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

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Collaboration with Prof. Chary Rangacharyulu

Available on:

http://www.yamadazaidan.jp/ys/apse2010/apse2010-18/18-12-25-Szpunar.pdf

October 7th, 2010, Eucera

1st Advanced FLUKA Workshop

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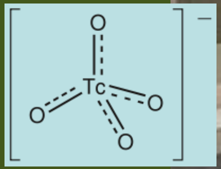
Purpose of this work and introduction

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

^{99m}Tc -> $^{99}Tc + \gamma$

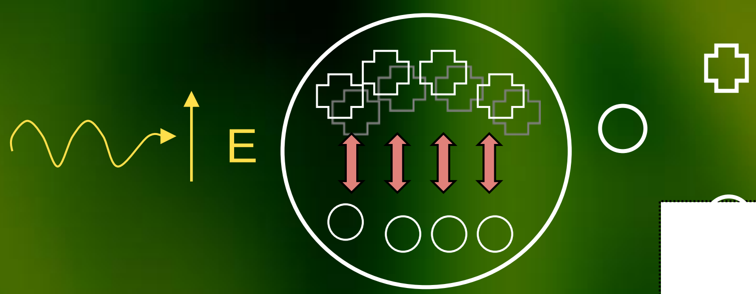
Each diagnostic uses few GBq

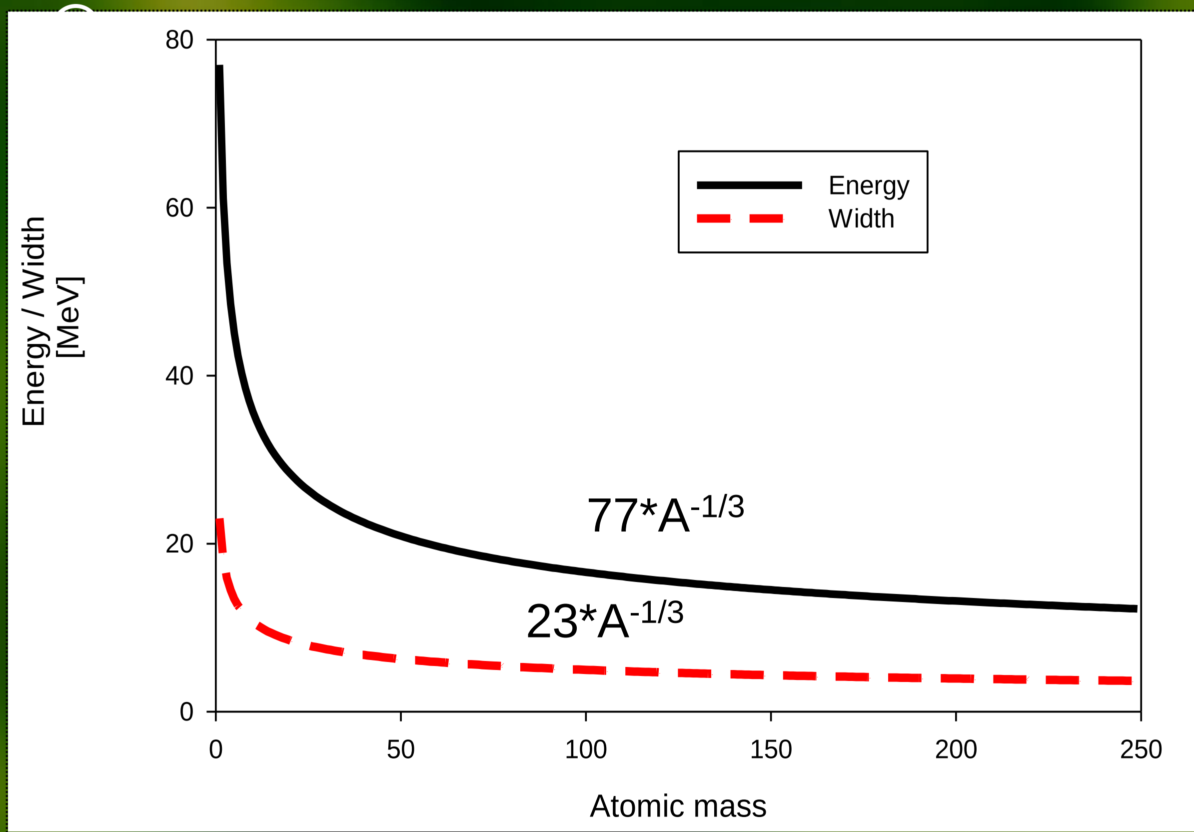
(1GBq = 0.027 Ci)



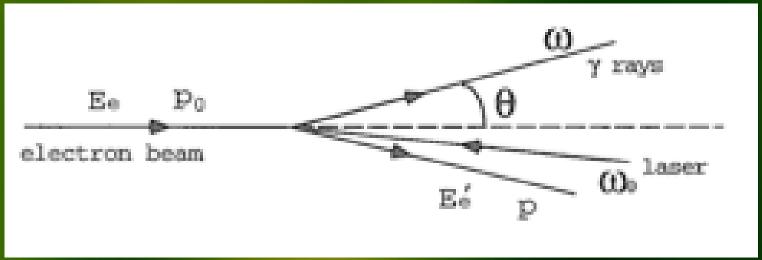
  - EPAC 2000 Vienna, world uses: 150,000 Ci/year
  - Proton cyclotrons located close to hospital can supply average usage: e.g. at CHUS, Sherbrooke 10 Ci/week of ^{99m}Tc
- ^{99m}Tc is short-lived ($T_{1/2}$ = 6.0058 h) therefore ^{99}Mo ($T_{1/2}$ = 65.94 h) needed for remote, small hospitals

^{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; input file

```
GLOBAL                                                1.0        1.0
DEFAULTS                                                                     PRECISIO
PHYSICS                1.                                                    COALESCE
PHYSICS                3.                                                    EVAPORAT
BEAM              -0.012       .006        0.0         .1        0.0      -1.0PHOTON
BEAMPOS              0.0        0.0      -20.1        0.0        0.0
GEOBEGIN                                                                     COMBNAME

…………..

GEOEND
PHOTONUC               5.        0.0                Mo100      Mo100
LAM-BIAS              0.0       0.02      Mo100    PHOTON                 0.0
LOW-MAT              LEAD        82.       208.      296.                    208-PB

…………..

USRBIN                10.     ENERGY       -50.       10.        10.       20.EneDep
USRBIN               -10.       -10.       -20.      200.       200.      200.&
RESNUCLE               3.       -61.                          TARGET       1.0Target
USRTRACK              -1.     PHOTON       -71.    TARGET        1.0      400.photon
USRTRACK               1.      1D-12                                         &
USRTRACK              -1.     PROTON       -71.    TARGET        1.0      400.proton
USRTRACK              1.0      1D-12                                         &
USRTRACK      -1.   NEUTRON    -71.   TARGET     1.0     400.neutron
USRTRACK      1.0    1.D-12                        &

………
```

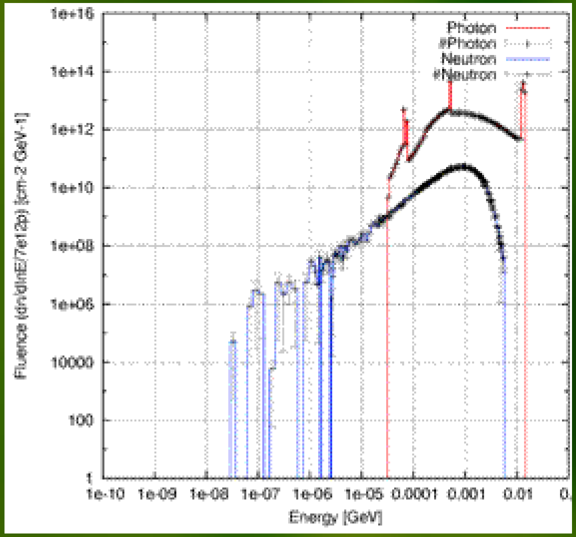
Photonuclear reaction; biasing

^{193}Ir of cylindrical shape (0.01 m diameter, 0.04 m height) in spherical lead container (0.3 m radius)

