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# DOE SNF Technology Development Necessary for Final Disposal

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## ABSTRACT

Existing technology is inadequate to allow safe disposal of the entire inventory of U.S. Department of Energy (DOE) spent nuclear fuel (SNF). Needs for SNF technology development were identified for each individual fuel type in the diverse inventory of SNF generated by past, current, and future DOE materials production, as well as SNF returned from domestic and foreign research reactors. This inventory consists of 259 fuel types with different matrices, cladding materials, meat composition, actinide content, and burnup.

Management options for disposal of SNF include direct repository disposal, possibly including some physical or chemical preparation, or processing to produce a qualified waste form by using existing aqueous processes or new treatment processes. Technology development needed for direct disposal includes drying, mitigating radionuclide release, canning, stabilization, and characterization technologies. While existing aqueous processing technology is fairly mature, technology development may be needed to apply one of these processes to SNF different than for which the process was originally developed. New processes to treat SNF not suitable for disposal in its current form were identified. These processes have several advantages over existing aqueous processes.

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# INTRODUCTION

As part of its role as DOE SNF lead laboratory, the Idaho National Engineering Laboratory (INEL) will help coordinate research, development, and testing of treatment, shipment, and disposal technologies for all DOE SNF (Program Plan, 1995). Fulfilling this role, the INEL has taken the lead to identify technology development needed for safe disposal of every type of DOE SNF. This does not include Naval SNF, since the Navy has a separate technology development program for issues related to disposal of Naval SNF.

Technology development embodies the scientific and engineering investigations required to ascertain basic principles of a process or concept. This includes basic experimentation for proof of principle, bench-scale and pilot-scale systems, experimental programs for data collection and analyses, development of prototypical designs and first-of-a-kind demonstration hardware, and/or application of known principles to new or differing conditions. It does not include application of off-the-shelf technology.

The DOE inventory consists of 259 fuel types with different fuel matrices, cladding materials, fuel meat composition, actinide content, and burnup. SNF is defined (Programmatic Environmental Impact Statement, 1995) as irradiated fuel, targets, or other material containing uranium, plutonium, or thorium that is permanently withdrawn from a nuclear reactor or other neutron irradiation facility following irradiation, the constituent elements of which have not been separated by chemical processing. Such materials include essentially intact fuel and disassembled or damaged units and pieces. Examples include irradiated reactor fuel, production targets, slugs and blankets that are presently in storage or that will be accepted for storage at DOE facilities. Table 1 lists principal generation sources of DOE SNF. Components or materials that have been historically retained with SNF (e.g., control rods and canisters), or for which comanagement with SNF is considered necessary for technical purposes (e.g., corrosion products retained on surfaces

**Table 1.** Principal generation sources of DOE SNF.

| | |
|---|---|
| Materials Production Fuels | This includes those nuclear fuels, drivers, and targets that historically were processed by DOE to recover valuable materials. Examples include N-Reactor SNF stored at Hanford and K/L/P reactor SNF and targets stored at the Savannah River Site (SRS). |
| Naval Nuclear Propulsion Fuels | This includes fuels irradiated in Naval nuclear reactors. Like production fuels, Naval propulsion SNF was processed to recover valuable materials. Some unprocessed Naval SNF remains in storage at the INEL. Additional quantities will be placed into storage as fuel is withdrawn from vessels. |
| Research Reactor Fuels | DOE has long supported nuclear research activities, both in the U.S. and overseas. SNF from research reactors is stored in the U.S. at a number of DOE sites (primarily Hanford, INEL, and SRS) and at numerous (primarily active) university and government research reactor sites within the U.S. Additional research reactor SNF is anticipated to be returned to the U.S. from foreign research reactors. Several examples of research reactor SNF stored within the DOE complex include Experimental Breeder Reactor-II at INEL, Molten Salt Reactor Experiment at Oak Ridge National Laboratory (ORNL), High Flux Beam Reactor at Brookhaven National Laboratory, university Materials Test Reactor-type at SRS, and High Flux Isotope Reactor at ORNL. |
| Specialty Fuels | This includes small quantities of unique SNF irradiated in small, specialty-type reactors. An example is SNAP fuel from the space program stored at INEL and ORNL. |
| Special Case Commercial Nuclear Power Reactor Fuels | Some SNF from early (or demonstration) commercial power reactors (Shippingport, Peach Bottom, Fort St. Vrain, etc.) is stored primarily at Hanford, INEL and ORNL. Also included is Three Mile Island debris stored at the INEL, and fuel procured for both the Materials Characterization Center at Hanford and dry storage demonstrations at INEL. |

of fuel assemblies), will be managed as SNF. Other nonfuel-bearing components or materials encountered with DOE-owned SNF, or colocated within SNF facilities, will not be managed within the SNF program. This recognizes the practicality of maintaining separate disposal paths for some components or materials and the existence of DOE organizations and programs more suitable for effective management of these materials [e.g., transuranic (TRU), low-level radioactive waste (LLW)].

