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

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Collaboration with C. Rangacharyulu, S. Daté, H. Ejiri

(Published: Szpunar B., Rangacharyulu C., Date' S. and Ejiri H., 33rd Annual Conference of the Canadian Nuclear Society, Saskatoon, Canada, 3A/138, June 10-13 (2012))

Earlier presentation available on:

September 20th, 2012, Vancouver,
2nd FLUKA Advanced Course and Workshop

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Abstract

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\textsubscript{2} laser system.

The photon intensity cannot be modeled in FLUKA yet. S. Daté from Spring 8, Japan, simulated the photon flux for the laser backscatter systems at the CLS. Klein-Nishina formula was used to calculate back scattered photons energy distribution.

This calculated parabolic shape of photon energy distribution is not included as a standard shape of source in Fluka. Therefore user routine needs to be used in modeling transmutations for this source. Alternatively one can use FLUKA to calculate transmutations for a narrow energy width (for which the standard source can be used) and integrate them over the whole energy range. The induced activities (of $^{99}$Mo, $^{192}$Ir and $^{196}$Au) were calculated using Bateman equations (implemented in EXCEL) and weighted according to the shape of the source residuals (obtained by FLUKA).
Content

• Purpose of this work and introduction
• FLUKA simulation
  • Photon induced artificial transmutation
    • medical isotope production
    • photofission
• Summary and conclusions
Purpose of this work and introduction

• Shortage of medical isotopes – call for alternative production methods
• Most widely used $^{99}\text{Mo} \rightarrow ^{99m}\text{Tc}$ (~35 common radiopharmaceuticals)

$^{99m}\text{Tc} \rightarrow ^{99}\text{Tc} + \gamma$

Each diagnostic uses few GBq
(1GBq = 0.027 Ci)

• EPAC 2000 Vienna, world uses: 150,000 Ci/year
• Proton cyclotrons located close to hospital can supply average usage: e.g. at CHUS, Sherbrooke 10 Ci/week of $^{99m}\text{Tc}$
• $^{99m}\text{Tc}$ is short-lived ($T_{1/2} = 6.0058$ h) therefore $^{99}\text{Mo}$ ($T_{1/2} = 65.94$ h) needed for remote, small hospitals
Artificial transmutation (photons)

\[
{}^{100}\text{Mo} \rightarrow {}^{99}\text{Mo} + {}^{0}\text{e} + \bar{\nu}
\]
Giant Dipole Resonance

Atomic mass
0 50 100 150 200 250
Energy / Width
[MeV]
0
20
40
60
80
Energy
Width

\[ 77A^{-1/3} \]

\[ 23A^{-1/3} \]
Medical Isotopes (photons)

Incident Energy [MeV]

Cross Sections [barns]

$^{100}$Mo ($\gamma,n$) $^{99}$Mo
$^{193}$Ir ($\gamma,n$) $^{192}$Ir
$^{197}$Au ($\gamma,n$) $^{196}$Au

LINAC ~ NRC (Bremsstrahlung)

Electron’s current:
43 μA: $27 \times 10^{13}$ e/s

Pin beam 35 MeV
• CLS equipped with a CO$_2$ laser back scatter system to test the feasibility of application of photo-nuclear transmutations.

• Discussion and collaboration with international community (Japan (JAEA), USA).

• Achievable at CLS maximum photon energy (Compton back-scattering at 0 degree incident angle): 15 MeV for 2.9 GeV electron beam energy.

• Intensity (Date'): $10^{10}$/s for 1 kW CO$_2$ laser, 250 mA electron beam current; maximal ($\sim P_L I_e$): $10^{12}$/s (50 kW, 500 mA). The highest ever reached by Duke HIGS FEL: $2 \times 10^9$/s.
GDR transmutation (CLS): maximum intensity limitation

Even maximal $10^{12}$/s photon intensity can only transmute very small fraction of atoms, therefore one can neglect interaction between particles produced in each transmutation!

$6 \times 10^{21}$ atoms in 1g of Mo
FLUKA; Photon induced activity (multiple targets by H. Ejiri)

Chary’s safe parameters for CLS used: 200 mA electrons and 1kW laser

S. Date’ photon flux simulations
Compton e-ph elastic scattering from resting electron

(Partially from Wikipedia)

Compton scattering formula:

$$\lambda' - \lambda = \frac{h}{m_e c} (1 - \cos \theta),$$

$\lambda$ is the initial wavelength, $\lambda'$ is the wavelength after scattering, $m_e$ is the mass of an electron.

Klein-Nishina formula (differential cross section of photons over solid angle: $d\Omega = 2\pi \sin \theta d\theta$):

$$\frac{d\sigma}{d\Omega} = \frac{1}{2} \alpha^2 r_e^2 P(E_\gamma, \theta)^2 (P(E_\gamma, \theta) + P(E_\gamma, \theta)^{-1} - 1 + \cos^2(\theta))$$

where $\alpha$ is the fine structure constant, $r_e = \frac{\hbar}{m_e c}$ is the Compton radius of the electron, $\theta$ is the scattering angle.

$$P(E_\gamma, \theta) = \frac{1}{1 + \frac{E_\gamma}{m_e c^2} (1 - \cos \theta)}$$

is the ratio of photon energy after and before ($E' = \frac{hc}{\lambda'}$) the collision.

