

SPENT FUEL FISSILE MASS VERIFICATION AT THE ANL-WEST RADIOACTIVE SCRAP AND WASTE FACILITY

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ABSTRACT

A fissile mass verification method is being developed for safeguarding nuclear spent fuel in dry storage. The Shielded Measurement System verification method will be tested using Experimental Breeder Reactor-II spent fuel currently in storage at the Argonne National Laboratory-West Radioactive Scrap and Waste Facility, a subsurface dry storage facility. This approach is simple, cost-effective and based on the availability of adequate process knowledge. A description and example of the verification method is provided, along with a description and status report of the Shielded Measurement System, the experimental platform for the confirmatory measurements. Finally, the applicability of the Shielded Measurement System method to the safeguarding and data qualification of other DOE fuels is considered.

INTRODUCTION

Recent DOE guidance has suggested that the fissile content of spent nuclear fuel must be verified. This guidance came in the form of a letter, "Recommendations on Safeguarding Irradiated Nuclear Fuels," issued by the Office of Security and Emergency Operations (General Eugene E. Habiger, August 31, 1999) stating that many aged DOE fuels "do not have well-characterized or known plutonium or enriched uranium content" and that "characterizing the fissile content of the aged irradiated nuclear materials through measurements or some other acceptable validation means is not only important to safeguards but is needed in support of safety and material management and dispositioning activities." Although the Habiger letter does not specifically define validation criteria, it is expected that any validation method will be based on the requirements set forth in DOE Manual 474.1-1, *Manual for Control and Accountability of Nuclear Materials*.

Argonne National Laboratory-West (ANL-W) is currently developing a spent fuel verification method, called the Shielded Measurement System (SMS) method, to meet the expected requirements for various types of spent fuel around the DOE complex. The basis for the SMS method is not new--previous measurements performed by other groups have used passive detection of neutron (both total and coincidence) and gross gamma emission rate to verify operator-declared attributes (e.g., exposure and plutonium content). However, these systems were usually designed to verify light water reactor fuel in underwater storage facilities (1,2). The SMS verification method, on the other hand, is primarily intended to verify fuel in dry storage. This paper includes the following:

- Description of the Radioactive Scrap and Waste Facility (RSWF) and the Experimental Breeder Reactor-II (EBR-II) spent fuel to be used for SMS verification method testing;
- Description of the SMS verification approach, a simple and cost-effective verification approach based on adequate process knowledge;
- Recent example of a similar verification approach successfully implemented in Kazakhstan at the BN-350 reactor;
- Description of the measurement platform used in the ANL-West approach, the Shielded Measurement System, and a status report on the method implementation at ANL-West;
- Preliminary analysis of the DOE spent fuel inventory to determine where this approach might be applied outside of ANL-West.

EBR-II SPENT FUEL AT THE RSWF

Over 30 years of EBR-II operation at Argonne National Laboratory-West produced a sizable and somewhat unique spent fuel inventory. Because elemental sodium provides the interface between fuel and cladding in the EBR-II fuel, it is not suitable for direct disposal in a repository—treatment is required. ANL is currently beginning a 10-12 year electrometallurgical spent fuel treatment program to ready the fuel for disposal. Prior to treatment, the EBR-II fuel is being stored at RSWF, a subsurface storage facility for spent fuel and remote-handled mixed and radioactive waste located on the ANL-West site near Idaho Falls, Idaho.

Currently, there are 1350 in-ground silo type storage locations of which 939 are full of remote handled waste or spent fuel. The loading practice for waste shipped to RSWF after 1978 includes remotely loading the waste into an inner can (29.5 cm diameter, 150 cm length) then placing this can in a second container (32.4 cm diameter, 187 cm length) using one of the ANL-West hot cells. This can arrangement is then transferred to the RSWF via a shielded cask where it is lowered into a carbon steel silo (40.6 cm diameter, 376 cm length) before placing appropriate shielding on top of the inner can, as shown in Figure 1.

The EBR-II fuel inventory consists of driver and blanket assemblies. Driver assemblies typically have 61 elements, each with a diameter of 0.58 cm and a fuel region of 34.3 cm. The 19 elements of the radial blanket assemblies have a 1.25 cm diameter and a 140 cm fuel region while the axial blanket assemblies (used only between 1966 and 1969) have 18 elements with a 0.80 cm diameter and 63.5 cm active length. In addition to the blanket and driver assemblies, the ANL-W inventory also includes a variety of EBR-II control and experimental assemblies. Table I summarizes the types of fuels that have been loaded into canisters and are currently in storage at ANL-West.

