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Photo-transmutation based on resonance γ-ray source*

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High intensity γ-ray source can be obtained through resonance reaction induced by protons. In this work, the possibility of using such high intensity MeV-range γ-ray source to transmute nuclear waste is investigated through Mont Carlo simulation. ^{197}Au(γ, n)^{196}Au experiment is performed to obtain the transmutation rate and compared with the simulation result. If the current of the proton beam is 10 mA at the resonance energy of 441 keV, with the γ photons emitted from ^{7}Li(p, γ)^{8}Be, then the corresponding transmutation yield for ^{129}I in 2π direction can reach 9.4×10^{9} per hour. The result is compared with that of LCS γ-ray source.

Keywords: high intensity γ-ray source, photo-transmutation, photonuclear physics

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1. Introduction

Transmutation by neutrons, produced by high flux accelerated protons bombarding a heavy metal target, is the primary method to deal with long-lived fission products (LLFPs) with large neutron capture cross-sections. However, for nuclides, whose neutron capture cross sections are small, this method is not very efficient. In addition, (*n*, γ) reactions may transmute stable nuclides into long-lived radiotoxic nuclides. For example, ^{133}Cs is stable, of which the (*n*, γ) transmutation product will be ^{134}Cs followed by ^{135}Cs. They are unstable and radiotoxic with long half-lives of 2.06 years and 2.3 million years, respectively.

Photo-transmutation can be a good supplement to neutron transmutation.[1] There are several methods to obtain γ rays: positron annihilation in flight, bremsstrahlung, laser-Compton scattering (LCS), and nuclear resonance reactions. γ-ray beams obtained by positron annihilation in flight is of low intensity, thus it is not appropriate for transmutations. Transmutations of ^{129}I based on a laser-induced bremsstrahlung (LIB) source have been already performed experimentally in Germany,[2–4] UK,[5] and Japan.[6] Theoretical investigations have been done on LLFPs such as ^{126}Sn,[7,8] ^{107}Pd,[9] ^{93}Zr,[10] ^{90}Sr,[11] ^{135}Cs,[12–14] and ^{137}Cs.[15] An LCS γ-ray source can produce γ-rays in a wide energy range, which can be utilized to transmute LLFPs. The Upgraded HIγS, operating since 2009, can produce up to 3×10^{9} photons/s. The energy of photons can be adjusted in 1 MeV–100 MeV. The energy of electron beam is 0.24 GeV–1.2 GeV and the laser wavelength is 1.064 μm. The rate of transmutation reaction of ^{135}Cs by the Upgraded HIγS is evaluated in Ref. [16] to reach 10^{11} per hour. Studies[17,18] show that the LCS-based photo-transmutation is more efficient than LIB. Many experimental[19,20] and theoretical[14,16–18,21] studies have been done using LCS γ-ray source to transmute LLFPs.

In the present paper, γ rays emitted from resonance reactions induced by proton is utilized to transmute LLFPs. Nuclear resonance reaction can produce mono-energetic γ rays, which can be used to transmute LLFPs. To do so, the energy of incident protons is supposed to be hundreds of keV to several MeV. Figure 1 shows high power proton beams worldwide (non exhaustive) crossing this region.[22–30] The mean beam current of high power proton beams that have already been constructed can reach 100 mA.

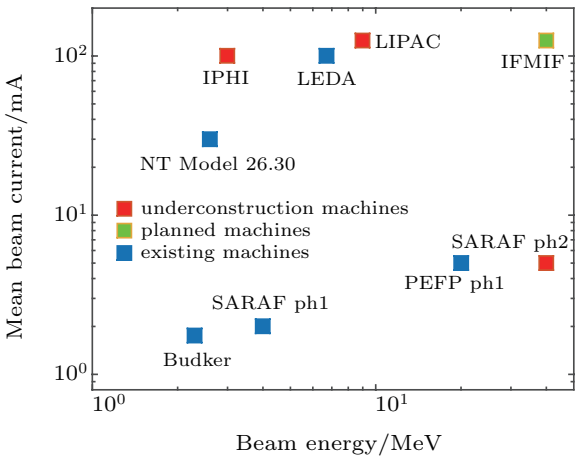


Fig. 1. High current proton beams worldwide (non exhaustive) in MeV region.

