

Electron linac based mixed-field transmutation of ^{129}I



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ABSTRACT

The goal of this work was to determine the feasibility of using electron linacs for transmutation of selective long-lived fission products (LLFPs), particularly ^{129}I . We have designed a converter system that produces both photons and neutrons, thus creating a mixed radiation field. The strength of the field depends on electron beam energy and current, as well as the converter material and geometry. The converter was optimized to maximize transmutation rate of ^{129}I via two channels: $^{129}\text{I}(\gamma, n)^{128}\text{I}$ and $^{129}\text{I}(n, \gamma)^{130}\text{I}$. The optimum electron beam energy was found to be around 100 MeV. Computer simulations were benchmarked by experimental data on ^{133}Cs (^{129}I surrogate) photon-induced and neutron-induced transmutation rates. While further optimization of the system is necessary, we have demonstrated that accelerator-driven photonuclear reactions can be practical for ^{129}I transmutation.

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1. Introduction

Used nuclear fuels and related recycling products contain transuranics (TRU), uranium, and fission products (FP). Unlike the first two, the FP do not hold any recycling value and are the major contributors to radiotoxicity levels in used fuel handling scenarios as they represent true nuclear waste in used fuel compositions [1,2]. For efficient nuclear waste storage strategies targeting transitions from geological time scales to engineering time scales, it is desirable to explore feasible ways to minimize the radiotoxicity of the FP stream. This can be accomplished when a significant amount of the most radiotoxic FP nuclides are transmuted into stable species or species with shorter half-lives. Numerous LLFP transmutation studies [3–11] have been performed over last several decades and most of them were focused on transmutation in critical or subcritical reactor systems. In this work, we would like to address LLFP transmutation methods based on photonuclear reactions using electron accelerators.

Photonuclear reactions, integrated over a broad bremsstrahlung spectrum, can transmute a significant fraction of the target material, as demonstrated by numerous reports on photonuclear production of isotopes [12–14]. Multi-kilowatt electron accelerators, with beam energies of 40 MeV or higher, produce extremely high GDR-photon flux (the term “GDR-photons” is used to refer to the photons with energies between 10 and 40 MeV). These photons excite the target nuclei causing particle emission and hence trans-

mutation reactions. A proper converter/target design can provide more than 10^{13} GDR-photons/cm²/s per kW of electron beam power. The total photo-absorption cross-section in the GDR region is typically dominated by neutron emission and yields up to 10^{12} fast fission spectrum neutrons/cm²/s per kW of electron beam power. As a result, radiation fields of photons and neutrons can be produced simultaneously. The ratio of neutron to photon fluxes and their energy distributions may be varied by adjusting the electron beam energy and the converter/target design.

In this work we will focus on transmutation of ^{129}I , which is one of the major risk components of the LLFP due to its high geochemical mobility and long half-life (10 million years). ^{129}I is the dominant consideration for sustainable long-term nuclear waste management schemes. It can be transmuted to shorter half-life species by (n, γ) and (γ, n) reactions; the $^{129}\text{I}(n, \gamma)^{130}\text{I}$ reaction results in ^{130}I which has a 12.36 h half-life, and the $^{129}\text{I}(\gamma, n)^{128}\text{I}$ reaction results in ^{128}I which has a 25 min half-life (Fig. 1).

2. Methods and materials

2.1. Mixed-field transmutation

Bremsstrahlung photons, produced by the interaction of electrons with a high Z material, have a unique continuous energy spectrum which can be “tuned” to some extent by electron beam energy and converter geometry. These photons can be copiously produced by electron accelerators. At relativistic electron energies (>1 MeV), highly forward directed photon beams are produced.

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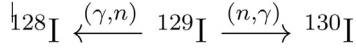


Fig. 1. Mixed-field transmutation scheme of ^{129}I .

