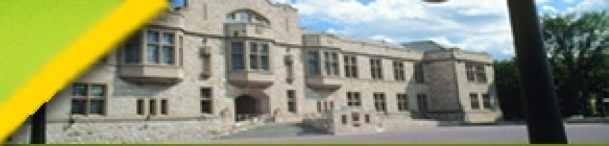

Simulation of photon-nuclear interaction in production of medical isotopes and transmutation of nuclear waste

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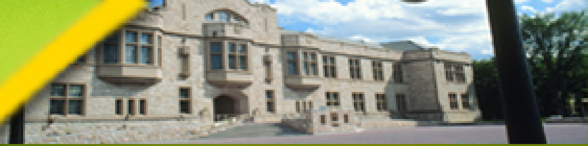
June 18th, 2010, Osaka, JICA

In memory of my mother
Anna
her unlimited love
and support!



Content

- **Purpose of this work and introduction**
- **Background on FLUKA simulation method**
- **Photon induced artificial transmutation**
    - Medical isotope production
    - Photofission
    - Treatment of long-lived isotopes
- **Summary and conclusions**

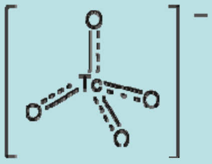
Purpose of this work and introduction

- Shortage of medical isotopes – call for alternative production methods
- Most widely used ^{99}Mo -> ^{99m}Tc (~35 common radiopharmaceuticals)

^{99m}Tc -> ^{99}Tc + γ

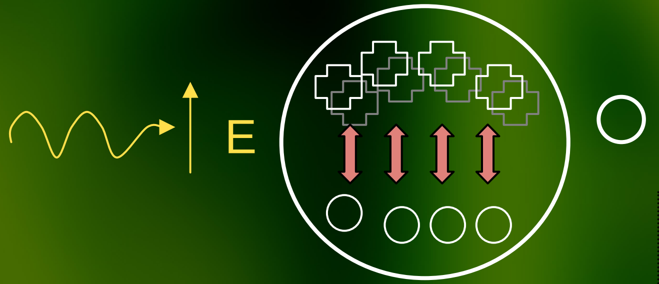
Each diagnostic uses few GBq

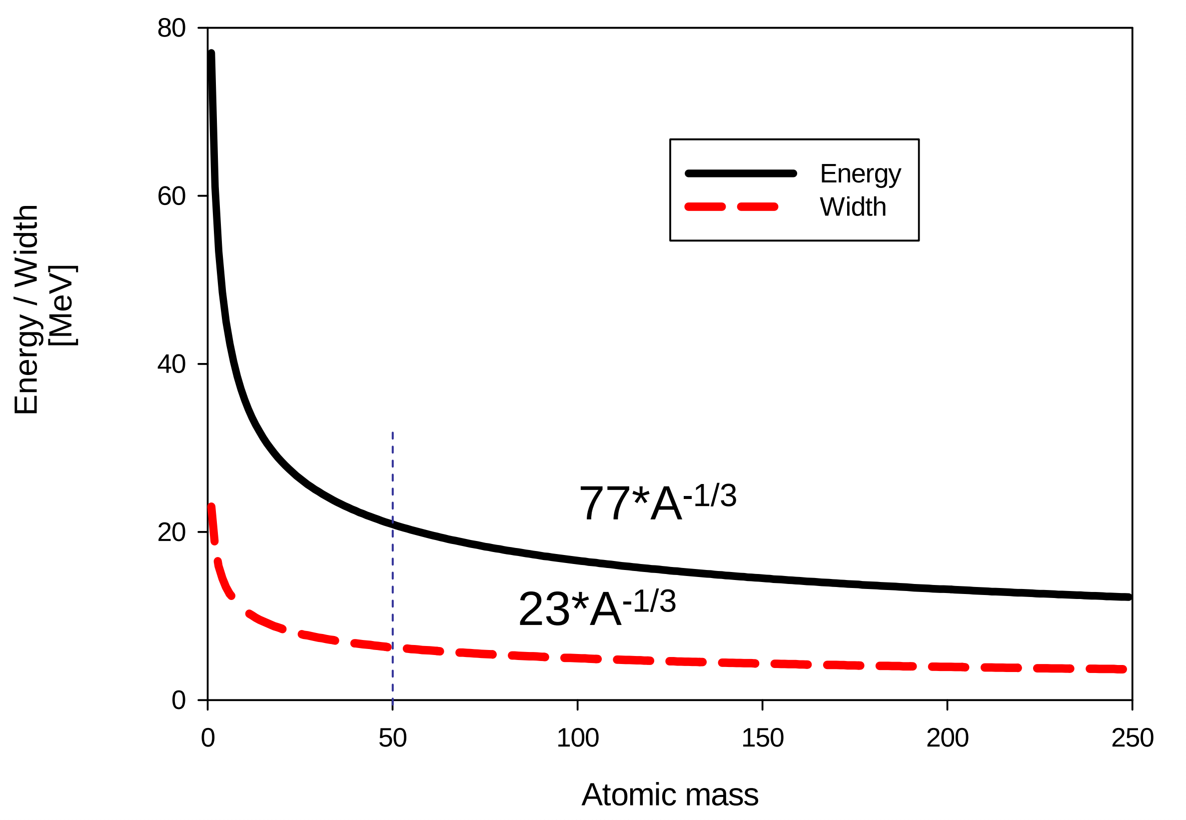
(1GBq = 0.027 Ci)



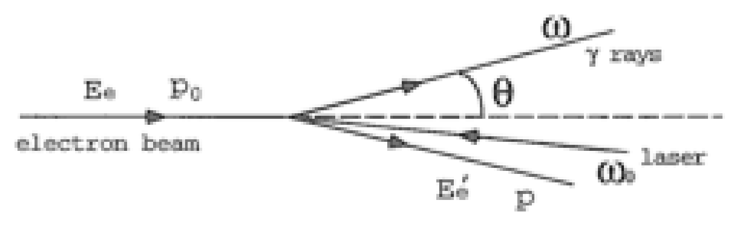
  - EPAC 2000 Vienna, world uses: 150,000 Ci/year
  - Proton cyclotrons located close to hospital can supply average usage: e.g. at CHUS, Sherbrooke 10 Ci/week of ^{99m}Tc
- ^{99m}Tc is short-lived ($T_{1/2}$ = 6.0058 h) therefore ^{99}Mo ($T_{1/2}$ = 65.94 h) needed for remote, small hospitals

^{99}Mo and other medical and industrial usage isotopes can be produced using photons at Giant Dipole Resonance energies

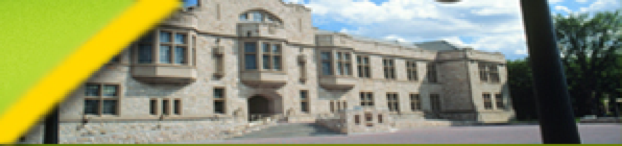




April 2010, Canadian Light Source, workshop



- 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: 15 MeV for 2.9 GeV electron beam energy (0 degree incident angle)
- Supportive FLUKA simulation: design of experiment, evaluation



FLUKA
http://www.fluka.org

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- **>2000 users**
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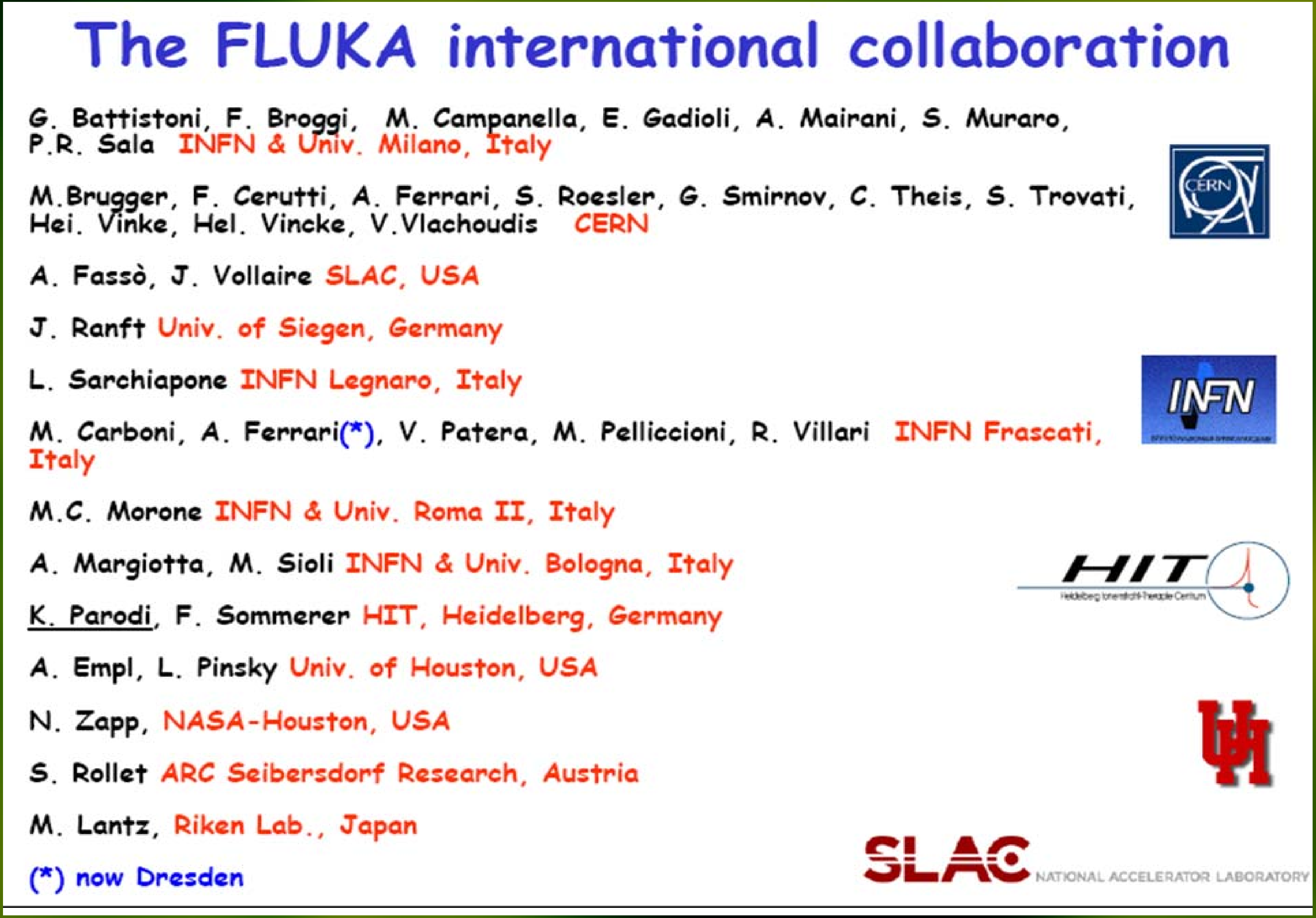