Table I shows that not all of the fuel in RSWF cans is suitable for a simple safeguards confirmatory method, like the SMS method, because prior to 1992 canister loading was not well-characterized in terms of specific subassemblies and storage geometry. The well-characterized canisters loaded after 1992 account for approximately 15.9 MTHM (Driver: 0.91 MTHM, Blanket: 15.0 MTHM) and are suitable for verification using confirmatory measurements.

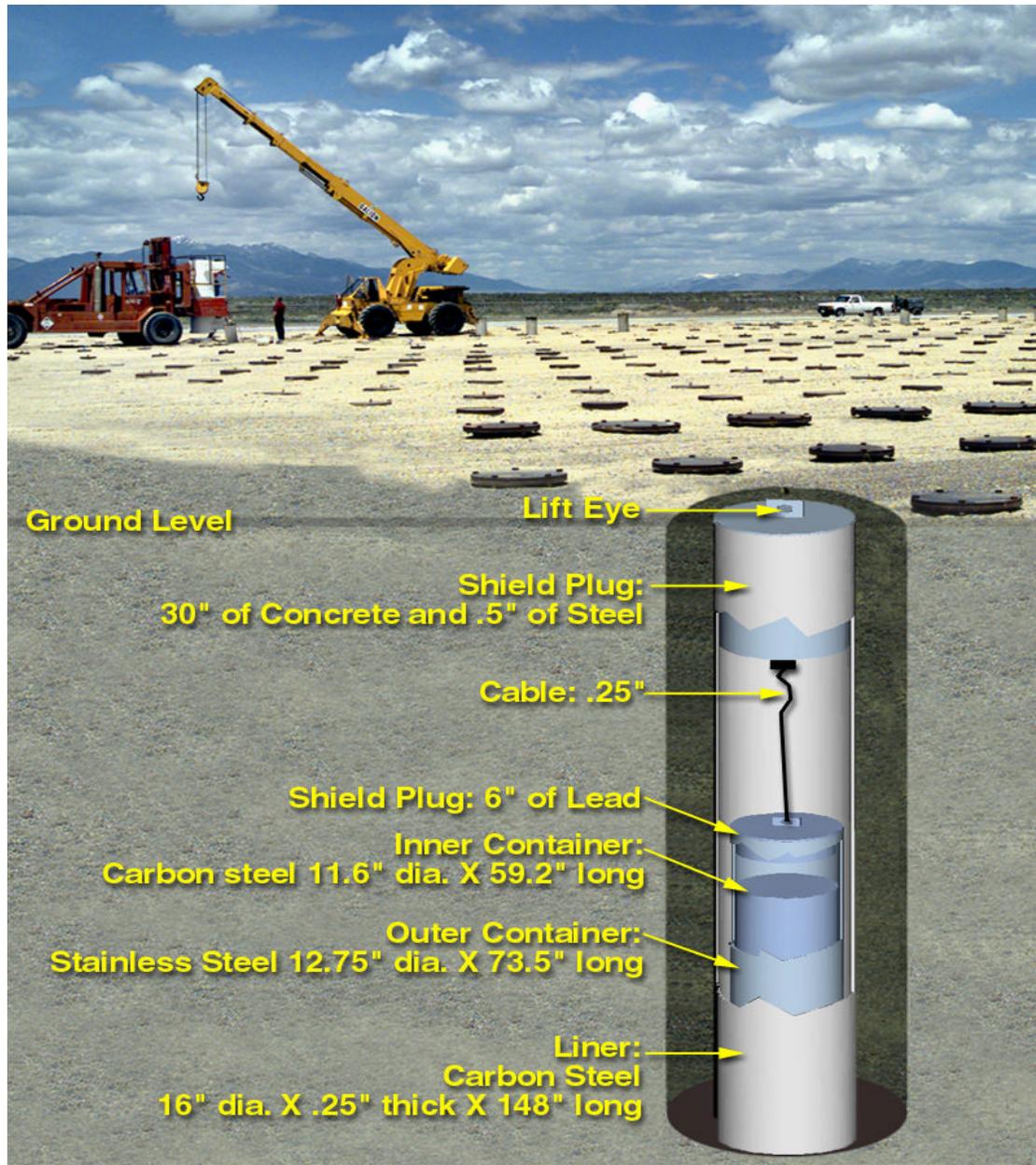


Fig. 1. Cutaway view of a spent fuel storage location at the ANL-West RSWF.

