A Simple Estimate of Production of Medical Isotopes by Photo-Neutron Reaction at the Canadian Light Source

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Abstract

In contrast to the conventional bremsstrahlung photon beam sources, the laser back scatter photon sources at electron synchrotrons provide the selective tuning capability of photons of energies of interest. This feature coupled with the ubiquitous giant dipole resonance excitations of atomic nuclei promise a fertile ground of nuclear isotope productions. In this article, we present the results of simulations of production of medical/industrial isotopes ^{196}Au, ^{192}Ir and ^{99}Mo by (γ,n) reactions. We employed FLUKA Monte Carlo code along with the simulated photon flux for a beamline at the Canadian Light Source in conjunction with a CO_2 laser system.

1. Introduction

In recent years, there has been a growing interest for the new methods of production of medical isotopes. This led to several proposals of new nuclear reactors and accelerator based isotope production facilities [1-3]. It is to be noted that *the University of Saskatchewan proposal [1] calls for a multipurpose reactor* which can take a few years before it is built and commissioned. *The Sherbrooke proposal [2] seeks to employ proton beams from a cyclotron facility.* They propose to make use of ^{100}Mo(p,2n) reaction to produce ^{99m}Tc of a short half-life $T_{1/2}$ = 6.6 hours.

We suggest that *the modern photon beam facilities such as the laser back scatter systems at the electron synchrotron sources* used in conjunction with salient nuclear excitations have a good potential to produce medical isotopes in a more cost effective way with minimal background radiations. The unique features of the resonant photonuclear isotope transmutations using the laser photons scattered off GeV electrons have been well described in Ref. [4]. This article describes the preliminary Monte Carlo simulations (using FLUKA [5,6] code) for the production of ^{99}Mo, ^{196}Au and ^{192}Ir isotopes, which find extensive applications in medicine. Also, simulations for a proposed laser back scatter parameters in context with the Canadian Light Source (CLS) are described on the example of production of ^{99}Mo, ^{192}Ir and ^{196}Au.

A glance at the nuclear data tables will convince one that several medical isotopes can be produced using photon-nuclear reactions via the giant dipole resonance (GDR) [7] decay by emitting neutrons. In Figure 1, the latest measured [8,9] cross sections for (p,2n) reaction (^{100}Mo→^{99m}Tc),

are compared with cross section for photo-nuclear (γ,n) reactions for ^{99}Mo (^{100}Mo→^{99}Mo) [10] and ^{192}Ir (^{193}Ir→^{192}Ir) [11]. Some earlier cross section data of ^{100}Mo(p,2n)^{99m}Tc differ from the data of ref. [8,9] by a factor of two.

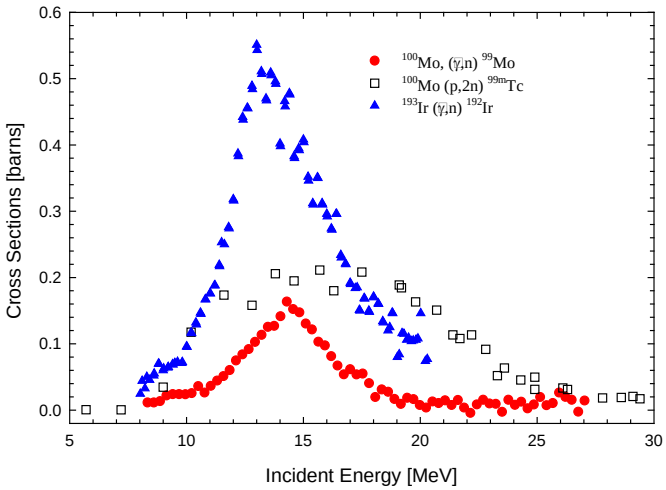

Figure 1 The cross sections for ^{100}Mo (γ,n) [10], (p,2n) [8,9] and ^{193}Ir (γ,n) [11] reactions versus incident beam energy. Data are taken from http://www.nndc.bnl.gov

The note worthy feature in Figure 1 is a maximum cross section at around 14 MeV (≈ $77*A^{1/3}$ MeV [7], where A is atomic mass) excitation with a width (FWHM) of about 5 MeV (≈ $23*A^{-1/3}$ MeV [7]). Thus, one should expect enhanced probabilities for the respective (γ,n) photo-neutron reactions to occur and both photonuclear reactions could be used in production of the respective medical isotopes.