## MANAGEMENT OPTIONS FOR DISPOSAL

DOE is planning for the ultimate disposition of DOE-owned SNF in a geologic repository. However, decisions have yet to be made regarding a final disposal site and form in which fuel will be disposed. Detailed technical acceptance criteria for DOE-owned SNF are also needed in order for SNF to be prepared for ultimate disposition. Currently, only preliminary requirements for DOE-owned SNF exist, and the current technical baseline only includes commercial SNF and vitrified high-level waste (HLW). The repository is scheduled to be ready for use by 2015 and will have received all of the DOE SNF by 2035.

Options for disposal include directly disposing SNF into a geologic repository or processing to produce a qualified waste form using either existing aqueous or new treatment processes. Each option has unique advantages and disadvantages relative to stakeholder support. However, not all SNF is currently in a form suitable for direct repository disposal and may require processing prior to disposal. Use of existing processing facilities has the disadvantage of separating weapons-grade material from waste material and producing large volumes of secondary waste. New processes may need to be developed to satisfy stakeholder concerns over nuclear proliferation and production of more waste.

## Direct Disposal

One of the disposal strategies is to build upon the precedence established by the Office of Civilian Radioactive Waste Management (RW) that allows direct disposal of qualified fuel types. It is probable that certain DOE SNF will prove acceptable for direct repository disposal. Technology may be needed in certain instances to mitigate risks associated with geologic disposal of certain SNF (criticality, releases to the environment, etc.).

Technology required for direct disposal depends on the state of the SNF, i.e. bare, canned, or stabilized (see Table 2). Questions listed in Table 3 were asked for each fuel type in the DOE SNF inventory. Drying, radionuclide release, canning, stabilization, and characterization are considered the primary parameters associated with direct disposal. *Drying* pertains to any additional drying research necessary to remove loose and/or bound water from the fuel to a specified level prior to placing the SNF into a multi-purpose canister (MPC) for direct disposal. This applies to SNF that has been previously stored wet or where moisture has entered into the SNF package. *Prediction of radionuclide release rates* and degradation of SNF in the repository is important regardless of whether SNF is bare, canned, or stabilized. Depending upon drivers, such as regulatory requirements, additional *characterization* may be required for disposal. Characterization should be confined to the minimum needed and performed according to guidelines given in the National Spent Nuclear Fuel Program Characterization Plan (Technology Integration Plan, 1994). *Canning* development refers to research that must be conducted to design, fabricate, or load a new can into an MPC. A can may be used to prevent SNF interaction with the storage environment, maintain the geometry of the SNF, or prevent the release of material into the environment. Note that canning development does not include engineering applications, but is meant to indicate where development of technology is necessary. *Stabilization* may be necessary to maintain configuration of the SNF for criticality control or to enhance the performance of the disposal form. Stabilization includes selecting a chemically and physically compatible stabilizer as well as how to emplace the stabilizer.

**Table 2**. Information needed for various states of direct disposal.

| BARE | CANNED | STABILIZED |
|---|---|---|
| Drying<br>Release rate<br>Characterization | Drying<br>Release rate<br>Canning technology<br>Characterization | Drying<br>Release rate<br>Canning technology<br>Stabilizer selection<br>Stabilizer emplacement<br>Characterization |

**Table 3**. Questions asked to gather information.

IDENTIFICATION OF DIRECT DISPOSAL TECHNOLOGY DEVELOPMENT NEEDS

- Is there any technology that must be developed to dry the SNF for disposal?
    - How will the SNF be dried?
    - How will the level of dryness be measured?
    - How dry is dry enough?
- Is there any technology that must be developed to mitigate radionuclide release?
- Is there any technology that must be developed to can the SNF?
- Is there any technology that must be developed to stabilize the SNF?
- Is there any technology that must be developed to characterize the SNF for final disposal?