Making the confirmatory measurements more challenging will be canister loading practices. First, the equivalent of two to four EBR-II assemblies are typically loaded in each canister, but this may be as a collection of dissimilar fuel elements packaged in a geometry less well-defined than an intact fuel subassembly. Second, other metal waste such as activated reactor hardware is commingled with fuel in many storage locations. Accurate canister loading information will be crucial to choice of the radiation signature detection method and to the reliability of the SMS verification approach.

Table I. RSWF inventory fuel attributes and quantities as of June 1, 2000.

Fuel Compositions	uranium-zirconium alloy, uranium-fissium alloy, uranium-plutonium-zirconium alloy, uranium oxide, uranium carbide, uranium nitride
Total Driver Assemblies	0.98 MTHM
SMS Driver Assembly Candidates	0.91 MTHM
Total Blanket Assemblies	20.1 MTHM
SMS Blanket Assembly Candidates	15.0 MTHM
Uranium Enrichment	0.2% - 93%
Plutonium Content	0% - 20%
Density	8.7 g/cm ³ - 18.9 g/cm ³

SMS FISSILE MASS VERIFICATION APPROACH

For purposes of fissile mass verification, spent fuel can be coarsely categorized according to the availability of process information (fuel fabrication data, irradiation history, etc.) since the difficulty of the verification task depends on this information. For fuels with adequate process information, an estimate of fissile mass inventory in each fuel assembly can be calculated using reactor physics, isotope depletion and other related computer codes. This estimate must then be confirmed in an acceptable manner. For fuels without adequate process knowledge, reliable fissile estimates cannot be computed and a direct measurement of the fissile isotopes is required. The ANL-West fuel materials to be measured with the SMS verification method fall into the first category: fuels with adequate a priori process information.

Two fissile mass verification options exist for fuels with sufficient process information. The first is the use of validated computational predictions based on this process information. Code validation often relies on wet chemistry analysis of spent fuel samples, a time-consuming and expensive process even when performed at specialized facilities such as those available at ANL-West.

The potential cost of the code validation process provides an incentive to confirm the necessary data using the second option: confirmatory nondestructive analysis measurements based on process records. That is, computational predictions of certain spent fuel parameters of interest are made using the process records. Simple nondestructive assay measurements are then used to detect the radiation signatures that will confirm those predictions. By using both physics analyses and direct radiological measurements, the SMS verification method uses all available information to confirm fissile material inventory.

This measurement-assisted calculation approach is the option of choice for ANL-West and will draw on Argonne's extensive experience in the computational prediction of the isotopic inventory of spent fuel, a natural outgrowth of its history in reactor development. Our SMS approach consists of four primary components discussed below (3).

Isotopic Inventory Prediction

The first step in the SMS verification approach is the computational prediction of the isotopic mass inventory for the spent fuel item in question. The calculations of isotopic inventory rely on process knowledge about the fuel, pertinent nuclear data, and specific information about the reactor in which it operated. The critical parameters include:

Fabricated Fuel

- Fabrication data, including detailed initial isotopic inventory
- Physical form

Fundamental Nuclear Data

- Neutron reaction cross section library for all isotopes of interest
- Radioactive branching ratios and transmutation functions

Reactor

- Core geometry (e.g., support structure, coolant composition, poisons, etc.)
- Spatial Flux Distribution
- Detailed operating history

With this information, a detailed spatial, multi-group reactor flux calculation is possible, which can then be used to generate appropriate one-group cross sections at specific reactor locations. These one-group cross sections are created by collapsing the energy-dependent cross sections into a single energy group.

The one-group spatial cross section and flux information are then used as input to an isotope depletion code such as ORIGEN to calculate the isotopic mass inventory for the fuel assembly at any specific time during or after irradiation (4). The ORIGEN code incorporates specific operating history data in order to predict the mass of fission products, actinides, activation products, fissile isotopes, and other nuclides of interest as a function of position for fuel elements and subassemblies. An extensive history of similar calculations at Argonne has shown that the ORIGEN estimates of nuclide inventory for EBR-II binary and ternary driver elements typically produce a one-sigma uncertainty of 6% for total burn-up and 1.06% for heavy metal mass (5).

Radiation Signature Prediction

The computational estimate of the spent fuel isotopic inventory is used along with spent fuel hardware composition and spent fuel geometry information to build a Monte Carlo N-particle model of the spent fuel in the Shielded Measurement System. This model is then used to run a Monte Carlo simulation of the specific radiation signature measurement to be performed (e.g., gross gamma, gross neutron, gamma spectroscopy, coincident neutron).

