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NON-NEUTRON TRANSMUTATION OF USED NUCLEAR FUEL

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PROJECT OVERVIEW

- **Non-neutron Transmutation of Used Nuclear Fuel**
  - ARPA-E 2022 OPEN/FOA: 2459-2951
  - Period: July 2022 – June 2025 (3 years)
- **Planned Work**
  - Develop a transformational long-lived fission product (LLFP) transmutation technology using incident energetic non-neutron particles (photons and protons)
  - Propose a national LLFP transmutation facility concept
- **Contributors**

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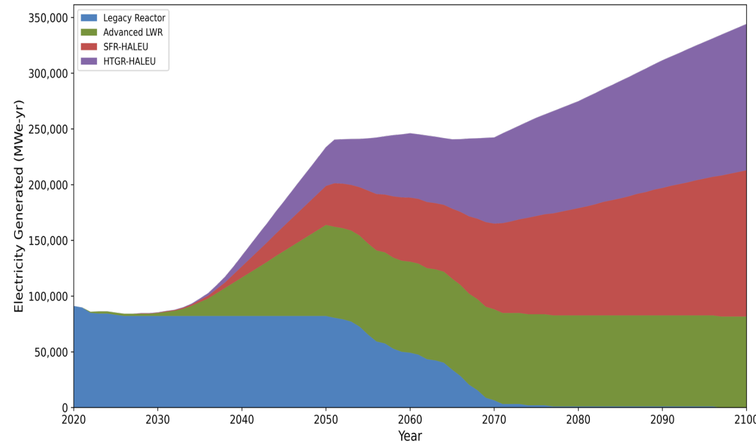
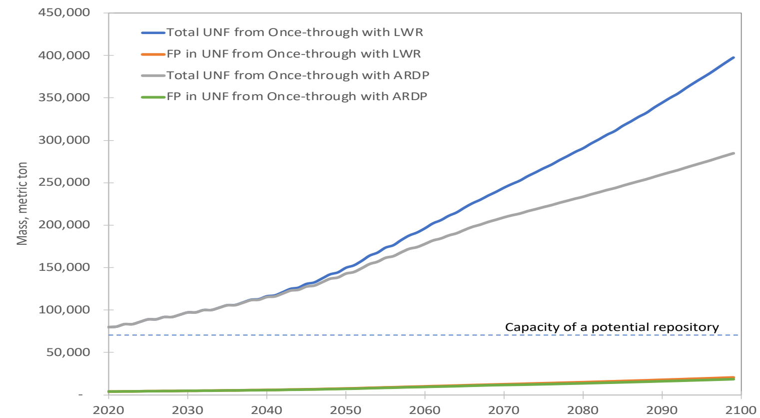
MOTIVATIONS (I)

- **Projection of nuclear energy demand for achieving net-zero emissions economy by 2050 and beyond**
  - ~250 GW in 2050 and ~340 GWe in 2100, consistent with projections by other studies such as NREL, OCED, etc.
  - See DOE SA&I Campaign report https://fuelcycleoptions.inl.gov/SiteAssets/SitePages/Home/NESP_Activity1_Scenario_Study_Final.pdf
- **Projection of Used Nuclear Fuel based on once-through fuel cycles**
  - Existing UNF: ~80,000 MT
  - Cumulative UNF from LWRs and ARDP reactors
    - Total UNF: ~150,000 MT by 2050; ~300,000 – 400,000 MT by 2100
    - FP only : ~7,000 MT by 2050; ~20,000 MT by 2100



Assumed projection of nuclear capacity



Projection of discharged fuel mass

MOTIVATIONS (II)

- **Need geologic repositories**
  - *"… would not eliminate the requirement for geologic repositories for some radioactive wastes…" by National Academies,* Merits and Viability of Different Nuclear Fuel Cycles and Technology Options and the Waste Aspects of Advanced Nuclear Reactors
- **Regarding the projection of nuclear demand, questions are how many repositories are needed and how long manage the repositories**
  - *Once-through fuel cycle*
    - *(how many) Need Yucca Mountain size (~70,000 MT capacity) repository every 20 years*
    - *(how long) Need to manage the repository for almost a million years*
  - *Recycling fuel cycle with the transmutation of long-lived isotopes*
    - *(how many) A single Yucca Mountain size repository is sufficient for ~200 years*
    - *(how long) Can close a repository after a few hundred years*
- **By assuming that the U.S. fuel cycle evolves into a recycling fuel cycle, this project focuses on the transmutation of LLFPs**

