Transmutation of nuclear wastes using photonuclear reactions triggered by Compton backscattering photons at the Shanghai laser electron gamma source^{*}

CHEN Jin-Gen(陈金根)^{1;1)} XU Wang(徐望)¹ WANG Hong-Wei(王宏伟)¹ GUO Wei(郭威)¹ MA Yu-Gang(马余刚)¹ CAI Xiang-Zhou(蔡翔舟)¹ LU Guang-Cheng(陆广成)¹ XU Yi(徐毅)^{1,2} PAN Qiang-Yan(潘强岩)¹ YUAN Ren-Yong(袁仁勇)¹ XU Jia-Qiang(徐加强)¹ YAN Zhe(阎喆)¹ FAN Gong-Tao(范功涛)^{1,2} SHEN Wen-Qing(沈文庆)¹

1 (Shanghai Institute of Applied Physics, CAS, Shanghai 201800, China)2 (Graduate University of Chinese Academy of Sciences, Beijing 100049, China)

Abstract Based on the facility of the Shanghai Laser Electron Gamma Source (SLEGS), the transmutation for nuclear wastes such as ¹³⁷Cs and ¹²⁹I is investigated. It is found that nuclear waste can be transmuted efficiently via photonuclear reaction triggered by gamma photons generated from Compton backscattering between CO₂ laser photons and 3.5 GeV electrons. The nuclear activities of ¹³⁷Cs and ¹²⁹I are evaluated and compared with the results of transmutation triggered by bremsstrahlung gamma photons driven by ultra intense laser. Due to the better character of gamma photon spectrum as well as the high brightness of gamma photons, the transmutation rate of Compton backscattering method is much higher than that of the bremsstrahlung method.

Key words radioactive wastes, photonuclear reactions, Compton backscattering

PACS 28.41.Kw, 25.20.Lj, 32.80.Cy

1 Introduction

Various long-lived radioactive nuclear wastes are generated in the courses of application of nuclear power^[1-3]</sup>. For example, there are 9 units of operational reactors in China with about only 2% of the nuclear electricity share in the total electricity generation of China. So the Chinese government has ratified recently a nuclear power plan named "Moderate Development of Nuclear Power" to improve the total installed nuclear capacity from 8700 MW at present to 40000 MW by 2020 by investing about 400 billion yuan. Accompanying this development project it is inevitable to produce more nuclear wastes in China. Although it has historically only produced a little (about 0.05%) volume of power production wastes. The very small proportion of radioactive wastes are extremely hazardous, compared with almost all other industrial wastes. The radioactive wastes from the nuclear industry have to therefore be managed responsibly. Transmutation by means of neutrons and/or charged particles, or γ rays becomes an important method for reducing the inventory of long-lived nuclear wastes. The aim is to transmute them by changing one nuclide into another via a nuclear reaction to produce shorter-lived or more stable nuclide.

Transmutation using bombardment with neutrons from a reactor or a particle accelerator has been discussed^[2, 4]. However, this method may not be the optimum approach for all nuclides. For example, it is impractical to transmute ¹³⁷Cs or ⁹⁰Sr with neutron bombardment because of the very low neutron capture cross section^[5, 6]. Thus transmutation via

Received 11 October 2007

^{*} Supported by Knowledge Innovation Project of Chinese Academy of Sciences (KJCX2-SW-N13), China Postdoctoral Science Foundation, National Natural Science Foundation of China (10475108, 10605036, 10405032), One Hundred Person Project of SINAP, and Shanghai Development Foundation for Science and Technology (06QA14062)

¹⁾ E-mail: chenjg@sinap.ac.cn

photonuclear reactions (γ, n) is a suggested option. Currently, there are two different feasible methods for producing γ -ray to trigger photonuclear reaction for transmutation of nuclear waste. One is to use a laser-driven γ -ray coming from the bremsstrahlung of electrons produced in ultra intense laser-solid interaction^[7, 8]. For the details of laser-driven technology one can refer to Refs. [9—15]. The other is to adopt γ -ray produced from Compton backscattering^[16] of laser photons on high energy electrons from synchrotron ring.