The FLUKA international collaboration

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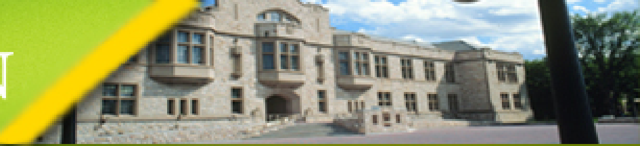
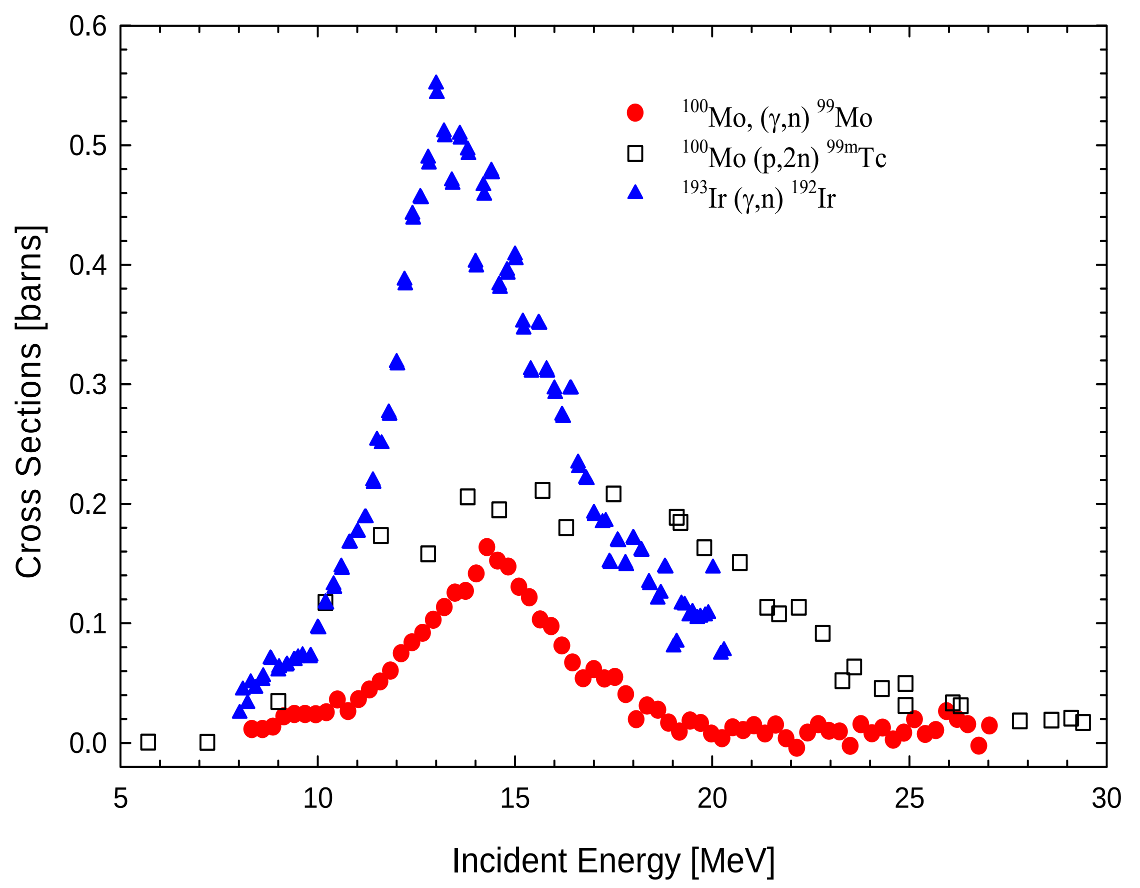


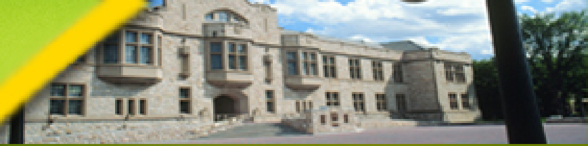
FLUKA code

- **FLUKA** (**FLU**ktuierende **KA**skade), fully integrated Monte Carlo simulation package for the interaction and transport of particles and nuclei in matter
- Well tested models
- Optimized by comparing with experimental data
- Fully analogue code or biased mode
- Double precision used, 10^{-10} energy conservation
- 60 particles modeled including polarized light
- First Monte Carlo particle transport code with photonuclear interactions: MeV – TeV energy range

Medical Isotopes (photons versus protons)



http://www.nndc.bnl.gov



I: FLUKA hybrid simulations

- Five runs (each 10^6 particles, GDR energy) ← **No time dependence**
  - •Energy deposition, equivalent dose
  - •Fluence ($n = L_{particle} / V_{target} \rightarrow dn/d\ln E$): neutron, photon, proton
  - •Residual nuclei (R_n- per particle)

Time dependence

- Calculate induced activity for a given beam intensity (I)

$$N(t \le t_i) = R\tau(1 - e^{-t/\tau})$$

$$\left|\frac{dN(t > t_i)}{dt}\right| = R(1 - e^{-t_i/\tau})e^{-(t-t_i)/\tau}$$

$R = I * R_n$

t_i – irradiation time

τ – mean life

$\text{Activity}_N(t) = N(t)\ \text{Activity}_{atom}$

II: FLUKA induced activity simulations

Time dependence

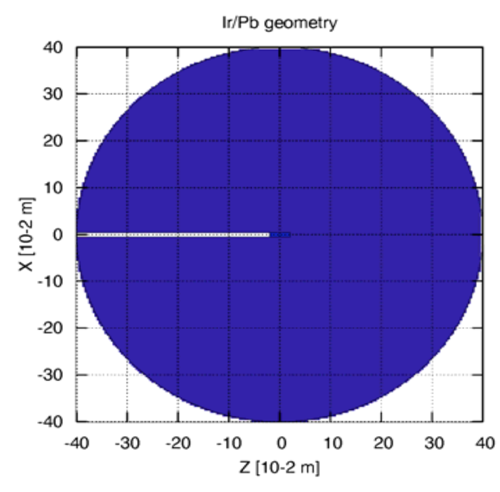
- Set irradiation and cooling time, beam intensity
- Perform five runs using FLUKA’s exact analytical implementation of Bateman equations for induced activity calculations
  - Total activity
  - Yield

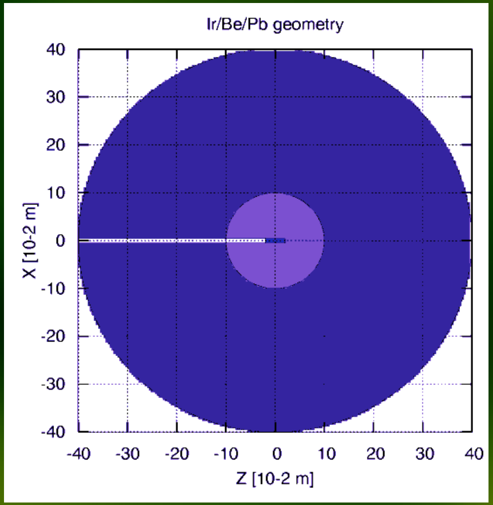
Photonuclear GDR interaction in natural Ir and ^{193}Ir (target: 1cm (d) x 4 cm (h)) (CNS 2010)

Natural Ir

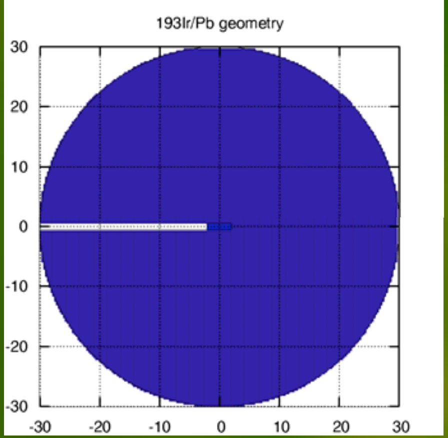
(37% of ^{191}Ir and 63% of ^{193}Ir)

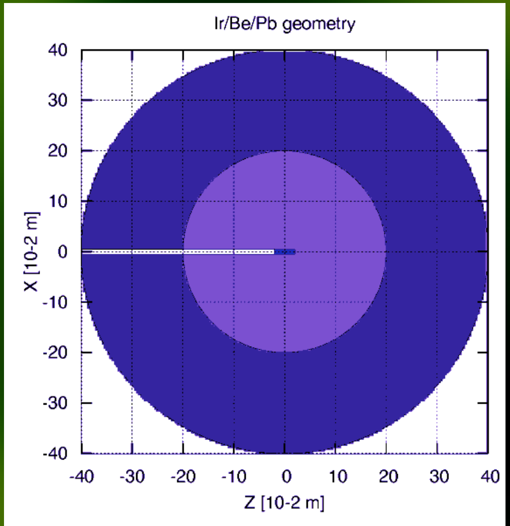
^{192}Ir, $T_{1/2}$ = 73.83 d

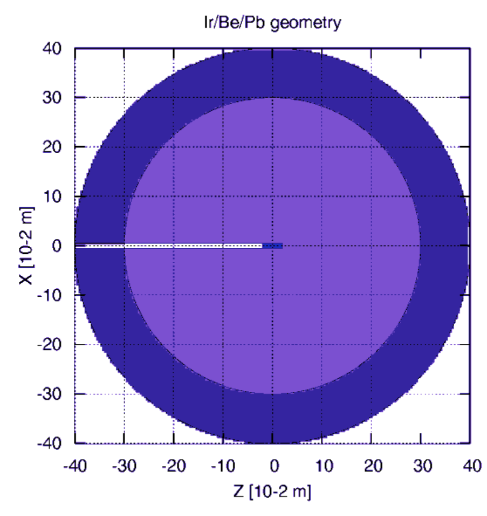


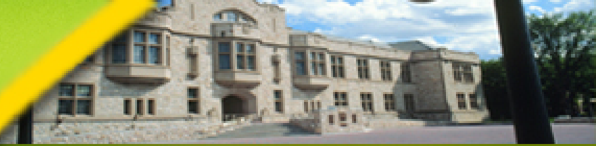


^{193}Ir target









Natural Ir, n reflected by Be

^{193}Ir target

Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
$(\gamma,n)\ ^{194}\mathrm{Ir}^{\#}$	1.33×10^{-4}	0.2
$(\gamma,e^+e^-)_{atomic}\ ^{193}\mathrm{Ir}$	1.28×10^{-3}	0.1
$(\gamma,n)\ ^{192}\mathrm{Ir}$	**2.04×10^{-2}**	**0.04**
$(\gamma,2n)\ ^{191}\mathrm{Ir}$	9.16×10^{-4}	0.1
$(\gamma,p)\ ^{192}\mathrm{Os}$	1.97×10^{-8}	42

#Secondary neutron capture

*** ^{191}Ir**

Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
$(n,\gamma)\ ^{194}\mathrm{Ir}^{\#}$	3.44×10^{-4}	1.2
$(\gamma,e^+e^-)_{atomic}\ ^{193}\mathrm{Ir}$	1.29×10^{-3}	1
(γ,n) or $(n,\gamma)^*\ ^{192}\mathrm{Ir}$	**1.32×10^{-2}**	**0.3**
$(\gamma,2n)\ ^{191}\mathrm{Ir}$	8.87×10^{-4}	0.4
$(\gamma,3n)\ ^{190}\mathrm{Ir}$	7.56×10^{-3}	0.9

Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
$(n,\gamma)\ ^{194}\mathrm{Ir}^{\#}$	3.19×10^{-4}	1.9
$(\gamma,e^+e^-)_{atomic}\ ^{193}\mathrm{Ir}$	1.29×10^{-3}	0.9
(γ,n) or $(n,\gamma)^*\ ^{192}\mathrm{Ir}$	**1.31×10^{-2}**	**0.4**
$(\gamma,2n)\ ^{191}\mathrm{Ir}$	9.06×10^{-4}	1.7
$(\gamma,3n)\ ^{190}\mathrm{Ir}$	7.51×10^{-3}	0.3
$(\gamma,p)\ ^{190}\mathrm{Os}$	2.0×10^{-7}	99

