U.S. Department of Energy
Advanced Research Projects Agency – Energy (ARPA-E)

Request for Information (RFI)
DE-FOA-0002533
on
Cleaning Up RadioIsotope Enventories (CURIE)

Introduction

The purpose of this RFI is to solicit input for a potential future ARPA-E research program focused on the development of technologies that would enable the effective management of the Nation's commercial used nuclear fuel (UNF). The goals of this RFI are to (1) solicit information about reactor fuel needs for both the current commercial light-water reactor (LWR) fleet and future advanced reactors, and (2) seek insights into technology gaps and/or cost drivers that may be hindering economical recycling of existing LWR UNF.[1] This information is needed to help ARPA-E identify ways in which the Nation's roughly 86,000 MTU[2] inventory of UNF, which has been increasing by approximately 2,000 MTU per year, can best be recycled to support current and advanced reactor fuel needs. Such activities are consistent with ARPA-E's statutory goals, which include supporting the development of transformative solutions for addressing UNF.[3]

ARPA-E is interested in information about technologies with the potential to make recycling UNF at least as economical, safe, and secure as the current once-through fuel cycle.[4] Such technologies would enable a UNF treatment facility to be economically constructed, managed, and operated; yield an actinide product that is cost-competitive with natural uranium (U) obtained from traditional mining and milling; and generate significantly lower waste volumes than those generated from existing commercial UNF treatment facilities. Implementation of advanced nuclear material accounting technologies and incorporation of a safeguards-by-design philosophy would support this objective by enabling precise, remote, near-real-time monitoring and accounting of special nuclear material[5]; decreasing hands-on-

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1 For the purposes of this RFI, recycling of UNF entails (1) fuel treatment to recover valuable actinides (and potentially fission products) from UNF and (2) subsequent reuse of the recovered materials for nuclear and other applications.

2 MTU=metric tons of uranium. According to the U.S. Energy Information Administration, approximately 79,825 MTU were discharged between 1968 and 2017. Approximately 2,000 MTU UNF are discharged per year, meaning that approximately 86,000 MTU have been discharged as of 2020. https://www.eia.gov/nuclear/spent_fuel/

3 ARPA-E was chartered by Congress in the America COMPETES Act of 2007 (P.L. 110-69), as amended by the America COMPETES Reauthorization Act of 2010 (P.L. 111-358), as further amended by the Energy Act of 2020 (P.L. 116-260) (codified at 42 U.S.C. § 16538). ARPA-E's statutory goals are found in 42 U.S.C. § 16538(c). The Energy Act of 2020 amended such goals to include "provid[ing] transformative solutions to improve the management, clean-up, and disposal of radioactive waste and spent nuclear fuel".

4 International Atomic Energy Agency (IAEA). 2011. "The nuclear fuel cycle". https://www.iaea.org/sites/default/files/nfc0811.pdf

5 U.S. Nuclear Regulatory Commission. "Special nuclear material". March 09, 2021. https://www.nrc.gov/reading-rm/basic-ref/glossary/special-nuclear-material.html

inspection requirements; and minimizing operational downtime to verify accuracy of material accounting. In aggregate, these innovations could substantially reduce the volume, heat load, and radiotoxicity of high-level waste requiring permanent disposal while providing a valuable and sustainable fuel feedstock for advanced fast reactors and the existing LWR fleet.

The questions in this RFI are intended to allow relevant stakeholders a mechanism to provide input on (i) the nature of a potential UNF recycling facility, (ii) UNF recycling technology gaps, (iii) existing LWR and future advanced reactor feedstock and fuel needs, and (iv) cost drivers for UNF recycling facility capital and management and operations (M&O) costs. Responses to these questions will enable ARPA-E to refine its success metrics for a potential program aimed at supporting the development of economical, safe, secure, and safeguarded recycling technologies. The questions posed in this section are classified into several different groups as appropriate. ARPA-E does not expect any one respondent to answer all, or even many, of these prompts. Simply indicate the group and question number in your response. Appropriate citations are encouraged. Respondents are also welcome to address other relevant avenues/technologies that are not outlined below.