Some examples of calculations were presented by us previously [12] to compare production of ^{99m}Tc isotope by proton beam with the application of gamma ray beam for ^{99}Mo production. The production of Iridium-192, used for industrial applications, can be done by (γ,n) reaction with ^{193}Ir (natural abundance ~ 63%) as the target material.

Along the same lines, the Prairie Isotope Production Enterprise (PIPE) [3] called for a photon induced reaction ^{100}Mo(γ,n)^{99}Mo but by using bremsstrahlung radiation from an electron linear accelerator. The idea is based on the work done at Idaho National Laboratory in the 1990s. Additionally preliminary experiments at NRC [13] show that a high-power 35 MeV electron accelerator could produce significant amount of ^{99}Mo for Canada. It is not the intention of this work to repeat simulation of this experiment. However for comparison we performed simplified simulation of bremsstrahlung production of photons in 0.7 cm thick, 7 cm diameter tungsten converter. We used 0.5 cm diameter pin electron beam with 35 MeV energy. In Figure 2 we show simulated density of photons produced per electron. The black horizontal lines indicate the possible ^{100}Mo target positions. They should be positioned very close to the tungsten converter (high photon density) but in real experiment there are restrictions, since the cooling system is required due to the excessive heat produced by the electron beam as shown in Figure 3.

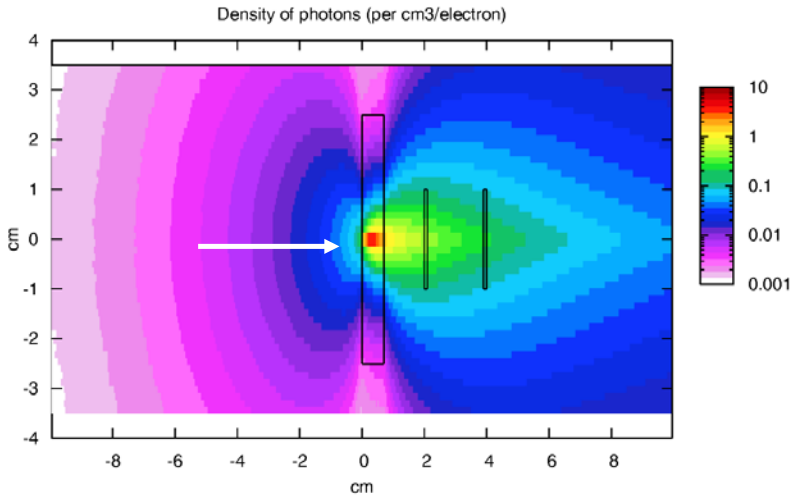

Figure 2 FLUKA simulation (with 1 keV lowest transport limit for photons and electrons) of bremsstrahlung production of photons in 0.7 cm thick, 7 cm diameter tungsten target by 0.5 cm diameter pin electron beam (35 MeV).

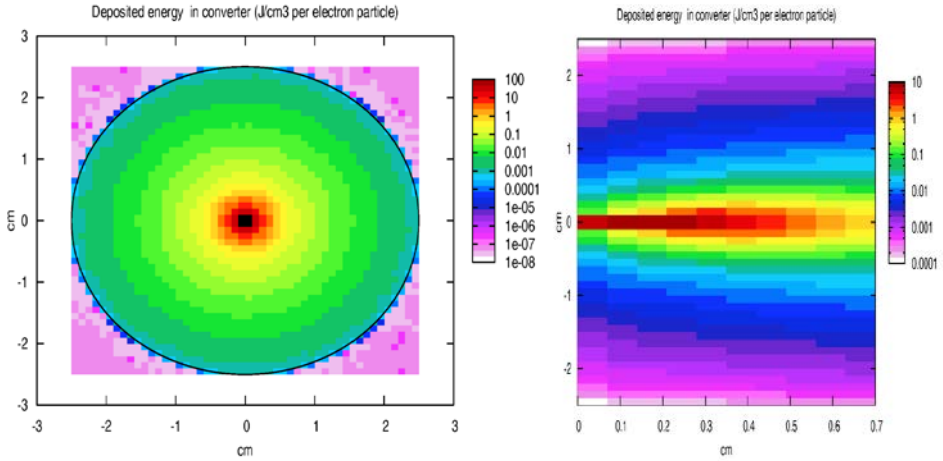

Figure 3 FLUKA simulation of energy density deposited per one electron of 0.5 cm diameter pin electron beam (35 MeV energy) in 0.7 cm thick, 7 cm diameter tungsten target.