PROJECT STRUCTURE

Task	Activities	Project Deliverables
Transmutation option study	• Non-neutron-based transmutation • Neutron-based transmutation (for comparison purpose)	• LLFP transmutation data • Transmutation parameters (energy, intensity, etc.) • Target/blanket transmutation concepts
External incident source option study	• Photon source option study • Proton source option study	• Energetic and high intensity accelerator concepts
National LLFP transmutation facility concept	• Fuel cycle scenario study • National transmutation concept study • Market survey	• Waste generation rates, based on fuel cycle options • National transmutation concept and overall cost

LONG-LIVED FISSION PRODUCTS

- **Long-lived fission products (LLFPs)**
  - LLFPs contribute more than 90% of long-term radiotoxicity of fission products at 1000 years after discharge (e.g., ingestion dose, ICRP-119)

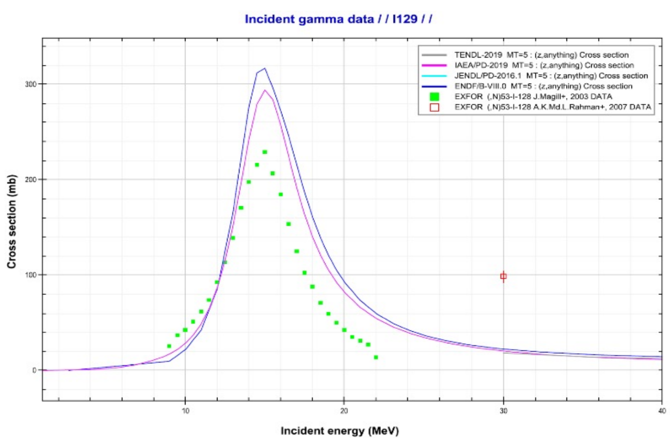
	Tc-99	I-129	Sn-126	Zr-93	Cs-135	Se-79	Total
Once-Through	42.6 [a] ±15.0%	22.1 ±4.4%	14.1 ±9.4%	11.3 ±2.7%	7.9 ±4.9%	1.8 ±0.8%	99.8
Limited-Recycle	46.3 ±4.7%	21.1 ±4.8%	11.9 ±4.7%	11.2 ±1.6%	7.3 ±3.5%	1.8 ±0.7%	99.6
Continuous-Recycle	44.1 ±6.3%	22.8 ±2.1%	12.4 ±5.2%	9.2 ±2.1%	9.4 ±4.2%	1.6 ±0.7%	99.5

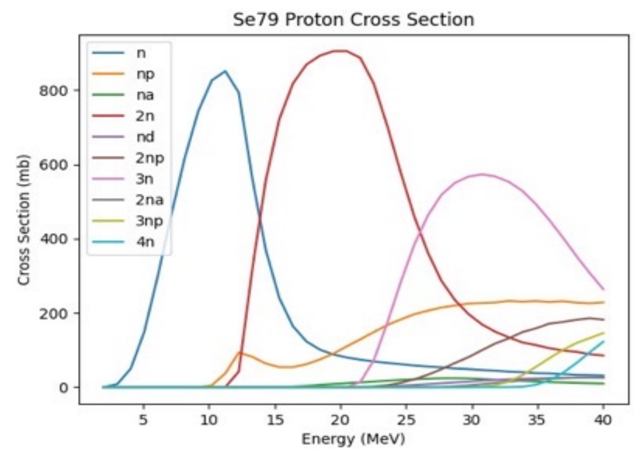
a) Variation from different reactor designs and fuel cycle options

  - LLFP is about 8% of total fission products in the PWR used nuclear fuel
  - Ignored long-lived activation products (such as C-14, Ni-59, etc.) due to low concentration in base materials

PHOTON AND PROTON CROSS SECTIONS

- **Cross-section libraries considered: EXFOR, ENDF/B-VIII, JENDL, TENDL, etc.**
- **Photonuclear and proton reactions**
  - There are threshold energy for photonuclear and proton reactions
  - Optimum reaction energy: 10 – 100 MeV (depending on incident particles and target isotopes)





- **Challenge #1: Cross-section uncertainties**

CROSS SECTION UNCERTAINTIES

- **Assessed cross-section uncertainties using two methods**
  - Total Monte Carlo (TMC) approach: Statistical analysis of stochastic simulations using randomly perturbed cross sections from TASMAN (nuclear data uncertainty generation code based on the TALYS nuclear reaction model code)
  - Uncertainty propagation using TENDL covariance data ($DR^2 = S^TCS$, C=covariance, S=sensitivity coefficient)

- **Cross section uncertainties**

Reaction Type	Tc-99	I-129	Sn-126	Zr-93	Cs-135	Se-79
Photonuclear reaction, total	N/A	[a] 2 – 14 %	~18 %	3 – 12 %	14 – 31 %	21 – 22 %
Total reaction with proton (p,xn)	~13%	~25%	~33%	~21%	~28%	~23%

a) Variation from different assessments

- **Cross-section uncertainties would not be a showstopper**
  - Estimated uncertainties in reaction rates are in the range of 10 - 30%

