Photo-transmutation of long-lived radionuclide $^{135}$Cs by laser–plasma driven electron source

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(RECEIVED 5 November 2015; ACCEPTED 20 May 2016)

Abstract

Laser-driven relativistic electrons can be focused onto a high-$Z$ convertor for generating high-brightness $\gamma$-rays, which in turn can be used to induce photonuclear reactions. In this work, photo-transmutation of long-lived radionuclide $^{135}$Cs induced by laser–plasma–interaction-driven electron source is demonstrated using Geant4 simulation (Agostinelli et al., 2003 Nucl. Instrum. Meth. A 506, 250). High-energy electron generation, bremsstrahlung, as well as photonuclear reaction are observed at four different laser intensities: $10^{20}$, $5 \times 10^{20}$, $10^{21}$, and $5 \times 10^{21}$ W/cm$^2$. The transmutation efficiency depends on the laser intensity and target size. An optimum laser intensity, namely $10^{20}$ W/cm$^2$, was found, with the corresponding photonuclear reaction yield reaching $10^5$ J$^{-1}$ of the laser energy. Laser-generated electrons can therefore be a promising tool for transmutation reactions. Potential application in nuclear waste management is suggested.

Keywords: Photo-transmutation; $^{135}$Cs; Bremsstrahlung $\gamma$-rays; Nuclear waste; Laser ponderomotive acceleration

1. INTRODUCTION

Beams of electrons, positrons, protons, and high-energy photons can result from the interaction of ultra-intense lasers with solid or gas targets. The process has received much attention because of its many potential applications (Ledingham et al., 2003; Mangles et al., 2004; Schwoerer et al., 2006; Luo et al., 2013, 2015; Hanus et al., 2014). Thanks to recent advances in laser technology, laser-driven electrons can be accelerated to hundreds MeVs. By focusing the resulting relativistic electrons onto a high-$Z$ metallic target, high-energy $\gamma$-rays can be generated through bremsstrahlung. Such radiation has a wide range of applications, such as activation (or transmutation), fission, and fusion (Ledingham et al., 2003; Schwoerer et al., 2003; Galy et al., 2007, 2009).

Photonuclear reaction induced by ultra-intense laser was first proposed by Shkolnikov et al. (1997), and bremsstrahlung $\gamma$-rays, positrons, and photoneutrons were obtained. Magill et al. (2003) performed a photo-transmutation experiment on the long-lived radionuclide $^{129}$I to confirm the existing reaction cross-sections for $^{129}$I ($\gamma$, n). Photo-transmutation of the radionuclides $^{135}$Cs, $^{137}$Cs, $^{90}$Sr, $^{93}$Zr, and $^{126}$Sn driven by laser-based electron-bremsstrahlung have also been considered (Takashima et al., 2005; Sadighi-Bonabi & Kokabee, 2006; Sadighi & Sadighi-Bonabi, 2010; Sadighi-Bonabi et al., 2013; Irani et al., 2012). These studies suggest that the number of photonuclear reactions is closely related to the laser intensity and irradiation time, and laser-based photo-transmutation of radioactive nuclear waste should be possible. However, these studies are limited to theoretical calculations for thin targets. They do not take into account $\gamma$-ray attenuation inside the targets, nor other reaction channels that can be competitive with the ($\gamma$, n) reactions. Furthermore, without considering the target geometry, transmutation of long-lived radionuclides cannot be optimized.

In this work, we report a proof-of-principle experiment on the transmutation of long-lived nuclear waste $^{135}$Cs by ultra-intense laser with intensity $(0.1 - 5.0) \times 10^{21}$ W/cm$^2$. The radionuclide $^{135}$Cs has high radio toxicity, long half-life ($T_{1/2} = 2.3$ million years), as well as geologic repository impact and inventory, so that it risks leakage into the biosphere (Yang et al., 2004). Using the photo-transmutation method, $^{135}$Cs can be transmuted into $^{133}$Cs through the ($\gamma$, n) reaction or into the stable nuclide $^{133}$Cs through the ($\gamma$, 2n) reaction. The $^{134}$Cs has a short half-life of 2.07 years as it beta decays into the stable nuclide $^{134}$Ba. These non-/low toxic or stable product nuclides can be easily handled. Although the transmutation of $^{135}$Cs by ultra-intense laser has been analytically demonstrated (Takashima et al., 2005; Sadighi-Bonabi & Kokabee, 2006; Sadighi & Sadighi-Bonabi, 2010; Sadighi-Bonabi et al., 2013; Irani et al., 2012), these studies are limited to theoretical calculations for thin targets. They do not take into account $\gamma$-ray attenuation inside the targets, nor other reaction channels that can be competitive with the ($\gamma$, n) reactions. Furthermore, without considering the target geometry, transmutation of long-lived radionuclides cannot be optimized.