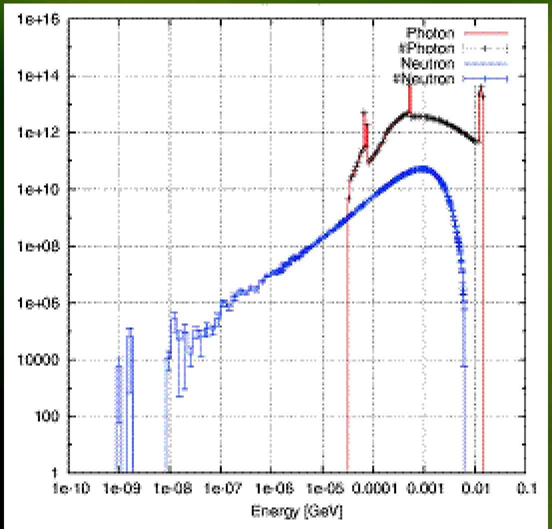
Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) ^{194}Ir	$1.36x10^{-4}$	3
(γ,e$^+$e$^-$)$_{atomic}$ ^{193}Ir	$1.28x10^{-3}$	0.8
(γ,n) ^{192}Ir	**$2.04x10^{-2}$**	**0.1**
(γ,2n) ^{191}Ir	$9.00x10^{-4}$	0.9

Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) ^{194}Ir	$1.33x10^{-4}$	0.2
(γ,e$^+$e$^-$)$_{atomic}$ ^{193}Ir	$1.28x10^{-3}$	0.1
(γ,n) ^{192}Ir	**$2.04x10^{-2}$**	**0.04**
(γ,2n) ^{191}Ir	$9.16x10^{-4}$	0.1
(γ,p) ^{192}Os	$1.97x10^{-8}$	42

No biasing



Hadronic interaction length reduced by a factor 0.02



Proceedings, CNS 31st Annual Conference, Montreal, May 24-27, 2010

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

$Activity_N(t) = N(t)\, Activity_{atom}$

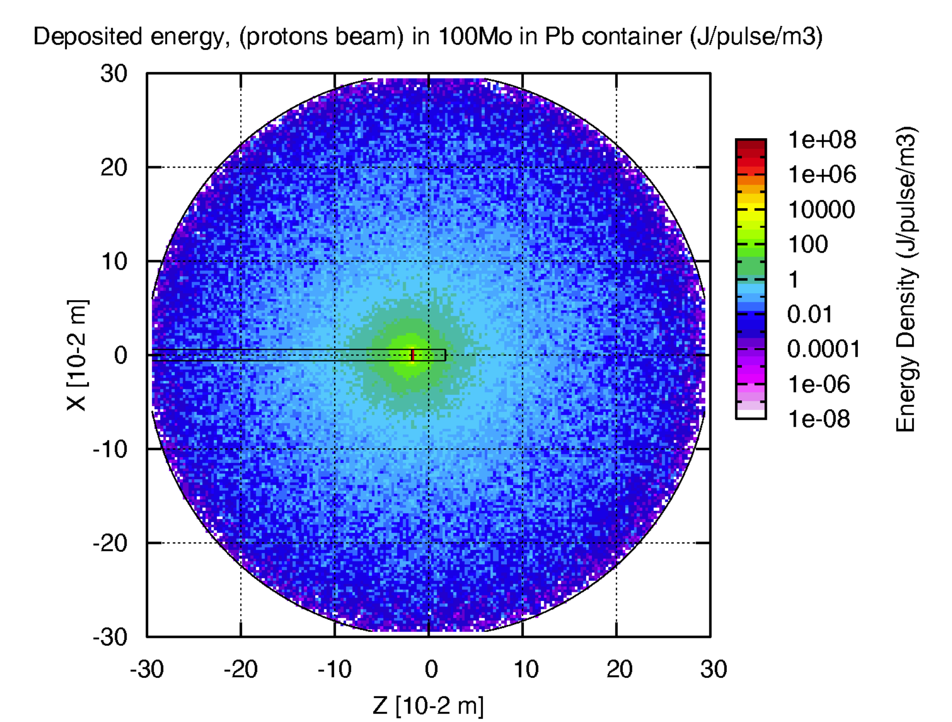
II: FLUKA induced activity simulations

Time dependence

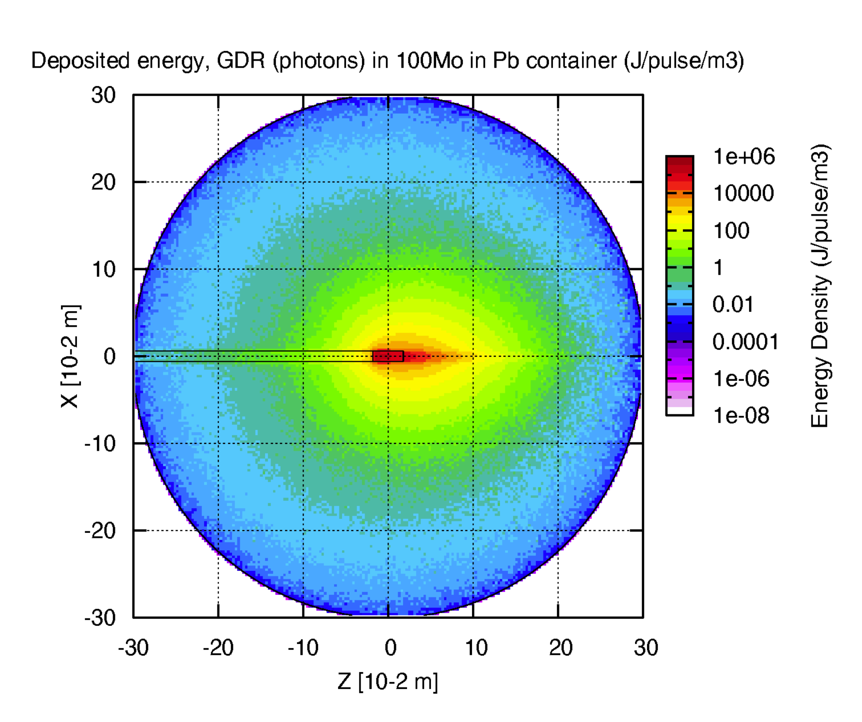
- Set irradiation and cooling time, beam intensity
- Perform five runs using FLUKA's exact analytical implementation of Bateman equations for induced activity calculations
  - Total activity
  - Yield
  - FLUKA ASCI files processed using Fortran routines with csv format to create directly EXCEL files for easy plotting