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2. Proton-induced resonance γ-ray source for transmutation

The excited levels of the compound nucleus are discrete and widely spaced when the bombarding proton energies are low. The so-called Breit–Wigner formula, as shown below, gives a good description for the cross section of a single resonance level,

$$\sigma(E_{\rm p}) = \frac{\pi\lambda_{\rm p}^2 g\Gamma_{\rm p}\Gamma_\gamma}{(E_{\rm cm,p}-E_0)^2+\left(\Gamma_{\rm T}/2\right)^2}, \quad (1)$$

where $\sigma(E_{\rm p})$ is the $({\rm p},\gamma)$ reaction cross section of the protons whose energy is $E_{\rm p}$ in laboratory frame, $E_{\rm cm,p}$ is the corresponding energy of protons in center-of-mass frame, $E_{\rm cm,p} = m_{\rm A}E_{\rm p}/(m_{\rm a}+m_{\rm A})$, E_0 is the resonance energy. $m_{\rm a}$, $m_{\rm A}$ are the mass number of the incident proton and target nucleus. $g = (2I_{\rm C}+1)/(2I_{\rm a}+1)(2I_{\rm A}+1)$ is the statistical factor, $I_{\rm C}$, $I_{\rm a}$, $I_{\rm A}$ are angular momentum of the compound nucleus, the proton and target nucleus, respectively. $\lambda_{\rm p}$ is the de Broglie wavelength of relative motion, $\Gamma_{\rm T}$ is the full width at half maximum (FWHM) of the compound nucleus level. Γ_γ is the partial width for the gamma ray transition to the ground state and $\Gamma_{\rm p}$ the partial width for proton emission. Thick target γ photon yields of some nuclides are shown in Table 1.[31–33]

The GDR peak energy of most LLFPs is about 15 MeV, therefore it would be better to use the reaction $^7{\rm Li}({\rm p},\gamma)^8{\rm Be}$ to obtain higher transmutation rates. According to the data listed in Table 1, the yield of 14.8 MeV (33%) and 17.6 MeV (67%) γ rays emitted from $^7{\rm Li}({\rm p},\gamma)$ when the energy of incident protons is 441.4 keV is 1.90×10^{-8} per proton in total.[31,32] The yield of 9.17-MeV γ-ray emitted from $^{13}{\rm C}({\rm p},\gamma)^{14}{\rm N}$ is 4.7×10^{-9}.[33]

Table 1. γ yields of some narrow resonance reactions.

Reactions	Resonance energy/keV	γ-ray energy/MeV	Thick target yield/proton	Width Γ/keV
$^7{\rm Li}({\rm p},\gamma)$	441.4 ± 0.5	14.8(33%) 17.6(67%)	1.9×10^{-8}	12
$^9{\rm Be}({\rm p},\gamma)$	998.0 ± 4 1087.0 ± 2	7.4 6.7, 0.72	1.78×10^{-8} 1.01×10^{-9}	94 4
$^{19}{\rm F}({\rm p},\alpha\gamma)$	340.0 ± 2	6.1(96%) 7.0(4%)	1.74×10^{-8}	3.2
	873.5 ± 1	6.1(72%) 7.0(28%)	3.6×10^{-7}	5.2
	935.3 ± 1	6.1(77%) 7.0(23%)	2.0×10^{-7}	8.0
	All ≤ 960	6.1(72%) 7.0(28%)	6.9×10^{-7}	–
$^{13}{\rm C}({\rm p},\gamma)$	1747	9.17	4.7×10^{-9}	–

Different compound nuclides have different energy levels ranging from several hundreds of keV to tens of MeV, which cover most Giant Dipole Resonance (GDR) regions. The yields of γ-radiation produced by proton beams in narrow resonance region follow the equation:

$$n_{\gamma,\Omega_0} = \int_0^{\Omega_0}\int_{E1}^{E2}\int_0^{l_1} I_{\rm p}(E(l))\sigma_{\rm p}(E,\Omega)n_1\,{\rm d}E\,{\rm d}l\,{\rm d}\Omega, \quad (2)$$

where n_{γ,Ω_0} is the number of resonance energy γ rays produced per second in the solid angle Ω_0, $I_{\rm p}(E(t))$ is the intensity distribution against the incident proton energy, which is a function of the depth of the target l, $\sigma(E,\Omega)$ is the differential cross-section when the proton energy is between E and $E+\Delta E$ and the emitted γ rays is in the solid angle Ω to $\Omega+\Delta\Omega$, n_1 is the number density of the target atom in the converter. l_1 is the thickness of the converter, E_1 and E_2 are the upper and lower bounds of the incident proton energy.