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2005) earlier, the effect of the laser intensity and target geometry on the details of the transmutation reactions is still unexplored.

Here, a photo-transmutation model, together with the Geant4 toolkit (Agostinelli et al., 2003), is developed for the transmutation of long-lived radionuclides using laser ponderomotive acceleration (LPA) of energetic electrons. In this model, the properties of the LPA produced electron beam (e-beam), such as the spectral and angular distributions, as well as competitive reaction channels that can result in additional contribution to the transmutation yield, are fully taken into account. Generation of intense bremsstrahlung γ-ray source driven by the laser-accelerated e-beam is investigated along with the photo-transmutation of $^{135}$Cs. Attention is also given to the dependence of the transmutation yield on the geometry of the converting target (CT) for bremsstrahlung generation and the adjacent transmuted target (TT), such as to optimize the number of transmutation reactions. It will be helpful for the similar photonuclear experiments performed by using high-peak power lasers.

Our study shows that the transmutation reaction yield can be enhanced more than three times by using an optimized target geometry and considering the contribution of electrons escaped from the rear side of the CT. It can reach $10^3$ J$^{-1}$ of laser pulse energy. This makes transmutation of nuclear wastes using state-of-the-art lasers quite promising. It should be reminded warmly that these phenomena can hardly be revealed according to the previous calculations.

2. PHOTO-TRANS_MUTATION MODEL

Currently, laser wakefield acceleration (LWFA) and LPA are the main table-top electron acceleration schemes (Esarey et al., 2009). LWFA can deliver high-quality relativistic ($\gtrsim 100$ MeV) e-beams with low (a few percent) energy spread and small (a few mrad) spatial divergence, but the beam current that can be accelerated is limited to tens pC. In contrast, LPA can generate relativistic e-beams up to a few nC (Glinec et al., 2005; Giulietti et al., 2008), which is useful for increasing the bremsstrahlung γ flux. Moreover, LPA e-beams have a wide bandwidth, and the need for narrowing their spectra is not needed since the bremsstrahlung γ source also has a continuous spectrum pattern. Accordingly, we shall use LPA e-beams to produce bremsstrahlung γ-rays, which in turn induce photo-transmutation of the cesium target.

A scheme for photo-transmutation of long-lived radionuclide $^{135}$Cs by the LPA e-beam is illustrated schematically in Figure 1. Because of its relative high conversion efficiency and more acceptable price than the more efficient but expensive Au target (Yan et al., 2012), metallic tantalum is used as the bremsstrahlung convertor. Both the convertor and the cesium target are assumed to have cylindrical structures with flexible radii and thicknesses. Since the dependence of the LPA e-beam spectra and angular distribution on the laser intensity has been well characterized, we can implement directly in the Geant4 simulations the characteristics of the LPA e-beams for the incident laser intensities $10^{20}$, $5 \times 10^{20}$, $10^{21}$, and $5 \times 10^{21}$ W/cm$^2$, with pulse energies 0.37, 1.86, 3.72, and 18.62 J, respectively and spot size 2.5 μm (full-width at half-maximum (FWHM)). The laser-to-electrons energy conversion efficiency is fixed at 30%, achieved by selecting appropriate acceleration lengths (Sentoku et al., 2002; Chen et al., 2009; Tanimoto et al., 2009; Hanus et al., 2014).

For the given energy conversion efficiency, the number of electrons can be related to the incident laser energy.

In order to reduce the computing time, a total of $10^8$ electrons are used in the Geant4 simulations and they have a Maxwellian energy distribution (Tanimoto et al., 2009; Antici et al., 2012)

$$f(E) = \frac{2}{\sqrt{\pi k T_h}} \sqrt{E_k} \exp \left( \frac{E_k}{T_h} \right),$$

where $E_k$ is the kinematic energy of the LPA electrons, $k$ is the Boltzmann constant and $T_h$ is the electron temperature (Wilks et al., 1992)

$$T_h = 0.511 \left[ \left( \frac{\lambda_\nu^2}{\lambda_\mu^2} \right)^{1/37} - 1 \right]$$

where $I$ is the laser intensity in W/cm$^2$ and $\lambda_\nu$ is the wavelength in μm. From Eq. (1) we can obtain the spectral distribution of the LPA e-beams for different laser intensities, as shown in Figure 2. We see that the laser intensity has an important effect on the e-beam spectrum: the higher the laser intensity, the larger the number of the energetic electrons.