Radiation Signature Measurements

The spent fuel item is placed in the SMS and the measurement procedure is completed. Specific types of measurements supported by the SMS are described later in "Description and Status of Shielded Measurement System."

Analysis

Uncertainties in both the radiation signature prediction method and the radiation signature measurement must be considered in the analysis of the fissile mass verification results. Since both of these uncertainties can be determined through appropriate analysis techniques, an acceptable safeguards criteria will be established for the calculated and experimental ratio (C/E). If the measurements meet the required C/E statistical criteria, yet to be defined by DOE, the spent fuel fissile mass inventory is considered qualified.

EXAMPLE OF VERIFICATION APPROACH

Recent Argonne and Los Alamos National Laboratory collaborative work at the BN-350 breeder reactor in Kazakhstan provides a good example for the use of confirmatory nondestructive analysis measurements to verify calculated estimates of spent fuel fissile mass. The fact that the BN-350 fuel is a fast breeder reactor fuel, similar in many ways to the ANL-West EBR-II inventory, extends the usefulness of this example.

The BN-350 project is driven by U.S. proliferation concerns that stem from the large inventory of plutonium-bearing fuel currently in storage at the reactor. In particular, the thick depleted uranium blanket region has relatively low burnup levels yet significant quantities of plutonium produced via neutron capture in U-238. The BN-350 driver assemblies also require fissile mass verification due to their inventory of highly enriched (17%-26%) uranium and plutonium. The verification of the plutonium content in the BN-350 blanket and driver assemblies was the primary goal of the project.

The confirmatory radiation signature in the BN-350 plutonium verification was singles and coincident detection of the spontaneous fission neutron emission from Pu-238 and Pu-240. The use of this signature is made possible by the fact that few higher actinides are generated by the hard energy spectrum in a fast reactor and consequently the spontaneous fission of the Cm-242 and Cm-244 isotopes is relatively small. In addition, the neutron emission rate from (α ,n) reactions is small compared to the spontaneous fission neutron rate from Pu-238 and Pu-240. The measurement of the Pu-238 and Pu-240 mass in the BN-350 fuel can be used along with the Pu-238/Pu-239 and Pu-240/Pu-239 ratios (predicted computationally with knowledge of irradiation history and local neutron energy spectrum) to infer, and consequently confirm, the predicted fissile Pu-239 concentration.

At BN-350, over 2000 assemblies were nondestructively assayed by Los Alamos National Laboratory using an underwater coincidence counter to verify the plutonium content predicted by Argonne calculations and declared by the facility. The root mean square error between the

measured and predicted plutonium inventories was 8% with none of the assemblies exhibiting an error greater than the IAEA verification criteria of 50% (6).

DESCRIPTION AND STATUS OF SHIELDED MEASUREMENT SYSTEM

The SMS will provide the remote handling and experimental platform for the confirmatory measurements required by the verification method. The fundamental objective of the SMS is to provide a portable, well-shielded space in which the radiation signatures from a highly radioactive item such as a spent fuel assembly can be measured using a variety of nondestructive tools. The cask-based SMS is designed to function primarily as an extension to the subsurface storage locations at RSWF but a flexible design ensures that interface with an ANL-West hot cell facility is also possible. Because the SMS attaches to these existing facilities, rather than being integral to them, impacts on other operations in the facility and radiation background are minimized.

As shown in Figures 2 and 3, the SMS consists of the following major components:

- Split-base shield ring for system leveling and interface to RSWF underground silos;
- Instrumented shield ring capable of housing a variety of radiation detectors including ion chambers, multiple neutron detectors with moderating materials, and gamma spectroscopy detectors;
- Scan shield with a 7.5-inch thick cylindrical lead wall to provide the interrogation chamber for the spent fuel item;
- Hoist system to withdraw the spent fuel item from a storage location, through the instrumented shield ring and into the scan shield. The hoist system accommodates both axial and azimuthal item scanning.

The SMS, when in operation at RSWF, will be used to interrogate spent fuel items in the waste cans described earlier (approximately 32.4 cm in diameter and 187 cm long). Once a waste can is drawn into the SMS, an operator will use the hoist system to position the waste in the axial measurement position, which is verified with an axial position transducer and computer position display. A rotational stage of the hoist system is connected to the top of the waste can by a vacuum torque coupling and allows the azimuthal scanning of the waste can for particular measurements (e.g., azimuthally averaged gamma-ray or neutron responses as a function of axial position).