Next, using the γ-radiation produced above, we obtain the transmutation rate without considering secondary process:

$$n_{\rm t} = \int_0^{l_2} n_{\gamma,\Omega_0}\,{\rm e}^{-\mu(E_0)l}\sigma_\gamma(E_0)n_2\,{\rm d}l, \quad (3)$$

where $n_{\rm t}$ is the transmutation rate, n_{γ,Ω_0} is the number of γ photons produced per second in the solid angle corresponding to the transmutation target, $\mu(E_0)$ is the attenuation coefficient of the target which is relevant to the γ photon energy E_0, l_2 is the thickness of the target, $\sigma_\gamma(E)$ is the cross-section of the photo-nuclear reactions when the energy of incident γ is E_0, n_2 is the number of nuclei to be transmuted. However, this is a complex process. γ photons can produce electrons and positrons through pair production process, these electrons and positrons produce photons again by bremsstrahlung, which can also add to transmutation rate. It is better to track all the particles with a Monte Carlo simulation.

From Eqs. (2) and (3), we can increase the transmutation rate $n_{\rm t}$ in the following ways: 1) higher proton beam intensity $I_{\rm p}$; 2) reactions with higher cross-section of proton induced γ-rays; 3) larger solid angle of the transmutation target.

3. $^{197}{\rm Au}(\gamma,{\rm n})^{196}{\rm Au}$ experiment

First, to ensure the accuracy of the calculation, the experiment of $^{197}{\rm Au}(\gamma,{\rm n})^{196}{\rm Au}$ was carried out in the 2×1.7-MV tandem accelerator of China Institute of Atomic Energy (CIAE).[33] Two gold targets of the same size are irradiated. Figure 2 shows the transmutation process. The γ photons are produced by $^{13}{\rm C}({\rm p},\gamma)^{14}{\rm N}$ reaction when the resonant energy of incident protons is 1.747 MeV. ^{13}C is evaporated onto a $\phi10\times2$ mm gold target up to a mass thickness of 100 μg/cm^2. Bombarded by the 9.17-MeV γ photons, ^{197}Au can be transmuted into ^{196}Au through $(\gamma,{\rm n})$ reaction. However, ^{196}Au is unstable, it decays to ^{196}Pt through electron capture process and to ^{196}Hg through β decay.

The first sample is radiated for 6 hours and the second for 5.5 hours. Then, the radioactivity of the irradiated gold samples is measured in a low-background lead chamber with anti-Compton method used to reduce the background radioactivity. The detected counts obey the exponential law given by

$$N_{\rm c} = \frac{P}{\lambda}(1-{\rm e}^{-\lambda t_1}){\rm e}^{-\lambda(t_2-t_1)}(1-{\rm e}^{-\lambda(t_3-t_2)}), \qquad (4)$$

where P is the generation rate during irradiation and λ the decay constant of of ^{196}Au. $0\rightarrow t_1$ is the irradiation time, $t_1\rightarrow t_2$ is the cooling time, and $t_2\rightarrow t_3$ is the measuring time. By measuring the decay counts $N_{\rm c}$, the generation rate P can be derived from Eq. (4), thus the transmutation counts N_0 can be obtained from $N_0 = P(t_2-t_1)$. The spectrum is presented in Fig. 3. Peaks at 355.73 keV (Iγ = 87%) and 333.03 keV (Iγ = 22.9%) originates from ^{196}Au. The result is shown in Table 2. In Table 2, the second column is the irradiation time, the third column shows the counts of peak at 355.73 keV, and the numbers of (n,γ) in ^{197}Au, calculated from experimental data, with self-absorption of the gold targets taken into consideration, are listed in the fourth column.

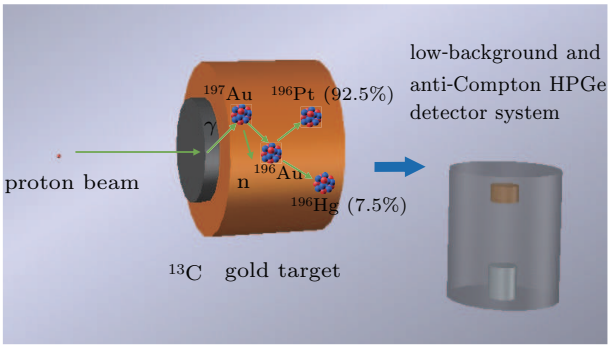


Fig. 2. ^{197}Au transmutation process.