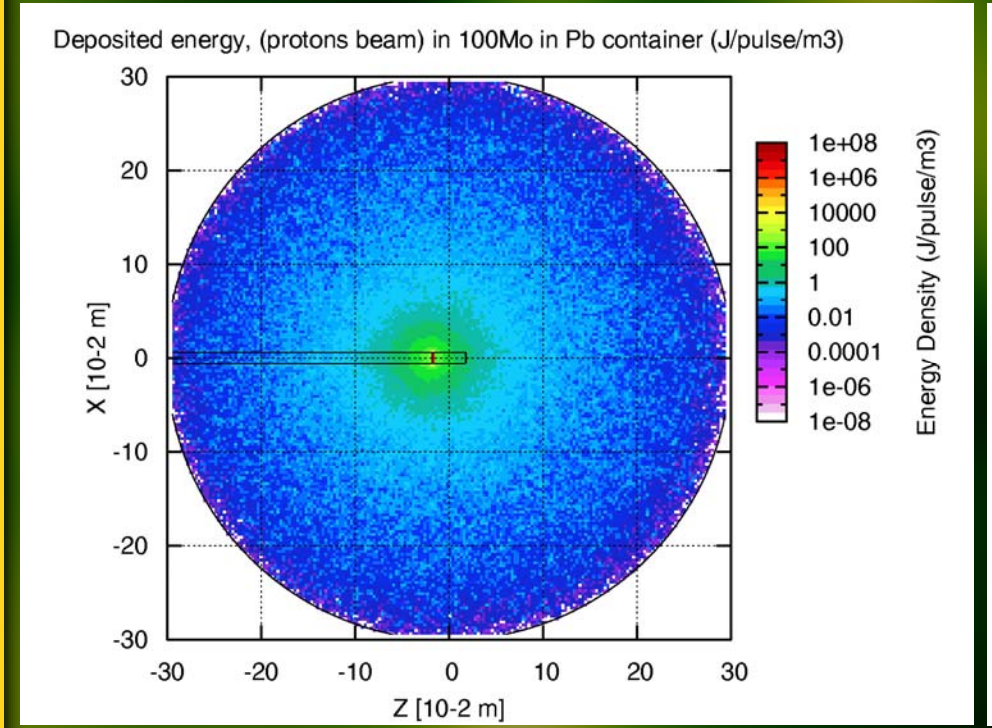
(37% of ^{191}Ir and 63% of ^{193}Ir)

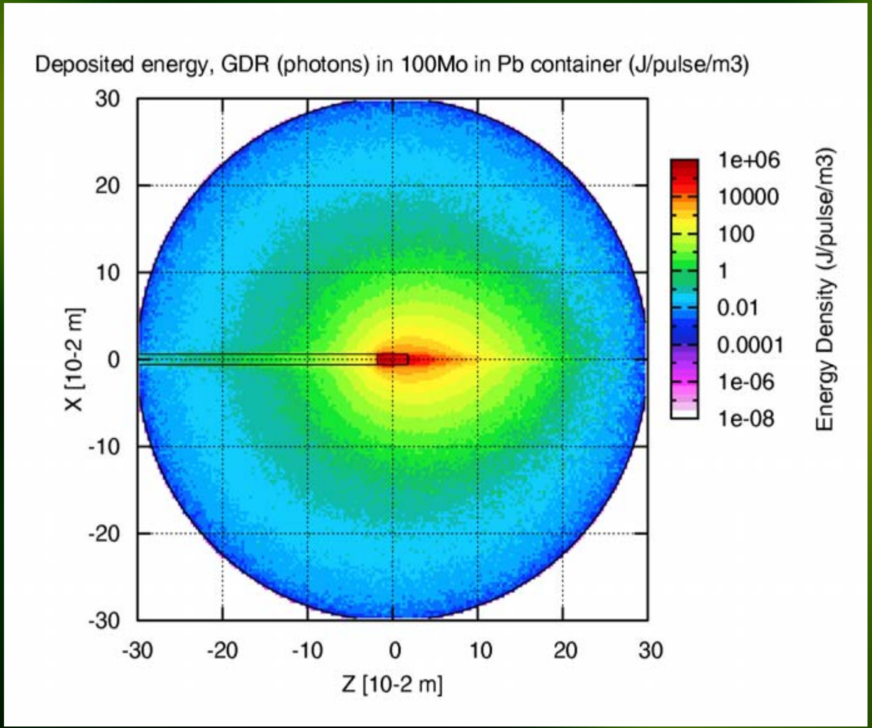
(Reaction) Produced Isotope	Yield [per one photon/cm^3]	Error [%]
$(n,\gamma)\ ^{194}\mathrm{Ir}^{\#}$	3.52×10^{-4}	1.7
$(\gamma,e^+e^-)_{atomic}\ ^{193}\mathrm{Ir}$	1.29×10^{-3}	0.7
(γ,n) or $(n,\gamma)^*\ ^{192}\mathrm{Ir}$	**1.33×10^{-2}**	**0.2**
$(\gamma,2n)\ ^{191}\mathrm{Ir}$	8.92×10^{-4}	1.1
$(\gamma,3n)\ ^{190}\mathrm{Ir}$	7.55×10^{-3}	0.3

FLUKA; Proton versus photon ^{100}Mo transmutation; Geometry (target: 1.2 cm (d) x 3.6 cm (h)), energy deposition

22 MeV (p,2n)

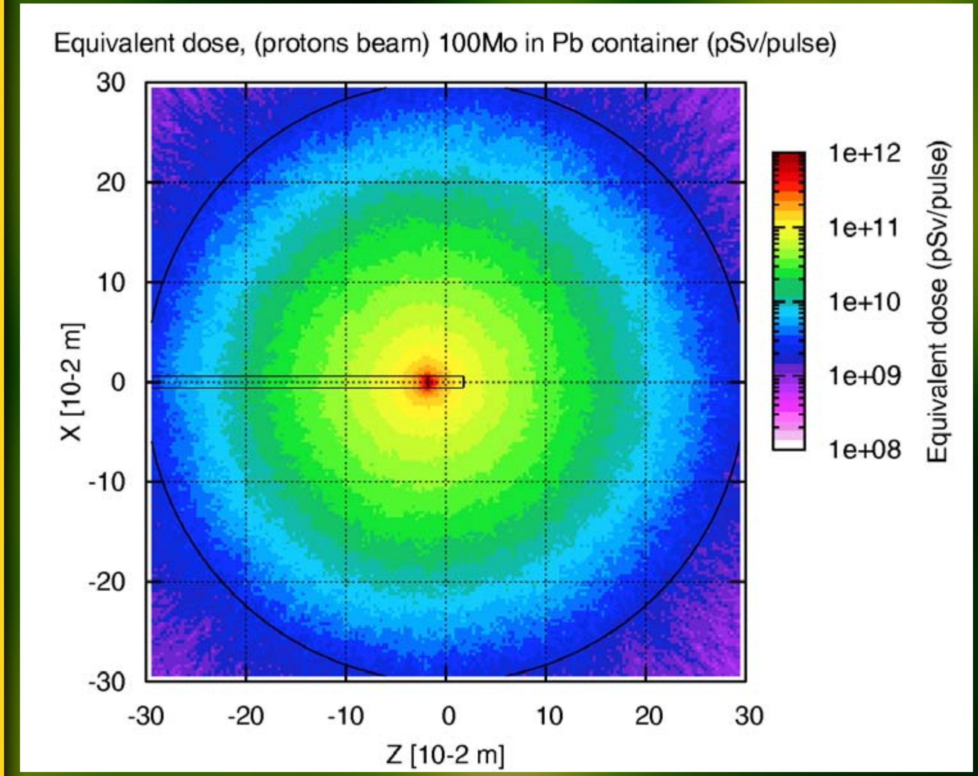
14.8 MeV (γ,n)


Deposited energy, (protons beam) in 100Mo in Pb container (J/pulse/m3)

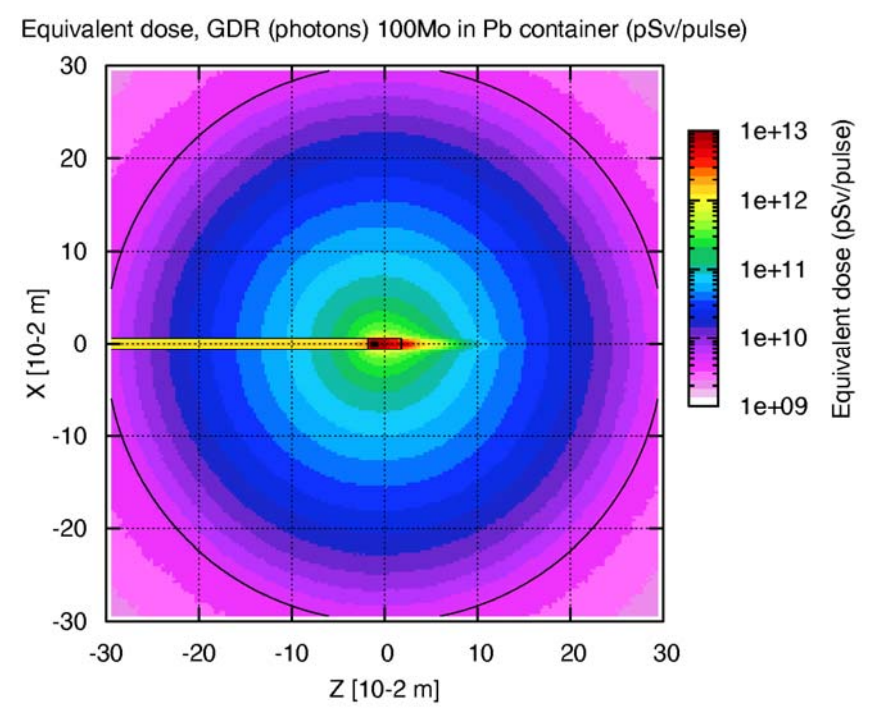

Deposited energy, GDR (photons) in 100Mo in Pb container (J/pulse/m3)

Natural Mo: 9.63% ^{100}Mo, 24.13% ^{98}Mo

FLUKA; Proton versus photon ^{100}Mo transmutation; Equivalent dose



(p,2n)



(γ,n)

W_n: 10 (2-20 MeV), 5 (above) and 20 (below this energy)

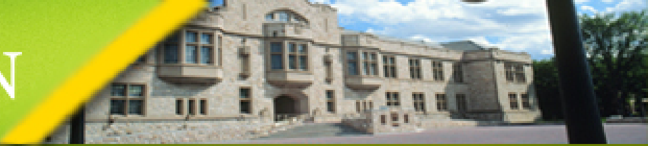
FLUKA; Proton versus photon ^{100}Mo transmutation

Produced Isotope (reaction)	Yield [per one proton/cm^3] ^{100}Mo target Pb container	Error [%]
(p,n) ^{100}Tc	5.44×10^{-4}	2.9
(p,2n) ^{99}Tc	**4.14×10^{-3}** (~ **2×10^{-3} ^{99m}Tc**)	**1**
(p,3n) ^{98}Tc	7.23×10^{-4}	1.6
(n,γ) ^{101}Mo#	1.00×10^{-6}	77.5
(p,p) ^{100}Mo	2.09×10^{-4}	1.1
(p,n&p) ^{99}Mo	9.42×10^{-h5}	4.8

