radiation protection, and other fields [?]. It enables accurate numerical simulation of particle-matter interactions, including particle transport, energy deposition, electromagnetic interactions, and nuclear reactions, while supporting extensions for particle types, geometry, material properties, detector types, and operating systems. GEANT4's primary advantages are its high reproducibility and reliability. Based on GEANT4, we developed a program to simulate medical isotope production, decay, and measurement, performing full-scale simulations.

## A. Medical Isotope Production

A Laser Compton Scattering (LCS) light source is a novel X-ray or gamma-ray source based on the interaction between a relativistic electron beam and laser photons. Characterized by high energy, short wavelength, fast pulses, and high peak brightness, it has become an important option for advanced light source technology worldwide [?]. SLEGS produces gamma rays in the energy range of 0.4-20 MeV, covering the nuclear structure to giant resonance region (keV to MeV) in nuclear physics research. The integrated gamma-ray flux across the entire spectrum can reach  $10^5$ - $10^7$  ph/s and is continuously adjustable. Collimation technology can achieve an energy spread better than 5% [?, ?]. Therefore, in our simulation we used a gamma beam with an intensity of  $10^7$  ph/s and energy spread of 5% to activate a Ti target and produce the medical isotope  $^{47}\text{Sc}$  via the  $^{48}\text{Ti}(\gamma, p)^{47}\text{Sc}$  reaction for further analysis.

[Figure 1: see original paper] shows that within the first half-day of irradiation, the yield of  $^{47}\text{Sc}$  increases rapidly, gradually approaching a saturation value after approximately 12 hours. The choice of target thickness relates to the penetration factor of the incident gamma beam ( $C_{\text{kbeam}}$ ). As target thickness increases, photons must travel a longer distance through the target material, decreasing the photon count while multiple scattering inside the target produces a broader photon energy distribution. Additionally, target thickness affects the penetration factor for gamma rays from target nuclear decay ( $C_{\text{k}\gamma}$ ), as these gamma rays attenuate with increasing target thickness, reducing the integral count of the full-energy peak and affecting measurement accuracy.

## B. Low-Background Gamma-Ray Spectrometer

Background radiation significantly impacts measured energy spectra, leading to unreliable analytical results. Background radiation originates primarily from three sources: natural environmental radiation in the laboratory, radioactive background from detector components and shielding materials, and cosmic ray contributions. Low-background gamma spectrometers require shielding against these radiation sources to achieve high-precision cross-section measurements for medical radioisotopes.

Numerous low-background gamma-ray spectrometers have been constructed domestically and internationally. Countries including China, France, the United States, and Japan have established underground laboratories at various depths to facilitate low-background measurement research. Representative international systems include those developed by Miley at the Pacific Naval Laboratory in 1991 [?], Byun at the Korea Atomic Energy Research Institute in 2001 [?], Semkow et al. in the United States in 2002 [?], and Heusser et al. in Germany [?, ?]. Similar devices have been built in Chinese laboratories, including the China Institute of Atomic Energy [?], the Institute of High Energy Physics of the Chinese Academy of Sciences, and the Third Institute of Oceanography of the State Oceanic Administration [?].

Based on existing low-background gamma spectrometers, we designed a shielded system using GEANT4 simulations. The shielding structure is illustrated in [Figure 2: see original paper] [?]. Using a high-purity germanium detector in a low-background gamma spectrometer requires careful consideration of shielding against various backgrounds. Gamma-ray intensity decreases drastically with increasing atomic number because materials with higher atomic numbers possess larger gamma attenuation coefficients than those with lower atomic numbers [?, ?]. Common high-Z materials include lead, oxygen-free copper, and steel; however, iron is easily contaminated with  $^{60}\text{Co}$  during smelting, and copper has a large thermal neutron capture cross section. Considering factors such as cost, atomic number, and mechanical properties, lead is the optimal choice for gamma shielding.

Cosmic rays produce secondary particles such as photons, neutrons, and electrons when undergoing muon capture in high-atomic-number materials like lead [32-36]. Therefore, the second shielding layer addresses neutron shielding, which requires both moderation and absorption. Water, paraffin, and polyethylene are widely used as moderating materials [?], with plastic scintillators or polyethylene serving as a second layer to slow fast neutrons. The third layer absorbs thermal neutrons.  $^{10}\text{B}$  is an excellent thermal neutron absorber, and  $\text{B}_4\text{C}$ —with a B/C ratio of 0.7828—has a very large neutron capture cross section, enabling effective absorption of moderated neutrons. Cadmium also has a large absorption cross section for thermal neutrons. Therefore, borated polyethylene can be used to moderate and absorb neutrons, while cadmium absorbs thermal neutrons [?]. However, cadmium atoms emit 2.3 MeV gamma rays after neutron capture, requiring additional low-background lead shielding to attenuate these gamma rays.