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

22 MeV (p,2n)

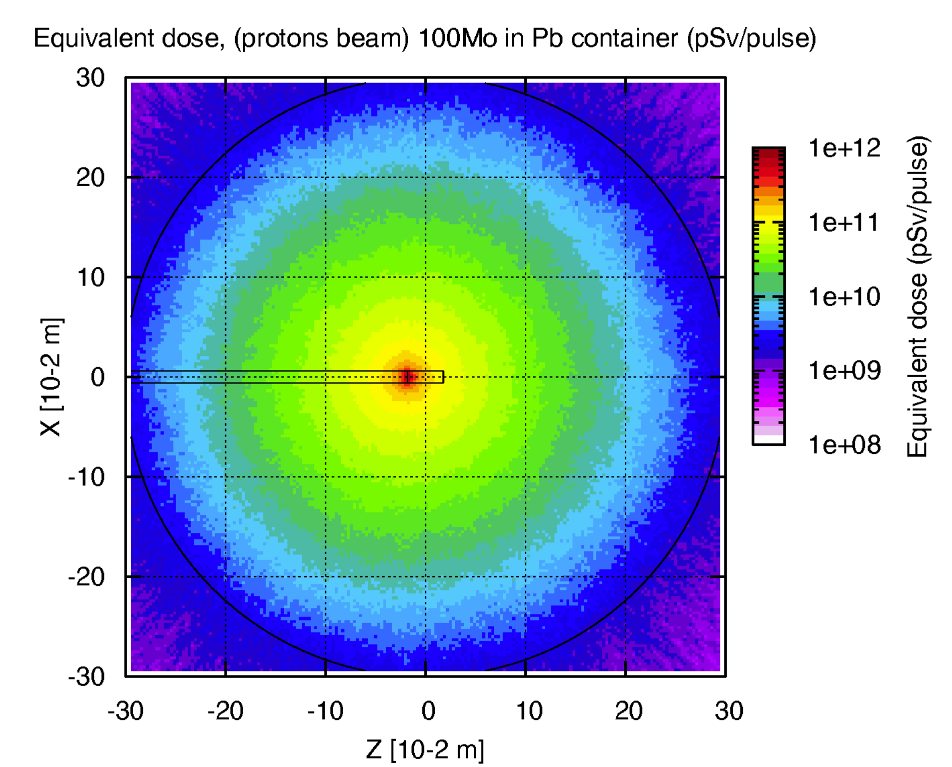


14.8 MeV (γ,n)

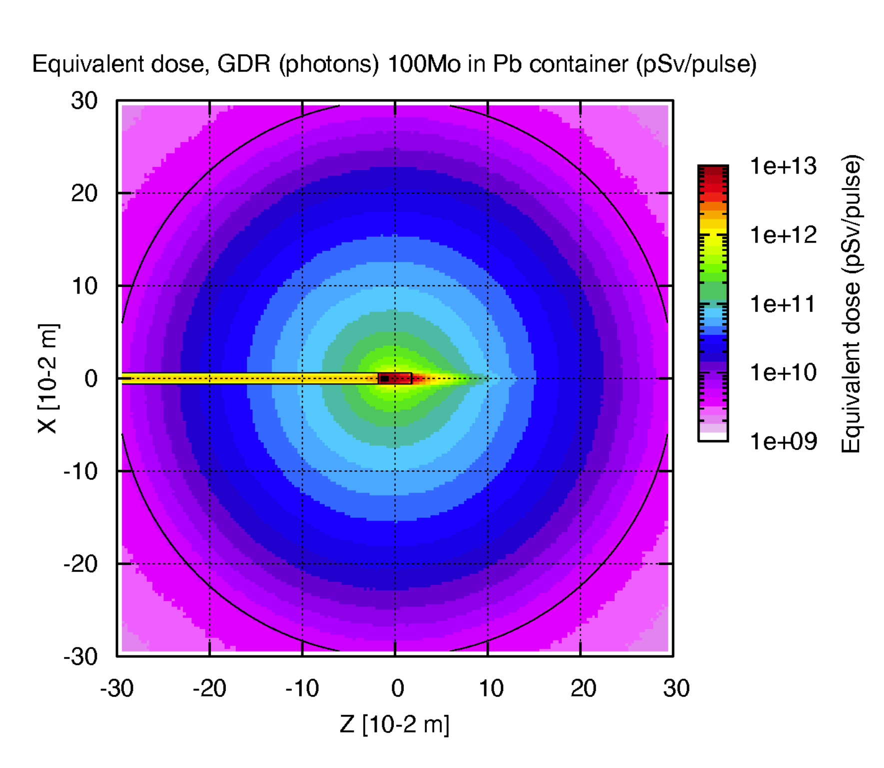


Natural Mo: 9.63% ^{100}Mo, 24.13% ^{98}Mo

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

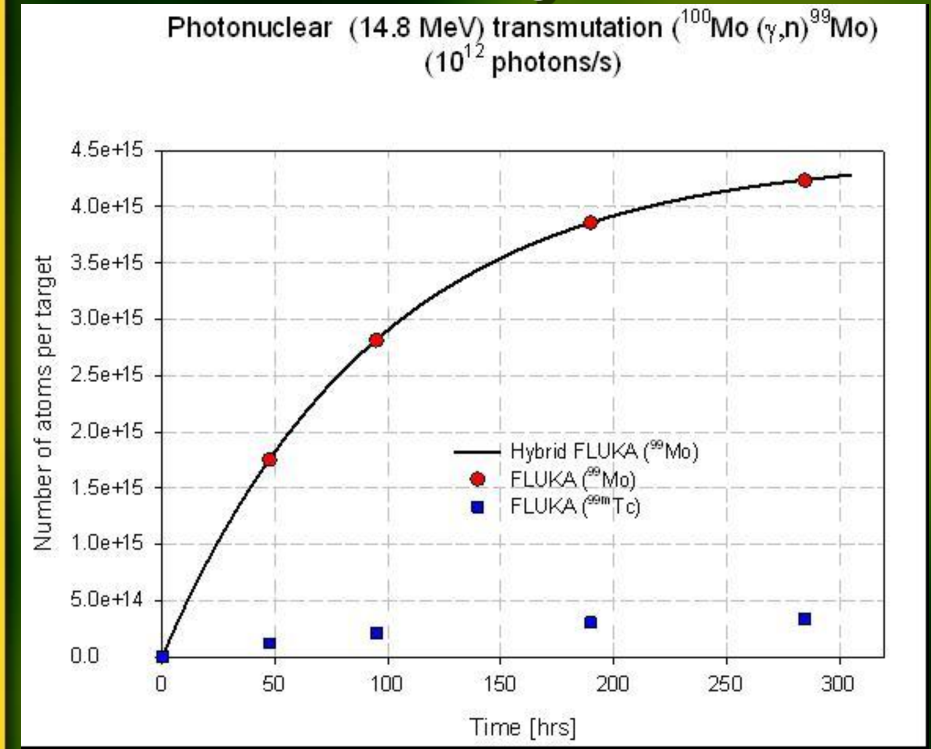
Produced Isotope (reaction)	Yield [per one proton/cm^3] ^{100}Mo target Pb container	Error [%]
(p,n) ^{100}Tc	5.44×10^{-4}	2.9
(p,2n) ^{99}Tc	**4.14×10^{-3}** (~ **2×10^{-3} ^{99m}Tc**)	**1**
(p,3n) ^{98}Tc	7.23×10^{-4}	1.6
(n,γ) ^{101}Mo#	1.00×10^{-6}	77.5
(p,p) ^{100}Mo	2.09×10^{-4}	1.1
(p,n&p) ^{99}Mo	9.42×10^{-5}	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
$(\gamma,e^+e^-)_{atomic}$ ^{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 hybrid versus FLUKA calculations