Produced Isotope (reaction)	Yield [per one photon/cm^3] ^{100}Mo target Pb container	Error [%]
(n,γ) ^{101}Mo#	3.65×10^{-6}	1.2
(γ,e^+e^-)$_{atomic}$ ^{100}Mo	7.85×10^{-5}	0.4
(γ,n) ^{99} Mo	**1.31×10^{-2}**	**0.03**
(γ,2n) ^{98}Mo	6.06×10^{-3}	0.1
(γ,3n) ^{97}Mo	9.27×10^{-5}	0.6
(γ,4n) ^{96}Mo	1.04×10^{-4}	0.3

#Secondary neutron capture; * ^{98}Mo

Natural Mo: 9.63% ^{100}Mo, 24.13% ^{98}Mo



FLUKA; Proton versus photon ^{100}Mo transmutation

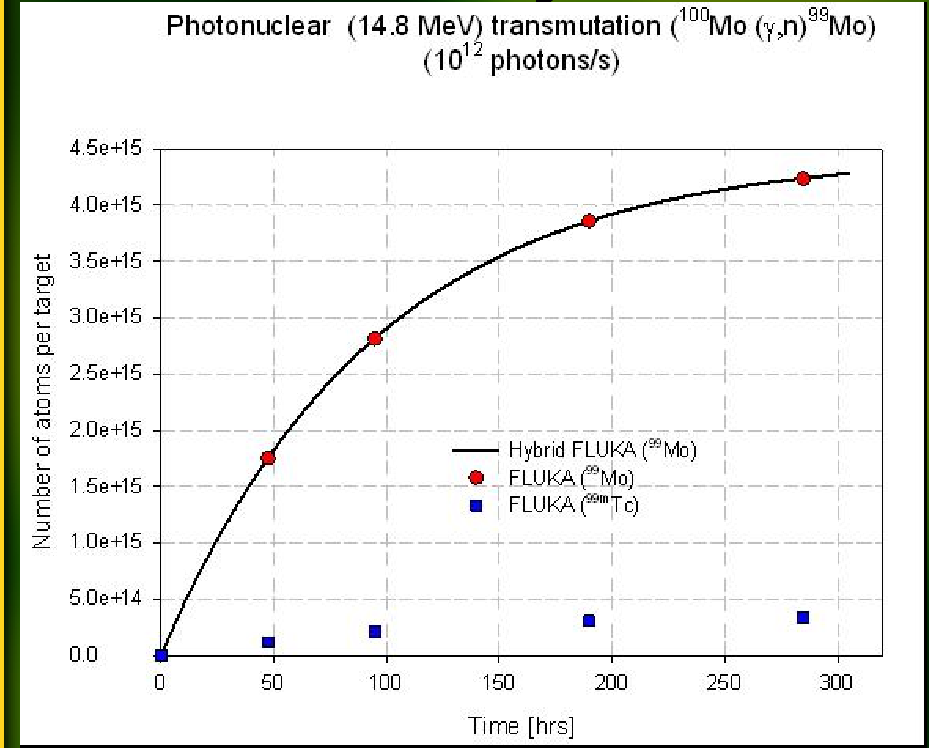
Produced Isotope (reaction)	Yield [per one photon/cm^3] Natural Mo target Be container	Error [%]
$(n,\gamma)\ ^{101}Mo^{\#}$	4.29×10^{-6}	0.9
$(\gamma,e^+e^-)_{atomic}\ ^{100}Mo$	6.32×10^{-5}	0.4
$(\gamma,n)\&(n,\gamma)\#\ ^{99}Mo$	**1.27×10^{-3}**	**0.01**
$(\gamma,2n)\&(\gamma,\gamma)^*\ ^{98}Mo$	7.07×10^{-4}	0.2
$(\gamma,3n)\&(\gamma,n)^*\ ^{97}Mo$	4.71×10^{-3}	0.1
$(\gamma,4n)\&(\gamma,2n)^*\ ^{96}Mo$	2.15×10^{-3}	0.1
$(\gamma,5n)\&(\gamma,3n)^*\ ^{95}Mo$	3.18×10^{-3}	0.1

Produced Isotope (reaction)	Yield [per one photon/cm^3] ^{100}Mo target Pb container	Error [%]
$(n,\gamma)\ ^{101}Mo^{\#}$	3.65×10^{-6}	1.2
$(\gamma,e^+e^-)_{atomic}\ ^{100}Mo$	7.85×10^{-5}	0.4
$(\gamma,n)\ ^{99}Mo$	**1.31×10^{-2}**	**0.03**
$(\gamma,2n)\ ^{98}Mo$	6.06×10^{-3}	0.1
$(\gamma,3n)\ ^{97}Mo$	9.27×10^{-5}	0.6
$(\gamma,4n)\ ^{96}Mo$	1.04×10^{-4}	0.3

#Secondary neutron capture; * ^{98}Mo

Natural Mo: 9.63% ^{100}Mo, 24.13% ^{98}Mo

FLUKA hybrid versus FLUKA calculations


Photonuclear (14.8 MeV) transmutation (^{100}Mo (γ,n) ^{99}Mo) (10^{12} photons/s)

6 x 10^{21} atoms in 1g of Mo

NRU produces (in reactor) per year :
~3.65 x 10^{22} ^{99}Mo atoms
~24 g Mo with 25% ^{99}Mo

6 g of ^{99}Mo: 2,887,993 Ci (decay) -> ~ 60,000 Ci

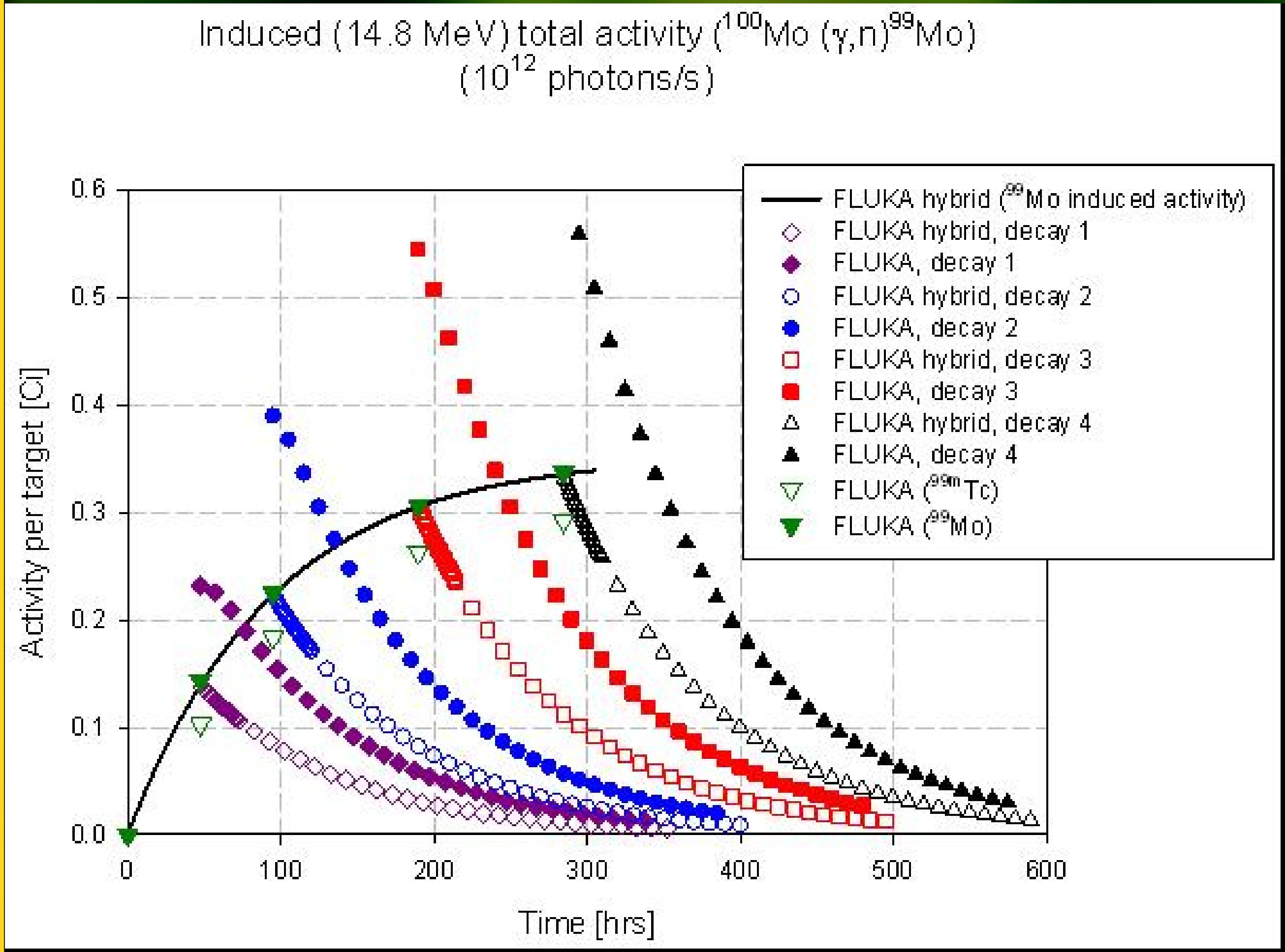
EPAC 2000 Vienna, used per year: 150,000 Ci

← $N(t) = Activity_g(t)/Activity_{atom}$

Target's weight: 41.6 g ↑

Target	Irradiation	^{99}Mo	^{99}Mo	^{99m}Tc	^{99m}T
^{100}Mo	Time [hrs]	Activity [Ci]	Specific Activity [Ci/g]	Activity [Ci]	Specific Activity [Ci/g]
^{99}Mo, $T_{1/2}$	284.67	0.337	0.008	0.296	0.007
65.94 h	189.78	0.307	0.007	0.266	0.006
^{99m}Tc, $T_{1/2}$	94.89	0.225	0.005	0.184	0.004
6.0058 h	47.44	0.143	0.003	0.102	0.002

