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A Comparison of Laser-induced Bremsstrahlung and Laser Compton Scattering for (γ, n) Photo-transmutation of Hazardous Nuclear Waste

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1. Introduction

Hazardous nuclear waste can pose serious problems due to high level of toxicity and very long half-life [1]. It can be transmuted into stable nuclides using photonuclear (γ, n) reactions. In this paper a detailed methodology to calculate transmutation reaction rates using Laser Induced Bremsstrahlung (LIB) and Laser Compton Scattering (LCS) has been discussed. The methodology was validated by comparing the calculated reaction rates against published data in publically-accessed literatures. In the second half of the paper, the authors present a novel concept to narrow down the LCS photon spectrum to an energy range that matches with the resonance region of a particular radionuclide. This is particularly useful considering hazardous waste is usually a mix of different isotopes. As such, being able to tune the LCS photon into any narrow energy range so as to selectively transmute any particular isotope of interest in the hazardous waste mixture would be very desirable. LCS spectrum is highly sensitive to the electron beam energy, laser power, laser luminosity and Compton backscattering angle [2]. This paper also presents sensitivity analysis to yield the maximum possible photo-transmutation rates.

2. Laser-Induced Bremsstrahlung (LIB) Transmutation Reaction Rates

It is a very well-known phenomenon that plasma is created when an optical laser beam having wavelength of 800 nm is imparted on a Ta-181 (2mm thick) target. Electron beam emerging from this plasma generally has Boltzmann-like distribution with hot electron temperature 'k_B T' [1] described as:

$$k_B T(\mathrm{MeV}) = 0.511 \times \left[\left(1 + \frac{I}{1.37 \times 10^{18}} \lambda^2\right)^{1/2} - 1\right], \quad (1)$$

where 'I' is the laser beam intensity and 'λ' is laser wavelength. The Boltzmann distribution for the calculated hot electron temperature is:

$$\frac{dN_e}{dE_e} = N_0 E_e^2 \exp\left(-\frac{E_e}{k_B T}\right), \quad (2)$$

where 'N_0'is normalization constant in MeV^{-3}. This electron beam subsequently generates bremsstrahlung photons with a cross-section of:

$$\frac{d\sigma}{dE_\gamma} = aZ^2(E_\gamma^{-1} - bE_e^{-1}), \quad (3)$$

where Z is the atomic number of the plasma-inducing material, 'a' is 11mb and 'b' is 0.83. Eq. (1) and (2) can be used to calculate the bremsstrahlung photons spectrum. The following Eq. (4) is meanwhile used to determine the bremsstrahlung photon spectrum [4]:

$$\frac{dN_\gamma}{dE_\gamma} = nd \int_{E_\gamma}^{Ec} \frac{d\sigma}{dE_\gamma} \frac{dN_e}{dE_e} dE_e, \quad (4)$$

where 'n' and 'd' is number density (atoms/cc) and thickness (cm) of the plasma-induced material. The limits of integration vary from the (generated) gamma-ray energy to electron cut-off energy in MeV. Using this bremsstrahlung photon spectrum and Giant Dipole Resonance (GDR) cross-section of the radionuclide, the transmutation reaction rate is therefore:

$$N_{reac} = n_{tar} d_{tar} \int_{Eth}^{Ec} \sigma_{reac} N_\gamma \, dE_\gamma, \quad (5)$$

where 'n_{tar}' and 'd_{tar}' is number density (atoms/cc) and thickness (cm) of the target radionuclide. 'σ_{reac}' is GDR cross-section of any radionuclide and is supposed to follow Lorentzian distribution described in Eq. (6). 'Eth' is threshold energy and 'Ec' is upper-cutoff defined by the maximum gamma-ray energy.

$$\sigma_{reac} = \sigma_{max} \left[4\left(\frac{E_{max} - E_\gamma}{\Gamma}\right)^2 + 1\right]^{-1}, \quad (6)$$

where 'σ_{max}' is maximum cross-section of the radionuclide with corresponding energy of 'Emax'. 'Γ' is Full Width Half Maximum of the radionuclide. All data related to the photo-neutron cross-section and reaction rate calculations are tabulated in Table I, which is used for the calculations of LIB and LCS transmutation reaction rates presented in this paper. Note that the listed cutoff energy in Table I is different in other sections since the value is based on the maximum gamma energy of the photon. This difference, when occurs, is nonetheless explicitly stated to avoid ambiguity.