2 Compton backscattering photon spectrum and calculations

Compton backscattering gamma photon spectrum has a rather flat energy distribution with small spreading compared with the bremsstrahlung spectrum, and which plays an important role in the transmutation of nuclear waste. With the construction and operation of synchrotron radiation facility around the world, transmutation based on laser Compton scattering process becomes practicable^[17].

In this paper, we will report on the evaluated results for transmutation of nuclear waste triggered by laser-electron Compton backscattering, which will be realized at the Shanghai Laser Electron Gamma Source (SLEGS) facility^[18]. The transmutation of ¹³⁷Cs and ¹²⁹I will be focused in this work. These two nuclides are the main long-lived fission products and are very hazardous for living beings. In addition, they are less mobile, soluble and highly absorbed by bloodstream and organs of human, and thus may cause many fatal diseases^[19]. More important is that these two nuclides cannot be transmuted efficiently by means of (n, γ) reactions with neutron bombardment. So it is timely and interesting to investigate the transmutation for 137 Cs and 129 I via (γ , n) reactions triggered by Compton backscattering gamma photons generated from SLEGS.

The SLEGS facility will be built as one of the beam lines at the Shanghai Synchrotron Radiation Facility (SSRF), which is the third generation synchrotron radiation facility and will function by $2009^{[20]}$. γ -ray with energy up to about 22 MeV can be produced by Compton backscattering of the far infrared CO₂ laser photons ($\lambda = 10.64 \ \mu\text{m}$) on the 3.5 GeV electrons circulating in the synchrotron ring of SSRF. The intensity of electrons stored in the synchrotron ring of SSRF can reach about 10⁷ s⁻¹ and the power of the selected CO₂ laser is 100 W. This results in the total brightness of γ -ray generated from the SLEGS facility to be the order of 10⁹ s^{-1[18]}. This high brightness γ -ray provides a favorable opportunity to transmute nuclear waste efficiently. The schematic illustration for transmutation of nuclear waste based on Compton backscattering is displayed in Fig. 1. In this figure we assume that ¹³⁷Cs is transmuted as an example. The scenario for transmutation of other nuclear wastes should be similar to that of ¹³⁷Cs. ¹³⁷Cs with a half-life of $T_{1/2}=30.17$ a in the radioactive target is transformed into ¹³⁶Cs due to a (γ , n) reaction. Then ¹³⁶Cs (with a half-life of $T_{1/2}=13.16$ d) beta decays to the stable nuclide ¹³⁶Ba. In parallel, the nuclide ¹²⁹I, which has a half-life of $T_{1/2}=1.577 \times 10^7$ a, can be transmuted into ¹²⁸I via (γ , n) reaction. Then ¹²⁸I ($T_{1/2}=25$ min) decays to ¹²⁸Xe with a ratio of 93%.



Fig. 1. The process of Compton scattering between a laser photon and an electron as well as the transmutation via (γ, n) reaction for ¹³⁷Cs as an example. If a laser photon has a head on collision with an electron, this process is named Compton backscattering.

As mentioned above, the energy spectrum of γ ray generated from Compton backscattering of laser photons and high energy electrons decides the transmutation rate and it can be derived from classical electromagnetic theory. Under the consideration of electron beam energy spread in the storage ring, the energy spectrum $n_{\gamma}(E_{\gamma})$ can be expressed as

$$\begin{split} n_{\gamma} &= \frac{\mathrm{d}N_{\gamma}}{\mathrm{d}E_{\gamma}} = \\ & \frac{N_{\gamma}}{\sigma_{\mathrm{t}}} \int \frac{\mathrm{d}\sigma}{\mathrm{d}E_{\gamma}} \frac{1}{\sqrt{2\pi}\delta_{E}} \exp\left[-\frac{(E_{\mathrm{e}} - E_{0})^{2}}{2\delta_{E}^{2}}\right] \mathrm{d}E_{\mathrm{e}}, \ (1) \end{split}$$