If the energy of the bremsstrahlung photon exceeds the threshold energy of single or multiple nucleon knock-out, nuclear excitations can occur. While the single-nucleon $((\gamma,n)$ or (γ,p)) reactions are the primary reaction types in the GDR region, other unavoidable higher-order reactions such as $(\gamma,2n)$, $(\gamma,3n)$, and (γ,np) are also present. However, the threshold energies for these reactions are usually higher and their integral cross sections are lower than those of single-nucleon emission reactions. Therefore the most important contribution to the total photo-absorption cross section is the single-neutron (γ,n) or the single-proton (γ,p) emission reaction. For the majority of the isotopes ($Z > 10$), the photoneutron reaction dominates over the photoproton, and as a result, an intense photoneutron field can be formed simultaneously with intense photon field. The transmutation rate can be found using:

$$Y_{(\gamma,n)} = N_o \cdot \int_{E_{th}}^{E_{max}} \phi_\gamma \cdot \sigma_{(\gamma,n)} dE$$

Similarly, neutron-induced transmutation rate can be found as:

$$Y_{(n,\gamma)} = N_o \cdot \int_0^{E_{max}} \phi_n \cdot \sigma_{(n,\gamma)} dE$$

Thus ^{129}I transmutation rate via $^{129}\text{I}(\gamma,n)^{128}\text{I}$ and $^{129}\text{I}(n,\gamma)^{130}\text{I}$ channels can be found using above two equations, where N_o is the number of target nuclides, ϕ_γ and ϕ_n are the photon and neu-

tron flux densities, and σ is the reaction cross section. In case of a mixed field of photons and neutrons the overall transmutation rate of ^{129}I can be found as a sum of the transmutation rates through each channel (Figs. 1 and 2):

$$Y = Y_{(\gamma,n)} + Y_{(n,\gamma)}$$

The transmutation rates were estimated using MCNPX, a general purpose particle transport Monte Carlo code, developed by the Los Alamos National Laboratory [15–17]. Both photon and neutron fluxes were simulated with F4 tallies. FM tally multiplier was used to multiplying them with ENDF/B-VII cross-sections [18] and obtaining the transmutation rates.

2.2. ^{129}I surrogate

Due to high radiotoxicity of ^{129}I we decided to use a surrogate material to benchmark our simulations of transmutation with a mixed field. A suitable surrogate material had to be found, with nuclear properties and cross-sections similar to ^{129}I (Fig. 3). Both (γ,n) and (n,γ) reactions on the surrogate had to produce isotopes whose yields are easy to measure with gamma-spectroscopy. In particular, these products need to have suitable half-lives (in the range of hours or days), distinct gamma-ray energies, and high transition branching ratios. ^{133}Cs was found to satisfy all the above criteria. MCNPX simulations confirmed that ^{133}Cs and ^{129}I have similar transmutation rates and thus ^{133}Cs can be an excellent surrogate for ^{129}I .

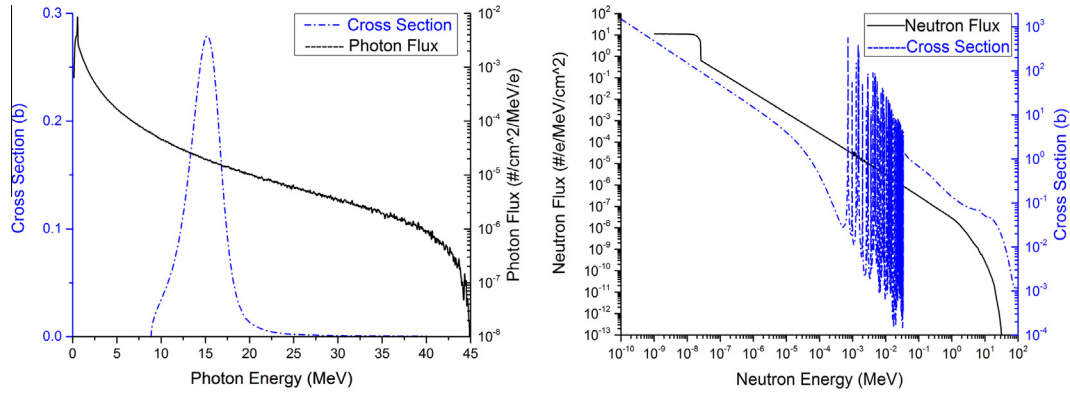


Fig. 2. $^{129}\text{I}(\gamma,n)^{128}\text{I}$ cross section and bremsstrahlung spectrum produced by a 45 MeV electron beam incident on a 0.35 cm W converter (left). $^{129}\text{I}(n,\gamma)^{130}\text{I}$ cross section and thermal photoneutron spectrum produced by a 45 MeV electron beam incident on a 0.35 cm W converter surrounded by water (right).