General UNF Treatment Facility Questions

Multiple factors, including the fuel treatment facility size (i.e., annual production throughput), number, location, and construction techniques, should be considered when determining the most efficient, cost-effective way to treat the Nation's existing UNF. For example, the optimal size of the facilities could be determined based on regional reactor fuel needs, UNF storage locations, and/or proximity to related fuel cycle facilities. In the U.S., since a majority of reactors and independent storage installations for existing UNF are clustered in the Midwest, South, and Northeast, a smaller, suitably-sized fuel treatment facility could be located in each region to accommodate that region's needs. Where possible, integrating or co-locating a UNF treatment facility with another fuel cycle facility (e.g., an enrichment, fuel fabrication, or consolidated interim storage facility) or reactor could further improve overall safeguardability, lessen regulatory costs, and reduce transportation-related burdens compared to a standalone fuel treatment facility. The fuel treatment facility could also be constructed using modular construction techniques or other design and construction philosophies. The questions below will help ARPA-E identify optimal ways in which a potential UNF recycling facility could be designed, built, and operated to the benefit of the nuclear industry and the government, where possible.

1. Discuss the opportunities and challenges of establishing a domestic commercial UNF treatment capability, with particular focus on fuel treatment as an international market enabler for domestic advanced reactor designers.
2. A new UNF treatment facility constructed in the U.S. could operate using one of the following corporate models: (a) government-owned, government-operated (GOGO), (b) government-owned, contractor-operated (GOCO), (c) privately-owned, or (d) public-private partnership. Which of these models would be the best option from a cost, safeguards, and/or any other perspective, and why?
3. Are there any currently existing technical, regulatory, or other bottlenecks that would make co-locating or integrating recycling facilities with fuel cycle facilities or reactors impractical?

4. Of the approximately 86,000 MTU of existing UNF, nearly 22,500 MTU of UNF is considered "older fuel,"[6] which is anticipated to have lower expected dose rates, decay heats, and fission gas releases but would also have relatively lower Pu-241 and higher Am-241 contents than newer fuel.[7,8] Such advantages should reduce shielding requirements and potentially simplify off-gas management, leading to potential M&O cost savings for a UNF treatment facility. Are there any considerations that would suggest that treating older fuel first would be more technically challenging or less economical than the current practice of treating newer fuel first?
5. Given the large UNF inventory currently in wet and dry storage across the U.S., would construction of several smaller, potentially regional treatment facilities be a better approach than constructing one large centralized facility? Why or why not? In either case, would the facilities benefit from modular construction techniques?

UNF Treatment Technologies

The solvent extraction-based Plutonium Uranium Reduction-Extraction[9] process (PUREX), developed in the 1950s, is the *de facto* international standard for commercial UNF treatment, with France's La Hague fuel treatment facility being the largest facility in the world (1700 MT/yr[10]). Although the PUREX process has decades of demonstrated commercial experience globally, PUREX-based facilities tend to have very large footprints. Further, the technology requires several process operations, is susceptible to radiolysis, and has costly head-end processing and waste management operations. As such, opportunities exist to improve fuel treatment economics by reducing the process footprint, reducing waste streams, facilitating regulatory compliance, and enabling accurate nuclear material accounting for each of the PUREX process operations.[11] For example, a new CoDCon process flowsheet has been developed that co-recovers uranium and plutonium (Pu) in defined ratios to accommodate various fuel compositions and increase proliferation resistance.[12] An integral part of the CoDCon flowsheet is online monitoring, which has also

[6] In this RFI, "older fuel" refers to fuel that was discharged from nuclear reactors at least 30 years ago. The estimated older fuel inventory was calculated using annual discharge data from the EIA UNF website. From 1968 and 1991, approximately 22,500 MTU UNF with an average burnup of 21.7 GWd/MTU was discharged from LWRs. www.eia.gov/nuclear/spent_fuel/ussnftab3.php.

[7] W.M. Nutt, Z. Duncan, T. Cotton, Paper No. 11008, "Prioritization Criteria for the Selection of Used Nuclear Fuel for Recycling," *Waste Management 2011 Conference*, Phoenix, AZ, February 27-March 3, 2011, pp. 11008_1-11008_13, Waste Management Symposia (2011).