Finally, other low-Z materials such as acrylic glass or copper can be added to the inner lining to shield against impurities like  $^{210}\text{Pb}$ , its daughter nucleus  $^{210}\text{Bi}$  in lead, and lead characteristic X-rays [?]. Through simulations to select appropriate shielding materials and optimize their thicknesses, the final shield structure from outermost to innermost layer was determined to be: 100 mm lead, 50 mm plastic scintillator, 100 mm borated polyethylene, 3 mm cadmium, 50 mm lead, and 2 mm oxygen-free copper, consistent with the low-background gamma spectrometer designed by Hu et al. [?].

[Figure 3: see original paper] compares the simulated HPGe energy deposition spectrum with results from Hu et al., revealing similar trends and comparable count rates for the 511 keV peak (approximately  $8 \times 10^{-3}$  cps). It is important to note that due to our shielding configuration—specifically the incorporation of an inner lead layer—the 478 keV peak is obscured and not evident in [Figure 3: see original paper]. In the low-energy region, the experimental spectrum shows higher count rates than the simulated spectrum due to differences in background and radioactivity from shielding materials and the detector itself, thereby verifying the reliability of our simulations.

### C. Medical Isotope Decay and Measurement

Under the passive shielding conditions shown in [Figure 2: see original paper], the simulated total integrated background count rate in the 100-2700 keV energy range is 0.22 c/s/100cm<sup>3</sup>Ge. Under better conditions using only lead shielding, this rate may reach 1 cps [?, ?, ?]. To reduce simulation complexity, we used lead shielding only in subsequent simulations. Based on the spectrometer constructed in our GEANT4 simulation program, we used the number of produced <sup>47</sup>Sc target nuclei as input for offline decay measurement simulations, assuming a 1-hour offline measurement duration and lead-only shielding.

[Figure 4: see original paper] presents spectra obtained under different conditions: Shanghai electron laser gamma source laboratory environmental background, low-background shielding device, and neglecting environmental background. After background subtraction, the integral count rate of the <sup>47</sup>Sc gamma full-energy peak at  $E_{\gamma} = 159$  keV is 0.16 c/s. When measured against environmental background alone, the <sup>47</sup>Sc full-energy peak is completely submerged. Low-background shielding effectively reduces the detection limit, making the integral count of the characteristic gamma full-energy peak identifiable. Thus, cross sections for medical radioisotopes produced by photonuclear reactions can be efficiently measured by reducing background. Additionally, factors such as irradiation time, measurement time, and beam intensity affect cross-section measurements, which we discuss in the next section through computational simulations.

### IV. Feasibility of Measurement

Our simulation program includes five control parameters: irradiation time, measurement time, time interval between irradiation end and measurement start, target thickness, and beam intensity. Considering real environmental background effects, we added background as a sixth control parameter. Based on SLEGS capabilities, the gamma beam intensity factor can be set to 10<sup>7</sup> ph/s [?].

First, we considered the background factor. According to Eq. (1),  $N$  represents the integral count of the gamma full-energy peak when background is neglected. However, the detector measures the combined integral count from target nucleus decay and background. Therefore:

$$N = C - C_0,$$

where  $C$  and  $C_0$  denote the integral count from target nucleus decay measured by the detector and the background count from the low-background detection system (considering only lead shielding), respectively. For realistic measurements, we set the relative statistical error of counts to 10%. According to error propagation, the relative statistical error is:

$$\frac{\Delta N}{N} = \frac{\sqrt{C + C_0}}{C - C_0}.$$

Next, we considered target thickness. According to Eqs. (1) and (2), target thickness primarily affects the attenuation coefficient and area density. We constructed a function  $f(x)$  that varies with target thickness:

$$f(x) = N_t C_{k\gamma} C_{k\text{beam}} = \frac{N_A \rho x}{M} e^{-(\mu/\rho)\rho x} e^{-(\mu'/\rho)\rho x},$$

where  $N_t = N_A x/M$ ,  $N_A$  is Avogadro's constant,  $M$  is molar mass,  $x$  is target mass thickness,  $\mu$  is the attenuation coefficient for the incident beam during online activation, and  $\mu'$  is the attenuation coefficient for de-excitation gamma rays escaping the target during offline measurement. Using  $^{69}\text{Ga}(\gamma, n)^{68}\text{Ga}$  as an example, the relationship between  $f(x)$  and  $x$  is shown in [Figure 5: see original paper].

