Feasibility of Photon-Neutron Hybrid Transmutation of Toxic Radioactive Fission Products

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Contents

Motivation

- Hazardous nuclear waste can pose serious problems due to:
  - High level of *toxicity.*
  - Long *half-life.*
- Technology development to convert it into
  - *stable nuclides* or *short living* radionuclides.

<u>Benefits</u>

Reduce global waste inventory of fission products.

Save millions of years repository management.

Solve one of the biggest challenges of nuclear waste handling**.**

Isotope	Transmutation Needs			
	Toxicity	Half-life (yrs.)	Repository Impact	Production[a] kg/(GW$_{th}$.yr)
^{99}Tc	Medium	Long (2.1×10^5)	High	8.54
^{107}Pd	Low	Long (6.5×10^6)	Low	2.34
^{129}I	Medium	Long (1.6×10^7)	Very high	1.96
^{135}Cs	Medium	Long (2.3×10^6)	Medium	4.00
^{137}Cs	High	Medium (30.0)	Low	8.52
^{151}Sm	High	Medium (88.8)	Low	0.15

[a] 3.2% ^{235}U enrichment, 33 GWd/MTU with 20-yr cooling time.

$$^{129}I\,(15.7\times10^{6}\ y)\xrightarrow{(\gamma,n)}{}^{128}I\,(T_{1/2}=24.99\,m)\begin{cases}\xrightarrow{\beta^-(93.1\%)}{}^{128}Xe\,(Stable)\\ \xrightarrow{\beta^+(6.90\%)}{}^{128}Te\,(Stable)\end{cases}$$

Transmutable in both photo and neutron fields

$$^{135}Cs\,(T_{1/2}=2.3\times10^{6}\ y)\xrightarrow{(\gamma,n)}{}^{134}Cs\,(2.07\ y)\xrightarrow{\beta^-(\sim100\%)}{}^{134}Ba\,(Stable)$$

$$^{137}Cs\,(T_{1/2}=30\ y)\xrightarrow{(\gamma,n)}{}^{136}Cs\,(13.16\,d)\xrightarrow{\beta^-(100\%)}{}^{136}Ba\,(Stable)$$

Cs-137 is non-transmutable in neutron field.

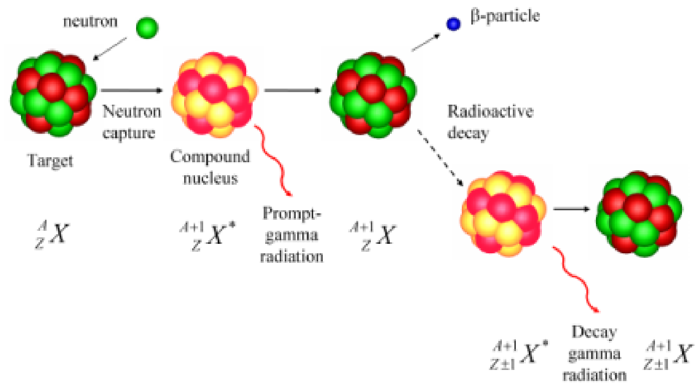
$$^{99}Tc\ (\mathrm{T}_{1/2}=2.1\times10^{5}\ \mathrm{y})\xrightarrow{(\gamma,n)}{}^{98}Tc\,(\mathrm{T}_{1/2}=4.2\times10^{6}\ \mathrm{y})$$

Tc-99 non-transmutable with photons

Introduction

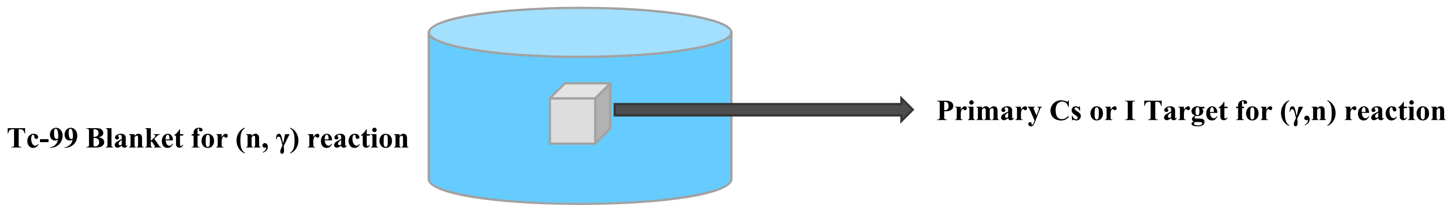
- **Transmutation of <u>LLFP's</u> with Neutrons and Photons:**

1. ***Neutron Transmutation***
   - Large capture cross section & high neutron flux
   - Costly nuclear reactors & secondary activation.
2. ***Photo-transmutation***
   - Photo-transmutation using (γ,n): a proven technology
   - Possibility of photo-transmutation using (γ,γ')
   - Rather compact and clean facility



- Photonuclear Giant Dipole Resonance (GDR) based (γ,n) cross section *is slowly varying function of mass number* (great advantage).
- However, Tc-99 cannot be transmuted with (γ,n) photonuclear reaction.

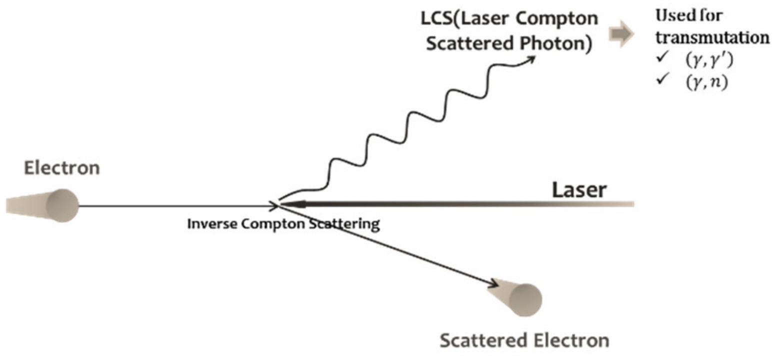
- ***Hybrid transmutation***
  - ***Hybrid transmutation utilize the energetic neutrons of photonuclear reaction to do the neutron capture transmutation.***



Laser-Compton Scattering

Laser-Compton Scattering (LCS) Process

- Gamma rays for the photonuclear GDR reaction can be produced using:
  - Conventional Bremsstrahlung.
  - Laser Induced Bremsstrahlung (LIB).
  - *Laser Compton Scattering* (LCS) etc.
- Laser Compton Scattering (LCS)
  - Interaction of very low energy photon with highly energetic electron beam.
  - Interaction is based on Klein-Nishina cross-section.



The Laser-Compton Scattering Process

$$\frac{d\sigma}{\sin\theta\, d\theta} = \pi r_0^2 \frac{(1-\beta^2)}{(1-\beta\cos\theta)^2} R^2 \left(R + \frac{1}{R} - 1 + \cos^2\theta'\right)$$

where,

$$r_0 = 2.818\, fm$$

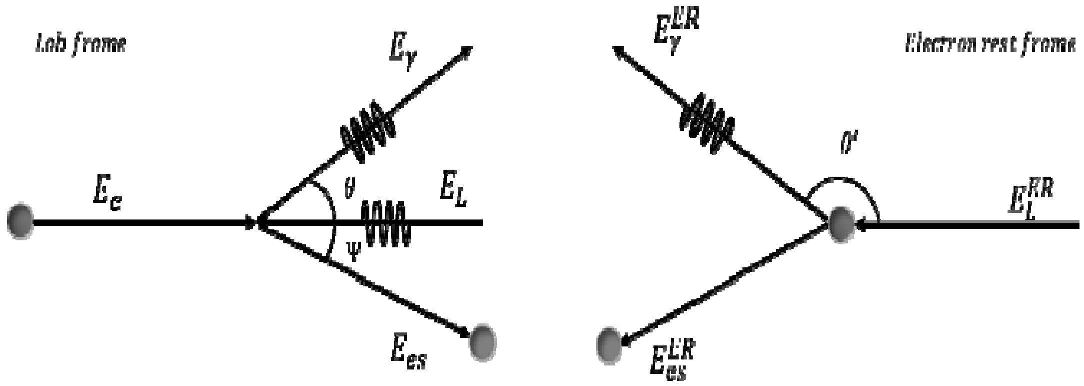
$$R = \frac{E_\gamma^{ER}}{E_L^{ER}} = \frac{1}{1 + \left(\frac{E_L^{ER}}{mc^2}\right)(1+\cos\theta')}$$

$$E_L^{ER} = \frac{\sqrt{1-\beta^2}}{1-\beta} E_L$$

$$\cos\theta' = \frac{\cos\theta - \beta}{1-\beta\cos\theta}$$

$$E_\gamma = \frac{(1+\beta)E_L}{1-\beta\cos\theta + E_L/mc^2\sqrt{1-\beta^2}\,(1+\cos\theta)}$$

$$\beta = \frac{\sqrt{T(T+2mc^2)}}{T+mc^2}$$

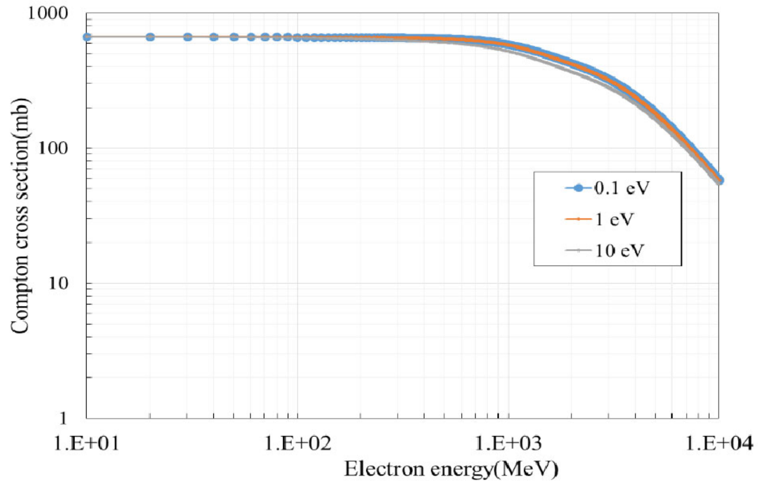


Laser-Compton Scattering (LCS) Process

The total Compton scattering cross-section for π-rad cone angle

$$\sigma(\theta_c) = \int_0^{\theta_c} \frac{d\sigma}{d\theta} d\theta.$$

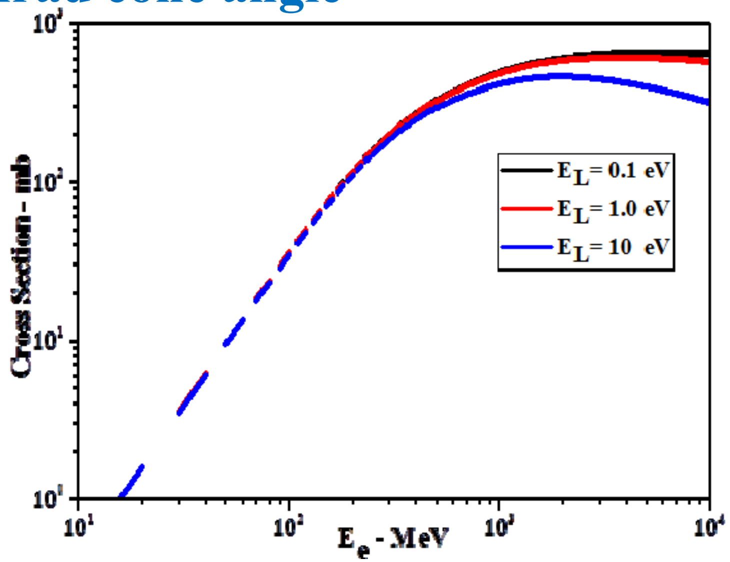
- Flat with electron energy up to 1 GeV.
- Beyond 1 GeV, the cross-section decreases regardless of laser energy due to quantum effect.
- Maximum cross-section is about 665 mb.



