Project No. 11-3054

Fission Product Transmutation in Mixed Radiation Fields

Fuel Cycle

Dr. Frank Harmon
Idaho State University

In collaboration with:
Texas A&M University

Dan Vega, Federal POC
James (Keith) Jewell, Technical POC

Nuclear Energy University Programs
U.S. Department of Energy

Final Summary Report Fission Product Transmutation in Mixed Radiation Fields
NEUP Proposal 11-3054

Summary:

Work under this grant addressed a part of the challenge facing the closure of the nuclear fuel cycle; reducing the radiotoxicity of lived fission products (LLFP). It was based on the possibility that partitioning of isotopes and accelerator-based transmutation on particular LLFP combined with geological disposal may lead to an acceptable societal solution to the problem of management. The feasibility of using photonuclear processes based on the excitation of the giant dipole resonance (GDR) by bremsstrahlung radiation as a cost effective transmutation method was accessed. The nuclear reactions of interest: (γ,xn), (n,γ), (γ,p) can be induced by bremsstrahlung radiation produced by high power electron accelerators. The driver of these processes would be an accelerator that produces a high energy and high power electron beam of ~ 100 MeV. The major advantages of such accelerators for this purpose are that they are essentially available "off the shelf" and potentially would be of reasonable cost for this application. Methods were examined that used photo produced neutrons or the bremsstrahlung photons only, or use both photons and neutrons in combination for irradiations of selected LLFP. Extrapolating the results to plausible engineering scale transmuters it was found that the energy cost for ^{129}I and ^{99}Tc transmutation by these methods are about 2 and 4%, respectively, of the energy produced from 1000MWe.

Introduction:

Used nuclear fuels and related recycling products contain uranium, TRU and fission products. Assuming that used nuclear fuel compositions are processed they can be partitioned and separated into three streams: uranium (possibly to be recycled in reactors), TRU and fission products. Unlike the first two streams, the fission product stream does not hold any recycling potential and, after the short lived species decay (~30 years), the LLFP contribute to radiotoxicity levels in geological storage scenarios. The only way the radiotoxicity of LLFP can be reduced in reasonable time is via transmutation of these nuclides into stable species or species with shorter half-lives. As ^{99}Tc and ^{129}I are major risk/dose components of LLFP, transmutation of these nuclides have been the principle focus of this work by examination of representative surrogate materials. It has been found that the concept of simultaneous irradiation by a combination of neutrons and high energy photons can improve transmutation yields of either alone for ^{129}I surrogates. Results of these surrogate studies also argue for dual irradiation transmutation of certain other LLFP species as well. An ancillary finding of this work is that the production of useful quantities of a class of medically interesting radioisotopes, that are difficult or impossible to produce with conventional means using reactors or cyclotrons, can be readily produced in quantity with photonuclear reactions.

Unlike the usual conception of accelerator transmutation where reductions of the transuranic components of used fuel are the major targets, here the aim is limited to a reduction of radiotoxicity of LLFP after partioning using no fission produced neutrons.

For example the two FP, ^{129}I and ^{99}Tc, have very long half-lives (>10^5 yr) and are highly mobile in the environment so they are major contributors to radiotoxicity. The procedure studied here for reduction in radiotoxicity is to transmute by photonuclear induced reactions the ^{129}I and ^{99}Tc into the shorter lived species ^{128}I, ^{130}I, and ^{100}Tc which have half-lives of less than a day.

An assessment of the potential utility of photonuclear processes has two major parts: a) What is the physical basis of the necessary processes; production of photons, adequacy of simulation tools, and experimental verification of calculations and b) is the process, in principle, scalable to a level that meets the requirements of the particular task. The emphasis in this work is on the former question, the physics part of the problem. Clearly the second part of the question emphasizes engineering and economic aspects that require the information from the first part. Thus the work done under this grant has been focused primarily on the physical basis of transmutation induced by photonuclear phenomena. We considered the following issues:

1. The properties of converters to take electron beam energy to photons and photo-neutron production.
2. The nuclear and thermal behavior of numerous photon and neutron converter designs.
3. Appropriate moderators for photo-neutrons
4. Evaluation of nuclear and chemical properties of surrogate materials for various LLFP.
5. Determination of the transmutation efficacy of; a) fast and moderated photo-neutrons b) photons c) mixed photons and fast and moderated neutrons combined.