FLUKA; Photon induced activity



^{99}Mo

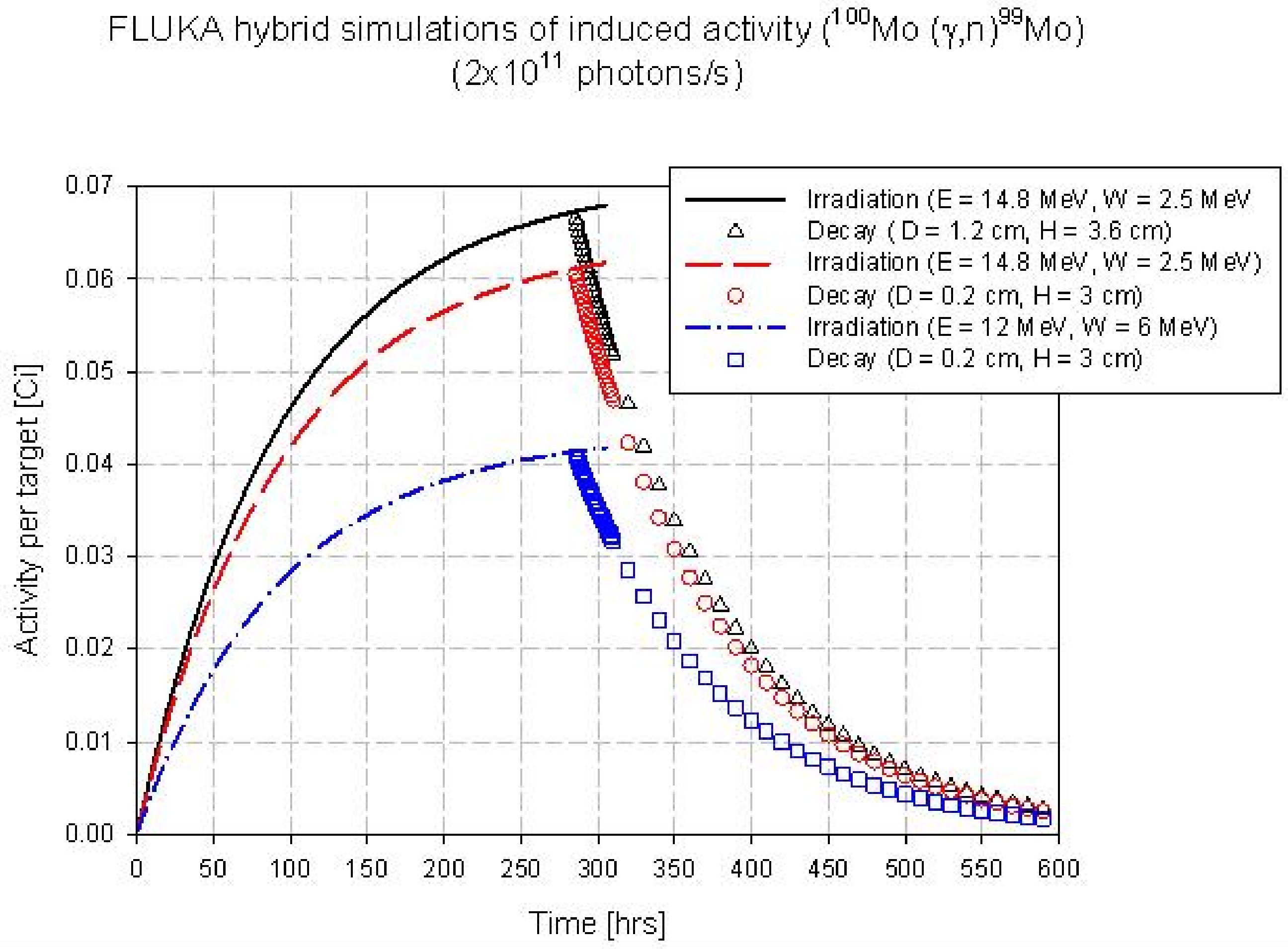
$T_{1/2}$ = 65.94 h

^{99m}Tc

$T_{1/2}$ = 6.0058 h

Target's weight:
41.6 g

FLUKA; Photon induced activity (Parameters by H. Ejiri, S. Date et al.)



^{99}Mo

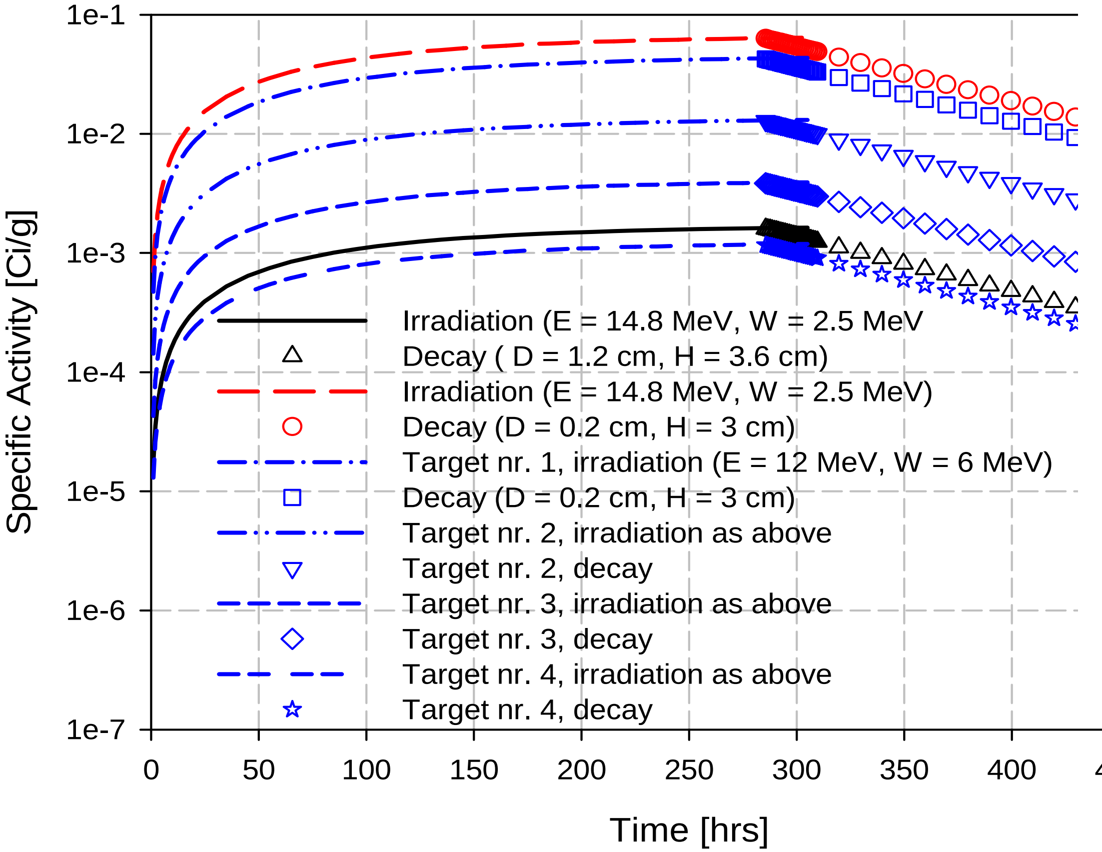
$T_{1/2} = 65.94$ h

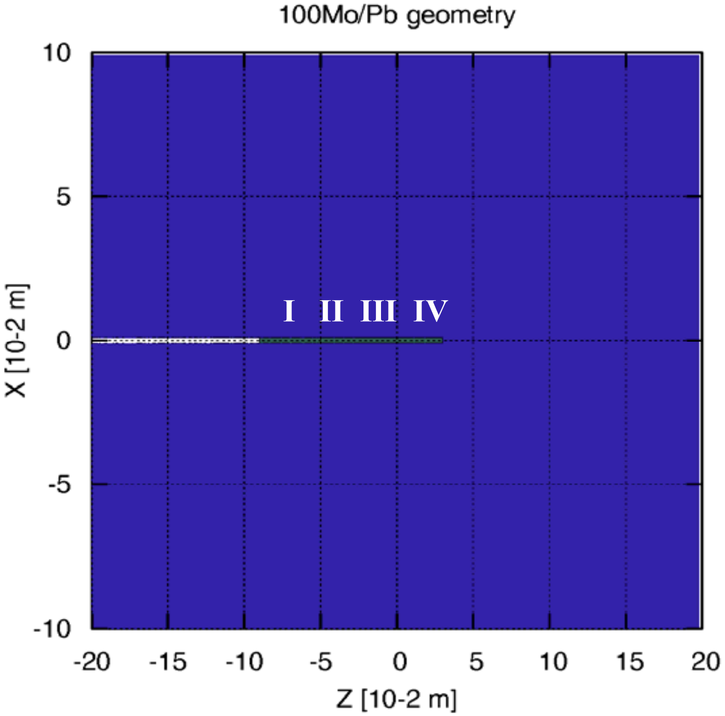
^{99m}Tc

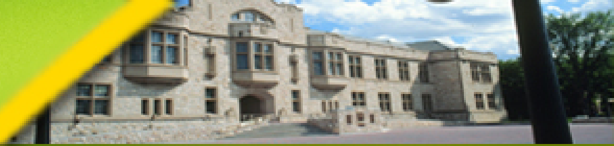
$T_{1/2} = 6.0058$ h

FLUKA; Photon induced activity (multiple targets by H. Ejiri)

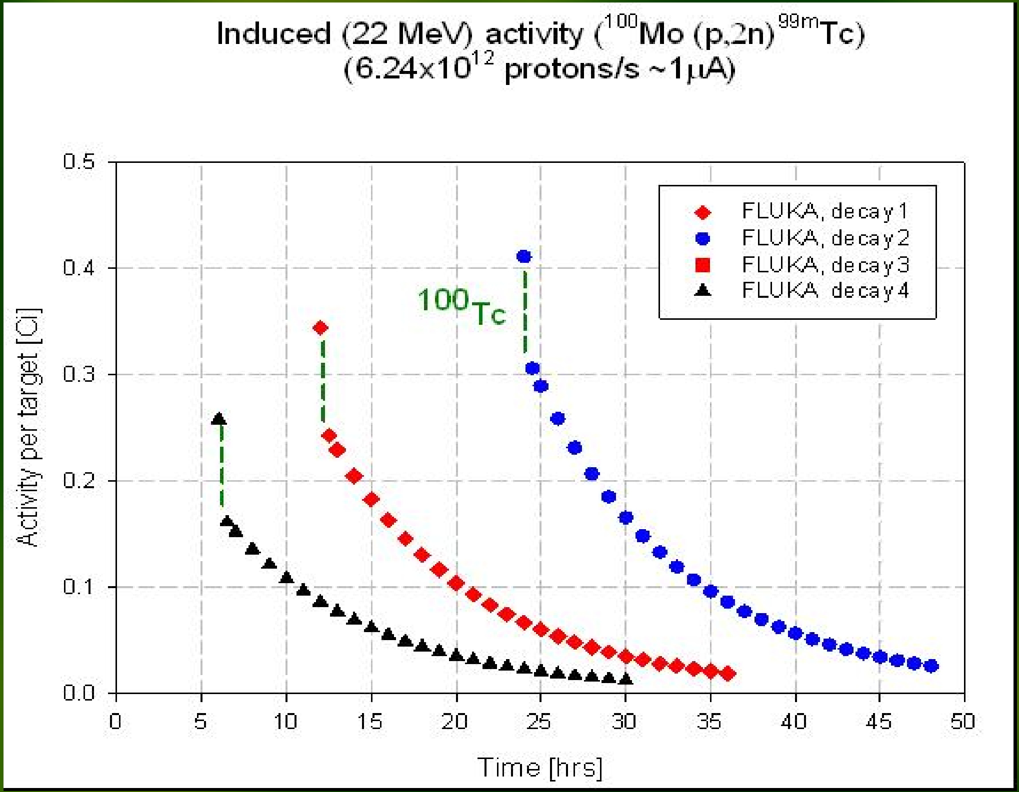
FLUKA hybrid simulations of induced activity (^{100}Mo (γ,n)^{99}Mo) (2×10^{11} photons/s)







FLUKA; Proton induced activity



Target	Irradiation	^{99m}Tc	^{99m}T	^{100}Tc	^{100}Tc
^{100}Mo	Time [hrs]	Activity [Ci]	Specific Activity [Ci/g]	Activity [Ci]	Specific Activity [Ci/g]
^{100}Tc, $T_{1/2}$ 15.46 s	24	0.317	0.008	0.087	0.002
	12	0.256	0.006	0.087	0.002
	6	0.174	0.004	0.087	0.002