The higher the electron current the more heat is created in the converter and therefore efficient cooling system is required. There is large number of photons produced per electron as shown in Figure 4, but only about 0.25 photons per electron for our parameters (35 MeV energy) are in the energy region (indicated by arrow) which can trigger GDR (compare Figure 1) and therefore the transmutation to ^{99}Mo may occur.

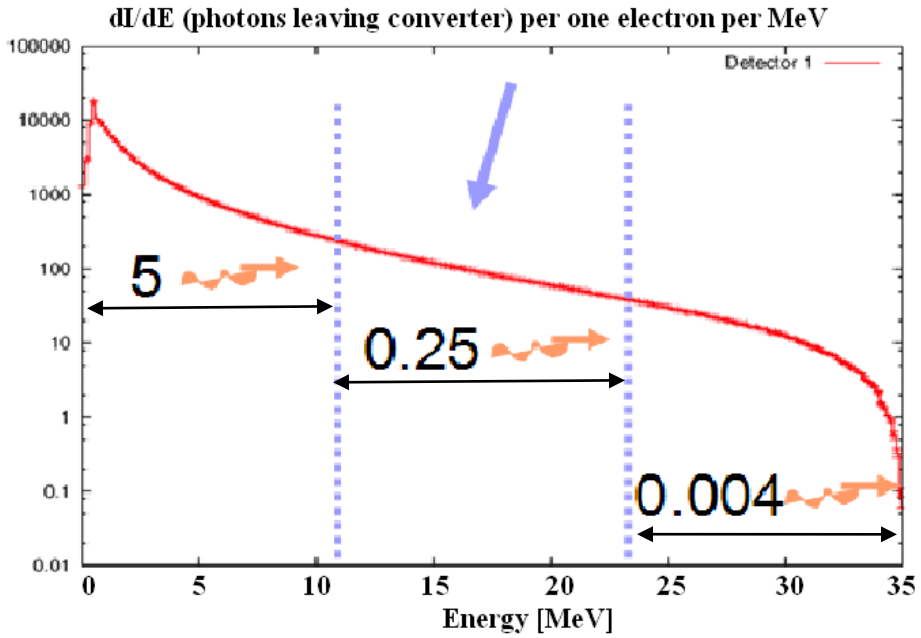

Figure 4 The bremsstrahlung spectrum calculated by FLUKA, plotted as number of photons produced per electron (of 35 MeV energy) in tungsten converter. The 0.25 photons are in the energy region where GDR occurs for the desired transmutation and as indicated by arrow.

The energy of photons is dependent on the setup of experiment and in this example there are a lot of them outside of the energy region of interest which create other isotopes or other undesirable radiation effects. In particular we calculated that there were 0.84 ^{98}Mo nuclei produced per one ^{99}Mo in the ^{100}Mo target, the closest to the converter, while the ratio was 0.93 for the second target.

Currently a high intensity bremsstrahlung radiation using the new electron linear accelerator is under construction at the CLS. More meaningful comparisons of experiment and simulations for production of ^{99}Mo by this method will be possible in the future.

2. Simulation of photon-nuclear interaction at Canadian Light Source

Recently, S. Daté simulated the photon flux for the laser back scatter systems at the CLS. The Klein–Nishina formula [14] was used to calculate back scattered photons energy distribution. We will use it here to estimate the exemplary production of ^{99}Mo, ^{192}Ir and ^{196}Au at CLS. A CO_2 laser used in conjunction with the 2.9 GeV electrons at the CLS will produce photon beams of up to 15 MeV. We will use here the same geometry of targets as proposed by H. Ejiri. To examine the transmutation as a function of depth and photon energy we will use four samples in one row of the same length (3 cm) and 0.2 cm diameter. In Figures 5 and 6 we show residuals and weighted residuals, respectively. They are calculated by FLUKA [5,6] with ^{197}Au, ^{193}Ir and ^{100}Mo targets per one photon, one cm^3 target and with the respective energies (per 1 MeV width) of the simulated

photon beam intensity of CLS as shown (Figures 5-6). The low energy cutoff for the transmutation of isotopes of interest is around 4 MeV below the maximum energy. The Figure 5 and 6 present calculations for the target (nr. 1), the closest to the irradiation source.