## Drying

It is important that water content remaining in a dry-stored container be maintained below a certain limit to minimize subsequent degradation of SNF. Several of the spent fuel types presently in basin storage, particularly aluminum plate fuels that have been stored bare, must be dried to remove water after retrieval from basin storage. Corroded SNF may contain a substantial amount of water, either as water of hydration of the corrosion product, as physisorbed water on the high surface area of the corrosion product, or as water inclusions. Amounts of residual water in SNF may impact prolonged safe interim storage. Radiolysis of residual water might present a risk of hydrogen embrittlement of containment, over-pressurization of the containment, or hydrogen ignition upon retrieval and opening of the canned SNF. In addition, any spent fuels that might be water-logged, such as bare plate fuels with failed cladding, will need special drying protocols to prevent further disruption of the fuel structure by swelling or cavitation that might occur during rapid or uncontrolled drying of the fuels. Such disruption might result in loss of configuration in fuels whose integrity has been compromised by extensive corrosion. Given the variety of fuel types, condition and configuration in the SNF inventory, and the undefined mix of sediment and corrosion products, appropriate protocols must be developed to dry corroded SNF safely to defined, acceptable levels of dryness. These technologies concern both trapped moisture in sludge and chemically or physically bound water in the fuel matrix.

Technology development is required for determining the level of moisture remaining within a complex geometry fuel element. Moisture measurement instrumentation must be: (a) able to accurately determine moisture content that remains within/on a fuel element while in a dryer; (b) operated remotely and with minimum maintenance; and (c) able to operate effectively within a high radiation field. Technology must be developed to dry a complex configured fuel element with corrosion product on and between the fuel plate structure. The technology must be able to quickly and thoroughly dry an entire batch of fuel elements. A combination of heating and vacuum procedures will be used to attain the desired level of dryness. The major challenge is removal of moisture from corrosion products without damaging the delicate plate structure of the elements.

## Radionuclide Release

It will be necessary to be able to predict release rates of radionuclides from SNF in the repository. Research must be conducted separately for the various fuel forms, i.e., metal, U-alloy, hydride, oxide, mixed oxide, and graphite. Radionuclide release rate will be dependent upon leach rates and resultant surface area. It may be possible to use some methodologies and data developed by RW for commercial SNF. The technology should be able to take a relatively short period of corrosion and leaching data and project it to 100,000 years. The leaching considered is from fission products, actinides, and activation products in the meat, and between the fuel meat and cladding.

Development of a standard container for disrupted SNF is needed. The purpose of this technology is to develop the standard design and materials to can SNF for placement into the repository. The materials must not have interactions with the SNF and should maintain SNF configuration for 100,000 years. Stabilizing materials may contain neutron poisons, if required. The neutron poisons should not leach differently or separate from the SNF.

## Canning Technology

MPC development is needed for all SNF at the INEL bound for the repository. Once development is complete, SNF can be placed into an MPC and be assigned a road-ready status. Technology associated with development of new canning prototype designs and techniques is needed.

## Stabilization

Development of stabilizer selection and emplacement technology may be necessary for criticality control of intact zirconium-clad, intact stainless-steel-clad, and intact aluminum-clad

SNF in the repository. Materials compatibility issues may dictate a different stabilizer for each of the different fuel types. The materials must not have interactions with the SNF and should maintain SNF configuration for 100,000 years. Stabilizing materials may contain neutron poisons if required. The neutron poisons should not leach differently or separate from the SNF. Many of the SNF types have unique geometries and configurations. How to emplace stabilizing materials in and around SNF must be determined.

Also, criticality verification and validation of the disposal package is needed. Computer codes have been proven for certain situations, but details incorporated into proposed methods for HEU fuel packaging (stacking, poisoned arrays, etc.) have yet to be verified.

Research is needed to evaluate several potential disposal forms for aluminum SNF and determine the best candidates for further development. Proposed forms include: chop and dilute with depleted uranium (DU), melt and dilute with DU, chemically dissolve and dilute with DU, consolidate and add neutron poison, limit fissile mass per container, chop and add neutron poison, melt and add neutron poison, or chemically dissolve and add neutron poison.