Compton Cross-section for π-rad cone angle

The Compton scattering cross-section for 1 mrad cone angle

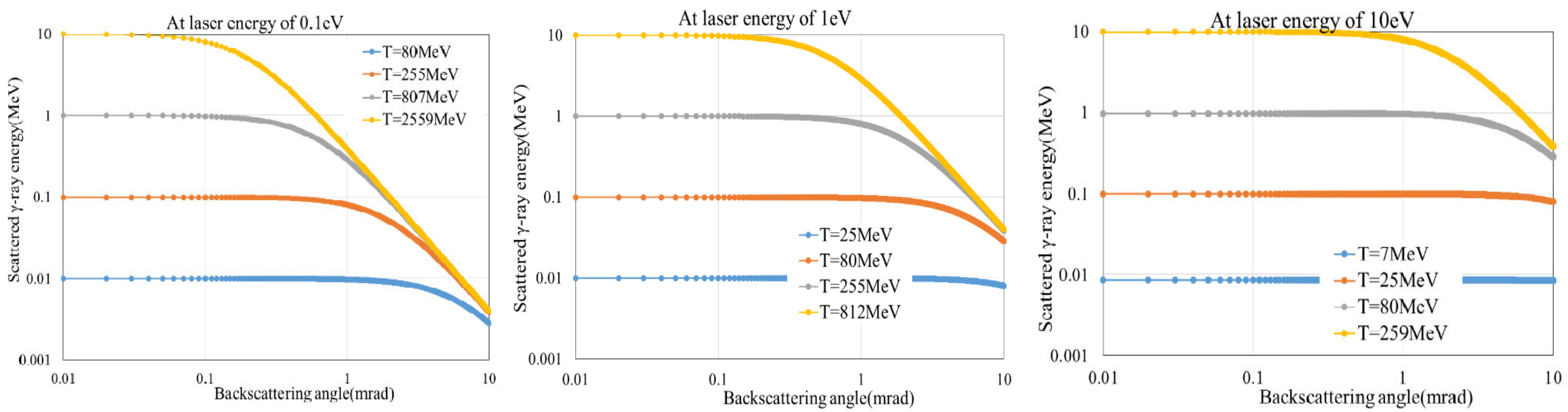
- The back scattering Compton cross-section increases rather linearly with electron energy up to near 1 GeV.
- Beyond 1 GeV, it is almost saturated for laser energy of 0.1 eV and 1 eV.
- However, clearly decreases for 10 eV laser energy.



Compton Cross-section for 1 mrad cone angle

Energy of scattered gamma rays:

$$E_\gamma = \frac{(1+\beta)\,\mathrm{E}_L}{1-\beta\cos\theta + \mathrm{E}_L/mc^2\sqrt{1-\beta^2}\,(1+\cos\theta)}$$



LCS photon energy as a function of scattering angles at laser energy of 0.1 eV, 1eV, and 10eV

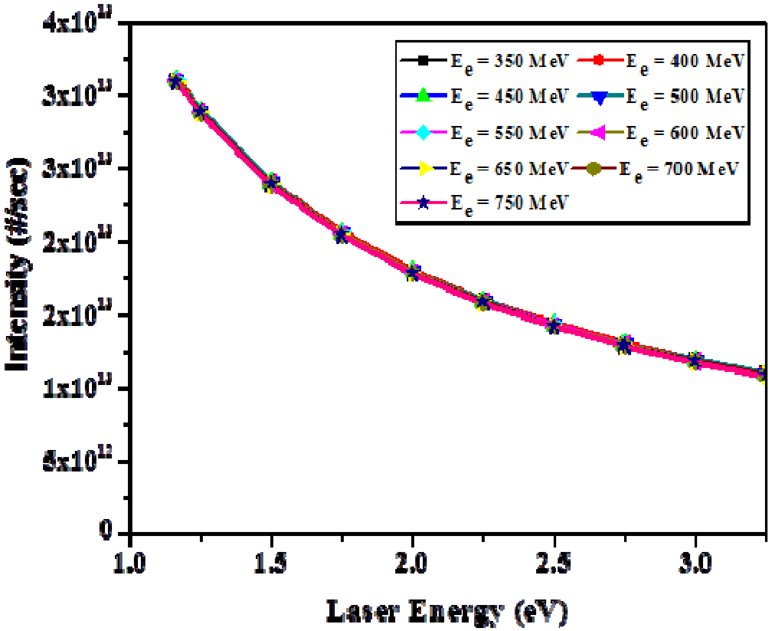
– Scattered gamma-ray energy in all cases are flat up to certain backscattering angles until rapid decrease.
– Largest backscattering angle in the flat γ-ray energy domain increases as the electron energy decreases.
  - No apparent relation between the largest backscattering angle in the flat γ-ray energy domain and laser energy.
– In order to yield high-intensity mono-chromatic LCS photons, the laser energy should be increased and the electron energy should be decreased for a targeted LCS photon energy.

Kinematics of LCS

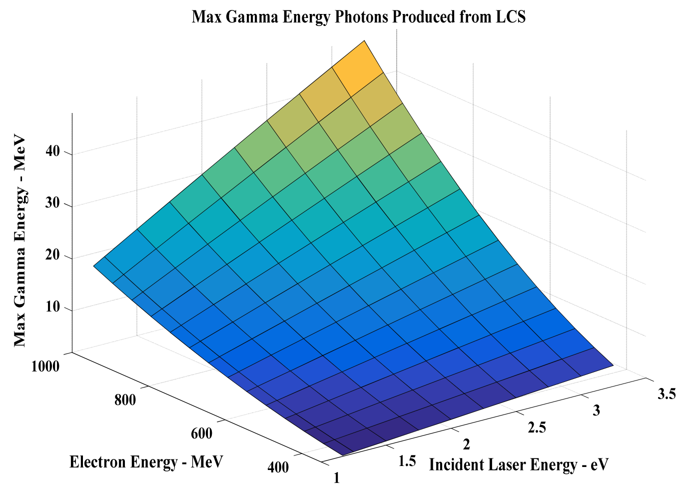
Optimization of electron and laser energy (Laser Power = 100 W)

- **Increasing the laser energy**
  - decreases the gamma intensity.
- **Increasing the electron energy**
  - has negligible effect on the gamma intensity.
- **Increasing the electron or laser energy**
  - increase the maximum gamma energy.
- Based on the cross-section the gamma flux is

$$N_\gamma(\gamma/\mathrm{s}) = \frac{f(Hz)\,N_e\,N_p\,\sigma(\mathrm{cm}^2)}{A(cm^2)}$$



E_e (MeV)	E_L (eV)	Cross- Section (mb)	E_m (MeV)	Intensity (γ/s)
750	1.25	655.89	10.63	2.876E+13
750	1.50	654.05	12.72	2.390E+13
750	1.75	652.23	14.80	2.043E+13
750	2.00	650.42	16.87	1.783E+13
750	2.25	648.63	18.92	1.580E+13
750	2.50	646.84	20.97	1.418E+13
750	2.75	645.07	23.00	1.286E+13
750	3.00	643.31	25.02	1.175E+13
750	3.25	641.57	27.03	1.082E+13



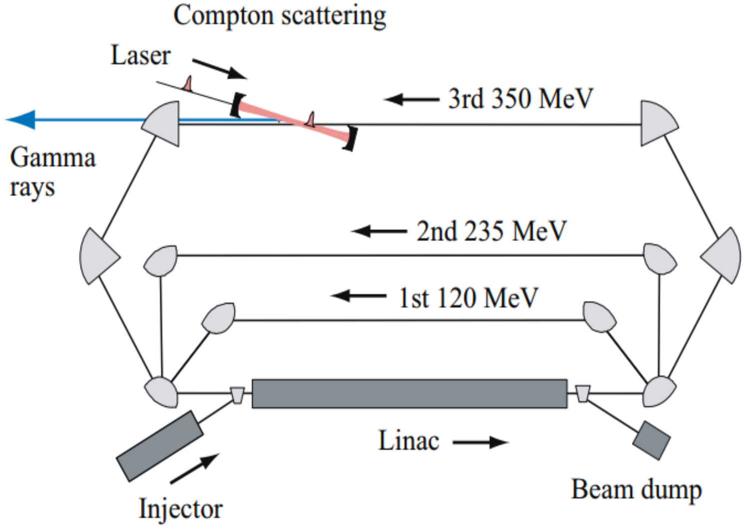
LCS Facilities

The laser-Compton scattering facilities around the world are:

Facility	Electron energy	Laser energy	LCS photon energy	Intensity (#/sec)
HIGS [In-service]	0.24 – 1.2 GeV	1.17–6.53 eV	1-100 MeV	10^7-$2x10^{10}$
MEGa-ray [Driving test]	250 MeV	2.33 eV	0.5-2.3 MeV	10^{12}
SLEGS [In-service]	3.5 GeV	0.117 eV	2-20 MeV	10^5-10^7
ELI-NP [Operation start in 2019]	600 MeV	2-5 eV	1-13 MeV	10^{13}

KEK, Japan LCS facility has gamma flux of 10^{13} γ/s.
Design is based on three loop energy recovery linac (ERL).

Design Parameter	Value
Electron beam energy	350 MeV
Incident laser energy	1.165 eV
Electron beam current	100 mA
Laser power	100 W
RMS of electron beam	70 µm
Collision frequency	80 MHz
Electron bunch charge	1 nC
Pulse energy	1.80 µJ
Amplification factor (Laser super cavity)	3000



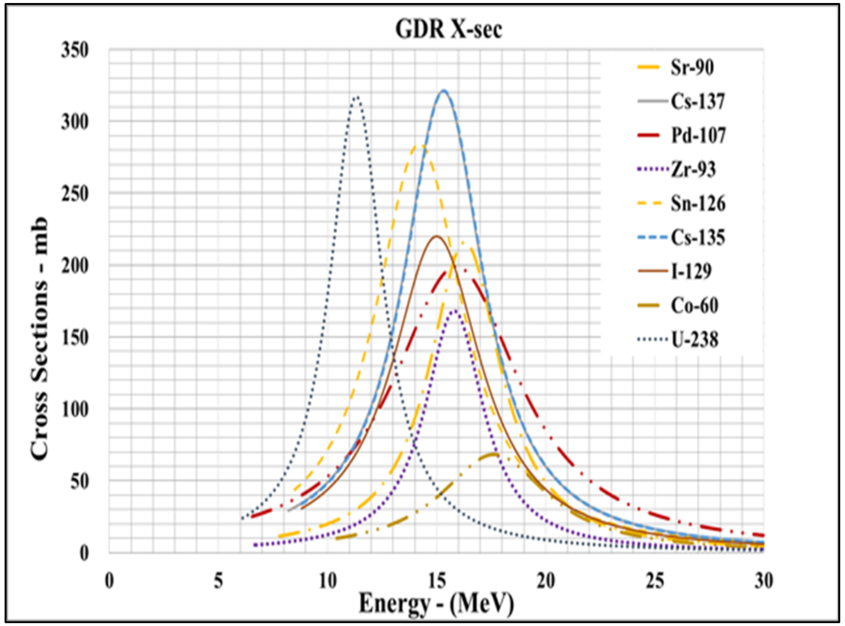
GDR-based Photonuclear Transmutation

Giant Dipole Resonance (GDR) Cross-section

- Giant Dipole Resonance is a dominant *excitation mechanism* ~ nuclei experience a *collective bulk oscillation* of neutrons against all the protons in the nuclei.
- (γ, n) reaction occurs at threshold energy 'E_{th}'
- Follows a Lorentzian distribution given by:

$$\sigma_{reac}(mb) = \sigma_{max}\left[4\left(\frac{E_{max} - E_{\gamma}}{\Gamma}\right)^2 + 1\right]^{-1}$$

  - 'σ_{max}' is the maximum cross-section in mb.
  - 'E_{max}' is the Energy at which max. x-sec appears.
  - 'E_{γ}' is the gamma ray energy.
  - 'Γ' is the FWHM (Full Width at Half Maximum)energy.



- GDR cross-section is largely in between 8 to 16 MeV.
- LCS spectrum should be optimized to maximize the transmutation.