By combining the Compton’s and Klein-Nishina’s formulas one can calculate the energy ($E' = \frac{hc}{\lambda'}$) distribution of scattered photons.

Compton e-ph backscattering from relativistic electrons (CLS)

Using invariant differential cross section for Compton scattering (Mandelstam variables) the energy distribution of backscattered photons was calculated by S. Date’ (Spring 8, Japan).
Simulated by S. Daté Spring 8, Japan, photon intensity for the CO$_2$ laser back scatter systems at the CLS

Beam intensity (flat rectangular: $\Delta x = \Delta y = 0$ cm) per 1 MeV photon

Energy (MeV)
0 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16

Beam intensity [photons/MeV/sec]
0 2.0x10$^8$ 4.0x10$^8$ 6.0x10$^8$ 8.0x10$^8$ 10$^9$ 1.2x10$^9$

- Beam
- SUBARU (CLS set up)
Residuals of target nr. 1 per cm$^3$ per 1 MeV photon

Energy (MeV)
0 2 4 6 8 10 12 14 16

Residuals per photon per cm$^3$

Beam intensity [photons/MeV/sec]

Cylindrical target nr. 1
(0.2 cm diameter, 3 cm height)
Residuals of target nr. 1 per cm$^3$ per 1 MeV photon times intensity weight

<table>
<thead>
<tr>
<th>Energy (MeV)</th>
<th>Residuals per photon per cm$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.00</td>
</tr>
<tr>
<td>2</td>
<td>0.02</td>
</tr>
<tr>
<td>4</td>
<td>0.04</td>
</tr>
<tr>
<td>6</td>
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</tr>
<tr>
<td>8</td>
<td>0.08</td>
</tr>
<tr>
<td>10</td>
<td>0.10</td>
</tr>
<tr>
<td>12</td>
<td>0.12</td>
</tr>
<tr>
<td>14</td>
<td>0.14</td>
</tr>
<tr>
<td>16</td>
<td>0.16</td>
</tr>
</tbody>
</table>

Beam intensity [photons/MeV/sec]

CLS

Cylindrical target nr. 1 (0.2 cm diameter, 3 cm height)
Estimated from FLUKA activity of $^{196}$Au isomers not included
FLUKA hybrid simulations of induced activity of $^{99}$Mo ($^{100}$Mo ($\gamma$,n)$^{99}$Mo) 
(Effective flux (4 MeV window) produced by 200 mA electrons (CLS) and 1 kW laser; $3.43 \times 10^9$ photons/s)

![Graph of 99Mo activity over time](image)

$3 \tau = 285$ hrs
Photofission versus GDR transmutation

Szpunar B., Rangacharyulu C., Date' S. and Ejiri H., 33rd Annual Conference of the Canadian Nuclear Society, Saskatoon, Canada, 3A/138, June 10-13 (2012)

<table>
<thead>
<tr>
<th>Photons (16 MeV)</th>
<th>Fission yield per one photon [Nuclei/cm³] (May be heavily underestimated by the FLUKA currently!)</th>
<th>β·yield</th>
</tr>
</thead>
<tbody>
<tr>
<td>Target</td>
<td>99Mo(42)</td>
<td>99Kr(36)</td>
</tr>
<tr>
<td>238U</td>
<td>1.17x10⁻⁷</td>
<td>8.32x10⁻⁸</td>
</tr>
<tr>
<td>Errors (%)</td>
<td>20.2</td>
<td>20.5</td>
</tr>
</tbody>
</table>

14.8 MeV

<table>
<thead>
<tr>
<th>Produced Isotope (reaction)</th>
<th>Yield [per one photon/cm³]</th>
<th>Error [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>(n,y) 101Mo#</td>
<td>3.65 x10⁻⁶</td>
<td>1.2</td>
</tr>
<tr>
<td>(γ,e⁺e⁻)atomic 100Mo</td>
<td>7.85 x10⁻⁵</td>
<td>0.4</td>
</tr>
<tr>
<td>(γ,n) 99 Mo</td>
<td>1.31 x10⁻²</td>
<td>0.03</td>
</tr>
<tr>
<td>(γ,2n) 98Mo</td>
<td>6.06 x10⁻³</td>
<td>0.1</td>
</tr>
</tbody>
</table>

#Secondary neutron capture
FLUKA simulations show that the production of the desired isotopes via GDR (photon) are orders of magnitude higher than the other isotopes, indicating this technique to be promising method for artificial transmutations.

Applications

- Production of medical and industrial isotopes
- Induced transmutation & photofission as a source of neutrons
Acknowledgement

- Access to FLUKA code
- FLUKA developers and support group (especially Francesco Cerutti)
- V. Vlachoudis for FLAIR graphic interface with FLUKA
- C. Cederstrand for technical support
- Science and Engineering division for start up funding and support
Thank you for your attention!