Data on these five points were obtained by extensive simulation and experiment using standard codes and IAC accelerators and nuclear support facilities. The results obtained allow a rough estimate of the economic feasibility of a large-scale transmutation system for selected LLFP. This extrapolation of experiment and simulation results to simplified engineering scale transmuter schemes also allows an estimate operating costs per transmuted mass in energy terms. The accelerator types needed for the irradiation processes are available from commercial vendors and cost estimates are known. Included in the analysis of the base engineering case are possible selections of the chemical form of the FP feed material based primarily on desirable nutronic and thermal properties of the form.

Discussion:

A list of long lived fission products in legacy pressurized water reactor fuel is shown in Table 1. These nuclides are candidates for transmutation in order to reduce their inventory within a relatively short time. The blue indicates ^{99}Tc for which photon irradiation will not lead to a shorter lived or stable isotopes but neutron irradiation will. The rest are isotopes that are amenable to both photon and neutron or mixed photon/neutron transmutation. As these nuclides are all roughly mid Z the irradiation yields will be similar and similar to the yields of the mid Z elements we selected and measured as surrogate materials selected for this work. Table 2 shows the amounts of the most radio toxic of the LLFP produced per 1000MW year.

Nuclide	**Isotope**	**Half-life (yr)**	**Atom % in Total FP inventory**
Selenium-79	Se-79	290,000.	1.0290
Zirconium-93	Zr-93	1,500,000.	27.1351
Niobium-94	Nb-94	20,000.	0.1809
Technetium-99	Tc-99	213,000.	23.7119
Palladium-107	Pd-107	6,500,000.	6.3315
Tin-126	Sn-126	230,000.	1.5683
Iodine-129	I-129	15,700,000.	4.2602
Cesium-135	Cs-135	2,300,000.	9.2816
Samarium-151	Sm-151	90.	0.2630

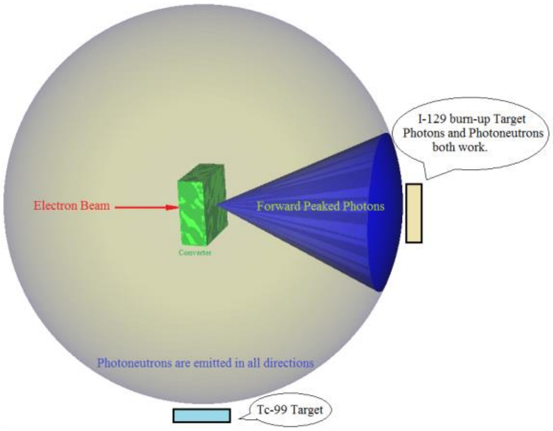
Table 1-Selected fission products in legacy PWR spent fuel. The isotopes listed would be amenable to transmutation by both photons and neutrons or a combination of both delivered simultaneously except for ^{99}Tc (blue) which is a case where only neutron induced transmutation is effective.

Nucleus	**T-1/2 year**	**Toxicity, microSv/Bq (ICRP61)**	**Mass, kg/TWh, 1000kWh**	**Mass, kg/year/ 1000MWe**
Tc-99	2.1x10e5	0.7x10e-3	3.2	19.5
I-129	1.6x10e7	0.1	0.87	5.32
Cs-135	2x10e6	0.2x10-2	1.23	7.53

Table 2- Amounts of the most radio-toxic of the LLFP produced per 1000MWe year.

Summary experiments and simulations:

The radiation fields in photonuclear driven gamma/neutron sources are depicted in the cartoon at the right. In general an energetic electron beam, > 20 MeV say, strikes a converter. And an electron/photon shower is produced in the material (X). A key characteristic of the bremsstrahlung photons produced by the relativistic electron beam is the pronounced on axis forward direction of the photons, photon intensity at large angles from the electron beam axis are down by one to two orders of magnitude or more depending on the converter properties. The photons have a broad energy spectrum extending up to the energy of the electron beam. However, photo-neutrons produced in the converter by high energy photons are emitted into roughly 4π steradians