Nuclide	E_{th} (MeV)	Γ (MeV)	E_{max} (MeV)	σ_{max} (mb)
I-129	8.80	5.0	15.0	220
Cs-135	8.83	4.5	15.3	321
Cs-137	8.27	4.5	15.3	321

Photonuclear Transmutation

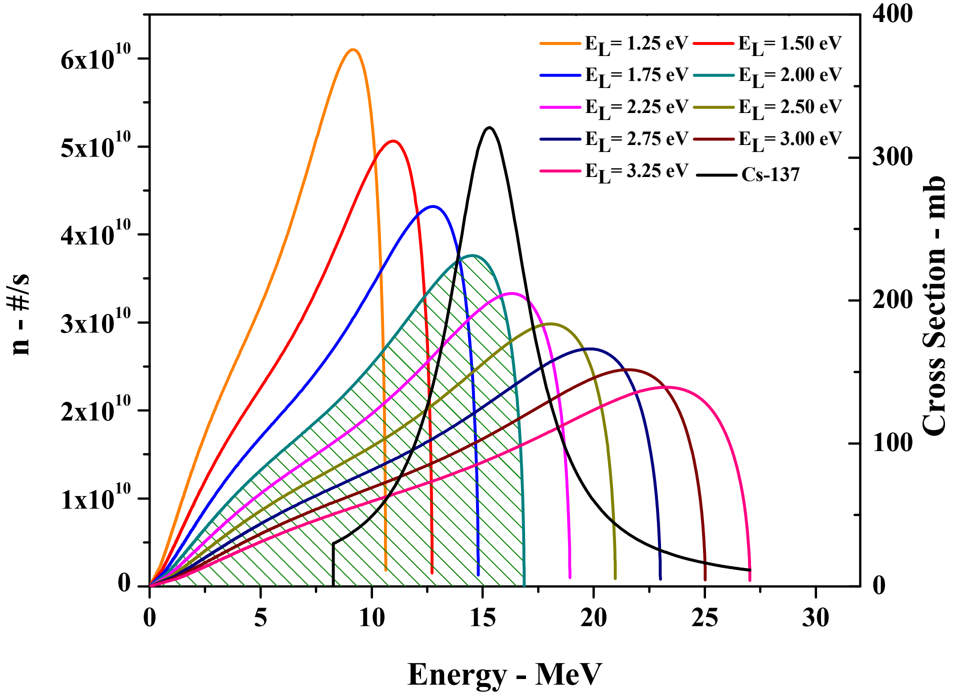
Photonuclear transmutation for I-129, Cs-135, and Cs-137 can be calculated as:

$$N_{reac} = n_{tar} d_{tar} \int_{E_{th}}^{E_C} \sigma_{reac} \frac{dN_\gamma}{dE_\gamma} dE_\gamma$$

where $\frac{dN_\gamma}{dE_\gamma}$ is the spectral density and can be calculated using:

$$\frac{dN_\gamma}{dE_\gamma} = \frac{N_\gamma}{\sigma_t} \int_{E_0-\delta_E}^{E_0+\delta_E} \frac{d\sigma}{dE_\gamma} \frac{1}{\sqrt{2\pi}\,\delta_E} exp\left[-\frac{(E_e-E_0)^2}{\delta_E^2}\right] dE_e$$

However, a rigorous optimization is needed to maximize the transmutation reaction rates. Numerical results of an in house developed code are as follows.



E_e (MeV)	E_L (eV)	Transmutation reaction rates (#/s)		
		^{129}I	^{135}Cs	^{137}Cs
	1.25	3.16E+09	2.61E+08	6.41E+08
	1.50	6.74E+09	5.66E+08	1.24E+09
	1.75	1.20E+10	1.09E+09	2.33E+09
	2.00	**1.42E+10**	**1.44E+09**	**3.05E+09**
750	2.25	1.23E+10	1.26E+09	2.66E+09
	2.50	9.90E+09	1.00E+09	2.13E+09
	2.75	8.00E+09	8.06E+08	1.70E+09
	3.00	6.60E+09	6.60E+08	1.40E+09
	3.25	5.56E+09	5.53E+08	1.17E+09

Photonuclear Transmutation

Requirement for Isotopic Separation

Iodine Case

Two isotopes in spent fuel inventory of LWR's

- I-127 with isotopic abundance of 22.98 %.
- I-129 with isotopic abundance of 77.02 %.

$$^{127}I\,(Stable) \xrightarrow{(\gamma,n)} {}^{126}I\,(T_{1/2} = 12.93\,d) \begin{cases} \xrightarrow{\beta^+ (56.3\%)} {}^{126}Te\,(Stable) \\ \xrightarrow{\beta^- (43.7\%)} {}^{126}Xe\,(Stable) \end{cases}$$

$$^{129}I\,(15.7\times10^6\,y) \xrightarrow{(\gamma,n)} {}^{128}I\,(T_{1/2} = 24.99\,m) \begin{cases} \xrightarrow{\beta^- (93.1\%)} {}^{128}Xe\,(Stable) \\ \xrightarrow{\beta^+ (6.90\%)} {}^{128}Te\,(Stable) \end{cases}$$

In case of Iodine no isotopic separation required

- I-127 abundance is comparatively much less as compared to I-129.
- I-129 with $(\gamma, 2n)$ is converted into stable I-127.

Cesium Case

$$^{133}Cs\,(Stable) \xrightarrow{(\gamma,n)} {}^{132}Cs\,(T_{1/2} = 6.48\,d) \begin{cases} \xrightarrow{\beta^+ (98.13\%)} {}^{132}Xe\,(Stable) \\ \xrightarrow{\beta^- (1.87\%)} {}^{132}Ba\,(Stable) \end{cases}$$

$$^{134}Cs\,(T_{1/2} = 2.07\,y) \xrightarrow{(\gamma,n)} {}^{133}Cs\,(Stable)$$

$$^{135}Cs\,(T_{1/2} = 2.3\times10^6\,y) \xrightarrow{(\gamma,n)} {}^{134}Cs\,(2.07\,y) \xrightarrow{\beta^- (\sim100\%)} {}^{134}Ba\,(Stable)$$

$$^{137}Cs\,(T_{1/2} = 30\,y) \xrightarrow{(\gamma,n)} {}^{136}Cs\,(13.16\,d) \xrightarrow{\beta^- (100\%)} {}^{136}Ba\,(Stable)$$

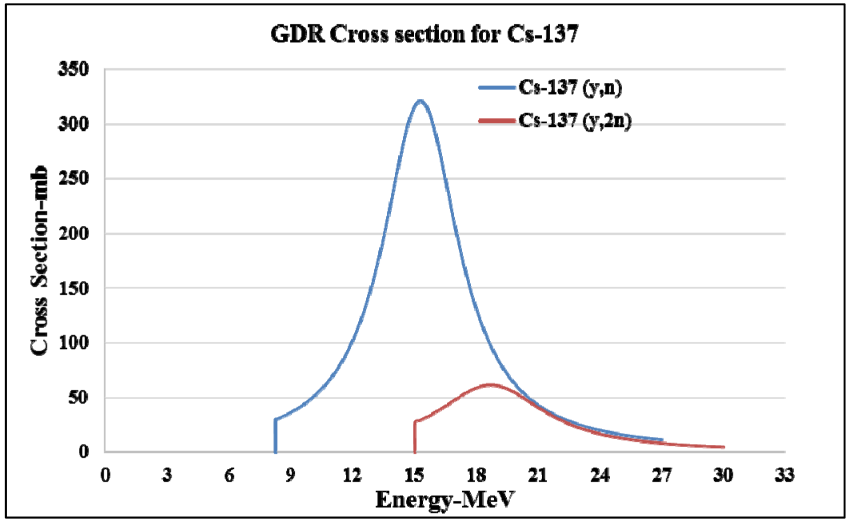
Destruction of Cs-135 > Production of Cs-135 via (γ,2n)

- **$(\gamma, 2n)$ reaction Cs-137 will convert it into Cs-135 with half life of 2.3 million years.**

Photonuclear Transmutation

Re-optimization of LCS spectrum

- Isotopic separation requirement.
- GDR based (γ, n) and $(\gamma, 2n)$ reactions are considered
- Combinations of electron and laser energy have been chosen and optimized.
- Amplification factor is taken to be 3000.



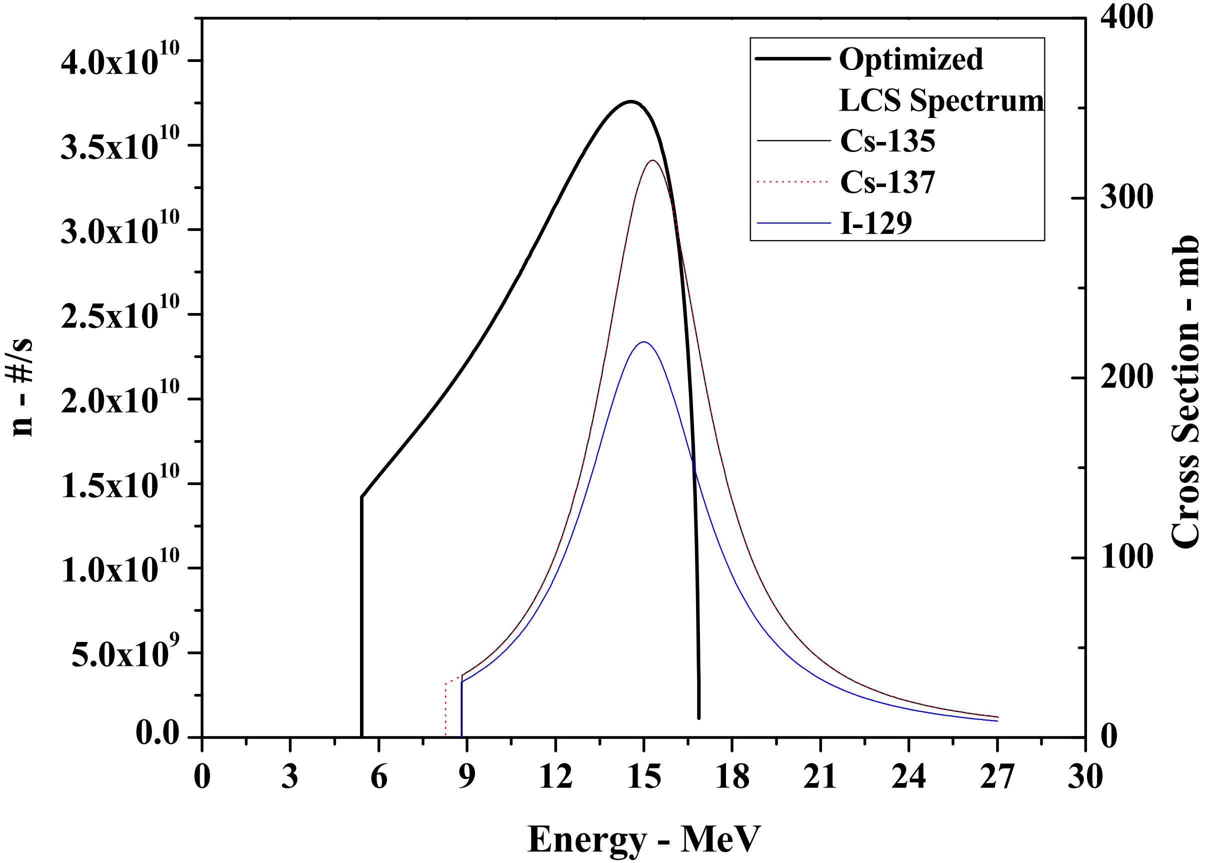
RESULTS

- The maximum possible reaction rates appear
  - Laser energy 2.0 eV
  - Electron energy 750 MeV.
- The $(\gamma, 2n)$ reaction at this particular combination is $3.45\times10^{7}(s^{-1})$.
- Laser energy of 1.75 eV $(\gamma, 2n)$ reaction of Cs-137 is theoretically zero
  - threshold energy is 15.04 MeV.
- Further optimization is required in between 1.75 and 2.0 eV
  - Maximize net destruction of Cs-135.