Table I. Data for the evaluation of various important parameters [6]

Nuclide	d (cm)	n(#/cc) ($\times 10^{22}$)	Eth (MeV)	Γ (MeV)	Emax (MeV)	σ_{max} (mb)
Co-60	1.0	8.93	10.4	6.0	17.6	68.5
Sr-90	1.0	1.76	7.80	4.0	16.3	215
Zr-93	1.0	4.21	6.73	3.3	15.8	168
Pd-107	1.0	6.80	6.54	7.1	15.9	199
Sn-126	1.0	3.49	8.49	4.9	14.2	284
I-129	1.0	2.30	8.80	5.0	15.0	220
Cs-135	1.0	0.83	8.83	4.5	15.3	321
Cs-137	1.0	0.83	8.27	4.5	15.3	321
U-238	1.0	4.80	6.15	3.0	11.3	317
Ta-181	0.2	5.54	7.58	5.0	12.7	367

3. Laser Compton Scattering (LCS) Transmutation Reaction Rates

In Laser Compton Scattering when high energy beam of electron interacts with relatively very low energy laser photon usually photon is scattered with very high energy and this scattered photon can be used for the radionuclide transmutation [7].

The photon spectrum of laser Compton scattering can be calculated using Eq. (7).

$$\frac{dN_\gamma}{dE_\gamma} = \frac{N_\gamma}{\sigma_t} \int_{E-\delta E}^{E+\delta E} \frac{d\sigma}{dE_\gamma} \frac{1}{\sqrt{2\pi}\,\delta_E} \exp\left[-\frac{(E_e-E_0)^2}{2\delta_E^2}\right] dE_e, \quad (7)$$

where 'N_γ' is the product of laser power, electron current and luminosity of the laser. 'E_0' is the electron beam central energy and 'δ_E' is the deviation of the electron beam central energy. 'σ_t' is the total differential energy cross section and can be calculated by the Eq. (8).

$$\sigma_t = \int \pi r_0^2 \frac{1-\beta^2}{(1-\beta\cos\theta)^2} \frac{(1+\beta)E_l}{E_\gamma^2\left(\beta-\frac{E_l}{E_0}\right)} R^2 \times (R + R^{-1} + \cos^2\theta_{ERF})\, dE\gamma, \quad (8)$$

where 'β' is the electron velocity in terms of speed of light, 'θ_{ERF}' is scattering angle in electron frame of reference and 'R' is ratio of photon energy before and after collision. Using the above expression the 'σ_t' is calculated to be 660 mb. Rest of the parameters in Eq. (7) depends on the experimental facility. In this paper all the calculations are done for the Shanghai Laser Electron Gamma Source (SLEGS) Facility. In Table II some important data used for the reaction rates calculation is given. SLEGS facility use SSRF (Shanghai Synchrotron Radiation Facility) that can run in multi batch mode [7].

Table II. SLEGS Facility Data [7]

Parameter	Value
Electron beam energy	3.5 GeV
Laser beam energy	0.11165 eV
Laser Power	100W
Luminosity	6.5×10^{7} $W^{-1}A^{-1}s^{-1}$
Current	300 mA
δ_E	3.5 MeV

Finally the transmutation reaction rate can be calculated using Eq. (5).

4. LCS and Resonance Region Interaction

LCS spectrum has a very wide energy distribution similar to the bremsstrahlung spectrum, but unique feature of LCS spectrum is that gamma ray yield is almost flat throughout the energy distribution as shown in Fig. 3. As the resonance region for (γ,n) reaction of every radionuclide varies a lot with respect to energy. In comparison LIB the yield of gamma ray decreases exponentially with high energies so the radionuclides having high threshold gamma energy for the reaction are less probable to be transmuted using this method. The most important thing noted during this study is that gamma ray spectrum can be narrow down by the use of collimators, controlling the backscattered angle. If the backscattering angle is small then the energy distribution will also have a sharp peak. The reduction depends on the possibility of reducing the backscattering angle. This may not look benefit at all as the reaction rate for the radionuclide may be somewhat small as the overlapping region reduces. The important application of this comes when there is a mixture of hazardous waste and useful radionuclides. If conventional broad energy spectrum of the gamma rays is used then the possibility of transmutation of useful radionuclide will be the same as the waste product regardless the GDR cross section threshold energies are different. In this case if we reduce the gamma spectrum to particular radionuclide energy range then only that radionuclide will be transmuted. From Eq. (7) it is very clear that we can increase the yield by increasing the laser power, electron beam current or luminosity of the laser. It is very easy to increase the laser power in case of LCS because there are lasers available commercially up to gigawatt power range. The luminosity depends on the horizontal, vertical dispersion of the electron beams and transversal dispersion of the laser beam if we can reduce these parameters laser luminosity can also be increased to give much higher yield of gamma ray flux in region of interest [7]. The preliminary results of this study are reported in section 7.