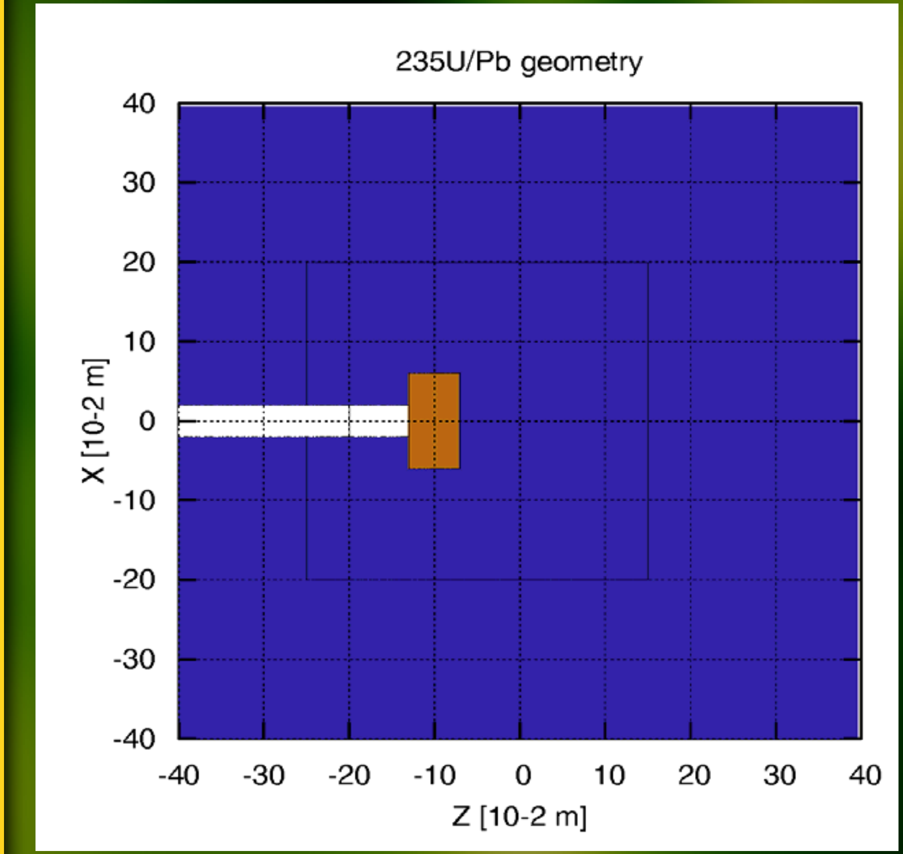
Photofission versus GDR transmutation

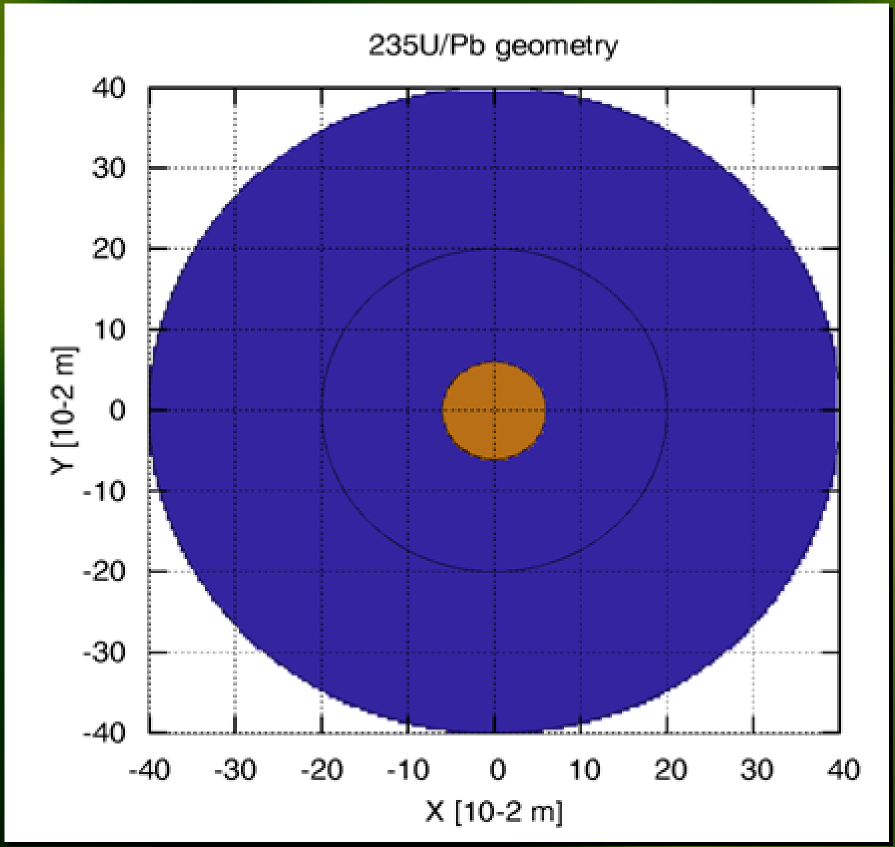
Photons (16 MeV)	**Fission yield per one photon [Nuclei/cm^3]** (May be heavily underestimated by the FLUKA currently!)							**β$^-$ yield**
Target	**^{99}Mo(42)**	^{99}Kr(36)	^{99}Rb(37)	^{99}Sr (38)	^{99}Y(39)	^{99}Zr(40)	^{99}Nb(41)	**^{99}Mo(42)**
^{238}U	**1.17x10^{-7}**	8.32x10^{-8}	3.94x10^{-6}	1.83x10^{-5}	6.47x10^{-5}	3.46x10^{-5}	6.15x10^{-6}	**1.28x10^{-4}**
Errors (%)	**20.2**	20.5	2.1	1.1	0.9	0.7	2.3	

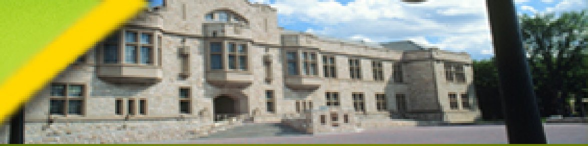
Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) ^{101}Mo#	3.65 x10^{-06}	1.2
$(\gamma,e^+e^-)_{atomic}$ ^{100}Mo	7.85 x10^{-05}	0.4
(γ,n) ^{99} Mo	**1.31 x10^{-02}**	**0.03**
$(\gamma,2n)$ ^{98}Mo	6.06 x10^{-03}	0.1

#Secondary neutron capture

Fission in FLUKA (geometry, target: 12 cm (d) x 6 cm (h))

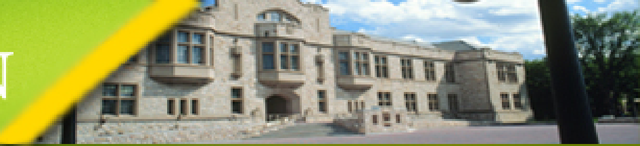






^{99}Mo production in photofission

Photons (12.5 MeV)	Subcritical fission yield per one particle [nuclei/cm^{3}] **May be heavily underestimated by FLUKA currently!**							**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	**$2.75x10^{-8}$**	$3.88x10^{-8}$	$1.74x10^{-6}$	$1.0x10^{-5}$	$3.65x10^{-5}$	$1.60x10^{-5}$	$1.62x10^{-6}$	**$6.63x10^{-5}$**
Errors (%)	**36.8**	15.0	2.9	1.6	0.6	0.7	6.5	
^{235}U	**$1.35x10^{-5}$**	$7.86x10^{-8}$	$4.40x10^{-6}$	$2.13x10^{-4}$	$2.23x10^{-3}$	$3.43x10^{-3}$	$3.08x10^{-4}$	**$6.19x10^{-3}$**
Errors (%)	**1.3**	22.2	1.3	0.3	0.2	0.1	0.2	
^{234}U	**$5.54x10^{-6}$**	$2.38x10^{-8}$	$9.83x10^{-7}$	$1.45x10^{-5}$	$2.39x10^{-4}$	$7.51x10^{-4}$	$1.51x10^{-4}$	**$1.16x10^{-3}$**
Errors (%)	**1.4**	40.8	4.6	1.1	0.2	0.2	0.2	
^{232}Th	**$4.00x10^{-0}$**	$3.92x10^{-9}$	$1.03x10^{-7}$	$6.62x10^{-7}$	$2.85x10^{-6}$	$1.52x10^{-6}$	$1.93x10^{-7}$	**$5.33x10^{-6}$**
Errors (%)	**99.0**	99.0	23.1	7.8	2.4	1.7	11.5	



^{99}Mo production comparison

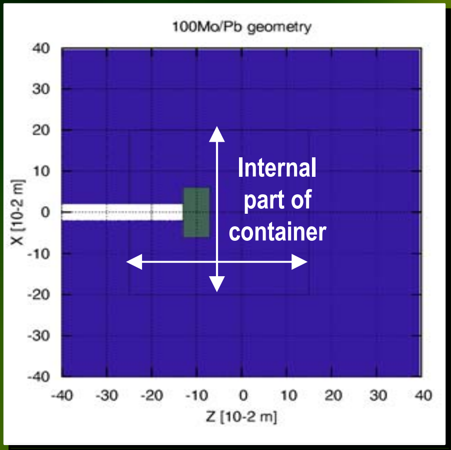
Target 0.12 m diameter and 0.06 m height

Thermal neutrons	**Fission (subcritical) yield per one neutron [nuclei/cm^3]**						**β$^-$ yield***	**Total**
Target	**^{99}Mo(42)**	^{99}Rb(37)	^{99}Sr (38)	^{99}Y(39)	^{99}Zr(40)	^{99}Nb(41)	**^{99}Mo(42)**	**^{99}Mo(42)**
^{235}U	**7.01 x10^{-4}**	3.40 x10^{-6}	0.00728	0.0854	0.139	0.0118	**0.2435**	**0.244**
Errors (%)	**1.2**	17.6	0.5	0.2	0.3	0.5		

GDR →

Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) ^{101}Mo$^{\#}$	6.88 x10^{-5}	0.2
(γ,e$^+$e$^-$)$_{atomic}$ ^{100}Mo	7.69 x10^{-4}	0.0
(γ,n) ^{99} Mo	**1.58 x10^{-2}**	**0.0**
(γ,2n) ^{98}Mo	8.47 x10^{-3}	0.1
(γ,3n) ^{97}Mo	9.71 x10^{-4}	0.0
(γ,4n) ^{96}Mo	1.11 x10^{-3}	0.1
(γ,5n) ^{95}Mo	1.75 x10^{-3}	0.1