E_e MeV	E_L (eV)	Transmutation reaction rates (#/s): Cs-137 $\xrightarrow{(\gamma,2n)}$ Cs-135	Cs-135 $\xrightarrow{(\gamma,n)}$ Cs-134	Net destruction of Cs-135
750	1.75	0.00E+00	1.09E+09	1.09E+09
	1.775	0.00E+00	1.15E+09	1.15E+09
	1.80	3.43E+06	1.21E+09	1.21E+09
	1.825	7.40E+06	1.26E+09	1.25E+09
	1.85	1.13E+07	1.31E+09	1.30E+09
	1.875	1.52E+07	1.35E+09	1.34E+09
	1.9	1.91E+07	1.38E+09	1.36E+09
	1.925	2.29E+07	1.41E+09	1.39E+09
	1.95	2.68E+07	1.42E+09	1.39E+09
	1.975	**3.06E+07**	**1.44E+09**	**1.41E+09**
	2.0	**3.45E+07**	**1.44E+09**	**1.41E+09**

Photonuclear Transmutation

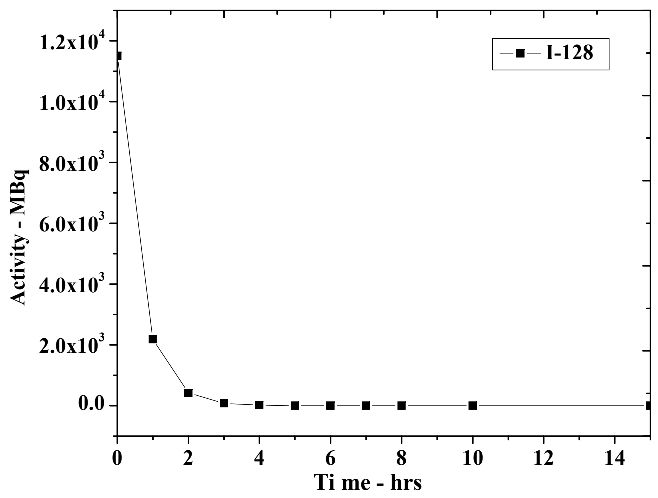
- *Cone Angle Based Optimization* is performed to avoid the *excessive heating* in target material while keeping the same transmutation reaction rate.
  - By eliminating the *unwanted reactions* of lower energy gamma rays.
- Practically, it is easy to make a collimator up to *1 mrad.*

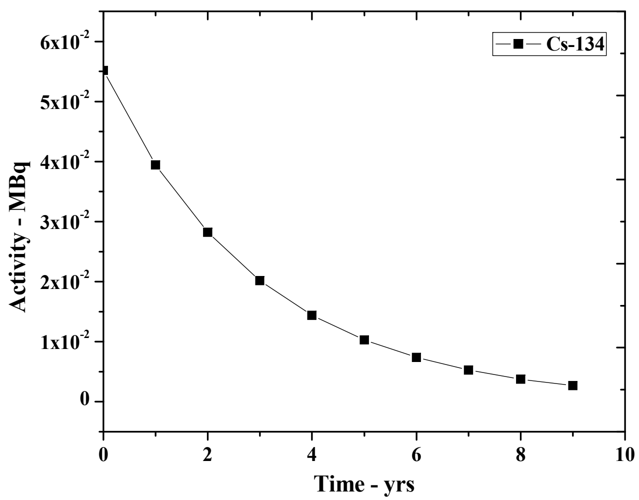


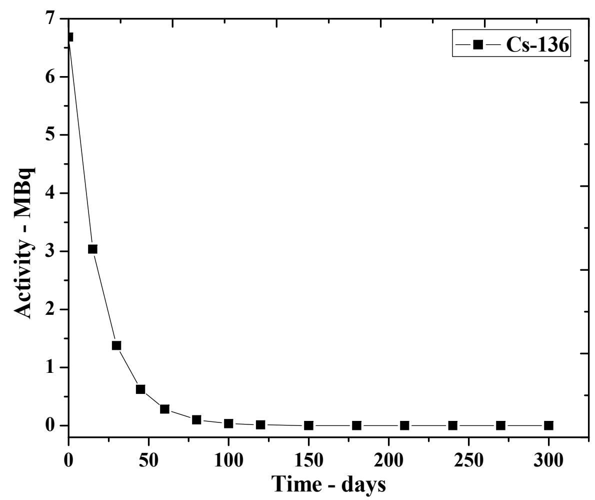
Photonuclear Transmutation

Activity Calculations for 1-hour irradiation period.

Reaction	Initial activity bef ore irradiation (MBq)	Activity after irradiation of 1 hour (MBq) $(A = A_0 e^{-\lambda t})$
I-129 $\xrightarrow{(\gamma,n)}$ I-128	2.48E+01	1.15E+04
Cs-135 $\xrightarrow{(\gamma,n)}$ Cs-134	1.34E+01	5.52E-02
Cs-137 $\xrightarrow{(\gamma,n)}$ Cs-136	3.90E+05	6.68E+00







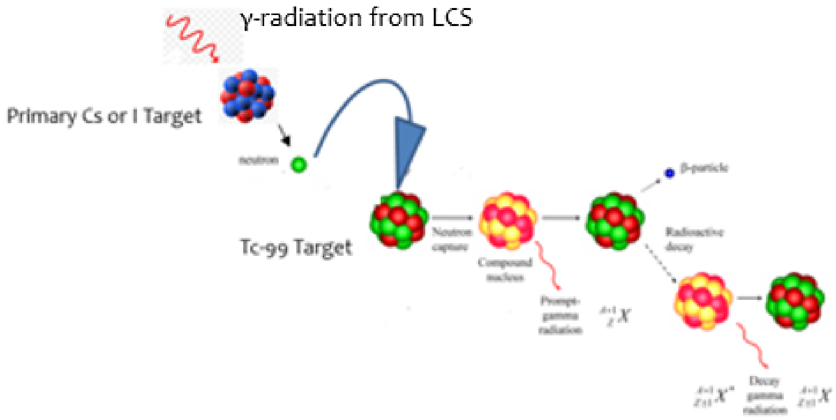
- Initial activities of the transmuted products (after irradiation) can be higher than those of the non-tra nsmuted radionuclides (before irradiation).
- However, their decays will be much faster and repository management will be much easier.

Hybrid Transmutation (Photon and Neutron)

Conceptual Details

The hybrid transmutation is based on concept of

– Utilizing the energetic neutrons from the photonuclear (γ,n) reaction.



Benefits

– Emitted neutron from the photonuclear reaction are energetic and hence results in more shielding requirements.

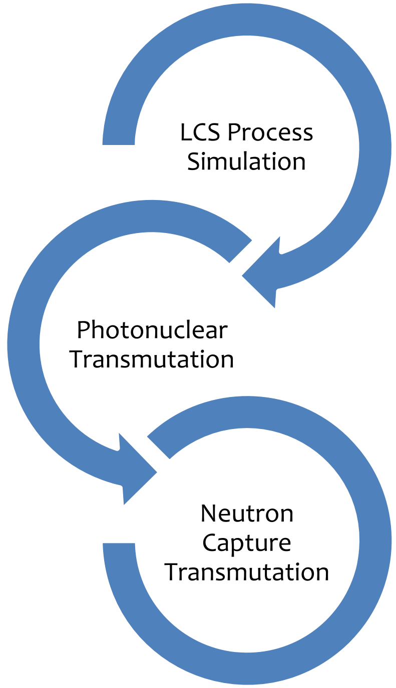
– Tc-99 cannot be transmuted with photonuclear reaction.

- Neutron capture reaction in the blanket can be utilized with energetic neutrons.

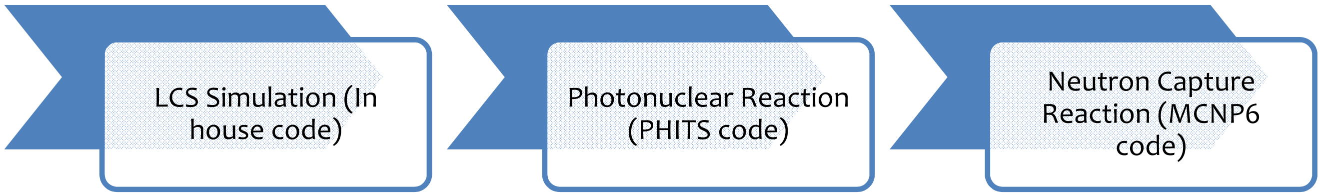
– Additional transmutation without any significant additional cost.

Hybrid Transmutation

- **The hybrid transmutation analysis is performed in a three step approach:**
  - 1st step: The production of optimized energetic neutrons using LCS process.
  - 2nd step: The calculation of photonuclear (γ,n) transmutation reaction rates.
  - 3rd step: The calculation of neutron capture (n,γ) transmutation reaction rates in the blanket.



- **Analysis Tools**



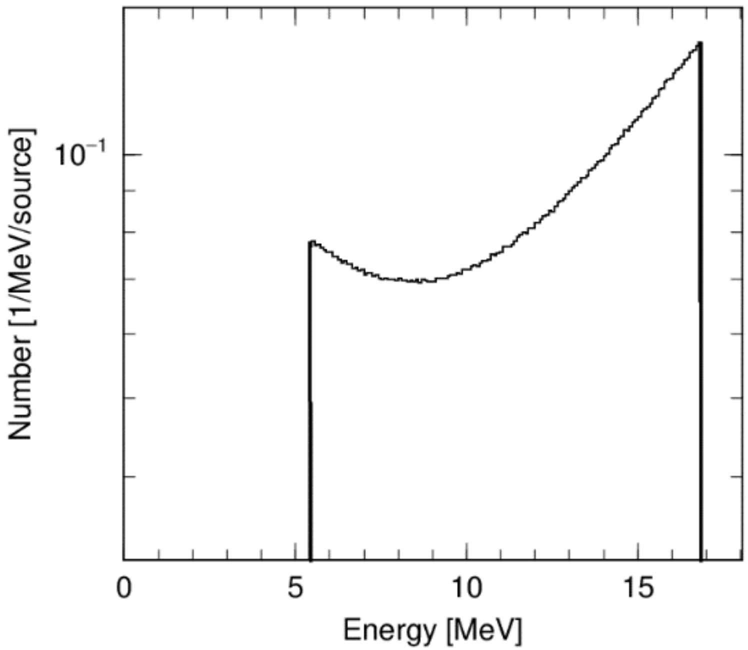
- *Details of the hybrid transmutation are explained in the coming slides.*
- *PHITS code is also used for the benchmarking of previously reported in house code results.*

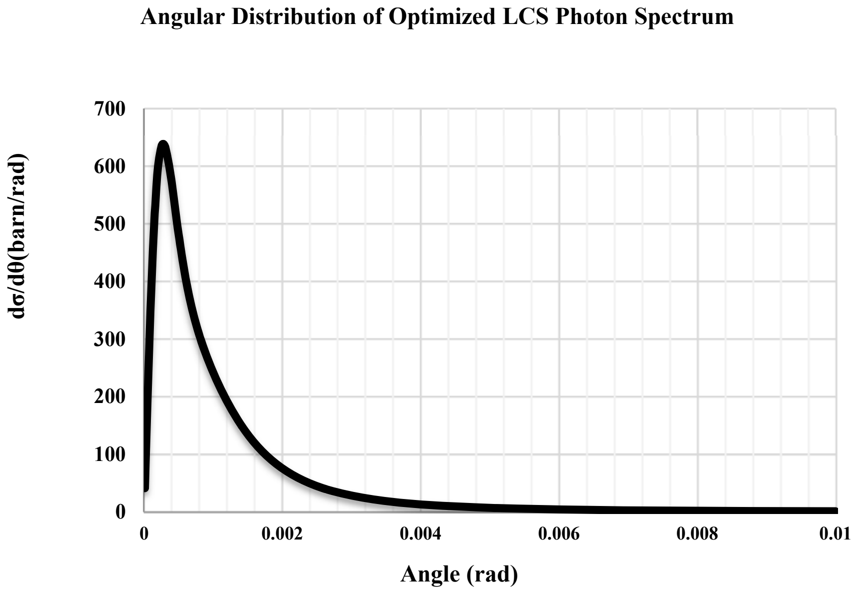
Hybrid Transmutation

LCS Process Simulation

- LCS process cannot be simulated using any commercially available Monte Carlo based code.
- For the photonuclear transmutation reaction rates calculation
  - Energy and angular distribution of the LCS photons are required.
- Using an in-house code, for an optimized LCS spectrum
  - Energy and angular distribution is given as input to **PHITS** code