5. Laser Induced Bremsstrahlung Results

In order to verify the calculation methodology first the results are verified with some of the reported results available. So, in section 5.1 the parameters used for the bremsstrahlung calculations are taken to be same as given in the reference. However in section 5.2 more calculated (more optimistic values) of these parameters are used in for the calculation of reaction rates. So the maximum reaction rate using bremsstrahlung can be calculated and compared with the LCS to assess which one is better.

5.1 Conservative reaction rate calculations

The parameters used for transmutation rate calculations are summarized in Table III.

Table III. Parameters used for validation [4]

Parameter	Value
Intensity (W/cm^2)	1.0×10^{20}
N_0 (MeV^{-3})	1.0×10^{7}
k_B T (MeV)	1.0
Electron cutoff energy(MeV)	20.0

These parameters are used for the conservative calculations. Otherwise hot electron temperature for the laser beam intensity value reaches to the almost 3 MeV. In section 5.2 the much optimistic values are used to obtain the reaction rate to the maximum.

Various hazardous radionuclides like Co-60, Sr-90, Zr-93, Pd-107, Sn-126, I-129, Cs-135 and U-238 are used for the photo-transmutation calculations.

Fig. 1 shows the bremsstrahlung spectrum and cross section of all the radionuclides. It can been seen that the bremsstrahlung yield is almost zero at 20MeV or higher energy. Also, there is a threshold value of the radionuclide transmutation cross section.

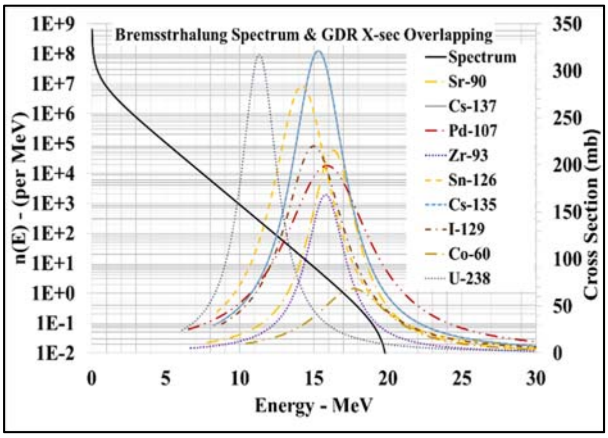

Fig. 1. Bremsstrahlung Spectrum and GDR Cross Section Overlapping (1st Approach)

5.2 Reaction rate calculations based on calculated hot electron temperature

Reaction rates calculated in section 5.1 are conservative. So, in this section all the parameters are taken as calculated by the methodology described in section 2. The parameters values are given in Table IV.

Table IV. Parameters calculated

Parameter	Value
Intensity (W/cm^2)	1.0×10^{20}
N_0 (MeV^{-3})	2.23×10^{7}
k_B T (MeV)	3.0
Electron cutoff energy(MeV)	27.8

The results of the above mentioned parameters are given in the Fig. 2. It is clear that the overall yield of the photons is increased.