SMS PHASE I

Phase I of the SMS project is currently underway and includes the design, fabrication and testing of not only the SMS, but the SMS verification method. As of this writing, system hardware fabrication is nearly complete and the system is being assembled for testing of the spent fuel handling components. Once an operational checkout of the system has been completed, the Phase I radiation detection modules will be installed in the Instrumented Shield Ring. This will include gross gamma detection via four ion chambers and eight He-3 neutron detectors

surrounded by moderating polyethylene matrices. Although complete isotope inventories will be computed for the prediction of radiation signatures, the primary isotopes of interest for EBR-II

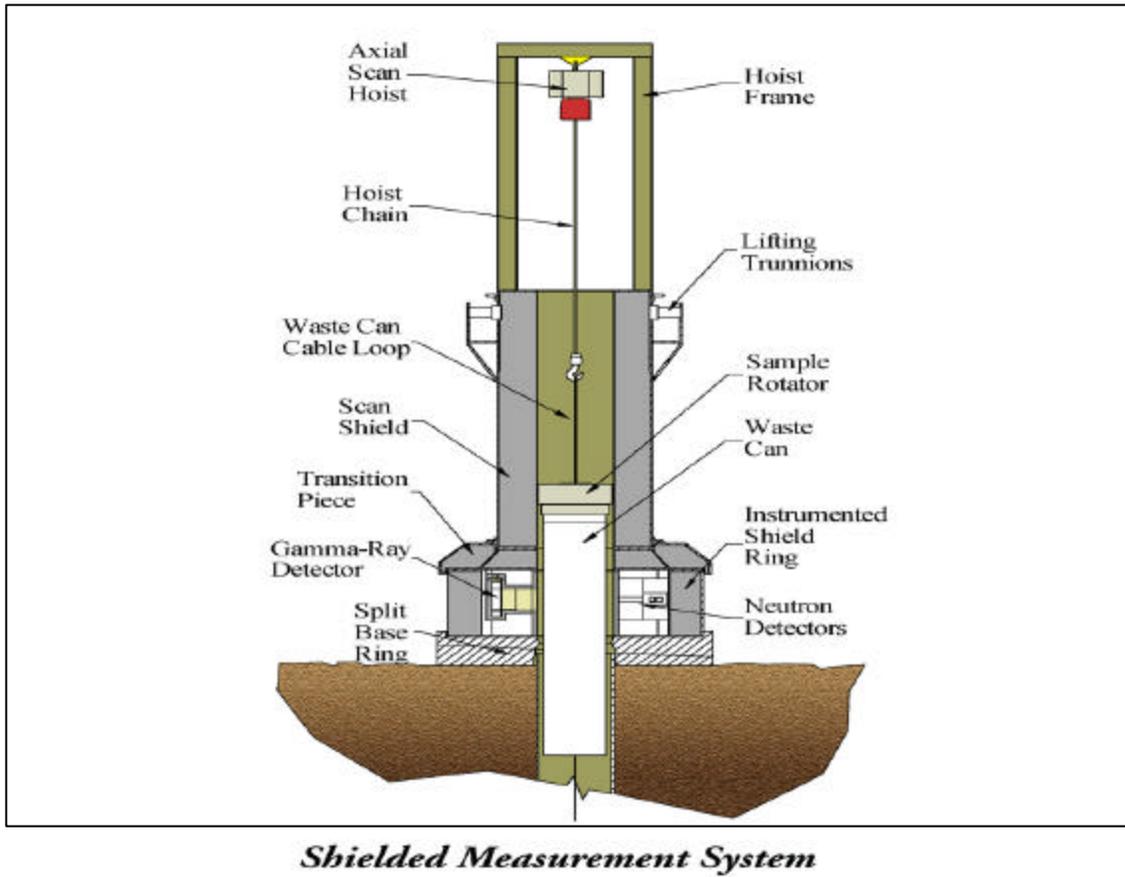


Fig. 2. Cross-sectional schematic of the SMS in use above an underground spent fuel storage silo at RSWF.

