Annals of Nuclear Energy 89 (2016) 109–114

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Photo-transmutation of long-lived nuclear waste ^{135}Cs by intense Compton γ-ray source

Zhi-Chao Zhu [a], Wen Luo [a,b,*], Zhuo-Cheng Li [a], Ying-Ming Song [a], Xiao-Dong Wang [a], Xing-Liu Wang [a], Gong-Tao Fan [c]

[a] School of Nuclear Science and Technology, University of South China, Hengyang 421001, China
[b] Extreme Light Infrastructure-Nuclear Physics (ELI-NP)/Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering (IFIN-HH), 30 Reactorului St., Bucharest-Magurele, jud. Ilfov, P.O.B. MG-6, RO-077125, Romania
[c] Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China

ARTICLE INFO

Article history:
Received 17 July 2015
Received in revised form 2 November 2015
Accepted 12 November 2015
Available online 11 December 2015

Keywords:
Photo-transmutation
Compton γ-ray source
Long-lived nuclear waste
^{135}Cs

ABSTRACT

MeV-range Compton γ-ray beam via Compton scattering of a laser pulse against a high-current relativistic electron beam can be used to induce photo-transmutation reactions. In this work, we investigate through Monte Carlo simulations the possibility of transmutation of radioactive nuclear waste, by high-intensity Compton γ-ray source, for the long-lived ^{135}Cs with a half-life of 2.3×10^6 years into short-lived ^{134}Cs with a half-life of 2.07 years, or into stable nuclide ^{133}Cs. Our simulation results show that the transmutation reaction yield is in the order of 6×10^9 per hour for a given γ-flux of 2×10^8 phs/s that can be obtained from the Shanghai Laser Electron Gamma-ray Source which is under construction. The transmutation capability of worldwide Compton γ-ray facilities is also evaluated, showing that the photo-transmutation induced by brilliant Compton γ-ray source, such as ELI-NP could be an efficient approach to transmute long-lived radioactive nuclear waste.

1. Introduction

Nuclear power is used extensively around the world and the disposal of long-lived nuclear waste have became a huge challenging problem in nuclear industry. From 1980s, underground burial became an official government policy in the disposal of nuclear waste, but this route requires millions of years for radioactive waste to decay to natural level. Once the radioactive nuclear waste is eroded by groundwater, or their surrounding geological conditions is changed due to unexpected factors or the validity period of the protective barriers expires, they will have a great potential to endanger the biosphere (Yang et al., 2004; Michel et al., 2005). Exploring potential methods to transmute the long-lived radiotoxic nuclides into stable or short-lived nuclides before burial is of great interest (European Commission, 2004; Boardon Radioactive Waste Management, 2001).

Recently several routes for nuclear waste transmutation have been proposed. Besides the transmutation of radioactive waste using bombardment from a fast neutron reactor or a particle accelerator (Baetsle et al., 2003; Bowman, 1998), the photo-transmutation induced by bremsstrahlung γ-source from ultra-intense laser (i.e. laser-driven bremsstrahlung source) may be considered as an alternative. Based on this approach, the long-lived nuclear waste such as ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, and ^{135}Cs have been investigated theoretically or experimentally (Irani et al., 2014, 2012; Sadighi-Bonabi et al., 2010; Ledingham et al., 2003; Magill et al., 2003; Liesfeld et al., 2004; Takashima et al., 2005). Since this method involves relativistic electron beam acceleration by ultra-intense laser pulse, followed by electron beam irradiation of high-atomic number target to generate bremsstrahlung source, however the energy conversion efficiency is not high (Giulietti et al., 2008). Further, the bremsstrahlung spectrum is wide and has a relatively low intensity in Giant Dipole Resonance (GDR) energy region. These results in a poor coupling of energy to the GDR for the transmutation.

With the rapid development of electron accelerators and laser techniques, Compton γ-ray source (CGS) (Litvinenko et al., 1997; Park et al., 2001; Weller and Ahmed, 2003; Miyamoto et al., 2007; Weller et al., 2009; Fujiwara, 2003; Aoki et al., 2004; Li et al., 2004; Luo et al., 2010a,b), via Compton scattering between a laser pulse and a high-current relativistic electron beam, can deliver a high-intensity, energy-tunable and collimated γ-ray beam

* Corresponding author at: School of Nuclear Science and Technology, University of South China, Hengyang 421001, China. Tel.: +86 15211819698.
E-mail address: wenluo-ok@163.com (W. Luo).

http://dx.doi.org/10.1016/j.anucene.2015.11.017
0306-4549/

ranging from a few MeV to tens of MeV, which covers effectively the GDR region and therefore has great potential for the transmutation of hazardous nuclear waste by photonuclear reaction (Imasaki and Moon, 2000; Takashima et al., 2005). It has been shown in Chen et al. (2009) and Irani et al. (2014) that photo-transmutation by the CGS has advantages in transmutation efficiency over the laser-driven bremsstrahlung γ-source.

In terms of reduction of the collective leakage-dose risks (product of toxicity and mobility) in the repository, nuclear waste as ^{129}I, ^{99}Tc and ^{135}Cs represent the priority. Among them, ^{129}I and ^{99}Tc can be transmuted to short-lived or stable isotopes via a single neutron capture reaction. ^{99}Tc is not suitable for photo-transmutation mechanism due to a small cross section for the photonuclear channel. In addition, laser-induced transmutation of ^{129}I and ^{99}Tc have already been explored experimentally (Ledingham et al., 2003; Magill et al., 2003).