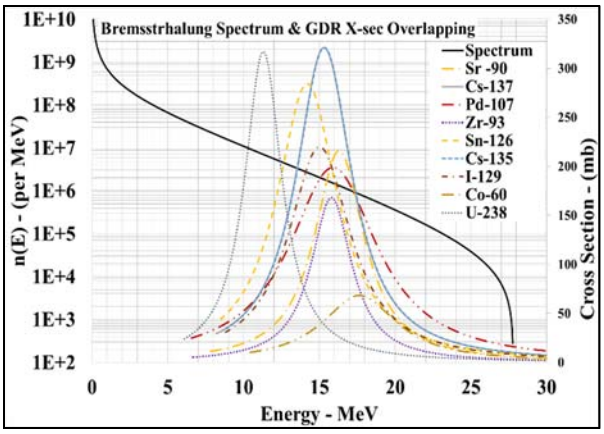

Fig. 2. Bremsstrahlung Spectrum and GDR Cross Section Overlapping (2nd Approach)

6. Laser Compton Scattering Results

Calculation methodology described in section 3 is used to develop a small computer program to study characteristic behavior of the LCS. The code is further used for the determination of gamma ray intensity and average gamma ray energy. The calculated gamma ray intensity reaches to the value 1.0×10^{8} s^{-1}. The energy spectrum of LCS gamma rays is given for all the radionuclides in the Fig. 3.

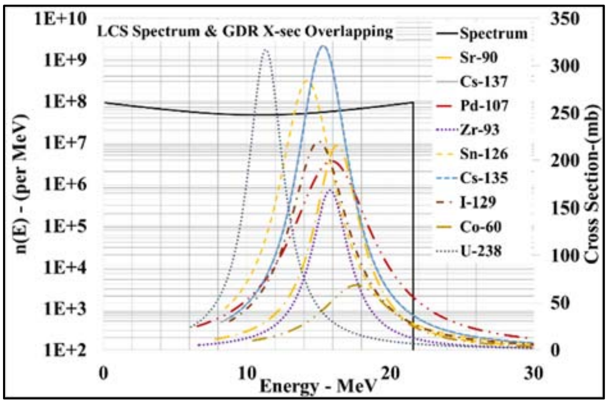

Fig. 3. LCS Spectrum and GDR Cross Section Overlapping.

7. Sensitivity Analysis of LCS Spectrum

In this section broad spectrum of the gamma ray energy is reduced by reducing the cone angle. The calculations shown in section 6 is for the broad energy spectrum if cone angle is 2mrad. Reduction in cone angle means that the emitted gamma rays scattered within that small cone angle. One more important thing is worth mentioning here as data of SLEGS facility is used in this study and the maximum energy is calculated as 21.76 MeV. Reduction in cone angle will shift the spectrum more towards the higher energy range. This means that radionuclides that have high energy for this particular facility will be transmuted. However, there is a possibility to tune this maximum energy. Maximum energy of the gamma spectrum is given by the Eq. (9). It depends both on the laser energy and electron beam energy so if somehow these parameters can be adjusted to get wishful maximum energy of the photons which further can be further tuned in to energy of GDR by adjusting the angle backscattered photons.

$$E_{max} = \frac{4\,\gamma^2 E_l}{1+\frac{4\gamma E_l}{mc^2}}, \quad (9)$$

where 'γ' is the ratio of electron beam kinetic energy to rest mass energy of the electron. So, by adjusting the laser and electron beam energy we can change the maximum energy of the photons with in a cone and hence can align it according to the resonance region of the radionuclide photo-neutron cross section.

In this study the electron beam energy and laser beam energy is adjusted to get shift the maximum photon energy near the energy of maximum cross section for most of the nuclide and then sufficiently small cone angle is taken to make the LCS spectrum as narrow as possible. Also the laser power is increased from 100W to 500W to see the effect of enhanced transmutation rate of most of the radio nuclides. Only Pd-107 and Cs-137 calculations are shown in this paper, however all the radionuclides can be treated in this manner to get reasonable transmutation rate with an added advantage that only hazardous waste products will be transmuted as discussed in the introduction.

From Table I, maximum cross section of Pd-107 occur at 15.9 MeV. Now as discussed above to shift the max energy of gamma ray in SLEGS facility which is 21.76 MeV to energy of 15.9 MeV a fine change in electron beam energy and laser energy is required. An optimization code can be written to choose to get parameters of interest. However in this paper hit and trial method is used to get optimized solution of the parameters.

For Pd-107 the electron beam energy changed from 3.5 GeV to 3.0 GeV and cone angle is taken as 2×10^{-5} rad. The Gamma spectrum along with the Pd-107 cross section are given in the Fig. 4. Similarly for Cs-137 the electron beam energy is adjusted to 2.95 GeV to get spectrum peak energy as 15.3 MeV.