Design Parameter	Value
Electron beam energy	750 MeV
Incident laser energy	2.0 eV
Electron beam current	100 mA
RMS of electron beam	70 µm
Pulse energy	1.80 µJ
Amplification factor (Laser super cavity)	3000





PHITS Code

Particle and Heavy Ion Transport code System

Capability

Transport and collision of nearly all particles over wide energy range

in 3D phase space
with magnetic field & gravity

neutron, proton, meson, baryon
electron, photon, heavy ions

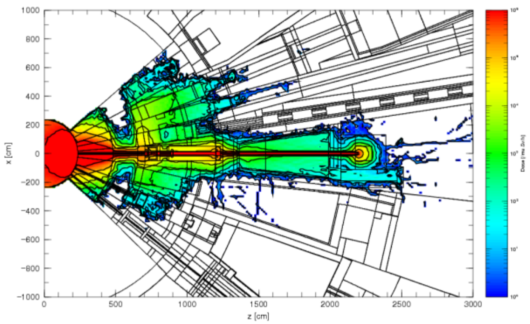
10^{-4} eV to 1 TeV/u

All-in-one-Package

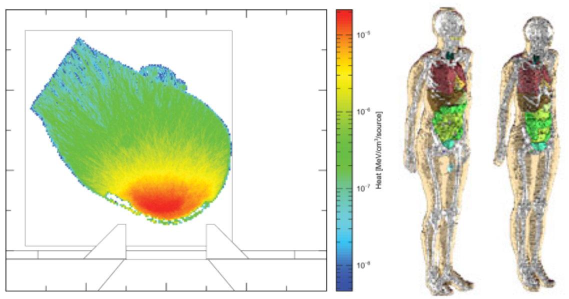
All contents of PHITS (source files, binary, data libraries, graphic utility etc.) are fully integrated in one package

OECD/NEA Databank, RSICC (USA, Canada) and RIST (Japan)

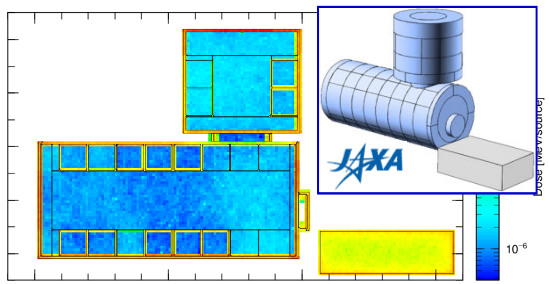
Applications



Accelerator Design



Radiation Therapy & Protection



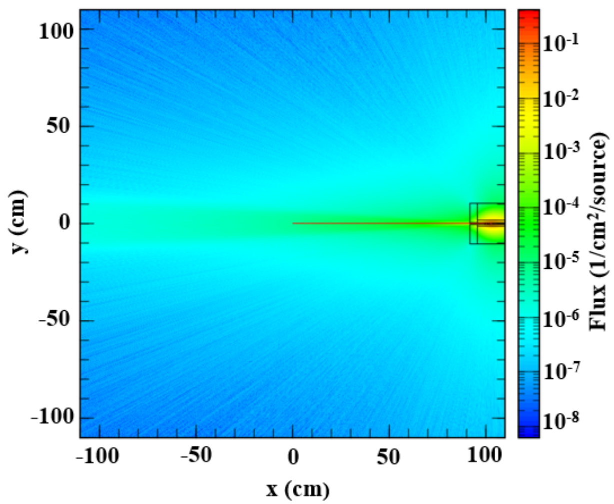
Space & Geoscience

Hybrid Transmutation

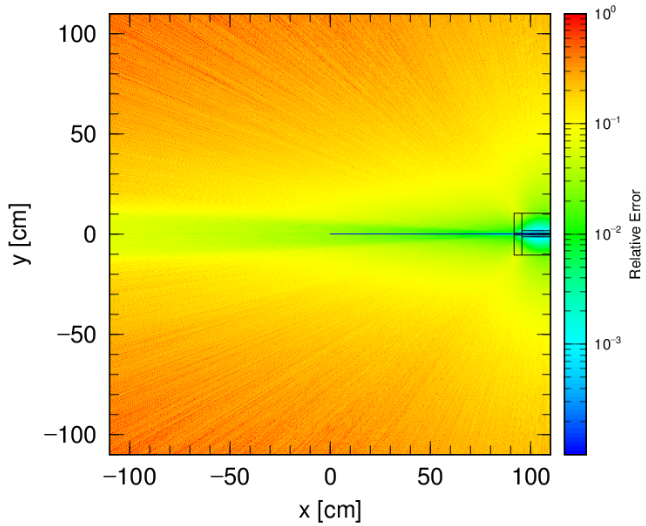
- **The photonuclear transmutation target is**
  - Chemically stable CsI with the isotopic composition of shown in the following table.

Element	Isotope	Isotopic Composition (wt. %)
Iodine	^{127}I	22.98
	^{129}I	77.02
Cesium	^{133}Cs	50.45
	^{134}Cs	0.01
	^{135}Cs	15.84
	^{137}Cs	33.71

  - The target is placed at 100 cm away from the photon source.

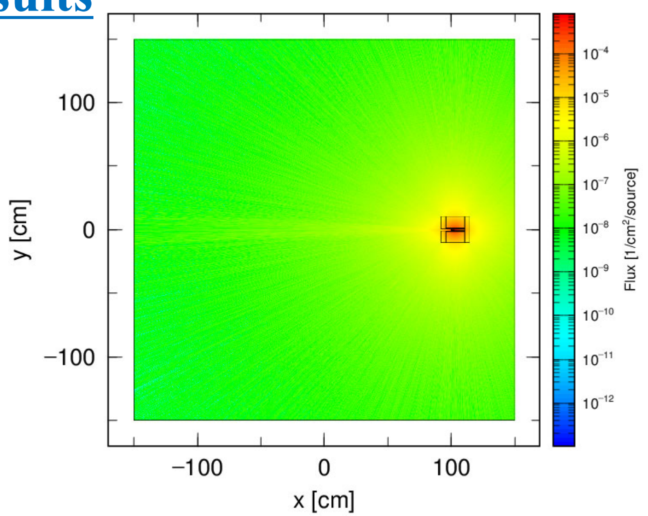


Photon Tracking results

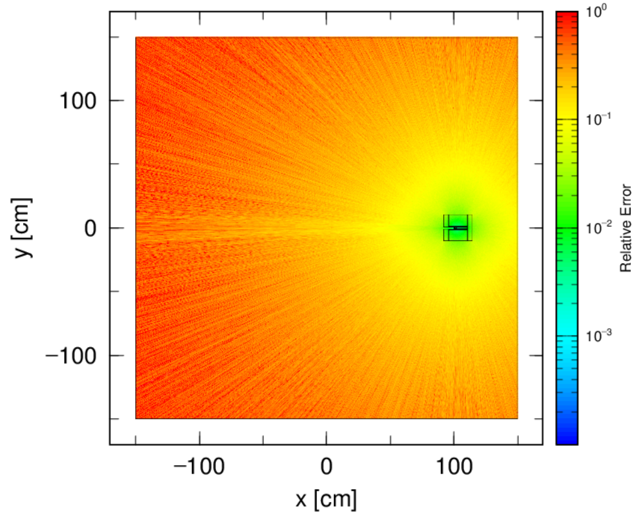


Relative error in Photon Tracking

- **Results**

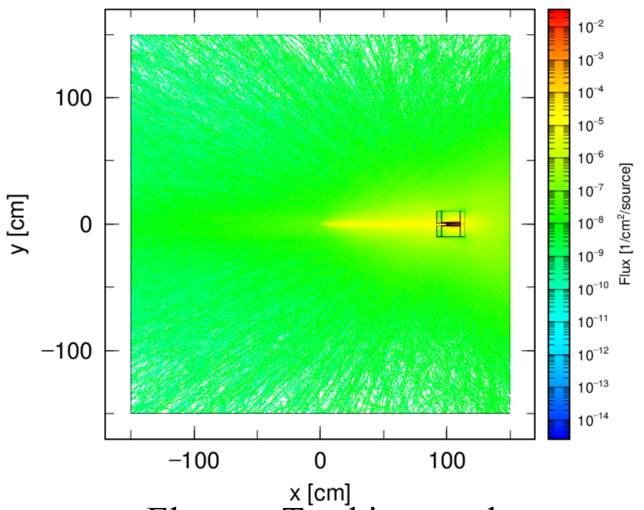


Neutron Tracking results

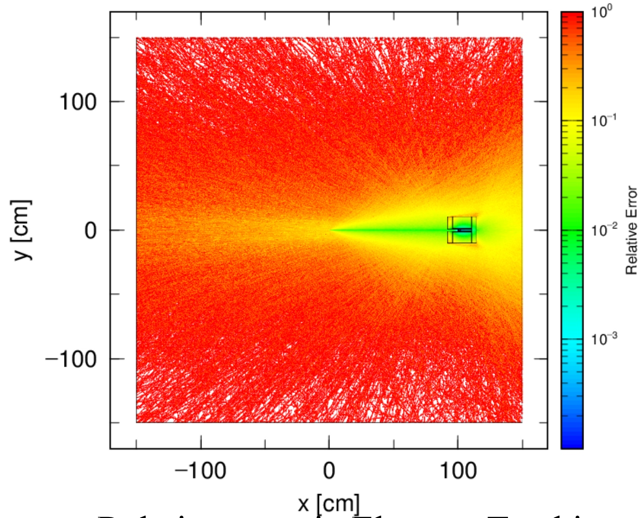


Relative error in Neutron Tracking

Results show that neutrons angular distribution of emitted neutrons is fairly isotropic.