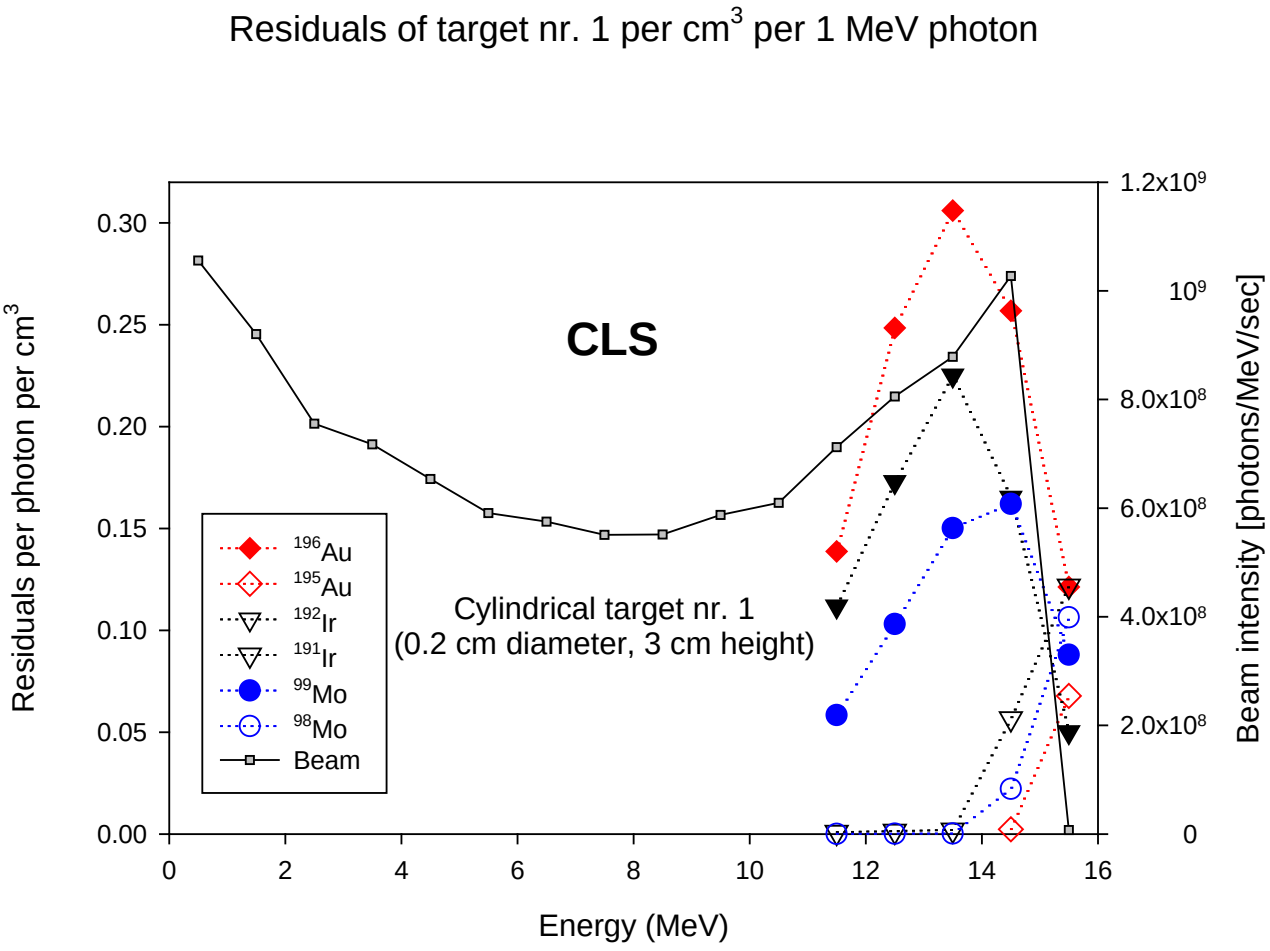

Figure 5 Residuals, calculated by FLUKA, in transmuted ^{197}Au, ^{193}Ir and ^{100}Mo targets per one photon per photon energy (1 MeV width) for given geometry. Also shown is the simulated photon beam intensity for the laser back scatter spectrum at the CLS with a CO_2 laser.

In Figure 5 one can see that at high energy of photon the ratio of production of ^{98}Mo to ^{99}Mo increases. However unlike the intensity distribution of photons produced by bremsstrahlung, the laser scatter photons shows a sharp cut off above 15 MeV and thus ^{98}Mo production is not significant (see Figure 6).