6×10^{21} atoms in 1g of Mo

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

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

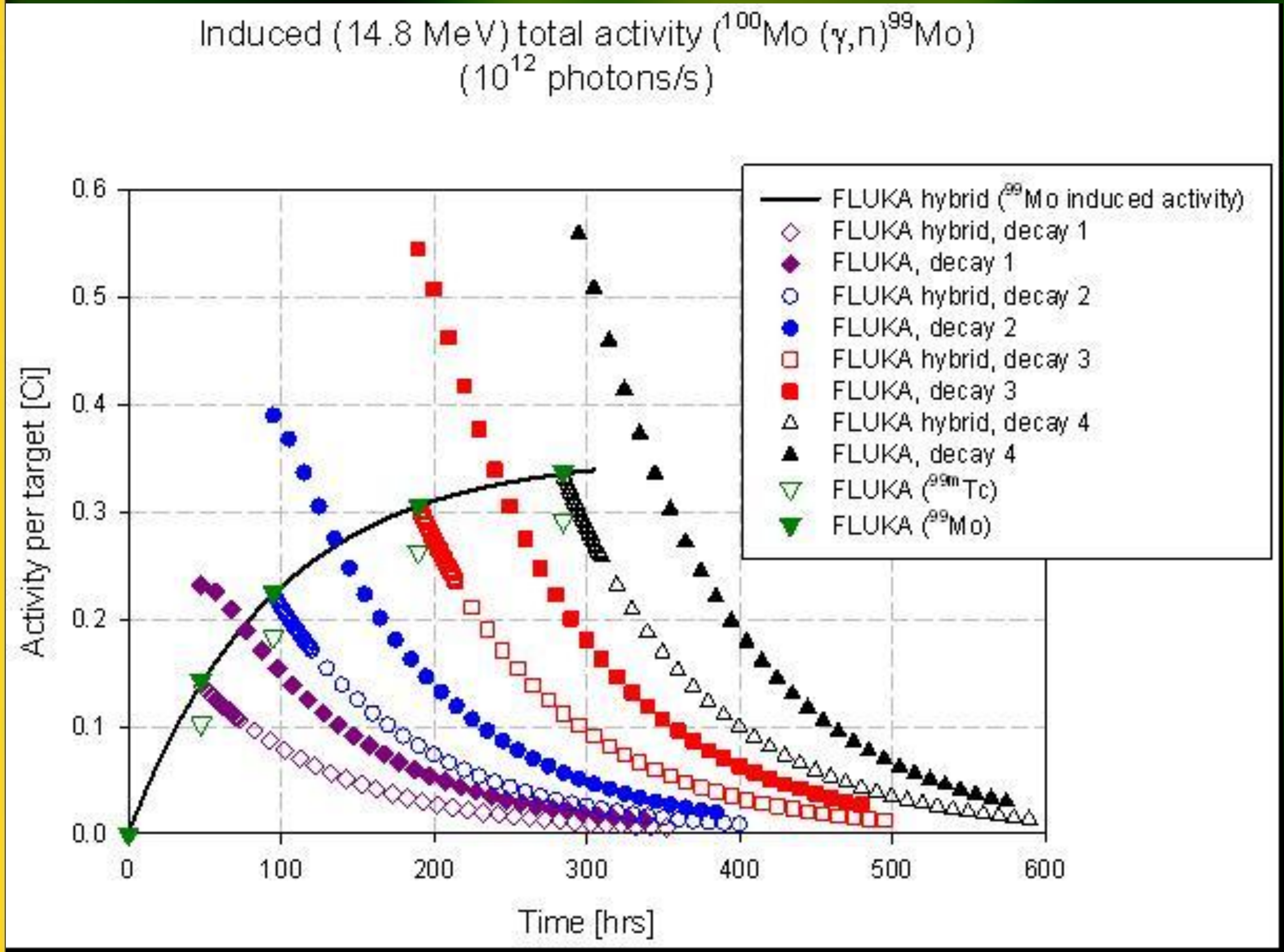
EPAC 2000 Vienna, used per year: 150,000 Ci

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

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

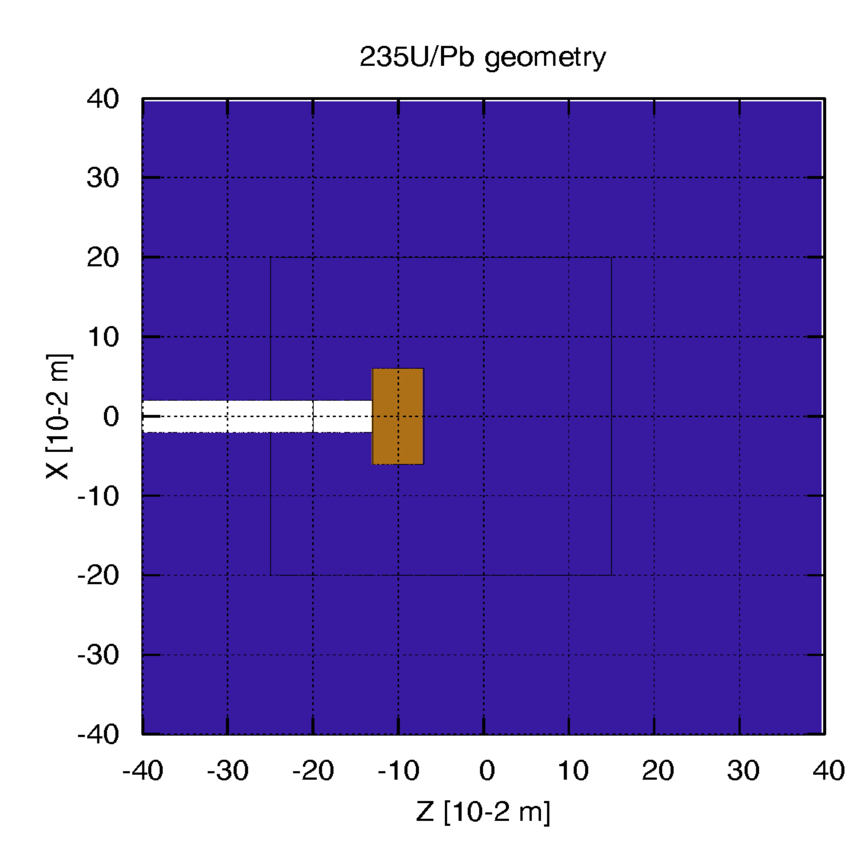
Photofission versus GDR transmutation

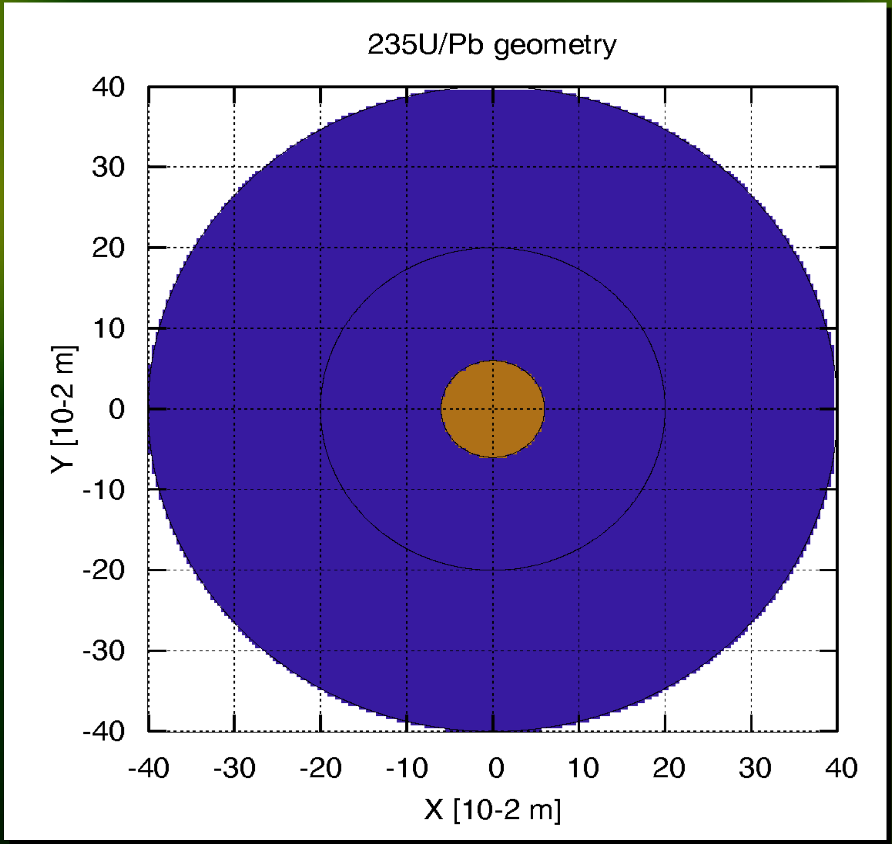
Photons (16 MeV)	Fission yield per one photon [Nuclei/cm^3] (May be heavily underestimated by the FLUKA currently!)							β^- yield
Target	**^{99}Mo(42)**	^{99}Kr(36)	^{99}Rb(37)	^{99}Sr (38)	^{99}Y(39)	^{99}Zr(40)	^{99}Nb(41)	**^{99}Mo(42)**
^{238}U	**1.17×10^{-7}**	8.32×10^{-8}	3.94×10^{-6}	1.83×10^{-5}	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	