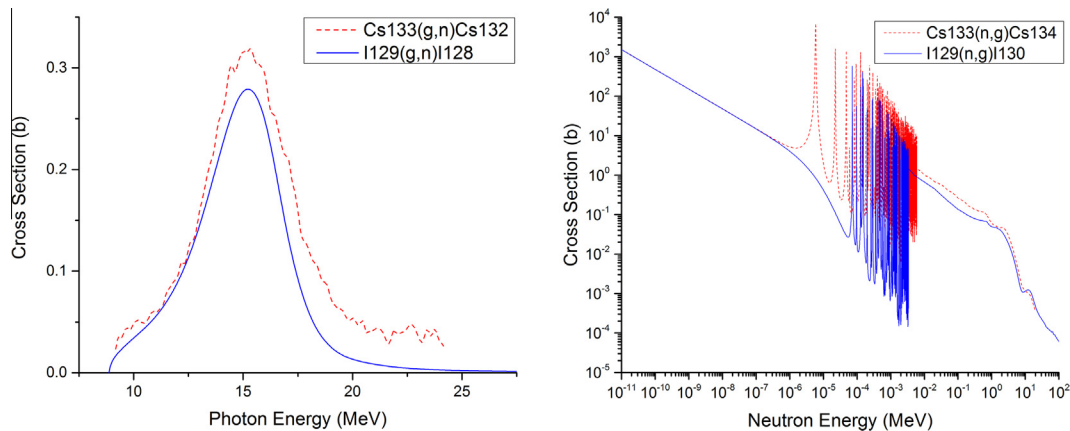


Fig. 3. (γ,n) and (n,γ) reaction cross section for ^{129}I and ^{133}Cs [21,22].

3. Results and discussion

3.1. Design

Maximizing photon flux and maximizing low-energy neutron flux are two mutually exclusive problems. High photon flux requires a relatively thin electron bremsstrahlung converter and the photons form a forward cone. Exploiting the high photon flux region requires placing samples very close to the converter. In contrast, high neutron flux with lower energy requires a thick converter and an even thicker moderator surrounding the converter. Thus, for each particular reaction we had to carefully examine tradeoffs in photon flux and neutron flux.

The (γ, n) reaction of ^{129}I has a threshold energy of 8.84 MeV (Fig. 3). The $^{129}\text{I}(\gamma, n)^{128}\text{I}$ cross-section peaks at 15 MeV and drops at 25 MeV. In the case of neutrons, the cross-section increases as neutron energy decreases (Fig. 3), so thermalizing the neutrons in the system can significantly enhance the ^{129}I transmutation rate. To create a mixed field of high energy photons and low energy neutrons, we used two models (Fig. 4). First model included a design of a single converter where the target material was distributed around the converter and surrounded by a neutron moderator (1 m^3 of H_2O). Second model involved two separate converters. Electron beam was incident on the photon converter, which was followed by the neutron converter surrounded by the target material and a moderator layer.

In the first design, a thick converter was required to generate significant neutron flux. However, thick converters limit the photon production. To find the optimum geometry of the converter a number of MCNPX simulations were performed and ^{129}I transmu-

tation rates were calculated for different converter and target layer thicknesses. The moderator layer was kept the same for all the versions of the setup. The optimum converter thickness was found to be 0.35 cm and this setup resulted in $3.48 \times 10^{-5} \text{ 1/cm}^2$ per source electron. The optimum iodine target layer thickness was 0.5 cm. The results of the simulations suggest that, for this design, the photon-induced transmutation is dominant and drives the geometry of the setup (Fig. 5 left).