[8] Oak Ridge National Laboratory, Report No. ORNL/TM-2012/308, "Categorization of Used Nuclear Fuel Inventory in Support of a Comprehensive National Nuclear Fuel Cycle Strategy," published December 2012.

[9] The Plutonium Uranium Reduction Extraction process has also been referred to as the "Plutonium URanium Extraction" or the "Plutonium Uranium Recovery by Extraction" process.

[10] https://www.world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/processing-of-used-nuclear-fuel.aspx. Accessed 04/23/2021.

[11] In general, standard process operations for UNF treatment facilities, regardless of the separations technology used, include fuel receipt and head-end processing (e.g., shearing, dissolution), separation (e.g., via solvent extraction or electrorefining), waste management (including vitrification and storage), off-gas capture and management, and conversion operations for future re-enrichment or fuel fabrication.

[12] G. J. Lumetta, et al., Simulant testing of a codecontamination (CoDCon) flowsheet for a product with a controlled uranium-to-plutonium ratio, *Sep. Sci. Technol.* 2019, *54*, (12), 1977-1984. DOI: 10.1080/01496395.2019.1594899

been shown to enable remote, near-real-time monitoring and control of uranium and plutonium extraction behavior during the process.[13]

Promising alternative treatment technologies to PUREX-like technologies include dry[14] processes, such as fluoride volatility, supercritical CO_2 (sCO_2) extractions or other potential technologies. Both processes use fewer process chemicals that are less prone to radiolysis than PUREX-like processes. Fluoride volatility is used to recover uranium and/or plutonium fluorides that are directly amenable to re-enrichment while producing a relatively small quantity of highly radioactive waste. Supercritical CO_2 extractions enable potential recovery of uranium or U/Pu directly from used oxide fuel, with the added benefit that sCO_2 is easily recyclable. Though these processes appear to be promising alternatives to PUREX-like processes, none of them have been commercialized. The questions below are intended to enable ARPA-E to obtain additional information about advances and potential areas of innovation for these UNF treatment technologies and identify other potential UNF treatment technologies or approaches to explore.

1. What are some technology challenges and opportunities for fluoride volatility that could improve the viability and cost-effectiveness of this fuel treatment technology?
2. Hybrid halide volatility-solvent extraction-based processes such as the chloride volatility-based Hybrid ZIRCEX and fluoride volatility-based FLUOREX processes have been developed as potential UNF treatment technologies. While these processes may simplify or improve head-end processing for UNF treatment, introduction of corrosive gases may complicate flowsheet design. What are some additional challenges associated with such hybrid processes, and do the benefits of hybridization outweigh the challenges compared to traditional head-end processing?
3. Briefly discuss the technology gaps for sCO_2 extractions that may impact its use as a cost-effective fuel treatment technology as well as any opportunities to improve the viability of this technology.
4. Briefly discuss how safeguards-by-design, security-by-design, and other modern advances in safeguards and security technologies can be incorporated into UNF treatment technologies, including potential synergies between such safeguards & security applications and improved overall cost-effectiveness. This topic may include areas such a machine learning and artificial intelligence capabilities for material accountancy applications, or other technologies that facilitate process monitoring opportunities to detect normal/off-normal process conditions.
5. Recent research on off-gas management has focused on the development of aerogels and silver sorbents to chemisorb iodine (I), metal organic frameworks and molecular sieves for krypton (Kr) capture, and engineered zeolites for Kr and xenon (Xe) capture. Which of these technologies has the greatest potential to simplify and improve the cost-effectiveness of off-gas management in future treatment facilities relative to the current state-of-the art approaches, and why? Are there any other off-gas treatment technologies worth exploring?
6. What are some other separations technologies that may simplify and lower the cost of recovering uranium (U) and transuranics[15] (TRU) from UNF? What are the remaining challenges, technical or

[13] A. M. Lines, et al., Sensor Fusion: Comprehensive Real-Time, On-Line Monitoring for Process Control via Visible, Near-Infrared, and Raman Spectroscopy, *ACS Sensors* 2020, *5*, (8), 2467-2475. DOI: 10.1021/acssensors.0c00659

[14] "Dry" processes are those that do not use water as a process solvent.