Produced Isotope (reaction)	Yield [per one photon/cm^3]	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

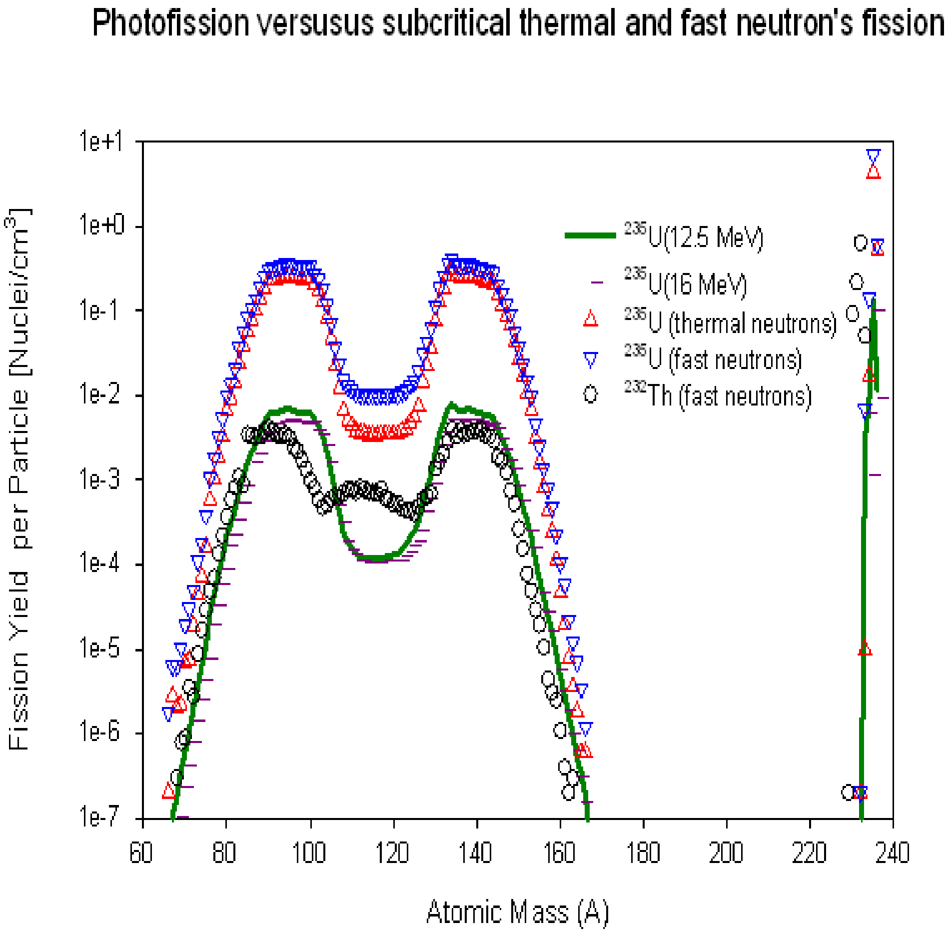
#Secondary neutron capture

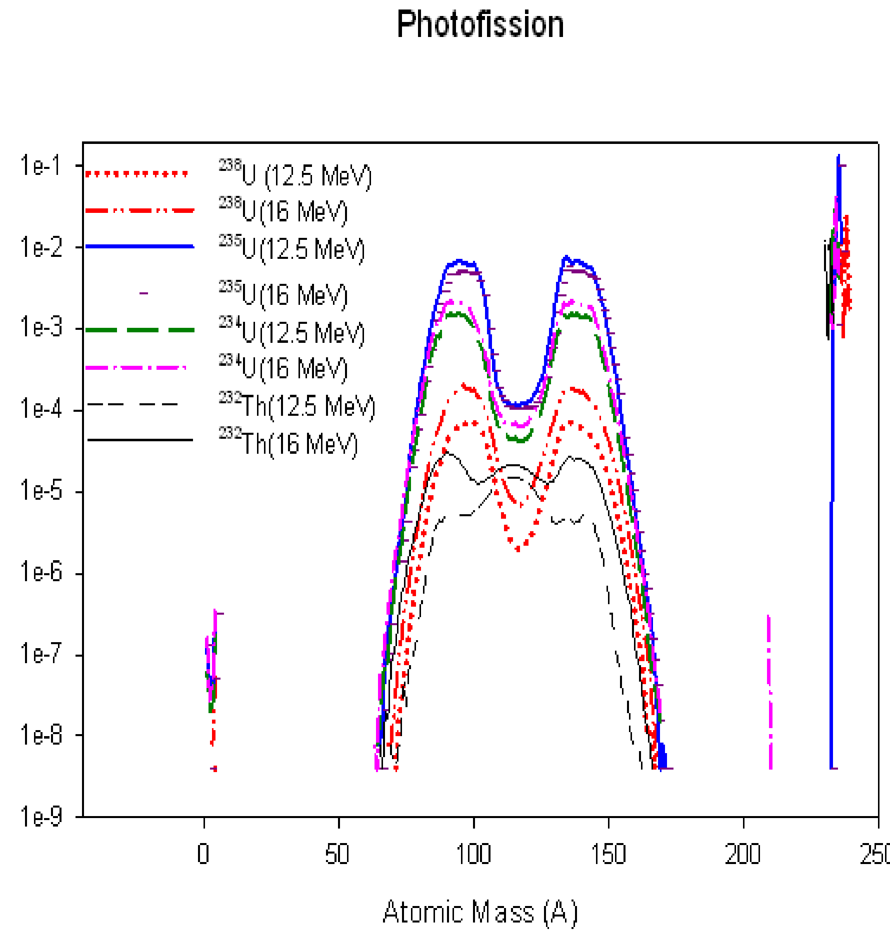
Fission in FLUKA (geometry, target: 12 cm (d) x 6 cm (h))