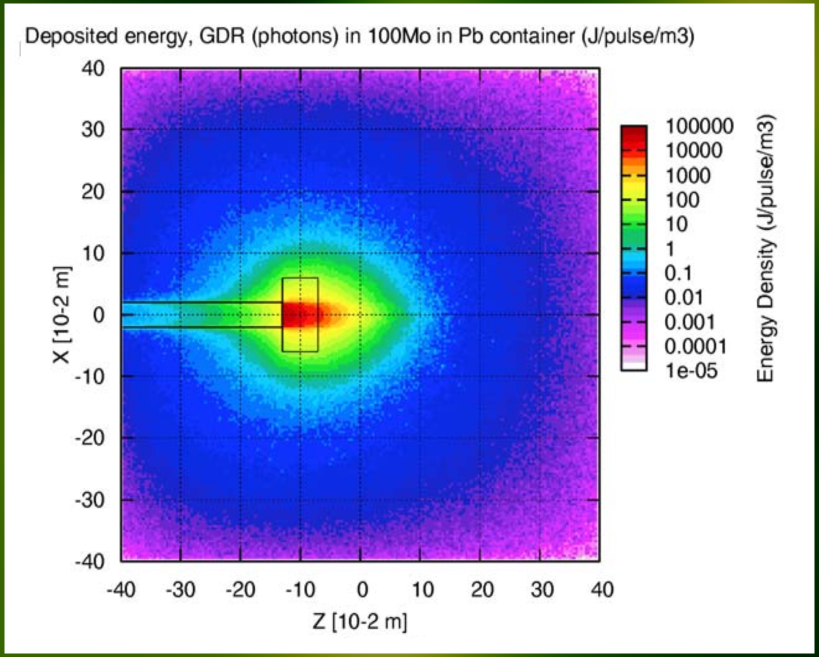
Energy deposition in ^{99}Mo production



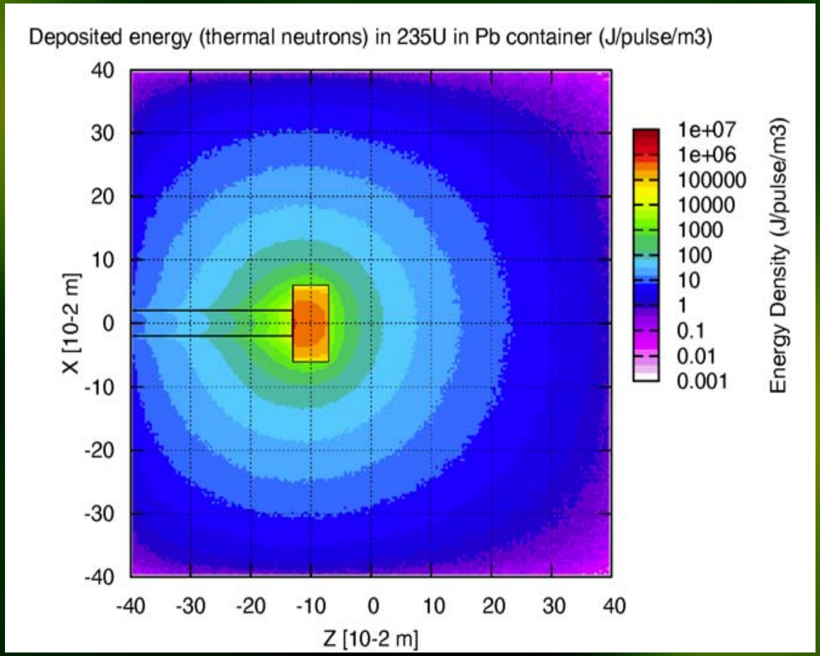
Thermal neutrons (0.025 eV)	Average Energy	Error	Standard Deviation
^{235}U Target	GeV/particle	[%]	GeV/particle
Target	0.7330	0.32	0.0023
Internal part of the lead container	0.0131	0.23	3.0509×10^{-5}
External part of the lead container	0.0036	0.29	1.0186×10^{-5}

GDR (0.0148 MeV)	Average Energy	Error	Standard Deviation
^{100}Mo Target	GeV/particle	[%]	GeV/particle
Target	0.01220	0.04	5.4589×10^{-6}
Internal part of the lead container	0.0002	0.19	4.5671×10^{-6}
External part of the lead container	1.29×10^{-5}	0.42	5.4807×10^{-8}

GDR, photofission versus thermal neutrons fission (subcritical); Energy deposition

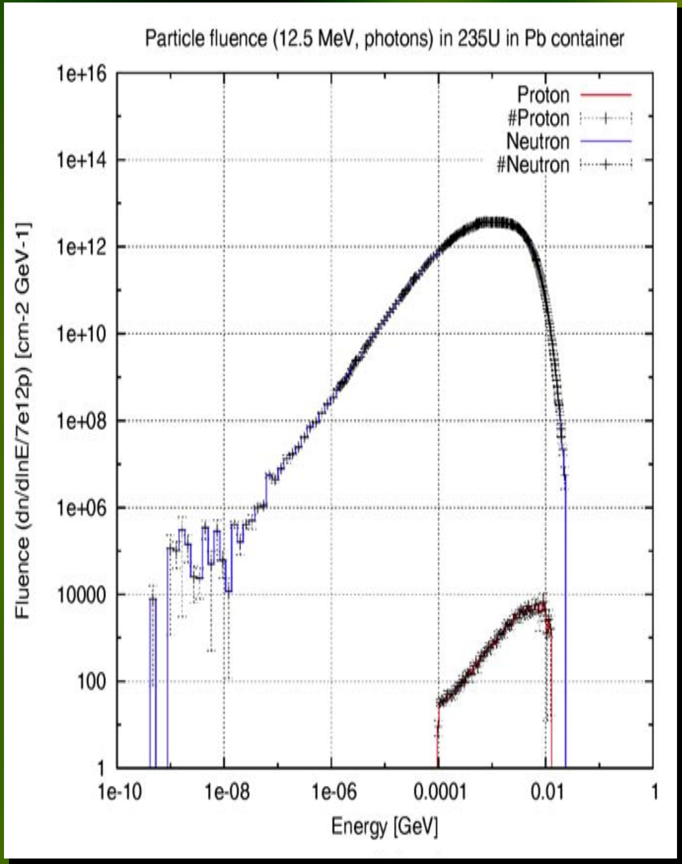


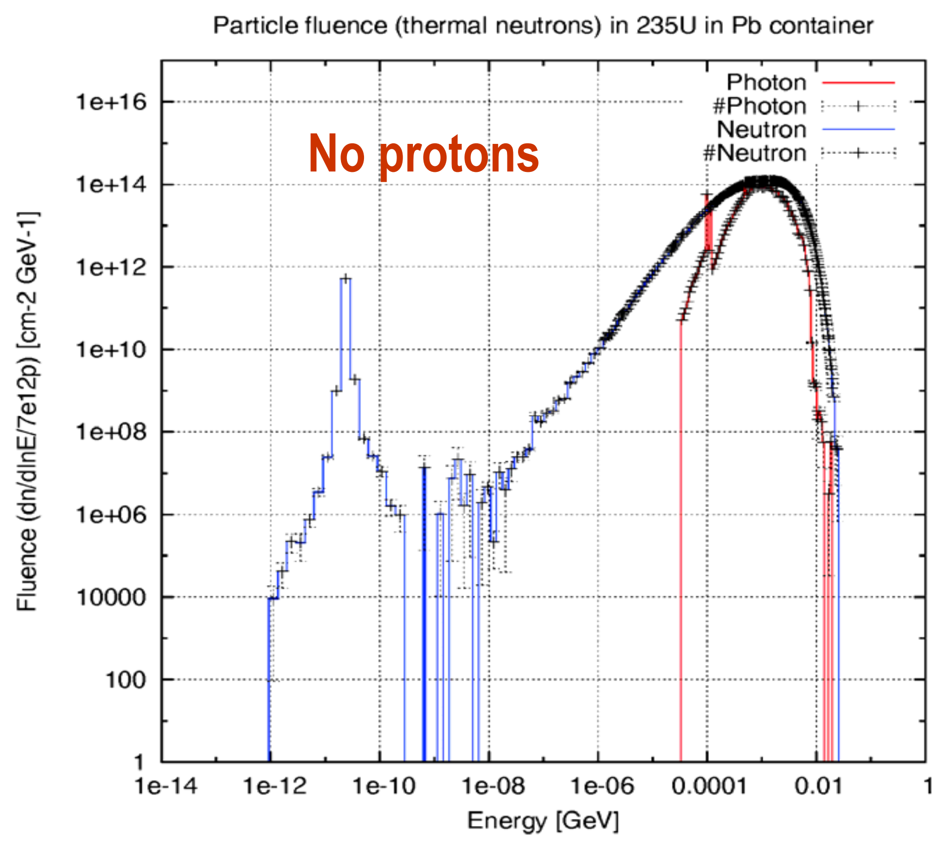
Deposited energy, GDR (photons) in 100Mo in Pb container (J/pulse/m3)



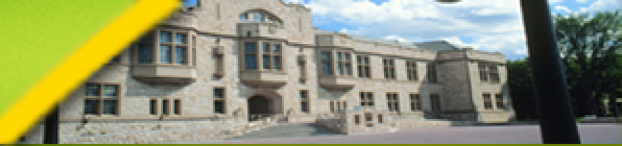
Deposited energy (thermal neutrons) in 235U in Pb container (J/pulse/m3)