The second design allows more effective use of both photon and neutron fields since each converter can be optimized separately. The optimum thickness of the photon converter was found to be 0.35 cm. We also compared this number with the average range of 40 MeV electrons in tungsten, which was found according to [19]:

$$R = \frac{E}{2\rho}$$

where R is electron range in cm, E is electron energy in MeV and ρ is material density in g/cm^3 . Our optimum thickness is slightly lower than the value reported by Berger and coworkers [20], who suggested that the optimum bremsstrahlung converter thickness corresponds to 40–50% of the mean range of the electrons in the material. Varying the thickness of the neutron converter and the thickness of the target layer, we simulated the transmutation rate for numerous geometries. Fig. 5 (right) shows that the $^{129}\text{I}(n, \gamma)^{130}\text{I}$ reaction is a major contributor to the total transmutation rate since the $^{129}\text{I}(\gamma, n)^{128}\text{I}$ reaction rate does not change significantly. The maximum total transmutation rate was found to be $7.5 \times 10^{-5} \text{ 1/cm}^2$ per source electron, which is almost twice as high as the trans-

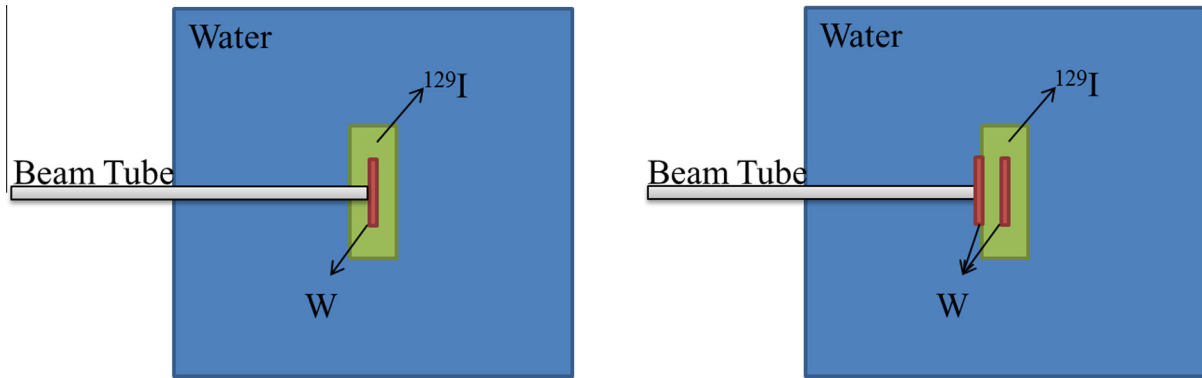


Fig. 4. MCNPX setup for ^{129}I transmutation.

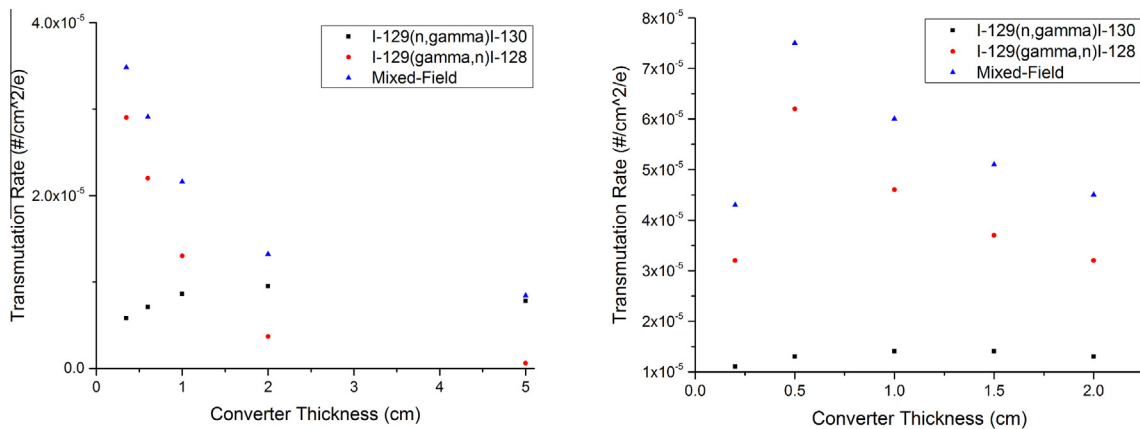


Fig. 5. Transmutation rates for two different designs.