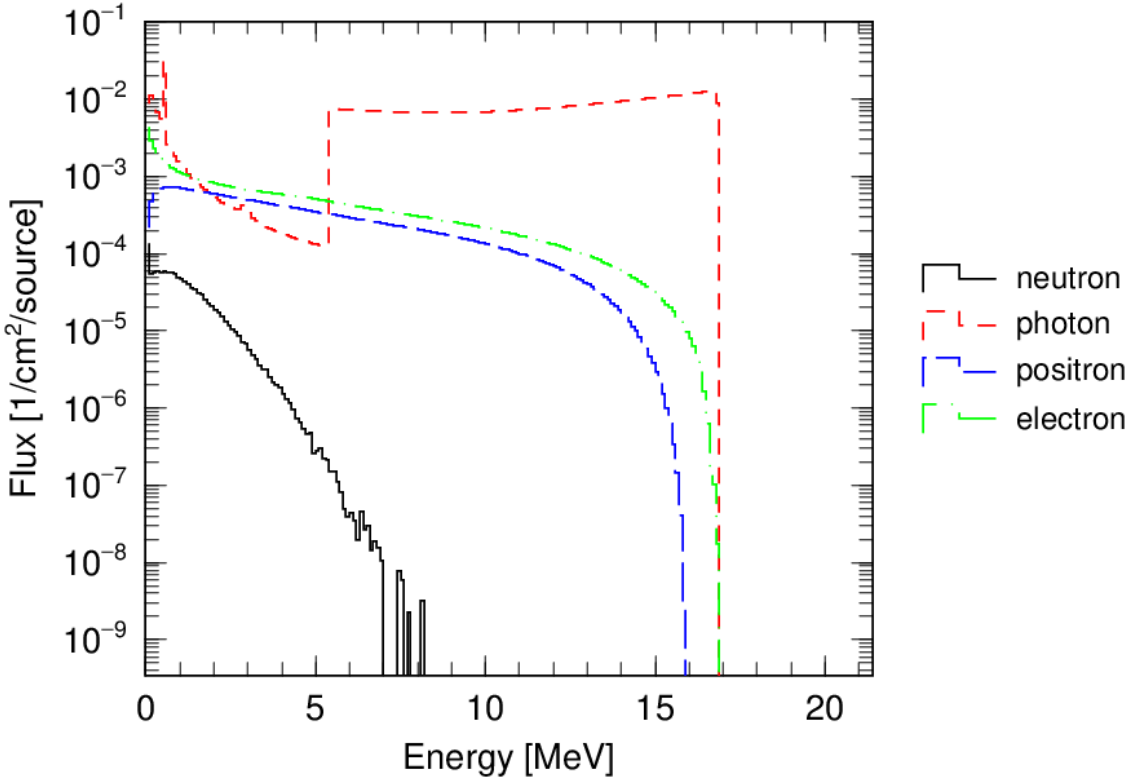
Electron Tracking results



Relative error in Electron Tracking

Hybrid Transmutation

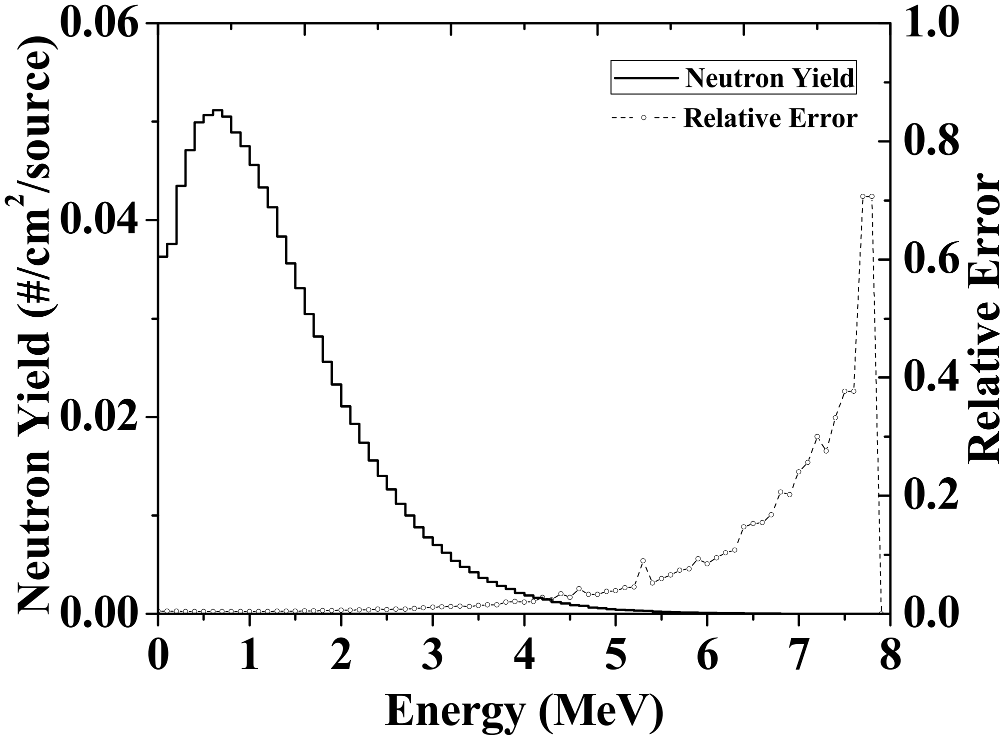
Spectrum in the primary CsI target



- The tail of photon spectrum is due to the reactions other than photonuclear GDR reaction.
- The electrons are produced because of the photonuclear reaction and positron is produced because of pair production.

Hybrid Transmutation

- **Emitted neutrons from the photonuclear reactions has**
  - Fairly isotropic angular distribution and energy distribution from the PHITS code is as follows.



  - Neutron source is modelled in MCNP6 with this energy distribution to calculate the secondary neutron transmutation reaction rates.

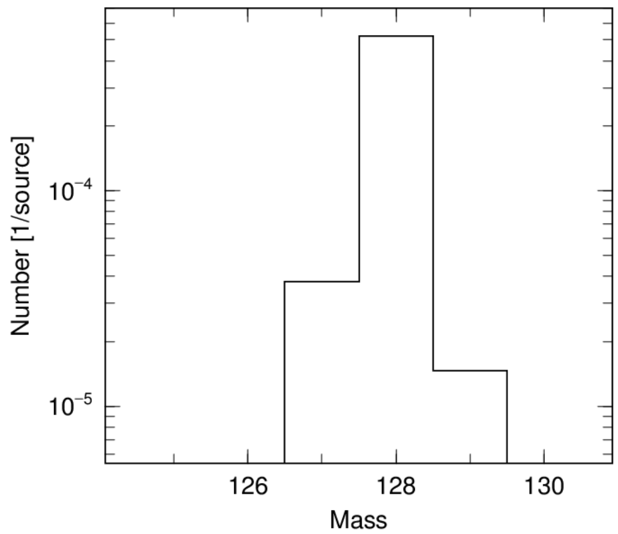
Hybrid Transmutation

Results of photonuclear transmutation in CsI target.

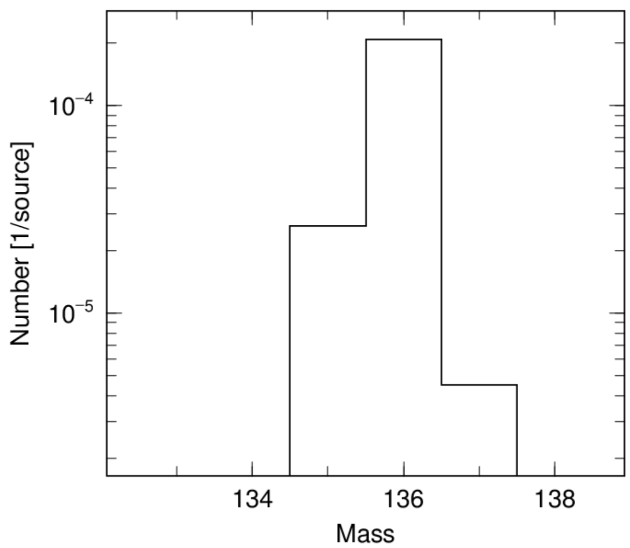
Radioisotope Conversion	Reaction rate (# /source/s)	Reaction rate (# /s)
Cs-137 $\xrightarrow{(\gamma,n)}$ Cs-136	2.09×10^{-4}	3.72×10^{9}
Cs-137 $\xrightarrow{(\gamma,2n)}$ Cs-135	2.69×10^{-5}	4.79×10^{8}
Cs-135 $\xrightarrow{(\gamma,n)}$ Cs-134	1.09×10^{-4}	1.94×10^{9}
Cs-135 $\xrightarrow{(\gamma,2n)}$ Cs-133	6.17×10^{-6}	1.10×10^{8}
I-129 $\xrightarrow{(\gamma,n)}$ I-128	5.25×10^{-4}	9.35×10^{9}
I-129 $\xrightarrow{(\gamma,2n)}$ I-127	3.91×10^{-5}	6.96×10^{8}

- *It is clear that (γ,2n) reaction is not significant and the net transmutation is much effective for both iodine and cesium.*
- *A similar run of PHITS code has been done to verify the the previously reported results.*

Iodine-129

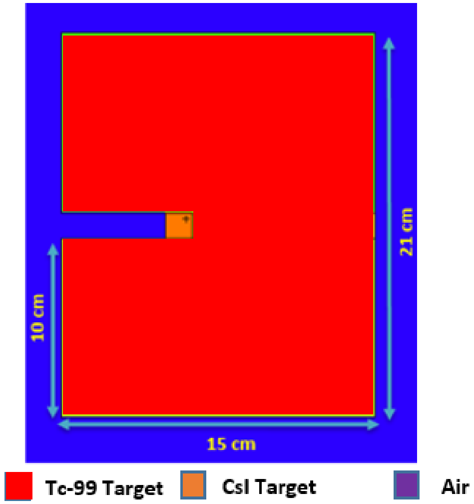


Cesium-137

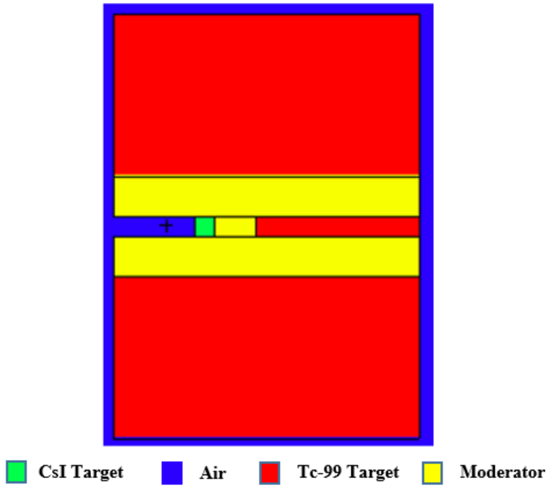


Hybrid Transmutation

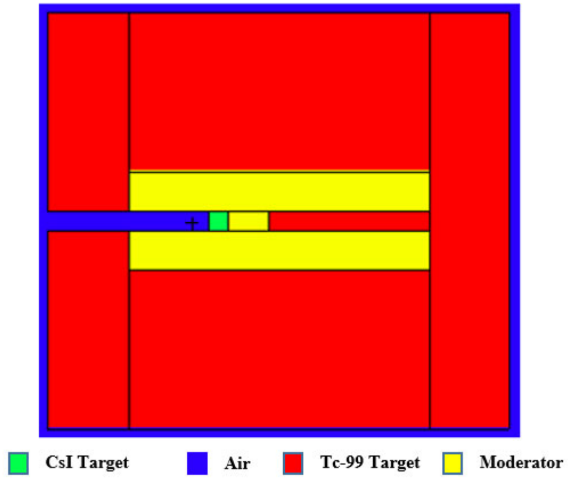
- **To simulate the results for neutron capture transmutation**
  - **A blanket of Tc-99 is surrounding CsI Target is modelled in MCNP.**



Case 1 (without moderator)



Case 2 (with moderator)



Case 3 (with moderator and additional Tc-99 on sides)

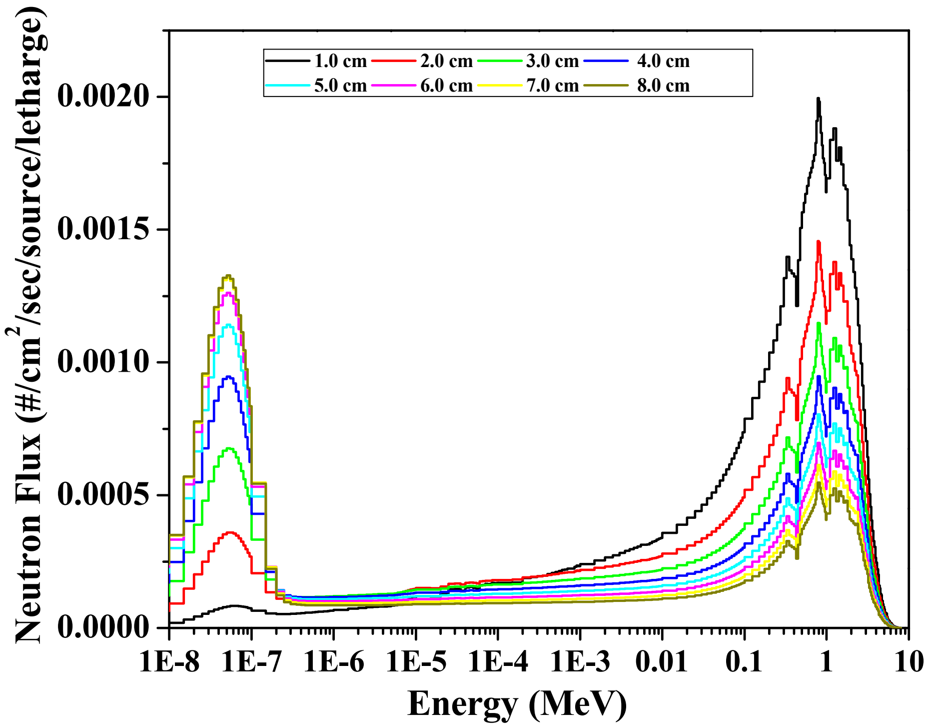
- **Case 1 Results**

W/o moderator case	RR ± % RE ($\#/cm^3/s$)	RR ± % RE ($\#/s$)
	$1.99\times10^5 \pm 0.01$	$1.32\times10^9 \pm 0.01$

Hybrid Transmutation

Case 2

- In case 2 addition of water moderator
  - Thermalize the neutron spectrum (more transmutation as Tc-99 has higher cross-section for thermal neutrons).
  - Provides additional cooling.
- Neutron flux is clearly thermalized
  - Total neutron yield is reduced with increased moderator thickness.
- This is due to the increased surface area and leakage of neutrons.