Different radionuclides have different energies for maximum cross sections. In this study only electron energy is changed to achieve the nearly mono energetic spectrum matched to the max energy of the two radionuclides Pd-107 and Cs-137. It should be noted that electron beam energies and laser beam energies can be tuned only within the permissible limits. These limits depends on the maximum allowable energies that can be achieved practically.

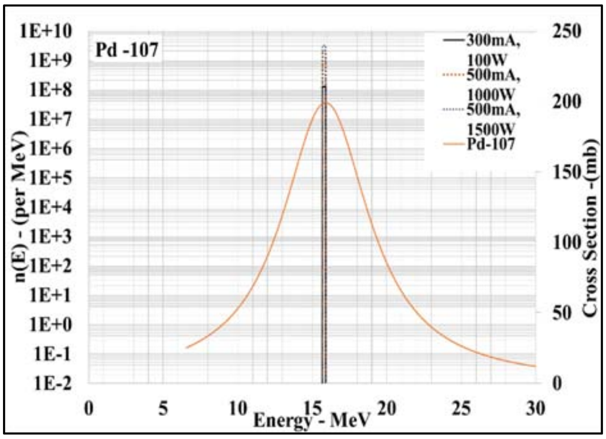

Fig. 4. Sharp resonance peak for 100 W laser power

It is quite obvious that transmutation rate will be much less as compared to broad spectrum but if we increase the laser power the spectrum position and width remain the same however yield will increase consequently results in to much higher reaction rates. The LCS spectrum for higher power and electron beam currents are shown in Fig. 5.

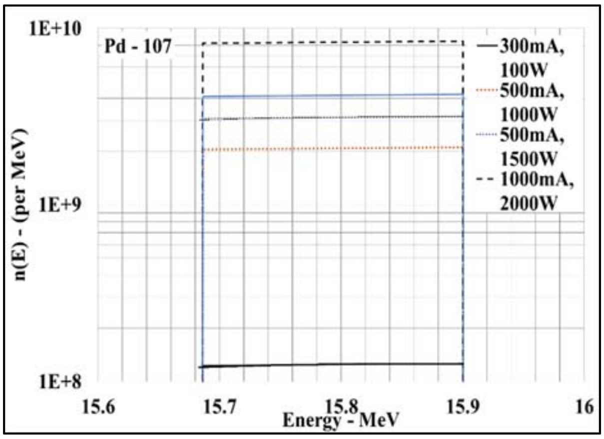

Fig. 5. Sharp resonance peaks for 100 W laser power

Table VII shows the results of all the calculations for Pd-107 and Cs-137.

8. Comparison of LIB and LCS Methods

Transmutation rate for all the above discussed radionuclides are given in the Table V & Table VI. Two different types of reaction rates are calculated for the LIB method for the similar problem. It is to be noted that reaction rates using conservative approach may underestimate the actual rate of transmutation whereas the reaction rates calculated by the alternative approach is purely based on the numerical figures obtained from the calculations. So the actual reaction rate using LIB method may be in between these two calculated reaction rates. Reaction rates calculated by LCS method is purely based in the on SLEGS facility available data in literature.

Table V. Comparison of reaction rates of LIB (conservative method) and LCS

Radionuclide	LIB (Conservative Approach)			LCS
		(10Hz)	(100Hz)	
	s^{-1}	s^{-1}	s^{-1}	s^{-1}
Co-60	0.77	7.7	77	2.7×10^6
Sr-90	2.08	20.8	208	1.2×10^6
Zr-93	6.80	68	680	2.0×10^6
Pd-107	54.1	541	5410	6.9×10^6
Sn-126	9.59	95.9	959	3.5×10^6
I-129	3.06	30.6	306	1.9×10^6
Cs-135	1.26*	12.6	126	9.3×10^5
Cs-137	1.84	18.4	184	9.3×10^5
U-238	81.3	813	8131	3.2×10^6

* Cs-135 reaction rate value is compared with the reported results of Ref [9] and rest of the values are calculated by the same procedure.