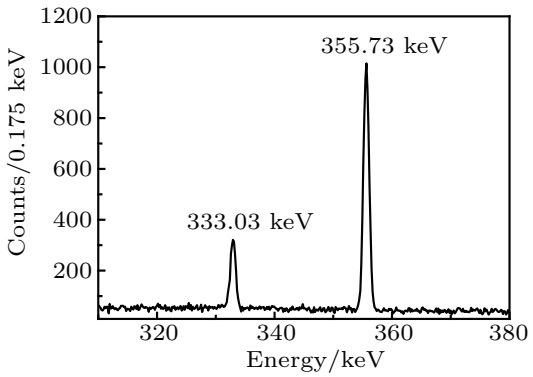


Fig. 3. γ spectrum of irradiated ^{197}Au, peaks at 355.73 keV (Iγ = 87%) and 333.03 keV (Iγ = 22.9%) originates from ^{196}Au.

Table 2. Transmutation numbers of different samples.

Sample	$T_{\rm irradiation}$/s	$N_{\rm Det}$	$N_{\rm tr}/10^6$
1	21600	1566 ± 49	1.01 ± 0.03
2	19800	6610 ± 100	1.09 ± 0.02
Average	20700	–	1.05 ± 0.03

The thick target γ photon yield of ^{13}C (100 μg/cm^2) was measured with an HPGe detector (GXM35P4-70). The maximum yield of 9.17-MeV γ photons is $(4.7\pm0.4)\times10^{-9}$/proton.[33] As the average current of the proton beam during the ^{197}Au transmutation experiment is 8 μA, the number of incident protons during 20700-s irradiation time is 1.04×10^{18}, producing $(4.9\pm0.4)\times10^9$ γ photons.

In the Geant4 simulation, 4.9×10^9 γ photons are assumed to be emitted in 4π direction from a point source. The point source is on the center of the front surface of a $\phi10\times2$ mm gold target, with zero distance. The number of transmutation is $(6.6\pm0.5)\times10^5$, which is 37% less than the experimental results but with no deviation in magnitude.

4. Geant4 simulation of photo-transmutation yields

In this section, we simulate photo-transmutation yields of nuclear waste like ^{129}I. The photonuclear cross sections of ^{129}I extracted from Geant4 packages are shown in Fig. 4.

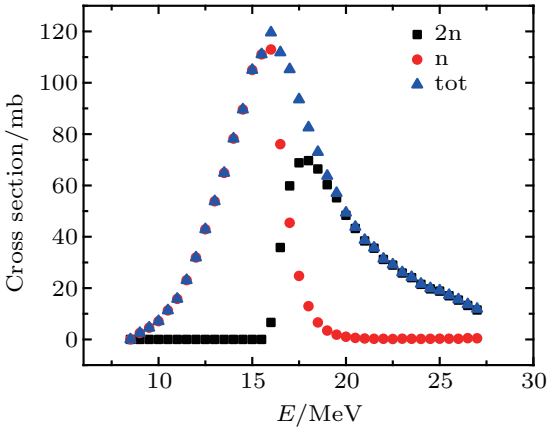


Fig. 4. Photonuclear cross section. The red circle, black square, and blue triangle present the cross section of ^{129}I$(\gamma,{\rm n})^{128}$I, ^{129}I$(\gamma,2{\rm n})^{127}$I, and in total, respectively. The unit 1 b = 10^{-28} m^2.

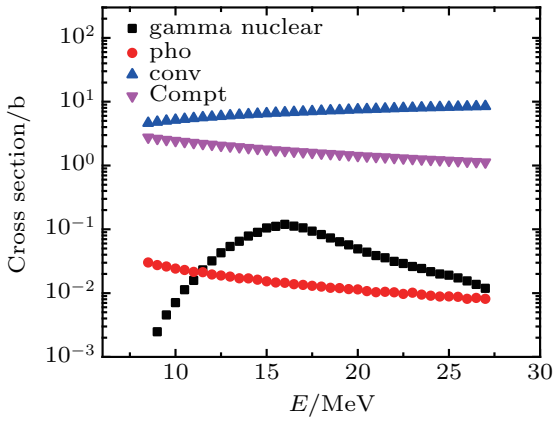


Fig. 5. ^{129}I cross section of different mechanisms, the black square, red circle, blue triangle, and pink inverted triangle show a cross section of the photo-nuclear reaction, photoelectric effect (pho), pair production (conv), and Compton effect (Compt), respectively.