Thus, together with the cross-sections of photonuclear...
reactions, one can optimize the number of reactions by varying the dimensions of the convertor and the cesium target, as discussed in Section 4.

An e-beam with spot size 3 μm (FWHM) impinges on the front surface of the convertor. It has a Gaussian energy distribution and angular spread (Moore et al., 1995; Quesnel & Mora, 1998; Debayle et al., 2010)

\[ \theta = \tan^{-1} \left( \frac{2}{\sqrt{\gamma - 1}} \right) \]  

where \( \gamma \) is the Lorentz factor of the relativistic electrons. The transverse profile of the e-beam from Eq. (3) is shown in Figure 3. Such a profile was recorded at 1 cm downstream of the initial position of the e-beam. We see that the e-beams produced by higher intensity lasers are more collimated and have higher energy.

3. SECONDARY SOURCES DRIVEN BY LPA ELECTRON BEAM

In general, the reaction yield depends on the convolution of the bremsstrahlung spectrum and the cross-sections of the photonuclear reactions. The interaction of the LPA electrons (see Figs 2 and 3) with the convertor is simulated for the laser intensities \( 10^{20}, 5 \times 10^{20}, 10^{21}, \text{ and } 5 \times 10^{21} \) W/cm², and secondary products such as electrons, positrons, and bremsstrahlung \( \gamma \)-rays are generated. Figure 4 shows the bremsstrahlung spectrum, produced by the LPA e-beam interacting with a 1.5 mm thick tantalum convertor. Also shown in Figure 4 is the total cross-sections of \( (\gamma, n) \) and \( (\gamma, 2n) \) reaction with \( ^{135}\text{Cs} \). Competitive reactions such as \( (\gamma, 3n), (\gamma, a), (\gamma, p), (\gamma, n + p), (\gamma, n + a), \) and \( (e, n) \) are not included in the figure because their reaction cross-sections are below 10 mbarn. The transmutation reaction has neutron separation energy of 8 MeV, its peaked cross-section occurs at about 15 MeV. At laser intensities below \( 10^{21} \) W/cm², the photonuclear reaction yield caused by the bremsstrahlung \( \gamma \)-rays increases with the laser intensity according to the convolution between the bremsstrahlung spectrum and the reaction cross-section, as shown in Figure 4. However, at laser intensities above \( 10^{21} \) W/cm², the reaction yield increases slowly.

Together with the bremsstrahlung \( \gamma \)-rays from the rear face of the convertor, the emitted secondary electrons and positrons can also irradiate the TT and produce high-energy bremsstrahlung \( \gamma \)-rays, which in turn trigger additional photonuclear reactions. The resulting electron and the positron spectra are shown in Figure 5. The target dimension is the same as that in Figure 4. It is found that both the electron and positron beams have Maxwellian-like spectral distributions. The numbers of high-energy electrons and positrons increase with the laser intensity. Due to the overlap of the energy spectra with the reaction cross-sections (see Fig. 4), similar to the bremsstrahlung \( \gamma \)-rays they can contribute to the transmutation yield. However, since the positrons are relatively few compared with the electrons, their contribution can be neglected.

Considering that the \( \gamma \)-rays and electrons with energies below the neutron separation energy cannot induce the photonuclear reaction, at four different laser intensities we counted the yield of electrons and \( \gamma \)-rays with energies above 6 MeV, as shown in Figure 6. As the CT thickness is increased, the secondary electrons decrease, but the \( \gamma \)-rays increase and become saturated for the few-mm thick target. The \( \gamma \)-ray yield is of order \( 10^{10} \) J⁻¹ (of laser energy). The peaked values \( 1.0 \times 10^{10}, 1.8 \times 10^{10}, 3.1 \times 10^{10}, \) and \( 3.7 \times 10^{10} \) J⁻¹ are obtained for the CT thicknesses 1.5, 2.5, 3.5, and 5.5 mm, respectively. That is, as the laser intensity increases, the thickness of the convertor should be increased.
We now consider the influence of the target parameters on the transmutation yield of $^{135}$Cs. We shall concentrate on the thickness of the convertor, the radius and thickness of the transmuted target, and the transmutation of $^{135}$Cs resulting from the dominant ($\gamma$, $n$) and ($\gamma$, 2$n$) reactions. It is found that other competitive reactions account for only 3% of the total product, so that they are neglected, even though the product nuclides such as $^{132}$Cs, $^{131}$I, $^{134}$Xe, and $^{133}$Xe are short-lived or stable. This can also be understood in terms of the reaction cross-sections, as discussed above.