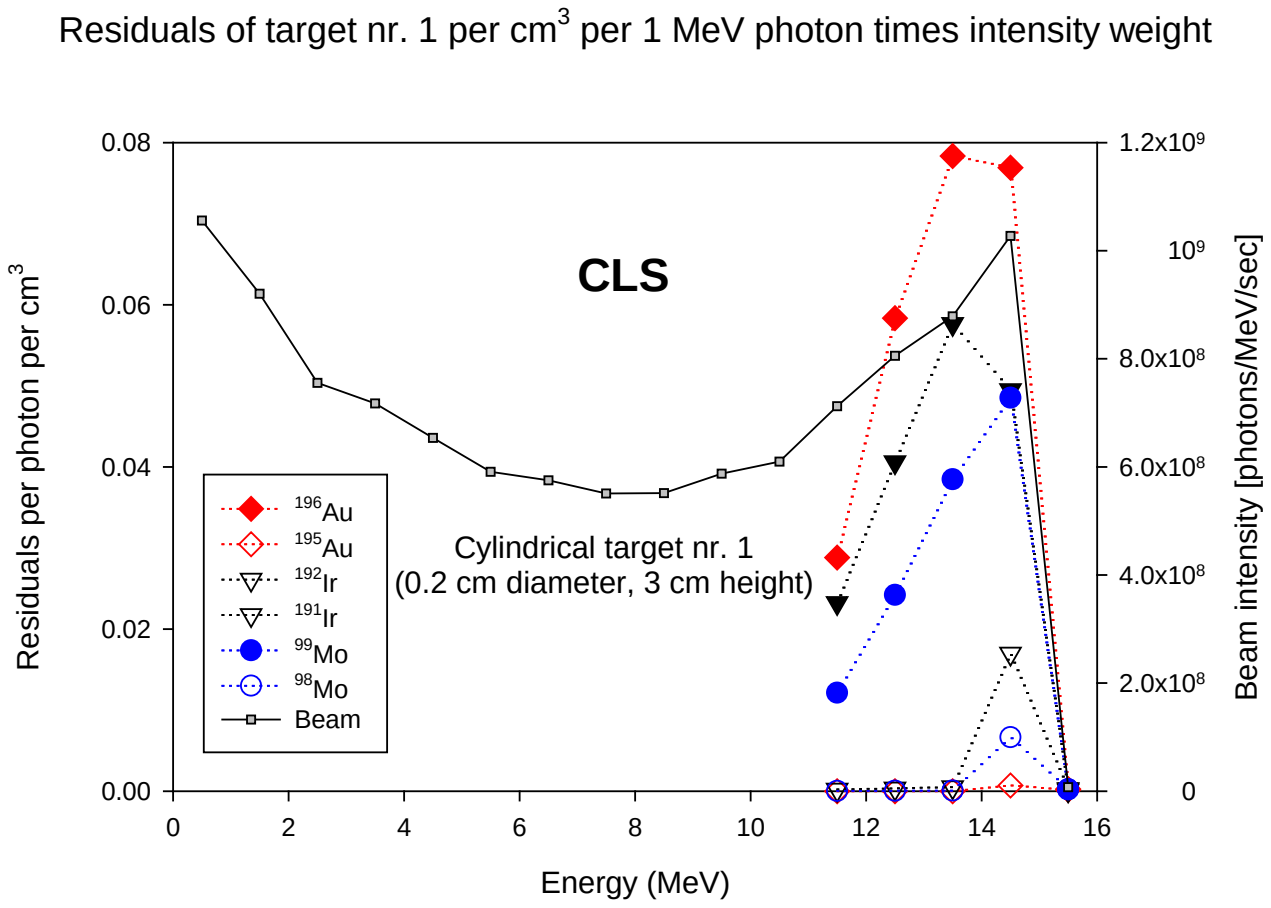

Figure 6 Residuals calculated by FLUKA in transmuted ^{197}Au, ^{193}Ir and ^{100}Mo targets per fraction of photon corresponding to this energy, per one cm^3 with the respective energies of the photon beam intensity as shown and assuming 1 MeV energy width.

Photon flux and the energy spectrum depend on the scattered angle and the target area. If the target is set far from the collision, and the target area is very small there are no low energy photons on the target as demonstrated by the NewSUBARU experiments [4]. While we do not know the total flux of photons produced at the CLS equipped with a CO_2 laser back scatter system, we may assume photon beam intensity of 11×10^9 photons per sec, of which 32% of photons (3.4×10^9 photons per sec) are within 4 MeV top energy window and it will be used to calculate photon induced activity. This spectrum feature is in contrast to bremsstrahlung spectra where we find less than 5% (see Figure 4) photon flux in the region of interest.

The induced activities, during irradiation and cooling time, for ^{100}Mo four all targets (described above) are presented in Figure 7. Using Bateman equations, we calculated weighted residuals (see e.g. Figure 6 for target nr. 1) for the proposed CLS flux and 2.74 days half life of ^{99}Mo.