[15] *Transuranic elements*, or *transuranics*, are actinide elements heavier than uranium. Relevant TRUs for nuclear applications and radioactive waste management typically include neptunium, plutonium, americium, and curium.

otherwise, that need to be considered for other potential technologies to enable scale-up and/or commercialization?

Reactor Fuel Needs

The nuclear reactor landscape has changed dramatically over the last decade to include a variety of advanced reactor designs, including, but not limited to, molten salt reactors, sodium fast reactors, high-temperature gas reactors, and microreactors. Although many of these reactor concepts are intended to consume HALEU fuel, fast-spectrum reactors in particular can also consume plutonium and other transuranic elements recovered from existing UNF, thereby reducing the long-term heat load and radiotoxicity of waste requiring geologic disposal and improving uranium utilization while producing electricity.[16] In addition, the slightly enriched (~1%) uranium (SEU), which comprises approximately 96% of UNF, represents a vast supply of uranium that could be reused after re-enrichment, combined with transuranics and burned in fast reactors, or used in Canadian Deuterium Uranium (CANDU) reactors under suitable market conditions. The questions below will enable ARPA-E to assess the market opportunities for SEU and/or TRU recovered from UNF. Generally speaking, U/Pu would be fuel intended for LWRs and/or advanced reactors, and U/TRU would be intended for advanced fast reactors.

1. What is the expected fuel burnup range of your reactor technology?
2. Briefly describe the opportunities and challenges associated with the availability of HALEU, U/Pu, and/or U/TRU on fuel procurement and reactor design.
3. How would the use of U/Pu and/or U/TRU fuel impact your reactor performance characteristics? For example, would it require substantial alterations to your reactor design?
4. Briefly discuss, from a reactor neutronics, radiation handling, or other relevant perspective, the range of desired fissile material (e.g., isotopes, weight percent) content in the your reactor's fuel, including any limitations on the content of other transuranic isotopes.
5. Do you perceive a market for SEU? If so, how large do you predict the market would be?
6. What are some other options for economically reusing SEU beyond those provided above?

Cost Drivers for SEU and UNF Treatment Facilities

The Nuclear Fuel Services plant, the only privately-owned UNF treatment facility to ever treat commercial UNF in the U.S., cost approximately $33 million[17] to construct in the 1960s. It used the PUREX process to treat 650 MTU of defense- and civilian UNF from 1966-1972 but ultimately shut down in 1976 because of new regulatory requirements and upgrades that proved too costly (~$600 million) to implement. It is estimated that any new UNF treatment facilities constructed in the U.S. today using a similar treatment technology are estimated to be very expensive, with capital costs ranging from an optimistic $250 million

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[16] A 2014 Nuclear Fuel Cycle Evaluation and Screening study, INL/EXT-14-31465, sponsored by the U.S. Department of Energy's Office of Nuclear Energy, proposed three fuel cycle scenarios that would potentially lead to improved nuclear waste management (including reduction of waste requiring geologic disposal by a factor of ten and uranium disposal needs by a factor of 100 or more), fuel resource utilization, and land requirements compared to the current once-through fuel cycle: continuous recycle of U/Pu with new natural-U fuel in fast critical reactors, continuous recycle of U/TRU with new natural-U fuel in fast reactors, and continuous recycle of U/TRU with new natural-U fuel in fast and thermal reactors.

[17] J. Blankenhorn and B. Bower, Paper No. 11203, "West Valley Demonstration Project – Past, Present, and Future," *Waste Management 2011 Conference*, Phoenix, AZ, February 27-March 3, 2011, pp. 11203_1-11203_13, Waste Management Symposia (2011).

to $20 billion, depending on throughput.[18] Moreover, the estimated price of new fuel produced from UNF ranges from approximately $3,000 to $8,500/kg for mixed oxide fuel, compared to $2100/kg for LEU and about $20,000/kg for HALEU produced from natural uranium.[19] Such prices would significantly impacts the revenue stream of a potential recycling facility.

Major cost drivers for fuel treatment facility capital costs include costs associated with head-end processing, separations, and waste management (including vitrification) operations, all of which could benefit from innovative design approaches to improve their efficiency and lower costs. In addition, there is the possibility of developing new revenue streams, including mixed U/Pu or U/TRU feedstocks to supply the existing LWR fleet and/or future advanced reactors, and commercially-valuable materials derived from UNF fission products (e.g., radiopharmaceuticals, industrial feedstock).