### 4. PHOTO-TRANS MUTATION OF $^{135}$CS

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#### 4.1. The Influence of CT Thickness

The secondary sources driven by the LPA $e$-beam are used to transmute the long-lived nuclear waste $^{135}$Cs. Figure 7 shows the contribution of the secondary particles to transmutation reactions together with the total contributions at different laser intensities. In the simulation, the radius of the CT is 2 cm, and the radius and thickness of the TT are 4 and 3 cm, respectively. For a thin CT, the electrons contribute much more to the transmutation reactions than the bremsstrahlung $\gamma$-rays. With increase of the CT thickness, the contribution of the electrons decreases but that of the $\gamma$-rays increases. However, as the CT thickness attains a certain value, the contribution of the $\gamma$-rays decreases because of their decreased yield. It is also shown in Figure 7 that due to the contribution of electrons the thickness of the CT that led to the maximum total reaction yield is slightly thinner than that led to the peak $\gamma$-ray yield. This suggests that the influence of the electrons should be taken into account in order to obtain reliable reaction yield. This effect has not been discussed in the existing literature. In addition, the contribution of the positrons is found to be much smaller than 7% and is thus not shown in the figure.

Figure 7 also shows the dependence of the total reaction yield on the CT thickness. As the value of the thickness of
the CT is set to 0, it means the case of “without CT”, from which the LPA \(e\)-beam irradiated on the transmuted target directly and then triggered the photonuclear reactions. One can see that with the help of the CT, the transmutation yield is enhanced. In order to obtain the maximum reaction yield, the optimized thickness for the CT is found to be 1.0, 1.5, 2.5, and 3.5 mm at laser intensities of \(10^{20}\), \(5 \times 10^{20}\), \(10^{21}\), and \(5 \times 10^{21}\) W/cm\(^2\), respectively. While the CT thickness below 1.5 mm, the reaction yield at laser intensity \(5 \times 10^{21}\) W/cm\(^2\) (see Fig. 7d) is slightly smaller than that at \(10^{21}\) W/cm\(^2\) (see Fig. 7c). This is mainly caused by the convolution of the \(\gamma\) spectrum with the profile of the photonuclear cross-section, as discussed above.

### 4.2. Effect of the Geometry of the Transmuted Target

Using the optimized thickness of the CT, the dependence of transmutation reactions on the TT geometry was investigated. The curve of the reaction yield as a function of the TT radius is investigated and is shown in Figure 8. The reaction yield enhanced rapidly when the target radius is relatively small, for example, \(\leq 1.0\) cm, meanwhile such enhancement ceased when the radius of the target larges 1.5 cm. Taking into account the volume of the TT, the radius of the TT is suggested to be 2 cm at different laser intensities. In the simulation, the thickness of the TT was set as 3 cm.

The dependence of the reaction yield on the TT thickness for different laser intensities is shown in Figure 9. The radii of the CT and TT are 2 cm, and the optimized thicknesses of the CT are used in the simulation, as discussed above. For \(\geq 10^{21}\) W/cm\(^2\) lasers, the reaction yield increases with the thickness of the TT. At the lower laser intensities, such increase is not obvious or even absent.

### 4.3. Discussion

At laser intensities ranging from \(10^{20}\) to \(5 \times 10^{21}\) W/cm\(^2\), the influence of the parameters for both the convertor and transmuted target has been demonstrated (see Figs 7–9). According to these simulations, the transmutation yield of \(^{135}\)Cs was...
produce about $3.7 \times 10^{21}$ reactions per second. The transmutation capability of intense laser-based electron source can therefore be comparable with that by photo-transmutation of long-lived radionuclides such as $^{133}$Cs using Compton scattering classical $\gamma$-ray sources (Imasaki et al., 2006; Shuji et al., 2007; Zhu et al., 2016).

5. SUMMARY

In this paper, the possibility of photo-transmutation of long-lived radionuclide $^{135}$Cs into the short-lived $^{134}$Cs or the stable nuclide $^{137}$Cs has been considered through Monte Carlo simulations. It is shown that the laser intensity and the geometry of both the converter and the cesium target have strong influence on the reaction yield of $^{135}$Cs. Moreover, proper choice of the target size for different laser intensities can enhance the transmutation efficiency by a factor of four. There is also an optimum laser intensity, namely $10^{21}$ W/cm$^2$, for producing maximum reaction yield. In view of the current advances in tabletop ultra-intense lasers, compact laser-based systems for photo-transmutation can be promising for nuclear waste management and medical isotope production.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (Grant Nos. 11405083 and 11347028) and the Research Foundation of Education Bureau of Hunan Province, China (Grant No. 14A120). W.L. appreciates the support from the Young Talent Project of the University of South China.

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