r

where N_{γ} is the total number of gamma photons generated from Compton scattering per second, which can be defined as $N_{\gamma} = PIL$. Here P and I are the laser power and the electron beam current (300 mA) for the SSRF storage ring, respectively. L is the Compton backscattering γ -ray luminosity which is about $6.5 \times 10^7 \text{ A}^{-1} \text{W}^{-1} \text{s}^{-1[18]}$. So N_{γ} can be determined to be $1.95 \times 10^9 \text{ s}^{-1}$. E_{γ} is the energy of gamma photons and $d\sigma/dE_{\gamma}$ is the differential Compton scattering cross section, which can be derived from Klein-Nishina formula^[21]. $\sigma_{\rm t}$ is the total Compton scattering cross section (660.58 mb), which can be obtained by integrating $d\sigma/dE_{\gamma}^{[18]}$. $E_{\rm e}$ and E_0 are the electron beam energy and the electron beam central energy (3.5 GeV) in the storage ring, respectively. δ_E is the corresponding energy deviation ($\delta_E = 3.5 \text{ GeV} \times 0.1\% = 3.5 \text{ MeV}$).

Now having the γ -ray spectrum, the number (N) of transmutation reactions per second can be evaluated with the help of the corresponding photonuclear cross section $\sigma(E_{\gamma})$ by the following expression

$$N = nd \int_{E_{\rm th}}^{E_{\rm up}} \sigma(E_{\gamma}) n_{\gamma}(E_{\gamma}) \mathrm{d}E_{\gamma}, \qquad (2)$$

where n and d are the density and thickness of target nuclide ¹³⁷Cs or ¹²⁹I, respectively. $E_{\rm th}$ and $E_{\rm up}$ are the energy threshold of photonuclear reaction and maximum energy of γ -ray, respectively. Generally, photonuclear cross section $\sigma(E_{\gamma})$ can be assumed as a Lorentzian-like shape, which has the form^[22]

$$\sigma(E_{\gamma}) \approx \sigma_{\max} / \left[4 \left(\frac{E_{\max} - E_{\gamma}}{\Gamma} \right)^2 + 1 \right], \qquad (3)$$

where σ_{\max} is the maximum cross section at E_{\max} and Γ the full width at half maximum cross section.

As we know, there are no available experimental data for 137 Cs and 129 I photo nuclear cross sections, so here we have to adopt the experimental data for 133 Cs and 127 I, respectively^[22]. This method has also been applied in some other work^[13, 15]. The related parameters to be input in the evaluation of transmutation are summarized in Table 1.

Table 1. Related parameters of ¹³⁷Cs and ¹²⁹I in calculations for transmutation.

nuclide	^{137}Cs	^{129}I
d/cm	1.0	1.0
n/cm^{-3}	8.36×10^{21}	2.30×10^{22}
$\sigma_{\rm max}/{ m mb}$	321	220
$E_{\rm max}/{\rm MeV}$	15.31	15.00
$\Gamma/{ m MeV}$	4.50	5.00
$E_{\rm th}/{\rm MeV}$	8.83	8.80
$E_{\rm up}/{\rm MeV}$	21.75	21.75

3 Results and discussion

Figure 2 shows the γ -ray spectrum produced from Compton backscattering of the 100 W CO₂ laser photons and the 3.5 GeV electrons, as well as the photo nuclear cross sections of $^{137}\mathrm{Cs}$ and $^{129}\mathrm{I}.$ One can see that the Compton backscattering γ photon spectrum has an excellent character. It can keep very high brightness (about 10^8 photons/MeV order) from 0 MeV to the upper limit of 21.75 MeV. This is significantly different from the bremsstrahlung γ photon spectrum, which has a high flux at low energy but drops rapidly with the increase of the photon energy^[7, 15]. In particular, the ratio of Compton backscattering and bremsstrahlung photon flux can reach 10^4 — 10^6 at the energy region close to $E_{\rm max}$, where the photon flux plays an important role in the transmutation of 137 Cs or 129 I. Therefore the flat γ ray spectrum from Compton backscattering will lead to a much higher transmutation rate than that via the bremsstrahlung method.