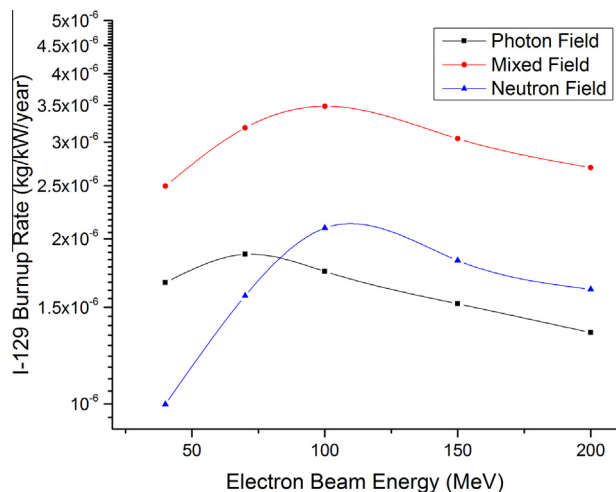


Fig. 6. ^{129}I burnup rate as a function of electron beam energy. Scattered points are connected with spline curve.

mutation rate for the single converter setup. The optimum thickness of the neutron converter was found to be 0.5 cm.

We also investigated the transmutation rate of ^{129}I as a function of the electron beam energy. As electron energy increases, the optimum thickness of each of the converters increases as well. The cost-effective transmutation rate using the photon field peaks at about 70 MeV and then slowly decreases (Fig. 6). This can be explained by the shape of the photoneutron cross-section, which has a pronounced peak in the GDR region but drops for higher energies. For the $^{129}\text{I}(n,\gamma)^{130}\text{I}$ channel, the optimum electron beam energy is about 100 MeV. The photon-induced and the neutron-induced transmutation rates are comparable. For energies below 80 MeV, photon-induced rates are slightly higher; for higher energies, neutron field starts to dominate. The optimum energy for the mixed-field transmutation was found to be around 100 MeV.

3.2. Experiment

To verify the simulated transmutation rates of ^{129}I we irradiated cesium molybdate (Cs_2MoO_4), a compound that contains ^{133}Cs , a surrogate material for ^{129}I . Both water solution of cesium molybdate and cesium molybdate powder were used to measure transmutation rate and validate the MCNPX simulations. Optimized converter was not available and the benchmarking was done using an un-optimized mixed-field setup, consisting of 1 cm thick photon converter and 6 cm thick neutron converter (Fig. 7). An aluminum tank (1 ft $\times$ 1 ft $\times$ 1 ft, 3 mm thick walls) was installed at the end of the 40 MeV electron linac beamline at the IAC. Photon and

Table 1

Activities of Cs samples.

Label	Experimental (μCi)		MCNPX (μCi)	
	^{132}Cs	^{134}Cs	^{132}Cs	^{134}Cs
1	21 ± 4	0.21 ± 0.05	32	0.4
2	0.02 ± 0.01	0.02 ± 0.01	0.08	0.05
3	0.3 ± 0.1	0.05 ± 0.02	0.4	0.09
4	0.25 ± 0.05	0.04 ± 0.02	0.4	0.09
5	0.015 ± 0.005	0.04 ± 0.02	0.05	0.08
6	0.015 ± 0.005	0.04 ± 0.02	0.05	0.08

neutron converters were placed inside the tank filled with water. Cesium molybdate samples were placed in different locations as shown in Fig. 7. After 3 h of irradiation with a 40 MeV 100 W electron beam and 1 h of cooling, the samples were transferred to the nuclear counting lab. A gamma spectroscopy system consisting of a HPGe detector with built-in preamplifier, amplifier, ADC, and a multi-channel analyzer was used for the spectroscopy measurement. MPA-3 software was used to plot, save, and analyze the spectra. Activities of ^{132}Cs and ^{134}Cs were calculated using 668 and 605 keV lines respectively.