Photofission versus subcritical thermal and fast neutron's fission





^{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	**2.75×10^{-8}**	3.88×10^{-8}	1.74×10^{-6}	1.0×10^{-5}	3.65×10^{-5}	1.60×10^{-5}	1.62×10^{-6}	**6.63×10^{-5}**
Errors (%)	**36.8**	15.0	2.9	1.6	0.6	0.7	6.5	
^{235}U	**1.35×10^{-5}**	7.86×10^{-8}	4.40×10^{-6}	2.13×10^{-4}	2.23×10^{-3}	3.43×10^{-3}	3.08×10^{-4}	**6.19×10^{-3}**
Errors (%)	**1.3**	22.2	1.3	0.3	0.2	0.1	0.2	
^{234}U	**5.54×10^{-6}**	2.38×10^{-8}	9.83×10^{-7}	1.45×10^{-5}	2.39×10^{-4}	7.51×10^{-4}	1.51×10^{-4}	**1.16×10^{-3}**
Errors (%)	**1.4**	40.8	4.6	1.1	0.2	0.2	0.2	
^{232}Th	**4.00×10^{-0}**	3.92×10^{-9}	1.03×10^{-7}	6.62×10^{-7}	2.85×10^{-6}	1.52×10^{-6}	1.93×10^{-7}	**5.33×10^{-6}**
Errors (%)	**99.0**	99.0	23.1	7.8	2.4	1.7	11.5	

^{99}Mo production comparison

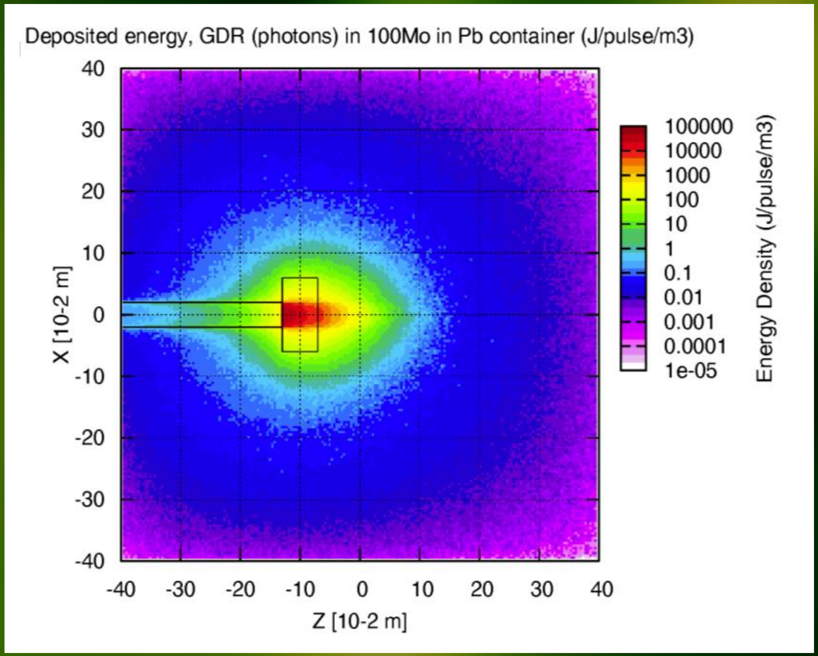
Target 0.12 m diameter and 0.06 m height

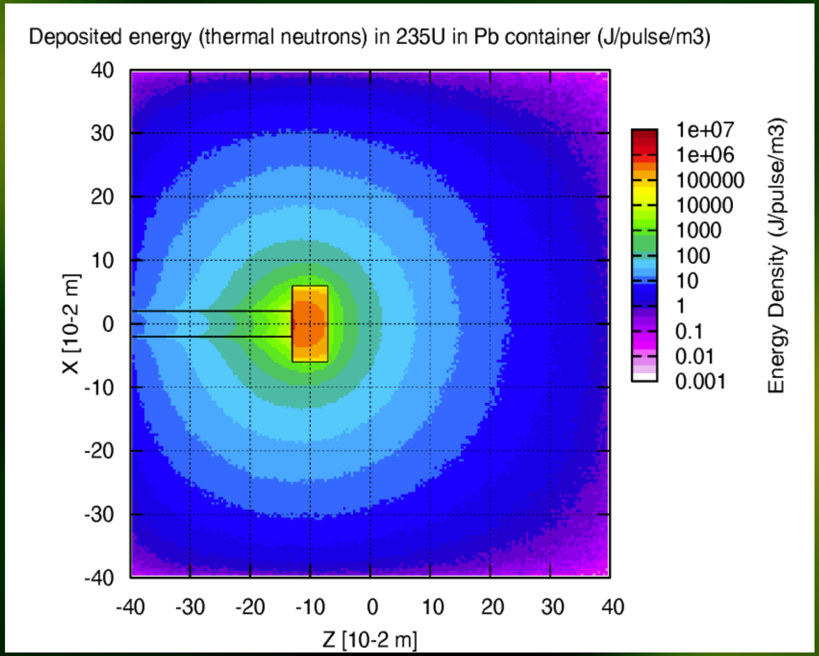
Thermal neutrons	Fission (subcritical) yield per one neutron [nuclei/cm^3]						β^- yield*	Total
Target	**^{99}Mo(42)**	^{99}Rb(37)	^{99}Sr (38)	^{99}Y(39)	^{99}Zr(40)	^{99}Nb(41)	**^{99}Mo(42)**	**^{99}Mo(42)**
^{235}U	**7.01 x10^{-4}**	3.40 x10^{-6}	0.00728	0.0854	0.139	0.0118	**0.2435**	**0.244**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

GDR → (γ,n) ^{99}Mo

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

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





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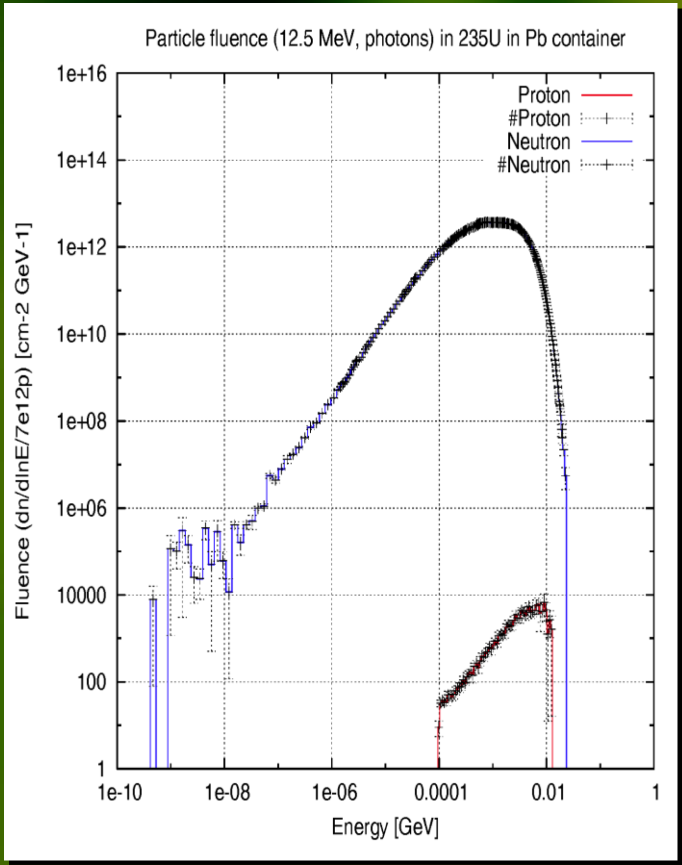
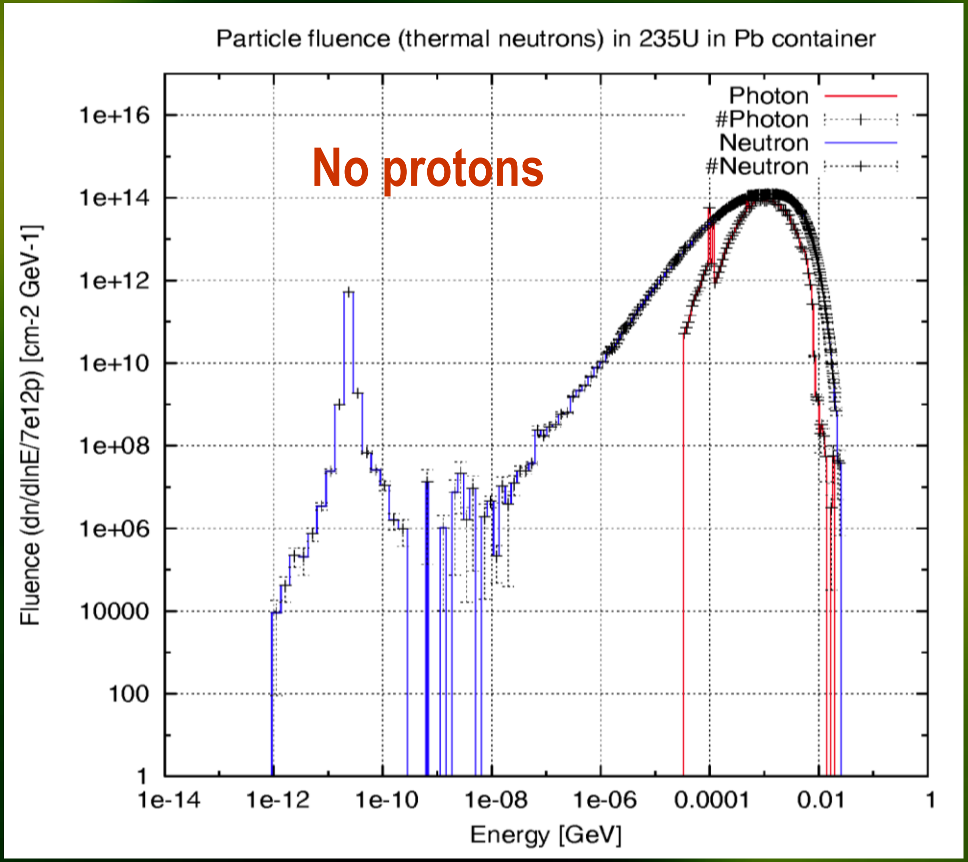
Photofission versus thermal neutrons fission (subcritical); Fluence