The calculated results for ¹³⁷Cs and ¹²⁹I are listed in Table 2. The results for transmutation triggered by the bremsstrahlung method at three laser repetition rates are also presented for comparison. The produced number (N) per second based on the Compton backscattering method is 1.26×10^6 for ¹³⁶Cs and 2.55×10^6 for ¹²⁸I, whereas the produced number per second based on the bremsstrahlung method is 2.20×10^4 for ¹³⁶Cs and 5.20×10^4 for ¹²⁸I if the laser repetition is assumed to be 1000 Hz. In practice, it is very difficult to obtain a 1000 Hz pulse repetition

Table 2. The calculated results for transmutation of 137 Cs and 129 I via Compton backscattering (denoted by Comp.). N (136 Cs) and N (128 I) denote the numbers of 136 Cs and 128 I generated from the photo nuclear reactions per second, respectively. A (t=30 min) represents the nuclear activity for 136 Cs or 128 I after being irradiated for 30 min. The results from the bremsstrahlung method (denoted by Brem.) are also displayed for comparison. The laser intensity is 10^{20} W/cm², and the laser repetition rates are assumed to be 10, 100 and 1000 Hz, respectively.

,	, 1	5
	^{137}Cs	
	Comp.	Brem.
$N(^{136}Cs)/s^{-1}$	1.26×10^{6}	$\begin{array}{c} 220 \ (10^1 \ \mathrm{Hz}) \\ 2200 \ (10^2 \ \mathrm{Hz}) \\ 22000 \ (10^3 \ \mathrm{Hz}) \end{array}$
A(t=30 min)/Bq	1388.19	$\begin{array}{c} 0.24 \ (10^1 \ \text{Hz}) \\ 2.4 \ (10^2 \ \text{Hz}) \\ 24 \ (10^3 \ \text{Hz}) \end{array}$
	^{129}I	
	Comp.	Brem.
$N (^{128}I)/s^{-1}$	2.55×10^6	$\begin{array}{c} 520 \ (10^1 \ \text{Hz}) \\ 5200 \ (10^2 \ \text{Hz}) \\ 52000 \ (10^3 \ \text{Hz}) \end{array}$
$A \ (t=30 \text{ min})/\text{Bq}$	1.44×10^{6}	$\begin{array}{c} 294.85~(10^1~{\rm Hz})\\ 2948.5~(10^2~{\rm Hz})\\ 29485~(10^3~{\rm Hz}) \end{array}$



Fig. 3. The number of reactions for 137 Cs (Upper panel) and 129 I (Lower panel) as a function of irradiation time. The solid line denotes the result for the Compton backscattering method at SLEGS. The dash, dot and dash-dot lines stand for the results for the bremsstrahlung method driven by the 10^{20} W/cm² laser at the laser repetition rates of 10, 100 and 1000 Hz, respectively.

rate if a laser intensity is increased to 10^{20} W/cm² at present. An ultra-intense laser can only operate at a repetition rate of 10 Hz order at present^[13]. Under this condition, the ratio of transmutation rate between the former method and the latter one can reach about 10^3 order for both ¹³⁷Cs and ¹²⁹I. Therefore, the transmutation rate based on Compton backscattering is much larger than that based on bremsstrahlung. In