- *Therefore, case 3 has additional Tc-99 on the sides to do additional transmutation.*

Hybrid Transmutation

Case 3 Results

Moderator Thickness (cm)	RR ± % RE ($\#/cm^3/s$)	RR ± % RE (#/s)
1.0	$3.69\times10^5 \pm 0.02$	$2.27\times10^9 \pm 0.02$
2.0	$4.19\times10^5 \pm 0.02$	$2.34\times10^9 \pm 0.02$
3.0	$4.66\times10^5 \pm 0.02$	$2.36\times10^9 \pm 0.02$
4.0	$4.81\times10^5 \pm 0.02$	$2.21\times10^9 \pm 0.02$
5.0	$5.01\times10^5 \pm 0.02$	$2.09\times10^9 \pm 0.02$
6.0	$5.19\times10^5 \pm 0.02$	$1.95\times10^9 \pm 0.02$
7.0	$5.45\times10^5 \pm 0.02$	$1.83\times10^9 \pm 0.02$
8.0	$5.94\times10^5 \pm 0.02$	$1.72\times10^9 \pm 0.02$

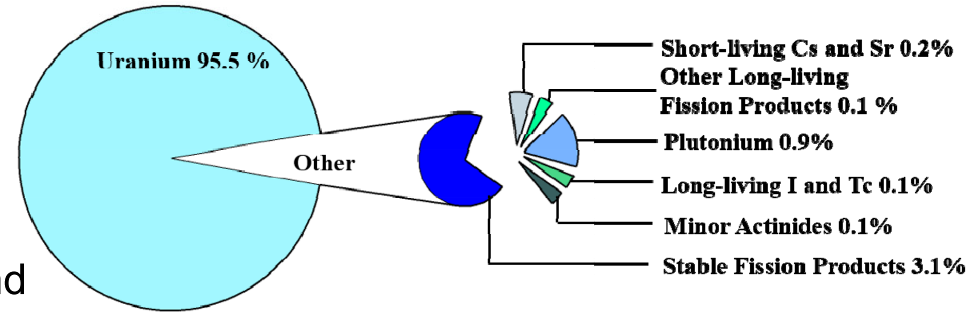
The percentage increase in reaction rates as compared to without moderator case is

Moderator Thickness (cm)	% increase in RR ($\#/cm^3/s$)	% increase in RR (#/s)
	$\frac{RR(with\ moderator) - RR({}^{w}/_{o}\ mod)}{RR({}^{w}/_{o}\ mod)} \times 100$	
1.0	85.0	71.9
2.0	110.1	77.5
3.0	133.7	78.8
4.0	141.1	67.7
5.0	150.9	58.3
6.0	160.2	48.2
7.0	173.0	39.0
8.0	197.5	30.3

Motivation

Long-lived fission product (LLFPs) in spent nuclear fuel

– LLFPs cause serious problems due to:
  - High level of toxicity
  - Long half-lives
  - Dangerous nature
    1) Cesium and Iodine: Volatile
    2) Technetium: Mobile in underground



<Composition of spent nuclear fuel for standard PWR>

– LLFPs cause serious problems due to:

Isotopes	Toxicity	Half-life (yrs.)	Repository Impact	Inventory	Production $kg/GW_{th}/yr$
^{99}Tc	Medium	Long (2.1×10^5)	High	High	8.54
^{107}Pd	Low	Long (6.5×10^6)	Low	Medium	2.34
^{129}I	Medium	Long (1.6×10^7)	Very high	Medium	1.96
^{135}Cs	Medium	Long (2.3×10^6)	Medium	Medium	4.00
^{137}Cs	High	Medium (30.0)	Low	High	8.52
^{151}Sm	High	Medium (88.8)	Low	Low	0.15

- Cs-135, Cs-137, I-129 and Tc-99 are regarded as problematic isotopes
- Those isotopes' mobility are still a big concern after Fukushima accident.

Hybrid transmutation

Concept of Hybrid transmutation

– Schematic diagram

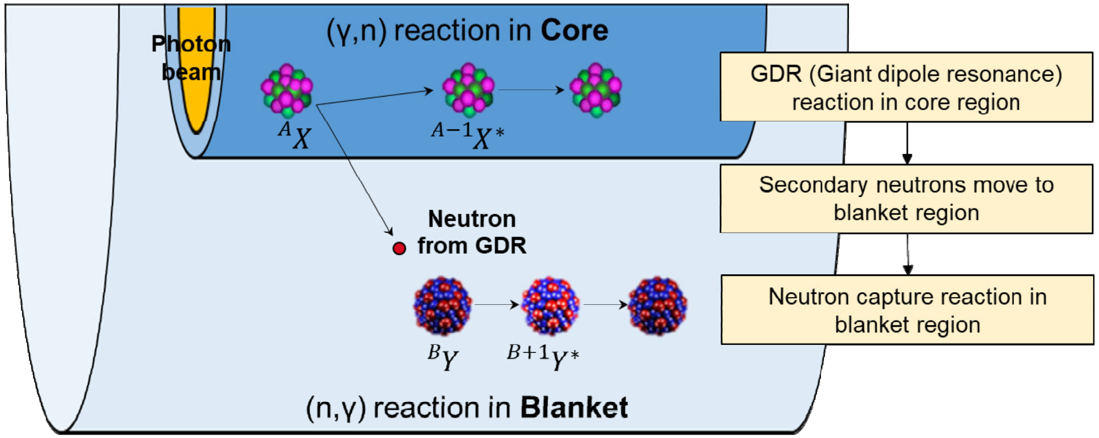


Table. Neutron capture cross-section and half-lives for important LLFPs

Isotopes (half-life)	Thermal flux	Fast flux
^{129}I (1.5E+7 yr)	3.12	0.35
^{135}Cs (2.3E+6 yr)	2.48	0.07
^{137}Cs (30.0 yr)	0.03	0.01
^{99}Tc (2.1E+5 yr)	9.32	0.45

– Main source
- Core region: LCS (Laser-Compton Scattering) Photon
- Blanket region: Neutron from (γ,n) reaction (=GDR reaction)

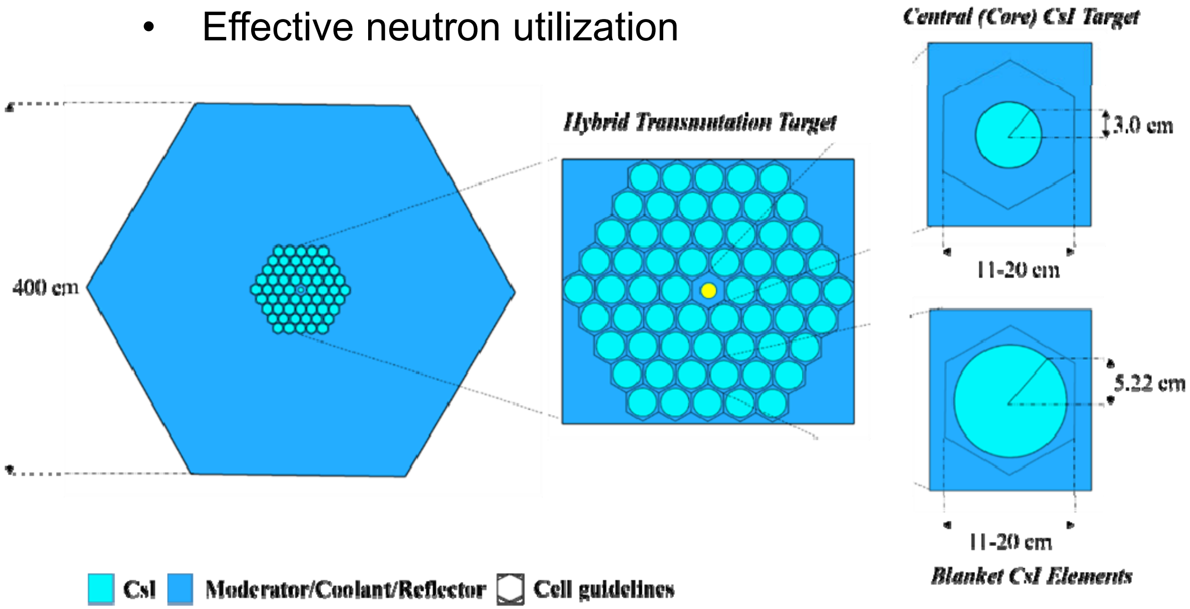
– Target material
- Core region: Isotopes with high XS in photon-field
- Blanket region: Isotopes with high XS in neutron field

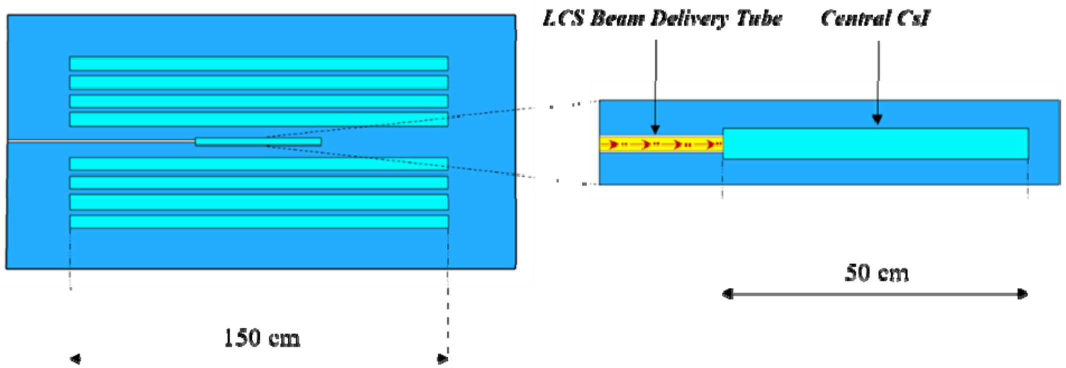
Isotopes difficult to remove in nuclear reactor
can be transmuted (e.g. ^{137}Cs and ^{135}Cs)

Hybrid transmutation

Geometry Modelling

- Advantages of hexagonal geometry
  - Tight coupling of CsI pins
  - Effective neutron utilization