The equilibrium state is reached at three times mean time value and as shown in Figures 7-8 stays almost unchanged. When the irradiation stops, the decay will occur with the characteristic mean life

(indicated as cooling). As can be seen in Figure 7 the maximum induced activity of ^{99}Mo is too small for commercial production of medical isotopes because of low intensity of photon flux.

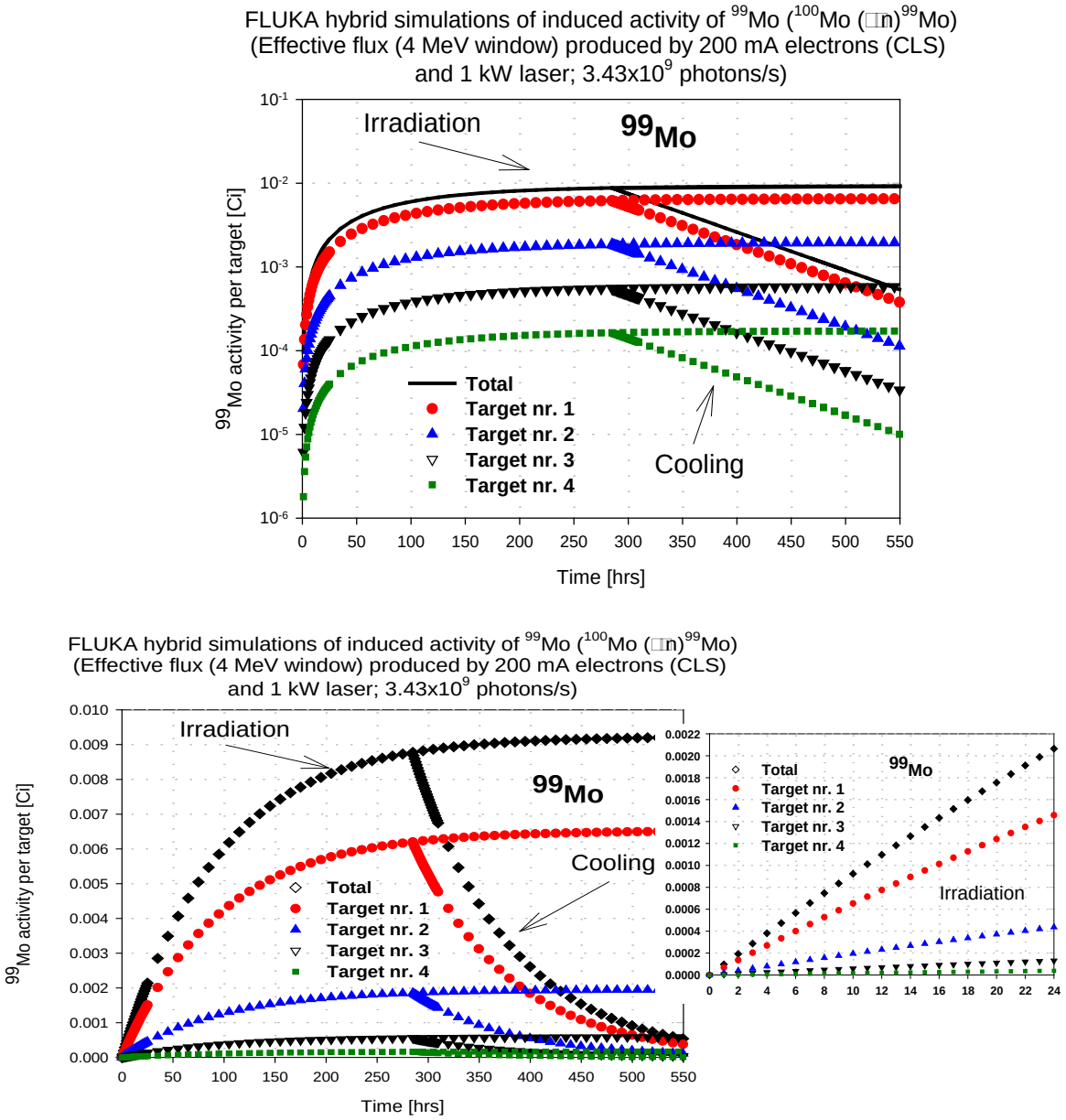

Figure 7 The induced activities of ^{100}Mo four targets (as described in text), calculated numerically using the calculated by FLUKA residuals and the predicted CLS flux and half life time ($T_{1/2}$) of 2.74 days of ^{99}Mo as a function of time. The activity during cooling time is also shown as indicated. The units on the axis of the embedded picture are the same as on the other figures.