Table VI. Comparison of reaction rates of LIB (calculated values) and LCS

Radionuclide	LIB (Calculated Values)			LCS
		(10Hz)	(100Hz)	
	s^{-1}	s^{-1}	s^{-1}	s^{-1}
Co-60	6×10^4	6×10^5	6×10^6	2.7×10^6
Sr-90	4×10^4	4×10^5	4×10^6	1.2×10^6
Zr-93	8×10^4	8×10^5	8×10^6	2.0×10^6
Pd-107	3×10^5	3×10^6	3×10^7	6.9×10^6
Sn-126	2×10^5	2×10^6	2×10^7	3.5×10^6
I-129	9×10^4	9×10^5	9×10^6	1.9×10^6
Cs-135	4×10^4	4×10^5	4×10^6	9.3×10^5
Cs-137	4×10^4	4×10^5	4×10^6	9.3×10^5
U-238	1×10^5	1×10^6	1×10^7	3.2×10^6

Results of sensitivity analysis on the transmutation rate of Pd-107 and Cs-137 are as follows.

Table VII. Reaction rates calculated for sharp resonance LCS spectrum

Parameters		Reaction Rates (s^{-1})	
L.Power (W)	**Current (mA)**	**Pd-107**	**Cs-137**
100	**300**	1.8×10^5	3.1×10^4
1000	**500**	2.7×10^6	4.9×10^5
1500	**500**	4.2×10^6	7.3×10^5
2000	**500**	5.6×10^6	9.7×10^5
2000	**1000**	1.1×10^7	1.9×10^6

9. Power Consumption per unit Transmutation Rate for LIB and LCS

The power consumption per unit transmutation rate is a very good indicative of the system effectiveness. Table VIII and Table IX shows power consumed per unit transmutation rate for both LIB and LCS methods. In LIB method maximum power consumed belongs to laser intensity that is 1×10^{20} W/cm^2 where the focal area is usually 5 μm^2. In case of LCS the total power consumption is not only limited to laser power but also the power required for producing high energy beams. The typical value for the electrical power required to generate the electrical beam of energy is 3 GeV in a storage ring is about 834 kW [9]. We assume the similar power is required for 3.5 GeV energy beam as well. So, total power will be the sum of laser power and electron beam power.

From the Table VIII it is quite evident that power consumption per unit transmutation decreases if we increase the repetition rate of laser.

Table VIII. Power consumption per unit transmutation (LIB)

Power Consumption per unit Transmutation rate (LIB)			
For I = 1×10^{20} W/cm^2, P = 2.5×10^9 W (focal area is 5 μm^2)			
Radionuclide	**1 Hz**	**10 Hz**	**100 Hz**
Co-60	3.25×10^9	3.25×10^8	3.25×10^7
Sr-90	1.20×10^9	1.20×10^8	1.20×10^7
Zr-93	3.68×10^8	3.68×10^7	3.68×10^6
Pd-107	4.62×10^7	4.62×10^6	4.62×10^5
Sn-126	2.61×10^8	2.61×10^7	2.61×10^6
I-129	8.17×10^8	8.17×10^7	8.17×10^6
Cs-135	1.98×10^9	1.98×10^8	1.98×10^7
Cs-137	1.36×10^9	1.36×10^8	1.36×10^7
U-238	3.08×10^7	3.08×10^6	3.07×10^5

Table IX. Power consumption per unit transmutation (LCS)

Power Consumption per unit Transmutation rate (LCS)	
For Laser Power = 100 W, Power Electron beam = 834 kW	
Radionuclide	**Power / Transmutation Rate**
Co-60	0.309
Sr-90	0.695
Zr-93	0.417
Pd-107	0.121
Sn-126	0.238
I-129	0.439
Cs-135	0.897
Cs-137	0.897
U-238	0.261

Power consumption per unit transmutation in LCS is significantly less than in case of LIB case even for higher frequencies.

10. Conclusions

In general the possibility of radionuclide transmutation using photo-neutron reaction is evaluated in this work. From the results it is quite evident that LCS is much better option for the radionuclide transmutation as reaction rates for the LCS is much higher than LIB method even for very small laser power. It can be seen even for the optimistic reaction rate calculations with bremsstrahlung method reaction rate is much lower than LCS case for 10 Hz repetition rate. If repetition rate of laser 100 Hz then LIB reaction rate has the same order of the magnitude as the reaction rate via LCS. Higher Laser Powers can yield very high transmutation rates. Using sharp spectrum of LCS it is quite possible to achieve same or even higher reaction rates as conventional LCS method with the added advantage that it can work in mixed radionuclide environments. If decay scheme of particular radionuclide is studied closely than photo transmutation is a very effective technique of radionuclide transmutation as a whole.

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