Photoelectric effect, Compton effect, pair production, and photo-nuclear reactions are the principal mechanisms by which γ rays interact with matter. Figure 5 shows cross section of each mechanism when γ rays interact with ^{129}I.

Assuming the intensity of the proton accelerator is 10 mA at 441 keV, according to the yields of 441-keV narrow resonance of reaction ^{7}Li(p,γ)^{8}Be in Table 1, γ-ray yield of 14.8 MeV and 17.6 MeV can reach 1.2×10^{9} per second, and the γ-ray source, produced by the reaction ^{7}Li(p,γ)^{8}Be, is assumed to be point shape and isotropic in 4π direction, of which 33% are 14.8 MeV and 67% are 17.6 MeV. Next, the ^{129}I target, with width and height are 2 cm, is placed 5 cm away from the point source. The ^{129}I(γ,n)^{128}I yields against target depth are shown in Fig. 6, the maximum yield can reach 5.7×10^{-5} per γ. Taking the γ-ray yield into consideration, there are 6.8×10^{4} ^{129}I(γ,p)^{128}I reactions per second.

Regarding transmutation of other LLFPs based on ^{7}Li(p,γ)^{8}Be 441-keV resonance γ-radiation in the same condition as mentioned earlier but the point source with zero distance is on the front surface center of the targets and the targets are as thick as 10 cm. The rates of transmutation reactions are calculated and shown in Table 3.

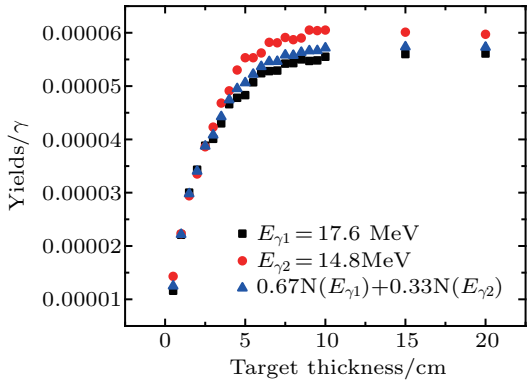


Fig. 6. Yields of ^{129}I(γ,n)^{128}I, the red circle, black square, and blue triangle show transmutations rates of 14.8-MeV γ photons, 17.6-MeV γ photons, and weighted average respectively.

Table 3. Transmutation rates of different LLFPs.

Reactions/hour	^{129}I	^{135}Cs	^{126}Sn	^{93}Zr	^{107}Pd	^{137}Cs	^{90}Sr
(γ,n)	4.7×10^{9}	2.1×10^{9}	2.8×10^{9}	8.6×10^{10}	9.7×10^{10}	1.8×10^{9}	2.8×10^{9}
(γ,2n)	4.7×10^{9}	2.2×10^{9}	7.9×10^{9}	7.9×10^{10}	5.4×10^{10}	2.5×10^{9}	6.1×10^{9}
Total	9.4×10^{9}	4.3×10^{9}	1.1×10^{10}	1.7×10^{11}	1.5×10^{11}	4.3×10^{9}	8.9×10^{9}

The transmutation rate of ^{135}Cs is 10^{9} per hour is shown in this chart in total. According to Ref. [16], among LCS facilities constructed around the world, the best transmutation ability is of the Upgraded HIγS, which can reach 10^{11} per hour, the HIγS is 10^{9} per hour, and the NewSUBARU is 10^{8} per hour.

5. Conclusion

Considering a 10-mA proton accelerator, nuclear waste transmutation driven by ^{7}Li(p,γ)^{8}Be 441-keV narrow resonance γ-rays have been investigated through Geant4 simulation. The maximum transmutation yield of ^{129}I is 9.4×10^{9} per hour. The transmutation yield of ^{135}Cs is obtained to be the same order as HIγS of 10^{9}, which is less than that of the upgraded HIγS by one order of magnitude but greater than that of the NewSUBARU by two orders of magnitude. In the future, we will improve the transmutation rates by improving the current of the proton accelerator, searching for higher yields of γ-radiation in GDR region, designing better target structure, and so on. Moreover, when compared with the good direction of LCS γ source, the γ-radiation emitted from narrow resonance is in 4π directions, which is appropriate for large areas of LLFP radiation.

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