In this work, we present the possibility of transmutation of the radioactive nuclear waste, by worldwide high-intensity CGS such as the Shanghai Laser Electron Gamma-ray Source (SLEGS) and the ELI-NP, for the long-lived ^{135}Cs with a half-life of 2.3×10^6 years into short-lived ^{134}Cs with a half-life of 2.07 years, or into stable nuclide ^{133}Cs through Monte Carlo simulations. ^{135}Cs was chosen due to its high radiotoxicity, the long half-life and for the visible geologic repository impact and inventory. Hence it leads to a huge risk of leakage-dose to biosphere. ^{135}Cs strongly requires isotope separation for effective neutron-induced transmutation, because of the high weight fractions and capture cross sections of ^{133}Cs and ^{134}Cs isotopes, leading to additional ^{135}Cs production (Yang et al., 2004). Furthermore, intense γ-rays emitted by ^{137}Cs would make handling and isotopic separation of cesium very difficult and costly. With these considerations, using intense γ-ray source to transmute ^{135}Cs is more feasible and economical than using high-flux neutron fields. Note that the transmutation of the ^{135}Cs by using the SLEGS γ-beam has been studied preliminarily (Chen et al., 2009). However, it is limited to the theoretical analysis by only considering a thin target with a specific size. Theoretical calculation can hardly take into account the γ-ray attenuation inside a thick target and the possible competitive reaction channels that could result in additional contribution to transmutation yield. Further, an obsolete parameter for the SLEGS was used in the calculation. Based to the previous studies, we use Geant4 simulation to address these issues, to further elucidate the details of photo-transmutation of ^{135}Cs according to the latest design of the SLEGS and then to obtain the optimum transmutation reaction yield. The potential transmutation of radionuclide ^{135}Cs triggered by worldwide CGS are also evaluated.

2. Methods

A schematic illustration of photo-transmutation of radioactive nuclide driven by the CGS, e.g. the SLEGS is displayed in Fig. 1. The two photonuclear reactions employed in the case of ^{135}Cs are: ^{135}Cs(γ,n) and ^{135}Cs(γ,2n), from which result the transmutation products ^{134}Cs and ^{133}Cs. ^{134}Cs has a half-time of 2.07 years and beta decays to the stable nuclide ^{134}Ba, whiles ^{133}Cs is a stable nuclide.

The SLEGS facility (Pan et al., 2009) has been proposed to be built at the Shanghai Synchrotron Radiation Facility (SSRF), which is an advanced third-generation synchrotron radiation facility. According to newest design version of the SELGS, it is expected to generate a polarized γ-ray beam of up to 22 MeV and 10^8–10^{10} phs/s by using a 10^2–10^4 W polarized CO_2 laser pulse backscattered off a 3.5 GeV, 300 mA relativistic electron beam produced at the SSRF storage ring (Pan et al., 2009). An adjustable aperture is collimating the beam approximately 10 meters downstream of the interaction point. The collimator will stop the scattered low-energy γ-rays that can hardly trigger photonuclear reactions. A cylindrical irradiation target with changeable radius and thickness is placed behind the collimator and aligned with the γ-ray flashes.

A 4D (three dimensional time and frequency domain) Monte Carlo laser-Compton scattering simulation (MCLCSS) code (Luo et al., 2011, 2013, 2014), benchmarked against laser-Compton scattering experiments performed at HIγS of Duke University and PLEIADES from LLNL, was used to predict the properties of the SLEGS γ-beam. Then the simulated γ-beam parameters including the spectral, transversal profiles and angular distributions were delivered to the particle gun of the Geant4. Accordingly, the simulation of the γ-ray beam irradiating the ^{135}Cs target, taking into account the standard electromagnetic physics list, particle transportation and the potential photonuclear reactions were performed by the Geant4 toolkit (Agostinelli et al., 2003).

To the best of our knowledge, many experimental cross sections for the photonuclear reactions are not currently available, for instance the ^{135}Cs(γ,n) and ^{135}Cs(γ,2n) are two of them. In Geant4, while the photonuclear cross sections are parameterized by combining the semi-empirical formula and interpolation algorithm, some physics models can also be employed to deal with the hadronic interactions required in simulation (Geant4 physic reference manual). In our case, the Geant4 Binary cascade physics model was chosen and the QGSP BIC HP physics list was utilized. Due to value of cross section dependent on some specific variables, the reliability of the simulation output should be verified further. An alternative approach to simulate the photonuclear processes is the direct implementation of the cross sections taken from the nuclear data library, such as TENDL (Koning and Rochman) or from Empire calculation (Herman et al., 2007), into Geant4 physics list. We also applied this algorithm to study the photo-transmutation of ^{135}Cs and then to crosscheck the results with the QGSP BIC HP physics list. Here the cross sections data for the dominant reactions (i.e. ^{135}Cs(γ,n) and ^{135}Cs(γ,2n)), available in TENDL evaluation, were utilized accordingly. The detailed comparison between the

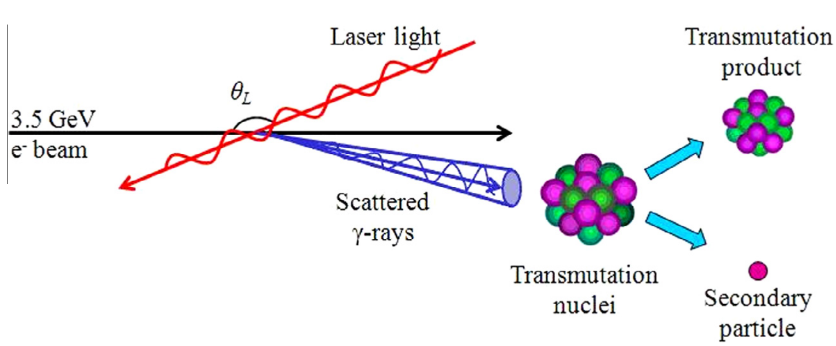

Fig. 1. Schematic of the transmutation of ^{135}Cs induced by the CGS.