and the neutrons produced have an evaporation spectrum similar to fission neutrons. A consequence of these properties is that it is possible to design a converter/moderator system that allows the use of photons only, neutrons only or both photons and neutrons simultaneously. It turns out that the latter scheme has a significant advantage for transmuting LLFP that can be transmuted to short lived species by both types of radiation, photons via (γ,xn,p) and neutrons via (n,γ) as noted in Table 1.Thus, multi-kilowatt, electron linear accelerator driven sources with beam energies of ~ 40 -150MeV, falling on appropriately designed converter targets, can produce copious high energy, broad spectrum, highly forward directed photons or evaporation spectrum neutrons or a combination of both. The converters intended to maximize photons and minimize neutrons are “thin”, about 1 radiation length in the converter material for high Z materials which provide the most efficient transformation of electron energy to photon energy, thickness is less than a cm. If the converter is made much thicker, about 10 radiation lengths, the photons produced couple strongly with the giant resonance in the converter nuclei to produce neutrons by (γ,xn) and the photons are largely absorbed. A compromise may be sought between these two extremes allowing high product on of both photons and neutrons. In the course of this work several designs for electron driven converters were studied; those optimized for photon production, those optimized for neutron production and combination converters. The yield, spectra, and thermal characteristics of these converters were studied by simulations and experiment. The transmutation yield of various surrogate materials were simulated and measured using simplified and easily fabricated converter types.

Neutron only transmutation, ^{99}Tc:

For ^{99}Tc burn-up where only neutrons via the ^{99}Tc(n,γ)^{100}Tc reaction are useful in producing a short lived result (^{100}Tc with a half-life of 15.8 seconds). Photons above the neutron emission threshold (~10MeV) drive the reaction ^{99}Tc(γ,n)^{98}Tc which is not helpful since it produces ^{98}Tc which has a half-life of 4.2 million years. This calls for a neutron source free of any excessive GDR-photons, i.e. a “thick” converter. The neutron cross section for ^{99}Tc is shown in figure 2 along with the water moderated neutron spectrum for an optimized neutron converter with a 40 MeV electron beam, note that this spectral shape will not change significantly for electrons up to ~150 MeV however the number of neutrons/electron will increase about 2 times.

Most neutron capture takes place in the resonance and thermal regions of the spectrum so that an appropriate moderator is essential for efficient transmutation as the converter produces mostly fast neutrons.

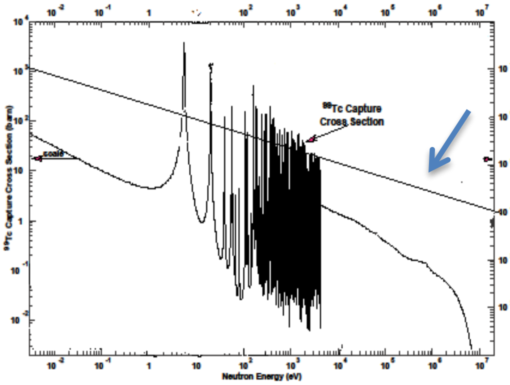


Figure 1- The ^{99}Tc(n,γ)^{100}Tc reaction cross section and the spectrum of a light water moderated photo-neutron source.

Engineering scale ^{99}Tc transmuter performance:

An estimate of the accelerator input energy cost for transmutation of ^{99}Tc via the ^{99}Tc(n,γ)^{100}Tc reaction with e beam produced neutrons was evaluated. Benchmarked simulations allow a reasonable calculation of the energy cost for transmutation driven by an e beam on a W converter target for e energies ~ 100MeV. We assume the accelerator to have 50% electrical efficiency, energy of 100MeV, a tungsten converter, light water moderator and metallic ^{99}Tc blanket placed around the converter, see figure 2. For maximum ^{99}Tc target loading we find a conversion rate of ~20kg ^{99}Tc per 40MWyr of electrical input to the accelerator. In one year the converted mass is about the annual production per 1000MWe fission produced power. The maximum beam power per accelerator that could reasonably be achieved would be about 5 MW so that several accelerators would be required. Aside from the capital cost the energy needed for 99Tc burn up would amount to about 4% of the produced energy.

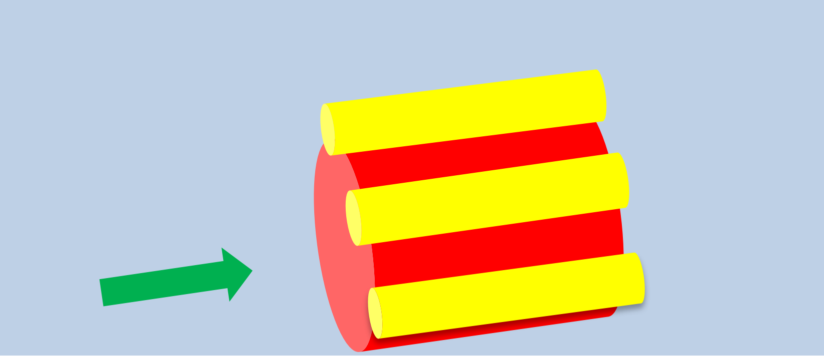


Figure 2- A cartoon figure of an engineering scale photo-neutron Tc transmuter. The moderated photo-neutron converter is surrounded by Tc (yellow) and water moderator (blue). The e beam comes in from the left through a vacuum beam pipe (green)

Photon and neutron transmutation:

For those LLFP in tables 1 and 2 that can be transmuted to stable or short lived by both photons and neutrons a simple mixed irradiation converter was designed and tested. The cross sections for (γ,n) and (n,γ) shown in figure 3 demonstrate the physics.