Photofission versus thermal neutrons fission (subcritical); Fluence


Particle fluence (12.5 MeV, photons) in 235U in Pb container


Particle fluence (thermal neutrons) in 235U in Pb container

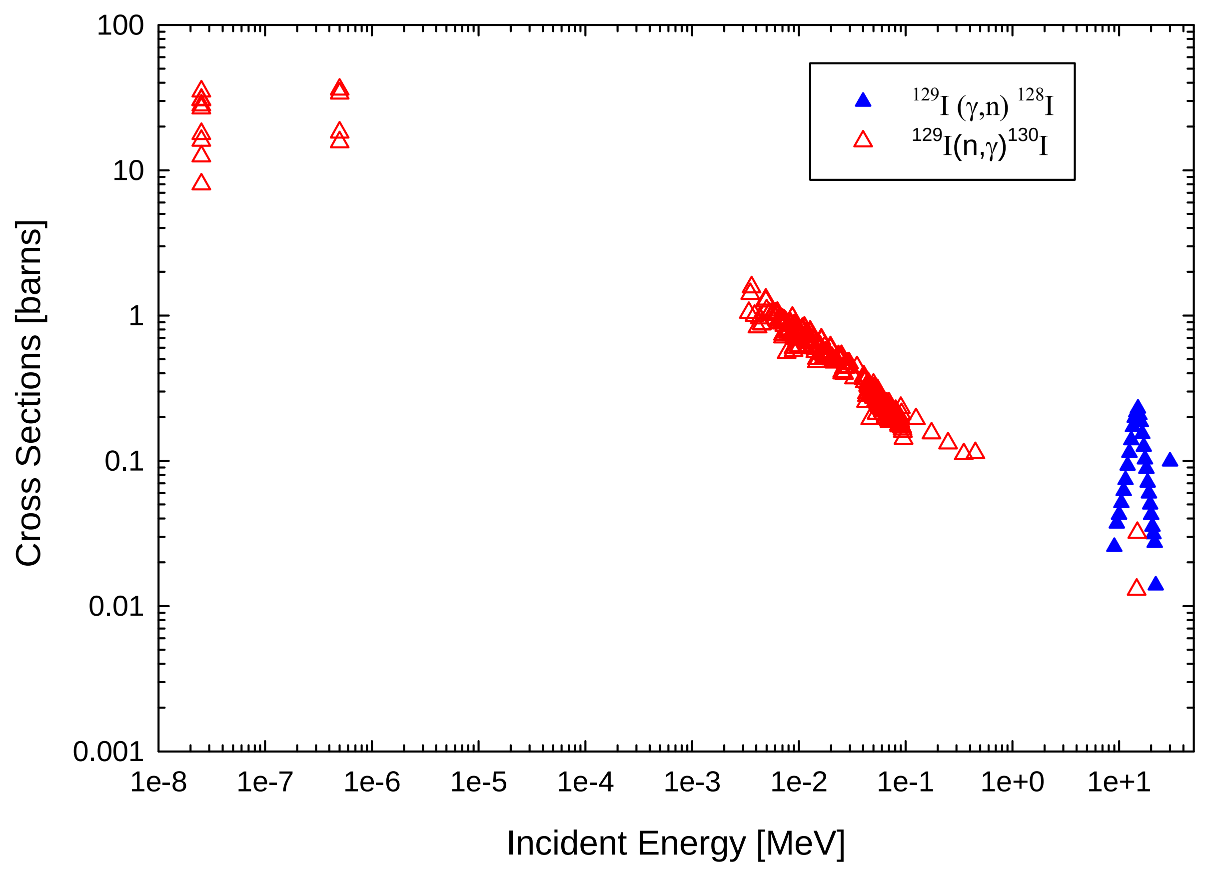
No protons



Waste management

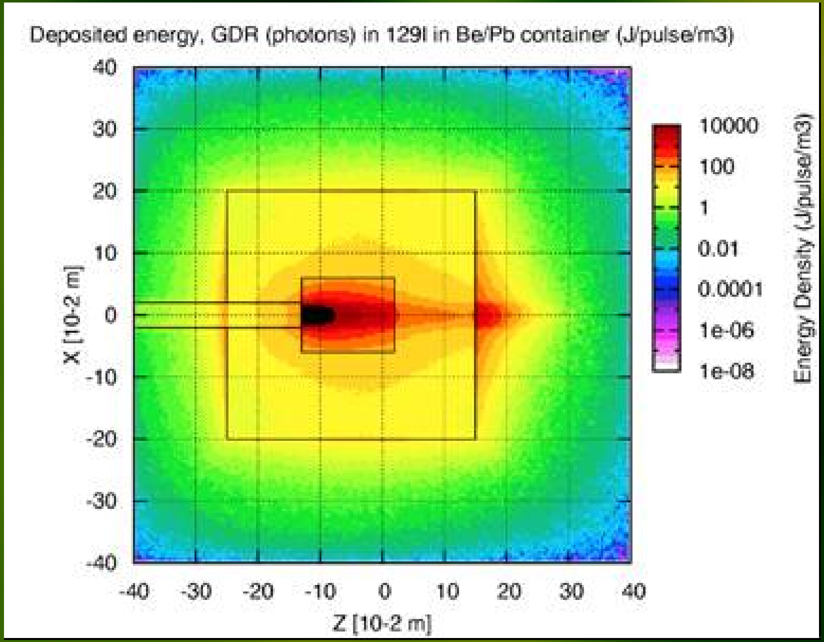
- Nuclear waste consists of 0.74% fission products and 99.26% actinides, with 98.81% uranium and **0.45% long-lived transuranic actinides** (Ottensmeyer, CNS 2010)
- Treatment of long-lived isotopes: ^{79}Se, ^{93}Zr, ^{107}Pd, ^{126}Sn, ^{129}I and ^{135}Cs; radio-toxic >10^5 years needed
- GDR transmutation to short-lived isotopes (^{99}Tc is not transformed to short-lived isotope)
  - 1.57 x10^7 years half-life time of ^{129}I to 24.99 m ^{128}I or 12.36 h ^{130}I (secondary neutron capture)

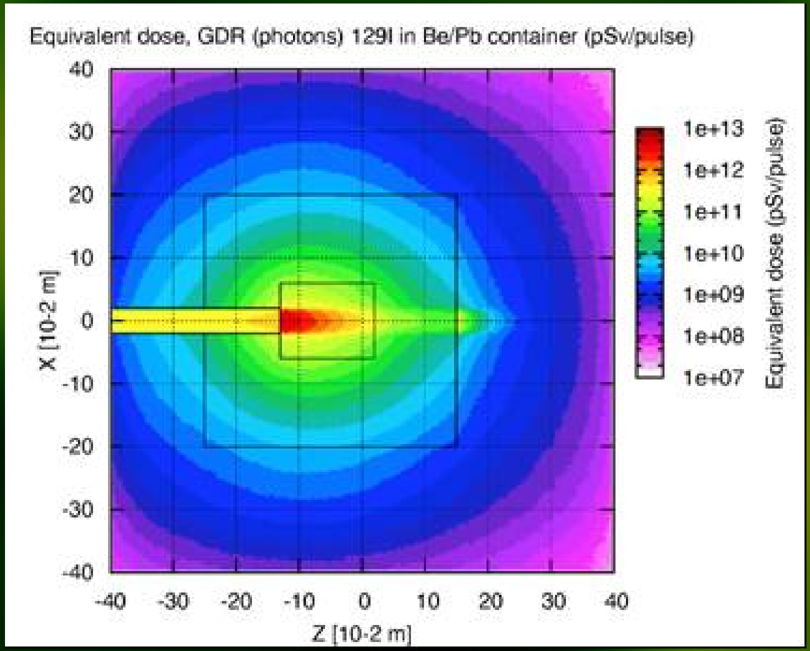
Photons versus neutrons



http://www.nndc.bnl.gov

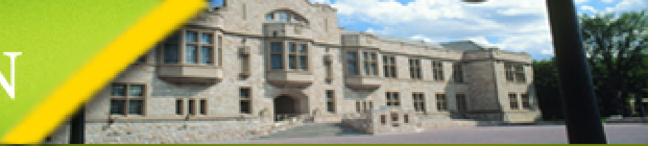
Long lived waste transmutation; energy deposition & equivalent dose





Wn: 10 (2-20 MeV), 5 (above),

20 (below this energy)



^{129}I transmutation by GDR (15.24 MeV)

Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) $^{130}I^{\#}$	9.22×10^{-03}	0.4
$(\gamma,e^+e^-)_{atomic}$ ^{129}I	6.15×10^{-03}	0.4
(γ,n) ^{128}I	**2.88×10^{-02}**	**0.3**
$(\gamma,2n)$ ^{127}I	9.88×10^{-04}	1.2
(γ,p) ^{128}Te	3.20×10^{-06}	33.4

Be container

Be/Pb container

Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) $^{130}I^{\#}$	7.91×10^{-03}	0.8
$(\gamma,e^+e^-)_{atomic}$ ^{129}I	6.17×10^{-03}	0.9
(γ,n) ^{128}I	**2.90×10^{-02}**	**0.2**
$(\gamma,2n)$ ^{127}I	9.87×10^{-04}	1.3
(γ,p) ^{128}Te	5.60×10^{-06}	26.2

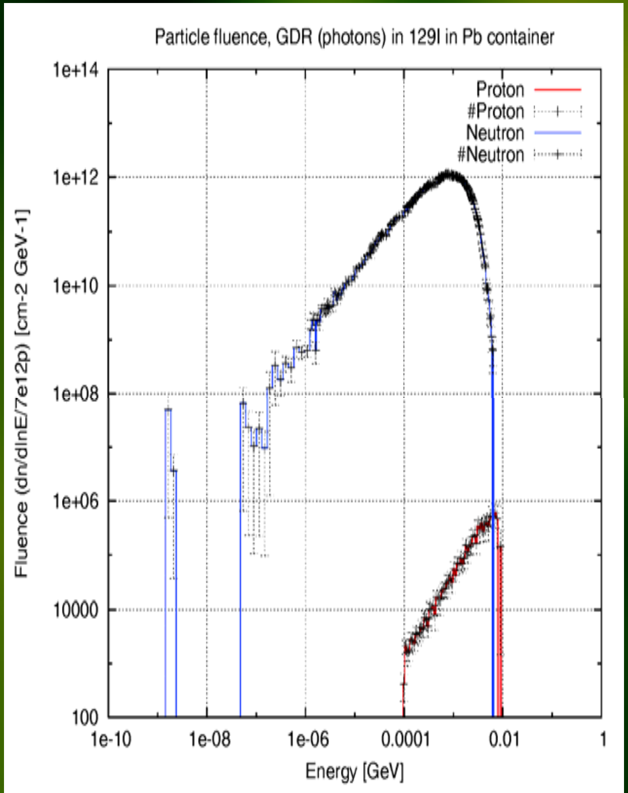
Pb container

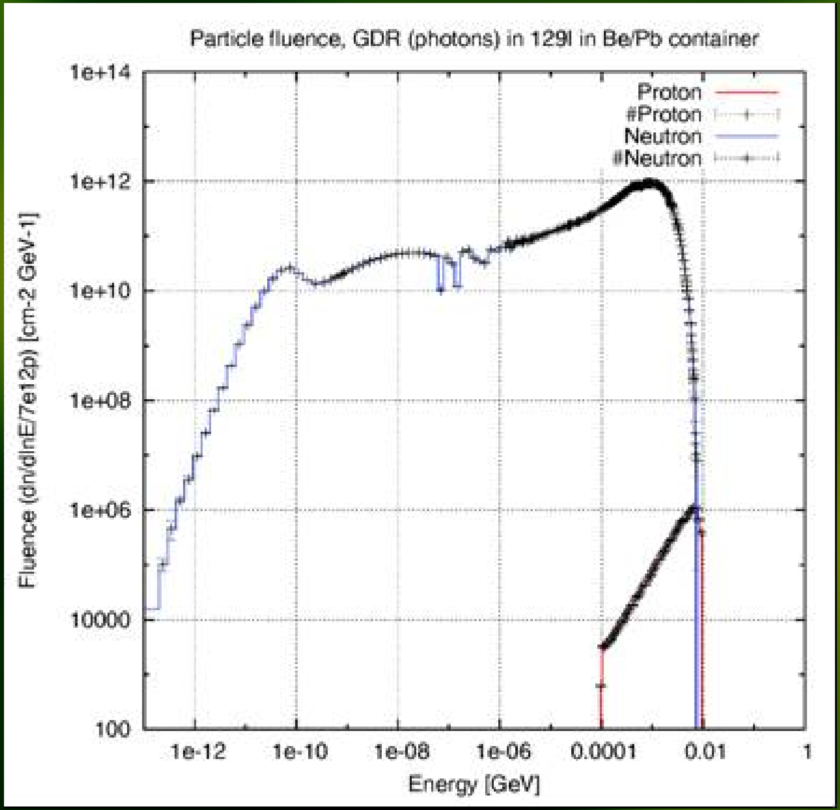
Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) $^{130}I^{\#}$	1.08×10^{-03}	0.8
$(\gamma,e^+e^-)_{atomic}$ ^{129}I	6.81×10^{-03}	0.5
(γ,n) ^{128}I	**2.88×10^{-02}**	**0.1**
$(\gamma,2n)$ ^{127}I	1.00×10^{-03}	1.7
(γ,p) ^{128}Te	2.80×10^{-06}	30.7

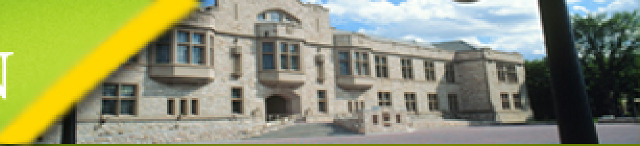
#Secondary neutron capture

1.57×10^7 years half-life time of $^{129}I \longrightarrow$ 24.99 min ^{128}I or 12.36 h ^{130}I (secondary neutron capture)

Long lived waste transmutation; fluence



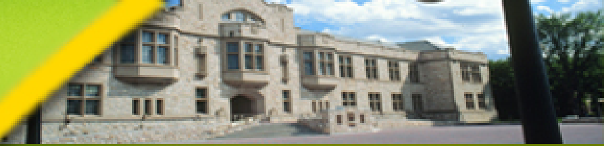




Summary and conclusion

- 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
  - Transmutation of long lived isotopes to short lived
  - Induced transmutation & photofission as a source of neutrons

Comment: Currently γ beams have too low intensity for some applications. While it is expected that the intensity will be increased sufficiently for a production of medical isotopes, the total transmutation of nuclear waste requires intensities that will be probably not possible to achieve in the near future.



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