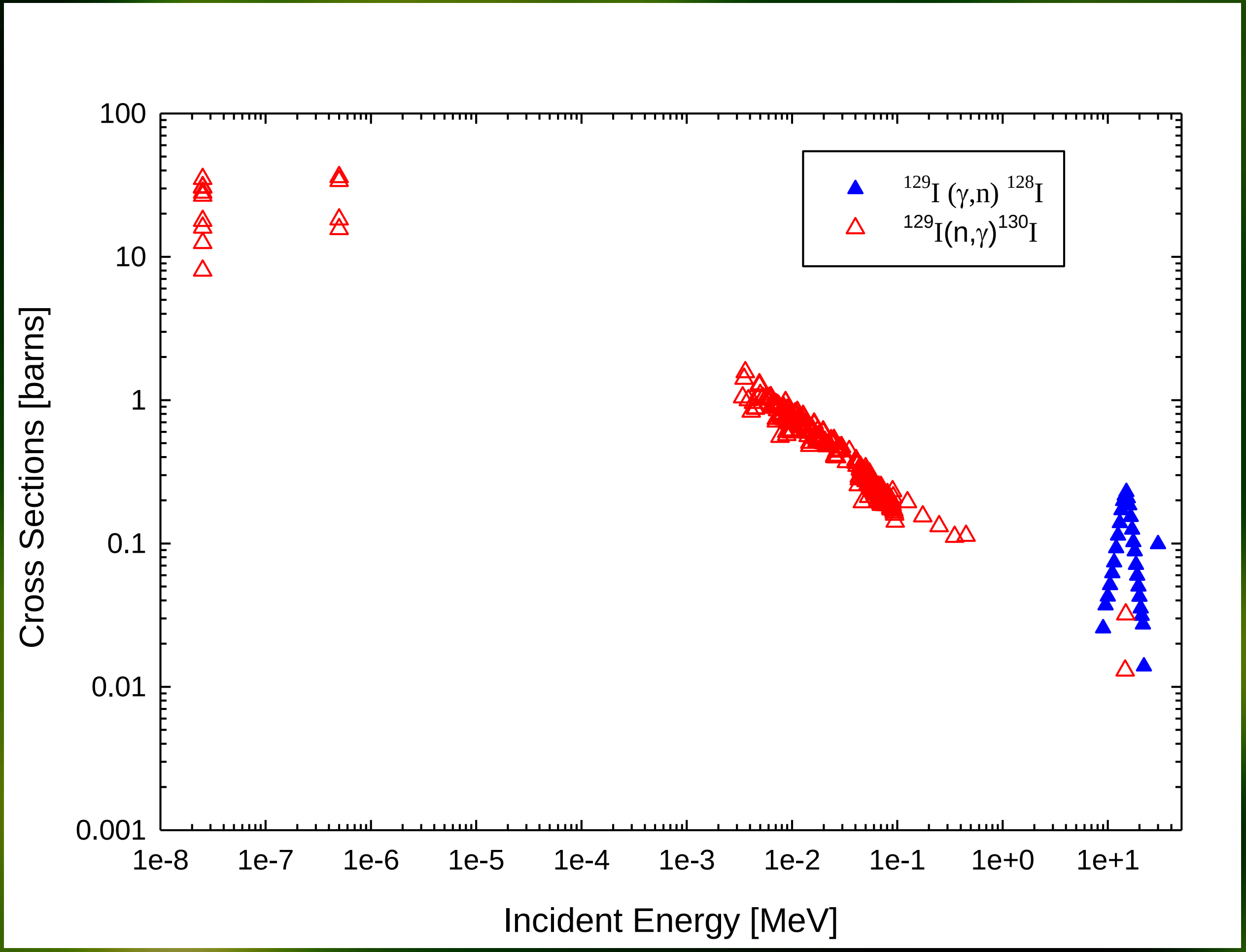




Waste management

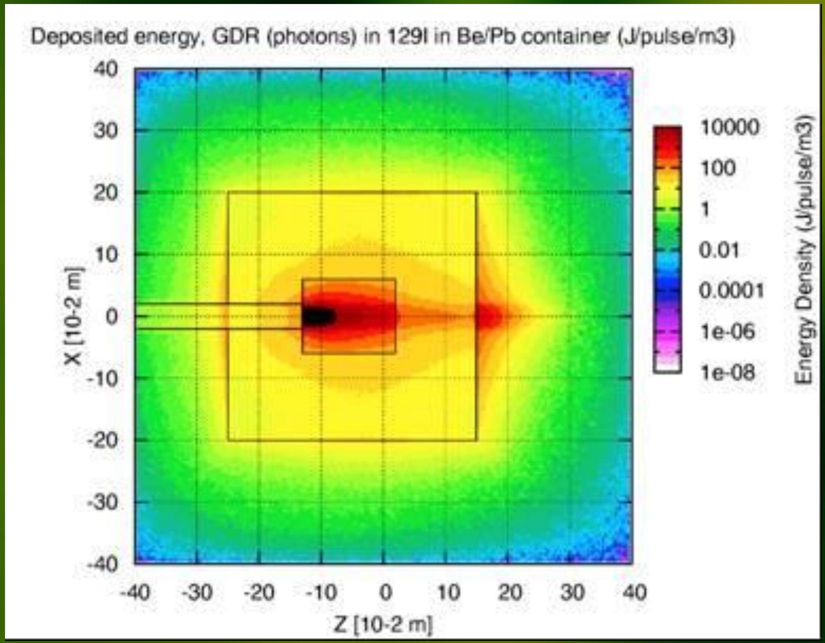
- Nuclear waste consists of 0.74% fission products and 99.26% actinides, with 98.81% uranium and **0.45% long-lived transuranic actinides** (Ottensmeyer, CNS 2010)
- Treatment of long-lived isotopes: ^{79}Se, ^{93}Zr, ^{107}Pd, ^{126}Sn, ^{129}I and ^{135}Cs; radio-toxic >10^5 years needed
- GDR transmutation 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 min ^{128}I or 12.36 h ^{130}I (secondary neutron capture)