## Characterization

Technology development is needed to determine fissile content of highly enriched uranium (HEU) SNF by nondestructive examination (NDE). It is essential to develop a device to characterize SNF without having to perform extensive recalibration for different shapes and forms. Neutron interrogation would use a neutron source to pulse segments of a sealed package, e.g., an irradiated fuel rod, or canister filled with scrap, to produce delayed neutrons (measured) that would be somewhat indicative of fissile atom density in the package. Known quantities of fissile materials in shapes or forms similar to existing fuels would have to be used to calibrate the interrogator. Such devices may have been tested, at least as prototypes, on perhaps one of a kind type packages. Relative accuracy would have to be determined, as would its suitability on different shapes. Detectors must also be able to function in a radiation field up to a specified Rad/hr level.

Since characterization of DOE SNF on a stick-by-stick basis is cost prohibitive, data regarding representative samples from each group of SNF should be obtained for planning purposes in preparation for repository disposal.

## New Applications of Existing Processes

Some of the existing SNF processing facilities can assist with DOE SNF management issues, e.g., modifying processes and equipment to stabilize SNF for storage and disposal. Processing is intended as a method to achieve safe storage and prepare for disposal by chemically dissolving SNF, then solidifying and immobilizing it into the disposal form. The question shown in Table 4 was asked to identify any new technology needed to modify existing aqueous process flowsheets. This is applicable only for those fuels where processing is considered to be necessary to produce an acceptable form for repository disposal.

**Table 4**. Question asked to gather information.

IDENTIFICATION OF TECHNOLOGY DEVELOPMENT NEEDS FOR PROCESS MODIFICATION

- Is there any technology that must be developed to modify existing flowsheets for other fuel types?

Aqueous processing technology is generally a mature technology. However, specific head-end flowsheets may have to be developed for dissolving certain SNF types in preparation for separation and/or solidification into a glass or glass-ceramic form. Several different fuel dissolution process flowsheets and types of equipment are needed to accommodate the variety of fuels in the inventory.

As a contingency to shipping, storage, and disposal of some stainless steel and zirconium-clad SNF, it may be in the best interest of DOE to process small amounts of these materials at existing processing facilities such as the canyons at SRS. Technology would have to be developed to allow these facilities, configured for processing aluminum-clad SNF, to process stainless steel or zirconium-clad SNF. This activity would involve research to verify the feasibility of new flowsheets to treat small volumes of SNF using existing SRS canyons.

## New Processing Technologies

New, advanced processes offer the potential advantage of separating HLW from fissile material without increasing the stockpile of nuclear weapons material or producing large volumes of secondary waste. This can substantially decrease total volume of SNF to a smaller volume of HLW that must be disposed in the repository, while producing quantities of LLW that must also be disposed. These new processing technologies would process all of the SNF that is not acceptable for direct disposal into a repository in its current form.

New processes (Technology Integration Plan, 1994), such as electrometallurgical treatment, GMODS, plasma arc, and chloride volatility, which can be used to treat DOE SNF, have been proposed. Each technology proposes to handle the entire inventory, with the exception of aluminum SNF for the electrometallurgical treatment and the MSRE SNF for the GMODS and chloride volatility processes. The chloride volatility process separates HLW from uranium and structural components of fuel, the electrometallurgical process separates uranium from fission products and transuranic elements, and the GMODS and plasma arc processes homogenize material into a disposal form that would go directly into the repository.

Evaluation of new processes requires careful evaluation of tradeoffs associated with each process. Technologies that yield a form acceptable for stable interim storage and disposal in a single process would be attractive.

## CONCLUSIONS

Technology development needs for disposal of DOE-owned SNF are summarized in Table 5. The number of disposal-related activities will probably increase as requirements for disposal are defined and if a new dispositioning facility is designed, built, and operational in the next 10–15 years. Strategies to regionalize DOE SNF inventory help to reduce the scope of the technology development program.

**Table 5.** Summary of technology development needs for DOE SNF disposal.

DIRECT DISPOSAL
- Drying technology
  - Measure moisture levels and determine best methods of drying SNF
- Radionuclide release
  - Predict release rates of radionuclides
  - Develop a standard container for disrupted SNF
- Canning technology
  - MPC development
- Stabilization
  - Neutron poisons and other stabilizing materials to maintain SNF configuration
  - Emplacement of SNF stabilizing materials
  - Criticality verification/validation of disposal form
  - Pulverizing and vitrifying SNF
- Characterization
  - Determine fissile content of HEU SNF by NDE

NEW APPLICATIONS OF EXISTING PROCESSES
- Process modification
  - Applicability of current processing to stainless steel or zirconium SNF

**Table 5.** (continued)

NEW PROCESSES

New processing technology
- Electrometallurgical treatment
- GMODS process
- Plasma arc process
- Chloride volatility process

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