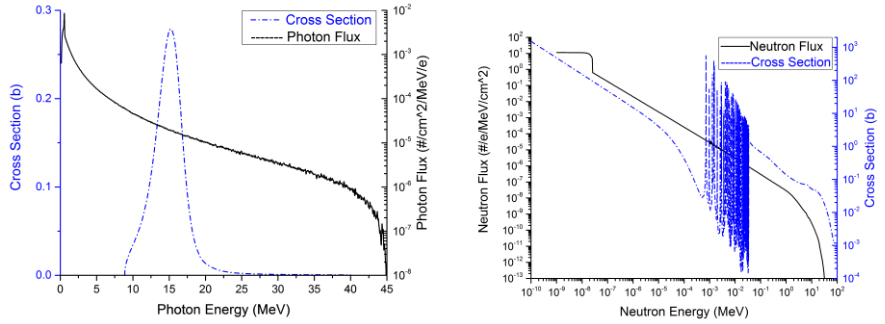
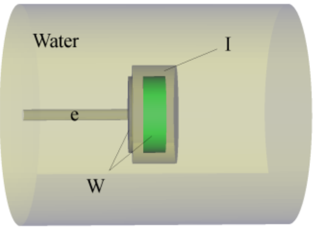


Figure 3- The graph on the left shows a 40 MeV bremsstrahlung spectrum and a representative (γ,xn) cross section. On the right is a moderated photo-neutron spectrum and a representative mid A nuclear (n,γ) cross section.

A converter designed to produce photons that can be directed into a mass of material and neutrons to be moderated and subsequently acting to transmute will be a compromise with production of either species optimized. Nevertheless our work has shown that there is an advantage in the dual scheme for those LLFP that are photon and neutron transmutable. This is shown for the case of ^{129}I in figure 4 for a divided converter as sketched on the right.



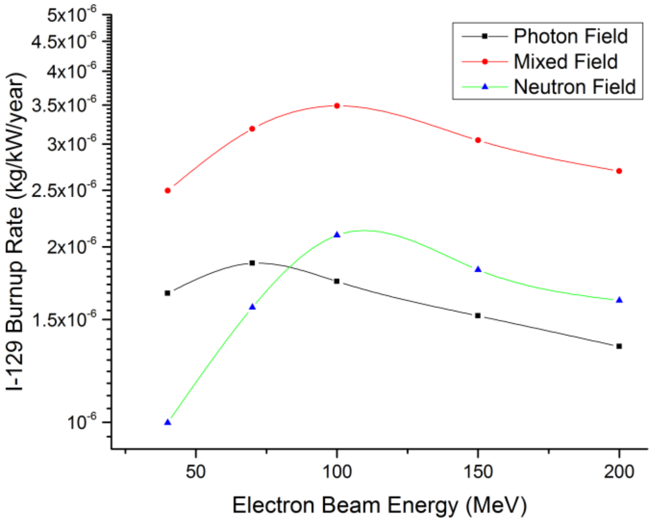


Figure 4- Simulation of the burn up rate of ^{129}I for mixed irradiation.

Engineering scale ^{129}I transmuter:

A converter with the simplified design shown would have poor thermal properties for a high powered transmuter. Analysis of potential engineering scale transmutation schemes requires methods of converter cooling and a choice of the LLPF chemical form. For the ^{129}I and similar FP in table 1 a molten salt containing the materials is assumed and the converter is to be circulating PbBi eutectic.

Shown below are two views of such a possible photon/neutron transmuter with the target structure being liquid PbBi eutectic (green and blue pipes, red discs), surrounded by a molten salt FP carrier (dark blue) and a water moderator/reflector. This device could transmute the FP in table 1 that are amenable to dual irradiation transmutation. This transmuter could convert 3.5 kg of ^{129}I /20 MW yr, the annual production for 1000 MWe, assuming 50% efficiency for the accelerators. This amounts to using 2% of the energy produced to burn up the ^{129}I from fission.