TRANSMUTATION BY PHOTONS OR PROTONS

- **Transmutation is dependent on energy and flux levels**

LLFP	Natural Decay Half-Life (years)	Photon			Proton		
		Energy, MeV	Flux, γ/cm^2s	Transmutation Half-life (years)	Energy, MeV	Flux, p/cm^2s	Reduction rate for 50 hours irradiation
^{129}I	$15,800 \times 10^3$	23.9	1.0×10^{18}	0.33	70	1.60×10^{18}	8%
^{135}Cs	$2,300 \times 10^3$			0.32	70	1.60×10^{18}	18%
^{126}Sn	230×10^3			0.34	70	1.73×10^{18}	7%
^{93}Zr	$1,500 \times 10^3$			0.44	70	1.85×10^{18}	17%
^{79}Se	327×10^3			0.52	50	1.87×10^{18}	13%
^{99}Tc	211×10^3	N/A			70	1.85×10^{18}	24%

  – Tc-99 is not transmutable by photons as it produces Tc-97 or -98, which have even longer half-lives than Tc-99

- **Challenge #2: Energetic and high-intensity photon/proton sources**

PHOTON & PROTON ACCELERATOR

- **Selected accelerators among more than 500 facilities worldwide**

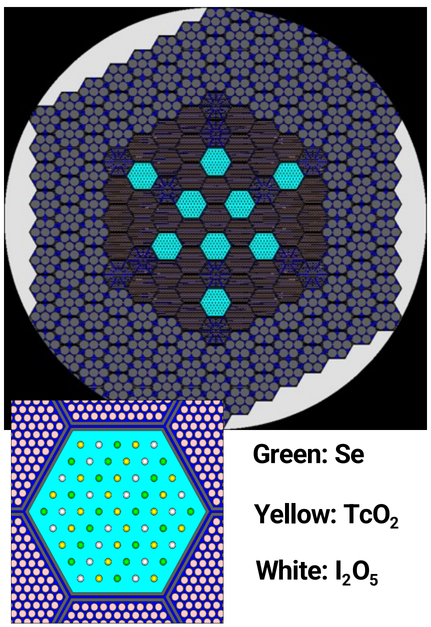
Photon Accelerator	Organization	Status	Energy, MeV	Intensity, γ/sec
VEGA	IFIN-HH, Romania	2025	1 – 19.5	10^{11} – 10^{13}
HIGS	Duke, USA	Operational	1 - 100	10^{11} @~10MeV
FACET-II	SLAC, USA	Operational	1 - 2000	10^{10} – 10^{11}
Gamma factory, CERN	CERN, Switzerland	2040	1 - 400	$6x10^{18}$

Proton Accelerator	Organization	Status	Energy	Flux, p/cm^2-sec
ION-12SC Superconducting Cyclotron	MIT, USA	Operational	12	$1.4x10^{14}$
VD-30	Sichuan Univ. China	TBD	14-26	$2.5x10^{14}$
PIF	PSI, Switzerland	Operational	6-230	$2.5x10^{9}$

- **High energy (10-100 MeV) accelerators are available, but intensity is much lower than the target level (which is ~ 10^{18} γ/sec or protons/cm^2-sec) and irradiation area is narrow (few square centimeters)**

TRANSMUTATION BY NEUTRONS

- **Explored transmutation performances with (1) fission neutrons and (2) spallation neutrons (without fission)**



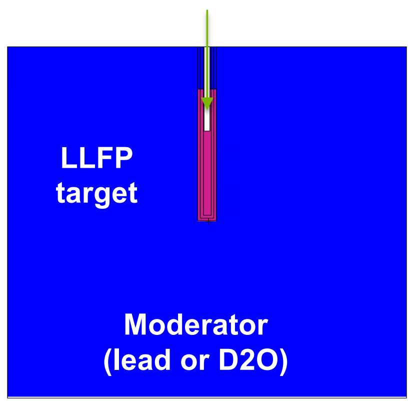
Green: Se

Yellow: TcO_2

White: I_2O_5

LLFP target in reactor

Proton beam



LLFP target in spallation system

	Fission neutrons in Reactor	Spallation neutrons
Spectrum	Thermal	Fast or thermal
Fuel	HALEU, UZr metal	No fuel (no fission)
Target	Se, TcO_2, I_2O_5	Mixture of Lead & LLFP
Moderator	D_2O	Lead (fast), D_2O (thermal)
Flux, n/cm^2-sec	4 x 10^{15}	1.0 – 6.4 x 10^{15}
Performance	Years for 50% net reduction [a)]	Reduction after 5-year irradiation [b)]
Se-79	1.6 years	49 – 89 %
Zr-93		42 – 58 %
Tc-99	5. 3 years	52 – 93 %
Sn-126		87 – 89 %
I-129	3.0 years	44 – 87 %
Cs-135		76 – 97 %