According to [Figure 5: see original paper],  $f(x)$  has a maximum value. Rewriting Eqs. (1) and (2) as functions of  $f(x)$  yields:

$$N = \sigma(E) \phi Q I_\gamma \epsilon_\gamma e^{-\lambda t_d} (1 - e^{-\lambda t_m}) (1 - e^{-\lambda t_i}) f(x),$$

where  $f(x)$  is proportional to  $N$ . The maximum integral count  $N$  can be determined from  $f(x)$  when other conditions are fixed. In realistic measurements, target thickness affects the characteristic gamma count rate due to inevitable attenuation of incident gamma rays and self-absorption during de-excitation. Therefore, thickness must be determined based on practical considerations among other factors. For thicknesses from zero to the optimal value,  $f(x)$  increases monotonically with  $x$ . Based on this property, the detection limit can be determined by combining time factors.

The time factors include irradiation time  $t_i$ , measurement time  $t_m$ , and time interval from irradiation end to measurement start  $t_d$ . Here,  $t_d$  was uniformly set to 5 minutes. The relationship between  $t_i$  and  $t_m$  was determined from Eq. (7): when other conditions are fixed, increasing one quantity decreases the other. We define  $A = t_i \times t_m$ . Considering target nucleus decay,  $t_m$  is limited to 900-3600 s, and  $t_i$  is limited to within 24 hours in realistic scenarios.

We considered three limiting factors:  $x$ ,  $t_m$ , and  $t_i$ . When  $x$  has a minimum value of 0.01 cm, offline measurements can determine the cross section for medical radioisotopes generated by photonuclear reactions if simulated values of  $t_m$  and  $t_i$  fall within their respective limits ( $t_m$  between 900-3600 s and  $t_i$  within 24 hours). Smaller  $A$  values indicate easier measurements because less time is required for irradiation and measurement. If  $t_m$  and  $t_i$  exceed their limits when  $x = 0.01$  cm, measurement is not feasible at this thickness.

The detection limit is determined by gradually increasing thickness in 0.01 cm steps, with the difficulty of detection indicated by the values of  $x$  and  $A$ .

[Figure 6: see original paper] shows simulation results for selected medical radioisotopes of interest. When  $x = 0.01$  cm, cross-section measurements are feasible for  $^{87}\text{Sr}$ ,  $^{44}\text{Sc}$ ,  $^{68}\text{Ga}$ ,  $^{186}\text{Re}$ ,  $^{153}\text{Sm}$ ,  $^{89}\text{Zr}$ ,  $^{47}\text{Sc}$ , and  $^{97}\text{Ru}$ . Smaller  $A$  values indicate shorter required irradiation and measurement times. However,  $^{45}\text{Ti}$ ,  $^{103}\text{Pd}$ ,  $^{193}\text{Pt}$ ,  $^{195}\text{Pt}$ , and  $^{111}\text{In}$  could not be measured within 24 hours of irradiation and 3600 s of detection time, even at optimal thickness.

Several methods can enhance measurement of these radioisotopes' reaction cross sections, including using stacked targets, enriched isotopic targets, increasing gamma beam intensity, and employing low-background gamma spectrometers with improved shielding. Stacked targets—formed by combining several thin targets—can increase characteristic gamma decay counts and are relatively easy to implement. Using  $^{112}\text{Sn}(\gamma, p)^{111}\text{In}$  as an example, we simulated both natural and stacked targets with the same total thickness (1 mm). The natural target was 1 mm thick, while the stacked target comprised five pieces each 0.2 mm thick. [Figure 7: see original paper] compares the results.

For the characteristic gamma at  $E_\gamma = 171.29$  keV, the full-energy peak count rate measured with natural targets was 0.027 c/s, while the stacked target measurement yielded a significantly higher rate of 0.039 c/s. However, the low natural abundance of  $^{112}\text{Sn}$  in Sn targets results in low  $^{111}\text{In}$  yield. In actual measurements, background effects cause relative errors to exceed 10% even with stacked targets. Therefore, enriched isotopic targets can improve feasibility for  $^{112}\text{Sn}(\gamma, p)^{111}\text{In}$  cross-section measurements. With 100% isotopic enrichment, the count rate at  $E_\gamma = 171.29$  keV increases to 2.409 c/s, and the characteristic gamma full-energy peak becomes detectable under low-background shielding, as shown in [Figure 8: see original paper].

## V. Conclusion

Available data on gamma transmutation cross sections for medical radioisotopes are scarce. Measuring photonuclear reaction cross sections is essential for filling data gaps from gamma transmutation experiments, reducing large data uncertainties, and resolving discrepancies between various theoretical calculations and experimental data. This study simulated the generation, decay, and measurement of medical radioisotopes using GEANT4 combined with a shielded gamma-ray spectrometer.

Low-background shielding improves the signal-to-noise ratio, enables high-precision measurements, and conserves valuable beam time. Based on SLEGS, we demonstrated the feasibility of measuring reaction cross sections for specific medical radioisotopes. As China's only available gamma facility capable of providing intermediate- to high-energy, high-intensity, quasi-monochromatic, continuously energy-tunable gamma beams, SLEGS is expected to provide

an important platform for studying photonuclear reaction cross sections of medically relevant radioisotopes.

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