References

- Michel R, Klipsch K, Ernst Th et al. Radioprotection, 2005, 40(Suppl.1): S269
- 2 Baetslé L H, Achuthan P V, Burcl R et al. Implications of Partitioning and Transmutation in Radioactive Waste Management. Vienna: International Atomic Energy Agency Press, 2003
- 3 Yiou F, Raisbeck G M, ZHOU Z Q et al. Nucl. Instrum. Methods B, 1994, **92**: 436
- 4 Rubbia C, Abderrahim H A, Björnberg M et al. A European Roadmap for Developing Accelerator Driven Systems for Nuclear Waste Incineration. Roma: ENEA Publishers, 2001
- 5 Harada H, Sekine T, Hatsukawa Y et al. J. Nucl. Sci. Tech., 1994, **31**: 173
- 6 Harada H, Watanabe H, Sekine T et al. J. Nucl. Sci. Tech., 1990, **27**: 577
- 7 CAI D F, GU Y Q, ZHENG Z J et al. Chin. Phys., 2006, 15: 2363
- 8 XU M H, LIANG T J, ZHANG J. Acta Phys. Sin., 2006, 55: 2357 (in Chinese)
- 9 Shkolnikova P L, Kaplan A E, Pukhov A et al. Appl. Phys. Lett., 1997, 71: 3473
- 10 Norreys P A, Santala M, Clark E et al. Phys. Plasmas, 1999, 6: 2150

addition, we present in Fig. 3 the number of reaction dependence on irradiation time. For comparison, the numbers of reaction coming from bremsstrahlung at three different laser repetition rates are also provided. The difference of the transmutation efficiencies between the Compton backscattering method and the bremsstrahlung method is remarkable if one sees the nuclear activities and the numbers of the transmuted ¹³⁶Cs and ¹²⁸I shown in Table 2 and Fig. 3, respectively. Here the irradiation time for the samples ¹³⁷Cs and ¹²⁹I are assumed to be 30 min.

4 Conclusion

In conclusion, the transmutation for ¹³⁷Cs and ¹²⁹I based on Compton backscattering to be performed at SLEGS is investigated. The transmutation rate and the nuclear activity of ¹³⁶Cs and ¹²⁸I are evaluated and compared with the results of the transmutation based on bremsstrahlung method. Thanks to the better character of the gamma photon spectrum and high brightness, the transmutation rate via the Compton backscattering method is much higher than that via the bremsstrahlung method. In the future, when a pulsed CO_2 laser with the power of GW order and higher is employed at SLEGS, its transmutation rate can be improved by 10^7 at least. Of course, the bremsstrahlung method can also promote its transmutation rate by increasing the laser intensity with the ongoing progress of laser technology. In any case, the transmutation based on the Compton backscattering method will be attractive as an alternative transmutation method.

- 11 Malka V, Fritzler S, Lefebvre E et al. Science, 2002, 22: 1596
- 12 Ledingham K W D, Magill J, McKenna P et al. J. Phys. D, 2003, 36: L79
- 13 Magill J, Schwoerer H, Ewald F et al. Appl. Phys. B, 2003, 77: 387
- 14 Liesfeld B, Amthor K U, Ewald F et al. Appl. Phys. B, 2004, **79**: 1047
- 15 Sadighi-bonabi R, Kokabee O. Chin. Phys. Lett., 2006, 23: 1434
- 16 Balakin V, Alexandrov V A, Mikhailichenko A et al. Phys. Rev. Lett., 1995, 74: 2479
- 17 Imasaki K, Moon A. Int. Soc. Opt. Eng., 2000, 3886: 721
- 18 GUO W, XU W, CHEN J G et al. Nucl. Instrum. Methods A, 2007, 578: 457
- Bandazhevskaya G S, Nesterenko V B, Babenko V I et al. Swiss Med Wkly, 2004, 134: 725
- 20 ZHAO Z T, XU H J. SSRF: A 3.5 GeV Synchrotron Light Source for China. In: Europen Physical Society Accelerator Group ed. Proceeding of EPAC 2004. Lucerne: Paul Scherrer Institute Press, 2004. 2368
- 21 Klein O, Nishina Y. Z. Physik, 1929, 52: 853
- 22 Handbook on Photonuclear Data for Applications-Cross Sections and Spectra. Vienna: International Atomic Energy Agency Press, 2000