In Figure 8 similar calculations of induced activity during irradiation and cooling time are presented for ^{197}Au. Since in this calculations RESNUCLEI detector is used, the contribution from the estimated from FLUKA activity of ^{196}Au isomers is not included. The half-life value equal to 6.17

days is used. When comparing with the previous figure for ^{99}Mo transmutation one can see that it is less useful to employ four targets for the transmutation of heavier elements as for example Au, due to the shorter penetration depth of photons in these targets as seen in the bottom of Figure 8.

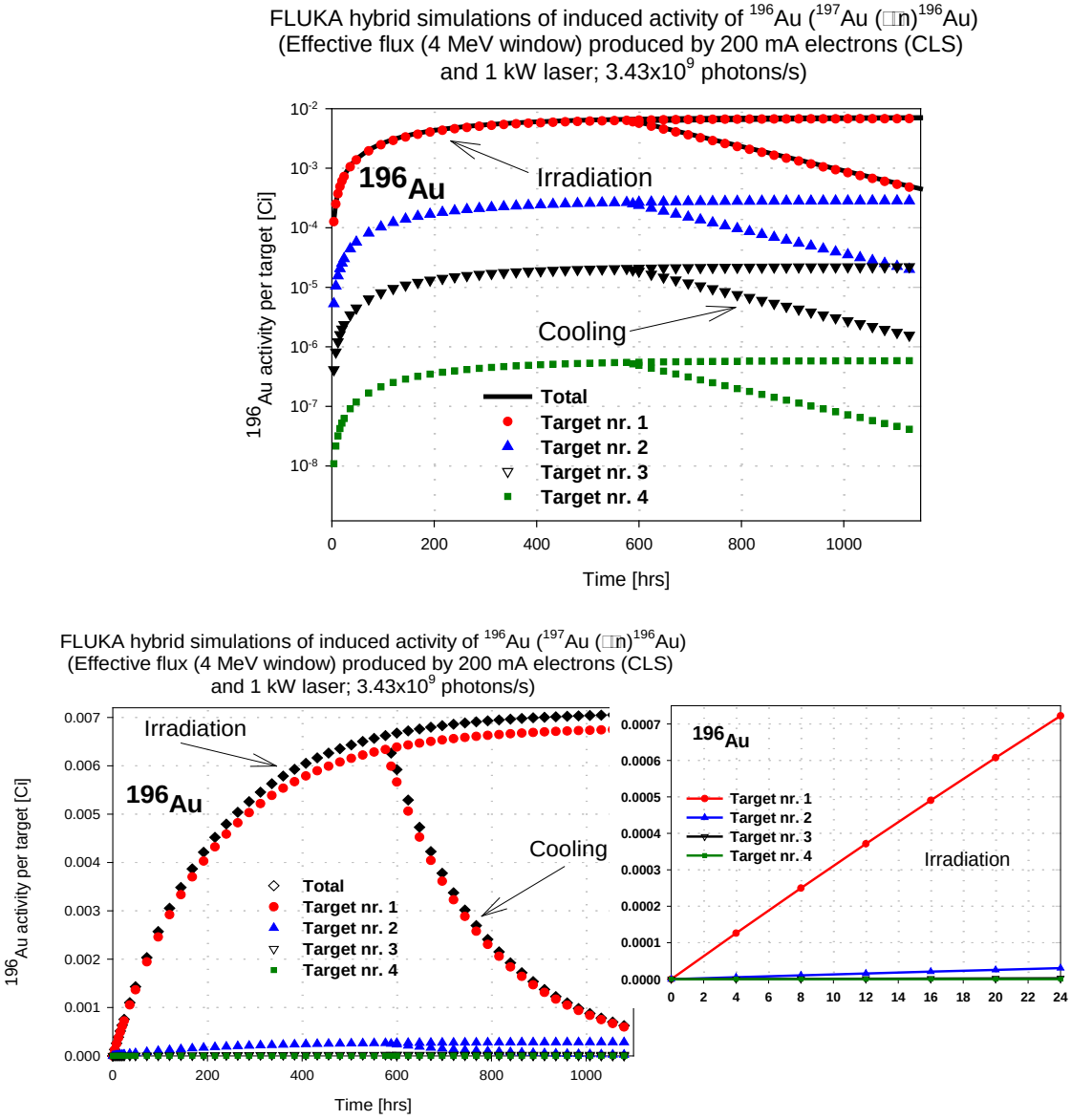

Figure 8 The induced activity per ^{197}Au targets (four targets as described in text), calculated analytically using the calculated by FLUKA residuals and the predicted CLS flux and half life time ($t_{1/2}$) of 6.17 days of ^{196}Au as a function of time. The activity during cooling time is also shown as indicated. The units on the axis of the embedded picture are the same as on the other figures.