MCNPX simulations were performed for the setup and the yields of ^{132}Cs and ^{134}Cs were calculated. The results of the experiment as well as the theoretical predictions of the activities are summarized in Table 1. We believe that a shifted electron beam (to the left), which has not been quantified and has not been considered during the MCNPX simulations, is the reason for larger variation. The transmutation rates of ^{133}Cs are higher if powder is used due to a higher concentration of ^{133}Cs . For both experiments, the measured yields of cesium isotopes are in a reasonable agreement with the MCNPX simulations.

4. Conclusion

In this work we investigated the feasibility of an accelerator-based nuclear waste transmutation method. With the aid of simulation codes, the optimum transmutation experimental design was obtained. Two separate converters are proposed for the photon and neutron production with target material (^{129}I) circulating through the channel between the converter and water moderator. Using such a design allows us to effectively utilize both photon and neutron fields. The burn-up rate of ^{129}I has been calculated for different electron beam energies, and the most cost effective transmutation of ^{129}I is achieved with a 100 MeV electron accelerator. Up to a gram of ^{129}I can be transmuted per year using a 100 kW linac. Our simulations were benchmarked by irradiating ^{133}Cs and inducing $^{133}\text{Cs}(\gamma,n)^{132}\text{Cs}$ and $^{133}\text{Cs}(n,\gamma)^{134}\text{Cs}$ reactions. The yields of ^{132}Cs and ^{134}Cs isotopes were measured and found

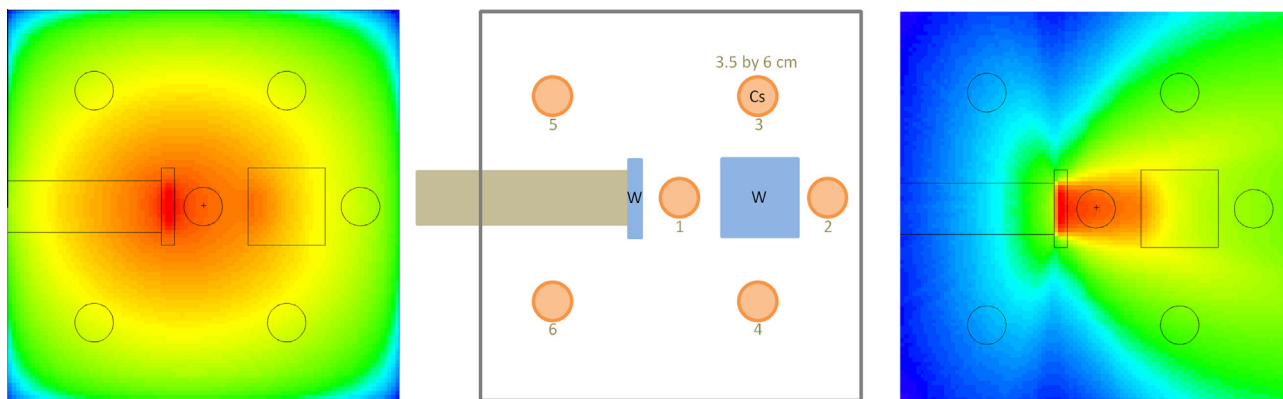


Fig. 7. Cesium Solution experimental setup (center); MCNPX modeled photon and neutron spatial flux distribution (left and right).

to be in a reasonable agreement with the predicted values by MCNPX.

Note that the system we optimized in this work was relatively simple. Even for such a basic geometry we were able to demonstrate significantly high transmutation yield. We believe that the yield can be further increased if a more complex design of the mixed-field converter is created. A moderator/reflector also needs to be optimized to increase the thermal neutron flux and, as a result, the transmutation rate. Finally, more work needs to be done to address the power deposition into the system and develop a proper cooling scheme so that high power beams can be used. Taking into account the advantages of using electron linacs in comparison to reactor systems, we believe that the mixed-field ^{129}I transmutation is highly viable.

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