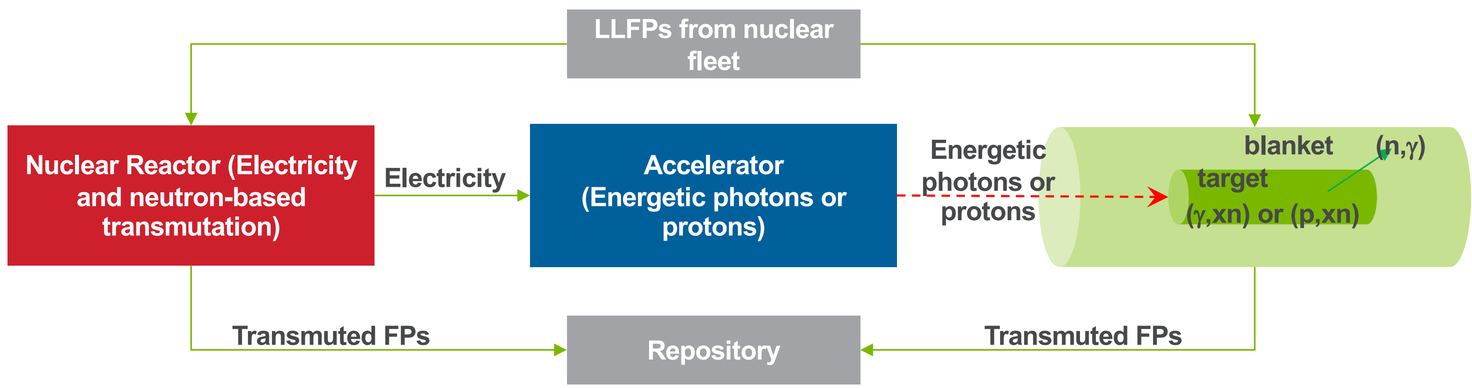
*) Results are preliminary (under review by team)
a) Net reduction = destruction – generation from fission
b) Variation depending on moderators

SUMMARIES OF 1ST YEAR ACTIVITIES

- **Identification of six LLFPs (Se-79, Zr-93, Tc-99, Sn-126, I-129, and Cs-135)**
- **Photon/Proton-based transmutations**
  - Assessment of transmutation conditions (energy and flux levels) for reasonable transmutation
    - Transmutation half-life (irradiation time till 50% reduction) could be less than 1 year with 20 - 100 MeV Photon or Protons with flux (intensity) level of ~10^{18} particles/cm^2-sec
    - Tc-99 is not transmutable with photon
  - Assessment of impacts of cross-section uncertainties on photonuclear and proton reaction rates
    - Impacts of cross-section uncertainties are 10 – 30%
- **Neutron-based transmutations**
  - Transmutation rates based on fission neutrons or spallation neutrons (without fission) are comparable
  - Transmutation half-life is dependent on target isotopes (Zr-93 may take longer)
- **May need a hybrid transmutation option (photon/proton + neutron) for compensating low reaction of specific isotopes (Tc, Sn, etc.)**

ONGOING ACTIVITIES

- **Exploring double transmutation option based on target and blanket concept**
  - Target = LLFPs having high reaction rate with photon or proton (such as Cs, I)
  - Blanket = LLPFs having low reaction rate with photon/proton, but high reaction rate with neutrons (Tc)
- **National transmutation facility concept based on three components (tentative)**
  - Nuclear reactor: electricity and potential transmutation of specific isotopes (Tc, Sn)
  - Accelerator: energetic photon/proton source
  - Transmuter: target and blanket concept



CHALLENGES

Challenges	Activities
LLFPs cross-section uncertainties	• Estimated uncertainties are in the range of 10 – 30% • Uncertainties of specific nuclides and reaction rates would not be a showstopper
Energetic, high-intensity (flux) photon or proton sources	• High energy (10 – 100 MeV) accelerators are available • Looking for accelerators that irradiate a wide area with high-intensity
Transmutation performance	• Develop target/blanket transmutation concepts, including potential hybrid (non-neutron and neutron) transmutation concepts
Design of national transmutation facility	• Predict wastes depending on fuel cycles • Develop a transmutation facility concept • Estimate techno-economic analysis of the transmutation facility

Q&A