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3. Comparison of ^{99}Mo yield from ^{100}Mo (γ,n) photonuclear reaction and ^{238}U photofission.

It is known that a ^{99}Mo can also be produced by photo-fission of ^{238}U. In Table 1 the simulated by FLUKA fission yield is shown per 16 MeV photons (the quarter value of the total gamma width of GDR resonance:1.89 eV is used). A cylindrical ^{238}U target of dimensions of 0.012 m diameter and 0.036 m height embedded in lead container is simulated. As shown in Table 1 only 1.17×10^{-7} nuclei of ^{99}Mo are produced directly through photo-fission in ^{238}U and most of the total number produced (1.28×10^{-4}) is produced via (β^-) decay. It is significantly lower amount than obtained by transmutation using GDR for the same geometry and the photon incident energy 14.8 MeV with the quarter value of the total width of GDR resonance (2.5 MeV) is assumed for both beams as presented in Table 2 (1.31×10^{-2}). Nevertheless, FLUKA may underestimate photo-fission [15] future experimental investigation at CLS may, thus, experiment would be useful.

Table 1
^{99}Mo nuclei produced in photofission reaction (see text for details). The last column shows the sum of β^- yield of isotopes (listed in columns 3-8) that lead to the production of ^{99}Mo, while the second column shows direct production of ^{99}Mo.

Photons (16 MeV)	**Fission yield* per one photon [Nuclei/ target]**							**β^- yield**
Target	**^{99}Mo (42)**	^{99}Kr (36)	^{99}Rb (37)	^{99}Sr (38)	^{99}Y (39)	^{99}Zr (40)	^{99}Nb (41)	**^{99}Mo (42)**
^{238}U	**1.17×10^{-7}**	8.32×10^{-8}	3.94×10^{-6}	1.83×10^{-5}	6.47×10^{-5}	3.46×10^{-5}	6.15×10^{-6}	**1.28×10^{-4}**
Errors (%)	**20.2**	20.5	2.1	1.1	0.9	0.7	2.3	

*This is an order of magnitude estimate as we neglect the neutron branchings of ^{99}Kr and ^{99}Rb.

Table 2
Isotopes produced by the photo nuclear reaction on ^{100}Mo. For target details see the text. The first column lists the reaction and the product nucleus.

(reaction) Product isotope	Yield per one photon	Error [%]
(n,γ) ^{101}Mo$^{\#}$	3.65×10^{-06}	1.2
$(\gamma,e^+e^-)_{atomic}$ ^{100}Mo	7.85×10^{-05}	0.4
(γ,n) ^{99}Mo	**1.31×10^{-02}**	**0.03**
(γ,2n) ^{98}Mo	6.06×10^{-03}	0.1
(γ,3n) ^{97}Mo	9.27×10^{-05}	0.6
(γ,4n) ^{96}Mo	1.04×10^{-04}	0.3
(γ,5n) ^{95}Mo	1.56×10^{-04}	0.2

For the production of ^{99}Mo at CLS natural Mo with 9.6 % of ^{100}Mo can be used because photonuclear reactions on other Mo isotopes produce stable Mo isotopes or the very long-lived ^{93}Mo or very short lived ^{91}Mo, all of which do not disturb ^{99}Mo activity. ^{100}Mo enriched isotopes are also quite realistic to produce by using MoF_6 centrifugal separation as used for example for double beta decays [16].

4. Conclusion

The idea of using MeV photon beams by taking advantage of an ubiquitous feature of nuclear excitations, known as Giant Dipole Resonance (GDR) is explored. The Monte Carlo simulations using the FLUKA demonstrate that the medical isotopes ^{99}Mo, ^{192}Ir and ^{196}Au are the main products of photonuclear reaction on ^{197}Au, ^{193}Ir and ^{100}Mo targets. The production of ^{99}Mo by GDR photoneutron reaction on ^{100}Mo target and ^{196}Au on ^{197}Au target are also explored using proposed parameters for experiment at CLS. It is pointed out that the CLS facility equipped with a CO_2 laser back scatter system will serve as a good testing ground to establish the feasibility of this technique. We conclude that this technique is promising for isotope production at future high intensity photon sources, even if the CLS would not become a commercial facility.

- **Acknowledgement**

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