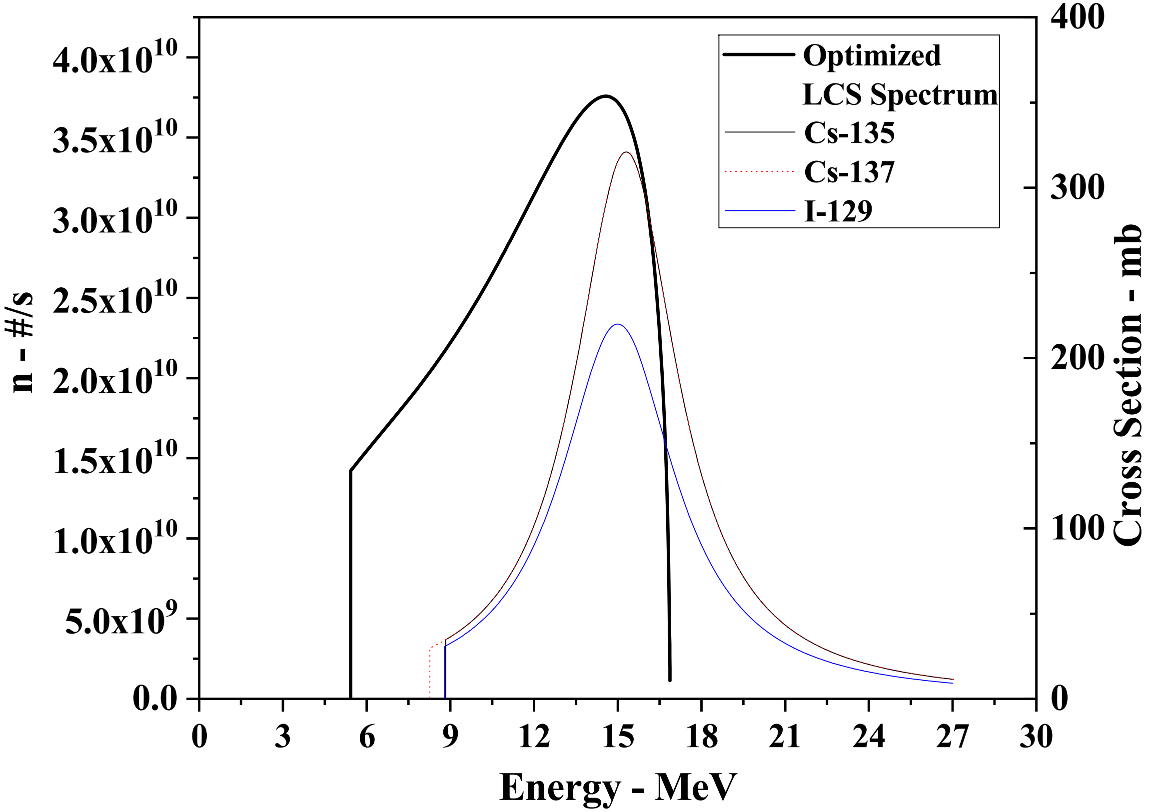


Major parameters			
Core	CsI pin	Length (cm)	49.9
		Radius (cm)	3
	Zircaloy clad	Thickness (cm)	0.05
Blanket	CsI pin	Length (cm)	19.9
		Radius (cm)	5.22
		No. of Rings	4
	Zircaloy clad	Thickness (cm)	0.05
Delivery tube	Air channel	Thickness (cm)	0.75/ 1.75
	Zircaloy clad	Thickness (cm)	0.25
Moderator/ Coolant		Light/Heavy water	
Reflector/ Shielding		Light/Heavy water	

Hybrid transmutation

How to maximize transmutation rate

– Optimization: LCS beam



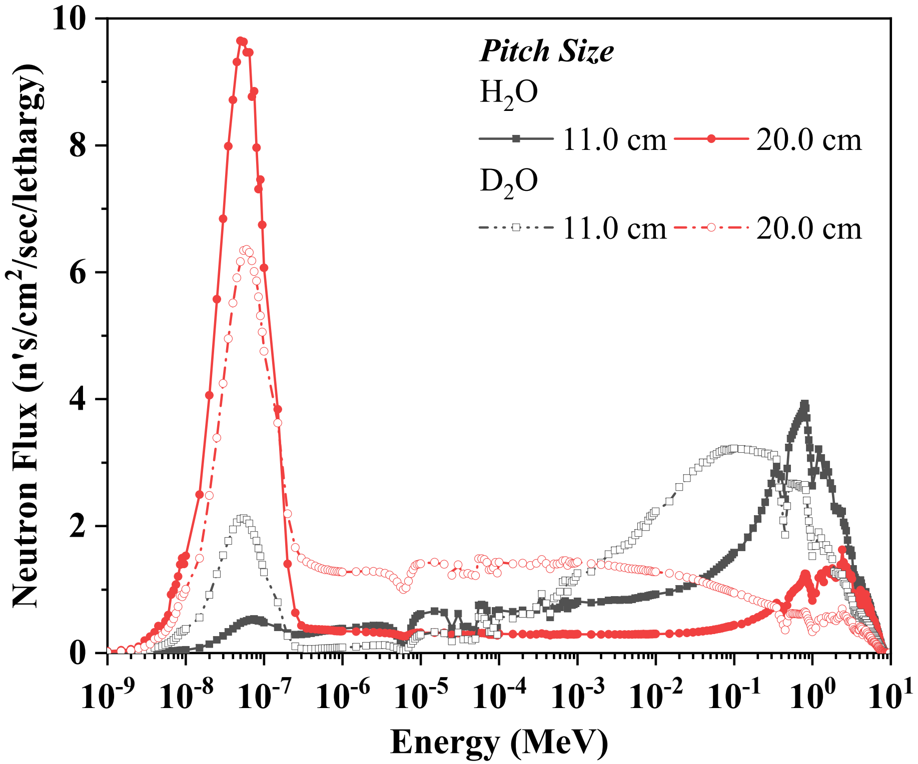
Design Parameter	
Electron beam energy	750 MeV
Incident laser energy	2.0 eV
Electron beam current	100 mA
RMS of electron beam	70 μm
Pulse energy	1.80 μJ
Amplification factor (Laser super cavity)	3000
Cone angle (Collimator)	1 mrad

- Cone angle to avoid excessive heating in target material
- Design parameters used in ERL (Slide 10)

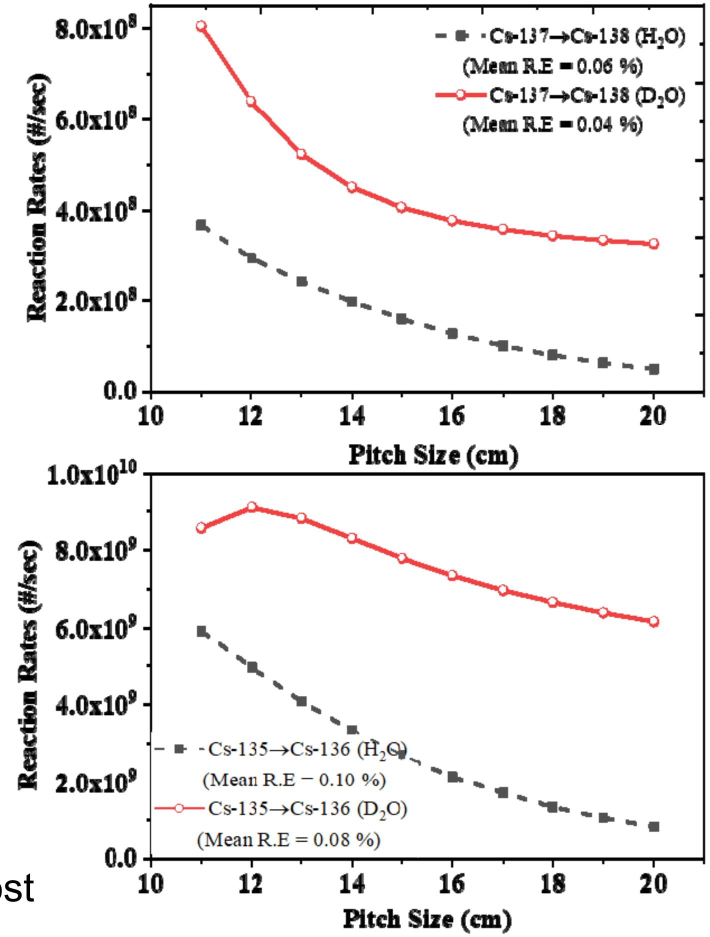
Hybrid transmutation

How to maximize transmutation rate

– Optimization: Pitch size of Blanket region



e.g.) Transmutation rate of Cesium isotopes with different pitch size



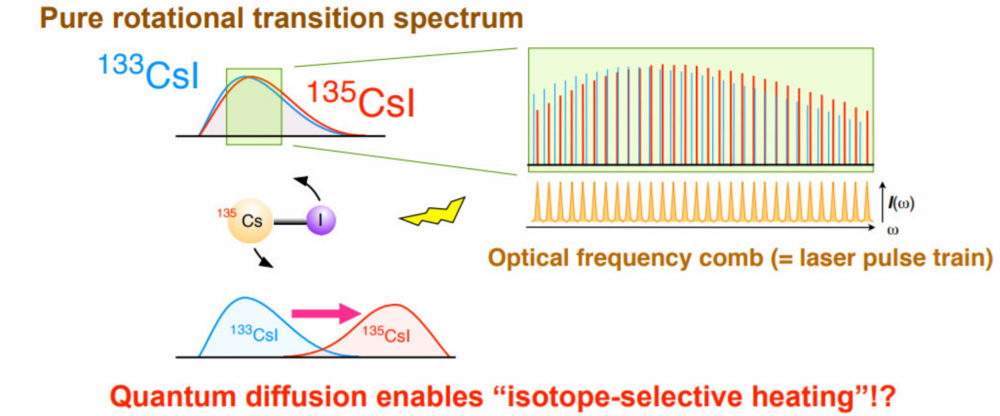
- It affects neutron moderation and absorption
- Reaction rate for the case of heavy water are almost 18.3 % higher in the blanket region.

Hybrid transmutation

Total transmutation rate

Reaction type		Transmutation rate (#/sec)	
Moderator		H_2O	D_2O
(γ,n)-reaction	Central CsI	1.55×10^{11}	1.55×10^{11}
	Blanket	4.50×10^{7}	4.04×10^{7}
(n,γ)-reaction	Central CsI	1.71×10^{10}	7.34×10^{9}
	Blanket	1.40×10^{11}	1.66×10^{11}
Total		3.12×10^{11}	3.28×10^{11}

– Potential enhancements
  - Isotopic separation can enhance transmutation rate in both regions.



Laser isotopic separation with quantum diffusion studied by Keiichi Yokoyama's research group in Japan

  - Beam intensity may be increased by technological shifts (ERL or XFELO).

Conclusions

1) LLFPs such as Cs-135, Cs-137, and I-129 can be transmuted by using the LCS-based GDR photo-nuclear reaction.

2) CsI can be a potential chemical form for Cs and I target in the photo-transmutation.

3) A photon and neutron hybrid transmutation can be a potential way for simultaneous transmutation of {Cs and I} or {Cs, I, and Tc}.

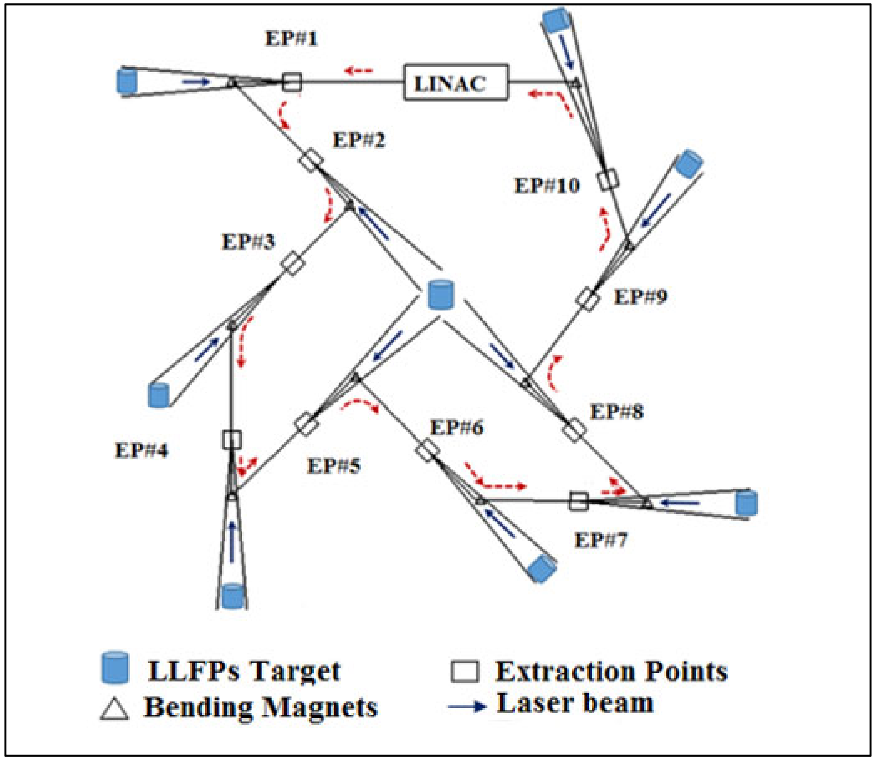
Backup Slide

MUltiple Laser-Compton scattering EXtraction (MULEX) Concept

MUltiple Laser-Compton scattering EXtraction (MULEX)

- Single LCS beam is produced from a single electron accelerator consumes higher electrical power.
- However, multiple LCS beams can be produced from single electron accelerator.

Design I



Design II