The questions in this section aim to (1) identify potential scenarios in which U/TRU fuel could become cost-competitive and (2) uncover potential areas of innovation in manufacturing or process designs that could reduce both the capital and M&O costs for a UNF recycling facility.

1. Considering future fuel cycle needs, briefly discuss the cost drivers – including HALEU availability – that could make U/TRU fuel economically attractive.
2. Briefly discuss the design philosophy, construction and manufacturing challenges, or improvements to UNF recycling equipment that could improve its economics.
3. What technical advances outside the nuclear field (e.g., the chemical or metallurgical industries) have occurred that could be applied to a UNF recycling facility to drive down costs, and to what extent?
4. Briefly discuss the opportunities and challenges of improving the cost-effectiveness of vitrification or other waste stabilization technologies.
5. Chloride volatility to chemically declad fuel and voloxidation to capture tritium and some volatile fission products have been investigated as potential processes that could improve head-end processing. What are the challenges in incorporating such technologies into a traditional UNF treatment facility, and what are the potential impacts of implementing these technologies on the facility footprint and off-gas/waste management costs?
6. Briefly discuss the benefits and challenges of co-recovering materials from UNF. This discussion should include economic, supply chain, technical, and/or other factors.

Approaches Not of Interest

This potential program is focused on supporting the development of innovative technology solutions that will enable cost-effective, safeguardable treatment of UNF. Approaches not of interest include

- Policy and legislative recommendations to address UNF
- General discourse on nuclear power or UNF treatment

[18] Idaho National Laboratory, Report No. NTRD-FCO-2017-000265, "Advanced Fuel Cycle Cost Basis – 2017 Edition," Module F1: Spent Nuclear Fuel Aqueous Reprocessing Facility, published September 29, 2017.

[19] Based on internal calculations for fuel costs, with recycling costs of $250-900/kgHM and uranium ore prices of $62/kg. Market prices for uranium, conversion, and enrichment current as of February 22, 2021. HALEU enrichment costs are higher relative to LEU and based on cost of the Centrus ACM pilot scale facility being developed at Piketon, Ohio.

- Technologies that are not effective for treatment of existing LWR used fuels (Note that this does not preclude technologies that are effective for both existing LWR and potential advanced reactor fuels.)

Please carefully review the REQUEST FOR INFORMATION GUIDELINES below. Please note, in particular, that the information you provide will be used by ARPA-E solely for program planning, without attribution. **THIS IS A REQUEST FOR INFORMATION ONLY. THIS NOTICE DOES NOT CONSTITUTE A FUNDING OPPORTUNITY ANNOUNCEMENT (FOA). NO FOA EXISTS AT THIS TIME.**

Purpose and Need for Information

The purpose of this RFI is solely to solicit input for ARPA-E consideration to inform the possible formulation of future research programs. ARPA-E will not provide funding or compensation for any information submitted in response to this RFI, and ARPA-E may use information submitted to this RFI without any attribution to the source. This RFI provides the broad research community with an opportunity to contribute views and opinions.

REQUEST FOR INFORMATION GUIDELINES

No material submitted for review will be returned and there will be no formal or informal debriefing concerning the review of any submitted material. ARPA-E may contact respondents to request clarification or seek additional information relevant to this RFI. All responses provided will be considered, but ARPA-E will not respond to individual submissions or publish publicly a compendium of responses. **Respondents shall not include any information in the response to this RFI that could be considered proprietary or confidential**.

Responses to this RFI should be submitted in PDF format to the email address **ARPA-E-RFI@hq.doe.gov** by **5:00 PM Eastern Time on Monday, June 14, 2021**. Emails should conform to the following guidelines:

- Please insert "Response to CURIE - <your organization name>" in the subject line of your email
- In the body of your email, include your name, title, organization, type of organization (e.g. university, non-governmental organization, small business, large business, federally funded research and development center (FFRDC), government-owned/government-operated (GOGO), etc.), email address, telephone number, and area of expertise.
- Responses to this RFI are limited to no more than 10 pages in length (12-point font size).
- Responders are strongly encouraged to include preliminary results, data, and figures that describe their potential processes.