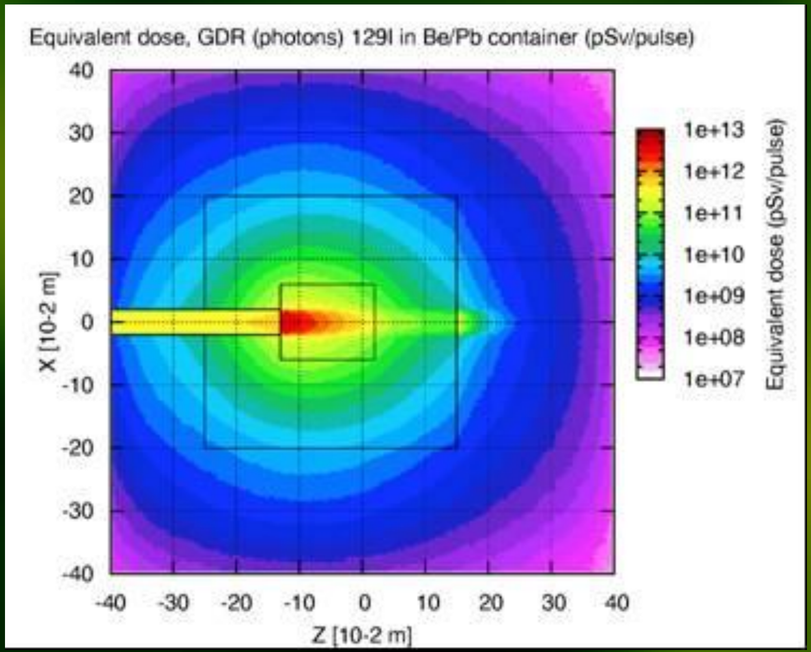
Photons versus neutrons



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 x10^{-03}	0.4
$(\gamma,e^+e^-)_{atomic}$ ^{129}I	6.15 x10^{-03}	0.4
(γ,n) ^{128}I	**2.88 x10^{-02}**	**0.3**
(γ,2n) ^{127}I	9.88 x10^{-04}	1.2
(γ,p) ^{128}Te	3.20 x10^{-06}	33.4

Be container

#Secondary neutron capture

1.57 x10^7 years half-life time of ^{129}I ⟶ 24.99 min ^{128}I or 12.36 h ^{130}I (secondary neutron capture)

Be/Pb container

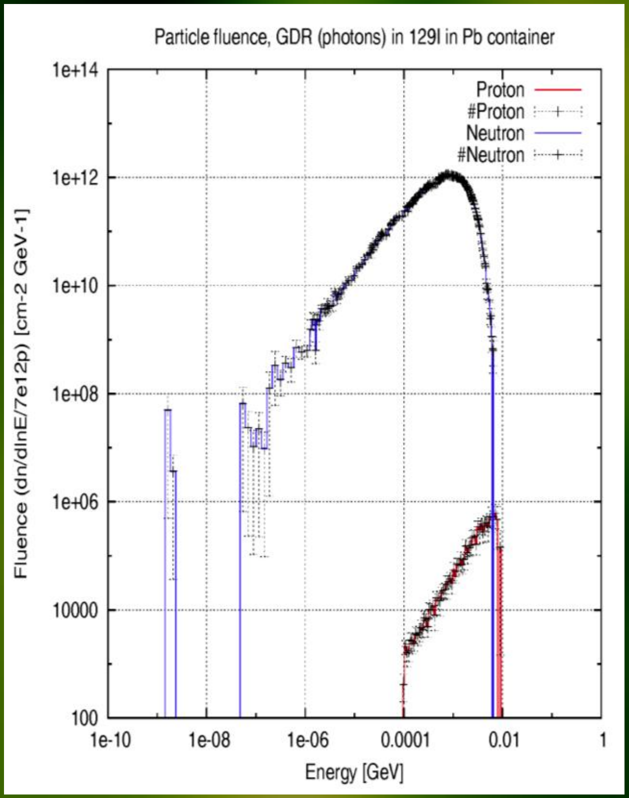
Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) ^{130}I#	7.91 x10^{-03}	0.8
$(\gamma,e^+e^-)_{atomic}$ ^{129}I	6.17 x10^{-03}	0.9
(γ,n) ^{128}I	**2.90 x10^{-02}**	**0.2**
(γ,2n) ^{127}I	9.87 x10^{-04}	1.3
(γ,p) ^{128}Te	5.60 x10^{-06}	26.2

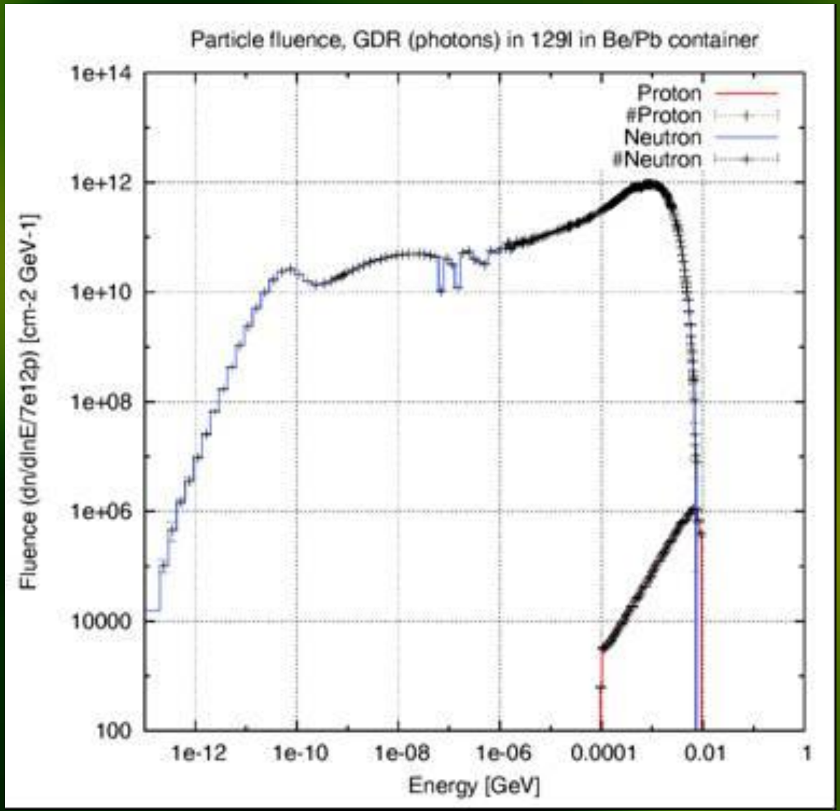
Pb container

Produced Isotope (reaction)	Yield [per one photon/cm^3]	Error [%]
(n,γ) ^{130}I#	1.08 x10^{-03}	0.8
$(\gamma,e^+e^-)_{atomic}$ ^{129}I	6.81 x10^{-03}	0.5
(γ,n) ^{128}I	**2.88 x10^{-02}**	**0.1**
(γ,2n) ^{127}I	1.00 x10^{-03}	1.7
(γ,p) ^{128}Te	2.80 x10^{-06}	30.7

University of Saskatchewan

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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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