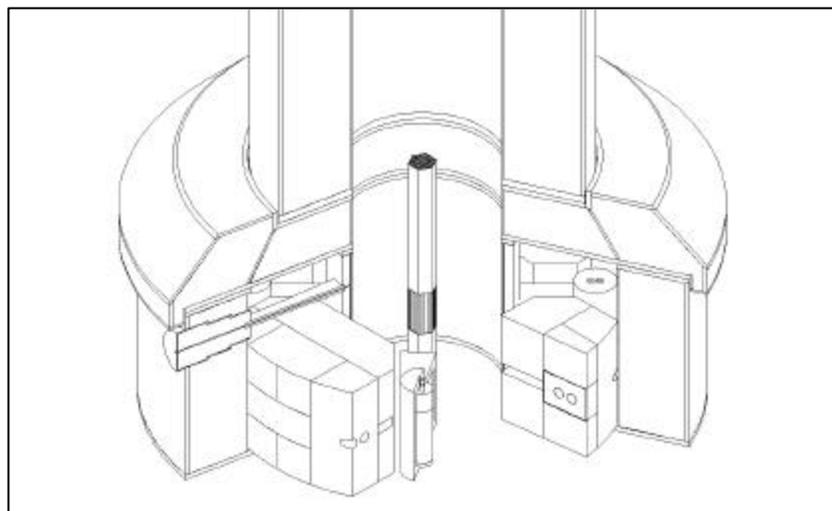


Fig. 3. Cutaway view of the SMS Instrumented Shield Ring depicting the measurement item, neutron and gamma-ray detector arrays as well as collimator penetrations for gamma-ray spectroscopy systems (waste can that contains the spent fuel is not shown).

fast reactor fuel are Cs-137 and Co-60 for the gross gamma signal and Pu-238 and Pu-240 for the gross neutron signal.

In order to test the suite of detection instruments and the various steps in the verification method, a series of simple measurements using calibrated gamma and neutron point sources will be completed. Any biases in the detection methods for the system can be determined in these simple measurements via the C/E ratio and either corrected or incorporated into the system model for radiation signature prediction. Finally, the SMS verification method will be used to provide confirmation of fissile mass for a series of well-characterized spent fuel reference items in the ANL-West inventory.

SMS PHASE II

Phase II of the SMS project will include expanded detection capabilities that provide more isotope-specific information. Additional neutron detector modules will be installed in the Instrumented Shield Ring in order to provide adequate counting efficiency for coincident neutron measurements. A coincident neutron capability will allow more accurate quantification of the Pu-238 and Pu-240 signatures by reducing the impact of singles neutrons produced in (α ,n) reactions. In addition, high-resolution gamma spectroscopy instrumentation will be installed so that isotope-specific signatures can be used to confirm burnup (e.g., the Cs-134/Cs-137 ratio) and cooling time (e.g., the Pr-144/Cs-137 ratio) as further confirmatory evidence of the fissile mass inventory declarations. Similar to Phase I, testing and calibration of the Phase II system will be completed using point sources and well-characterized spent fuel reference items.

EXTENSION OF SMS VERIFICATION METHOD TO THE DOE SPENT NUCLEAR FUEL INVENTORY

The SMS verification method is being developed at ANL-West and will be tested using EBR-II fuel, but it is intended to be applicable in safeguards measurements and in confirming repository data requirements such as burnup, cooling time and gamma/neutron radiation levels for other DOE fuels awaiting placement in a repository. By 2035, the Department of Energy will have over 200 different fuel types and 2470 MTHM of spent nuclear fuel stored at various sites around the United States (7). The question is, what portion of this DOE spent fuel inventory could benefit, in terms of either safeguards or repository data requirements, from the SMS data verification approach?

In order to answer this question, the database of the DOE National Spent Nuclear Fuel Program was reviewed to categorize candidates as strong, weak or unsuitable according to the availability of process knowledge (8,9). Table II provides a summary of the fuel mass and SMS method candidate ratings for various fuel types and groups (masses are those expected by the year 2035).

Table II. DOE spent fuel inventory categorization according to potential for verification using the SMS method.

Fuel Type or Group	SMS Method Candidate	Fuel Mass (MTHM)
EBR-II	Strong	15.9
Ft. St. Vraine	Strong	23.4
Shippingport	Strong	57.4
High Flux Isotope Reactor	Strong	3.5
Advanced Test Reactor	Strong	3.4
Fast Flux Test Facility, Fermi, Omega-West	Strong	49.1
Future Discharges	Strong	13.6
N-Reactor	Weak	2100
Transient Reactor Test Facility, VEPCO, High Flux Beam Reactor, National Institute of Standards and Technology, others	Weak	39.5
U.S. University	Weak	3.4
Foreign	Weak	0.8
Old (discharged before 1980)	Weak	67.1
Damaged and Scrap	Unsuitable	93.4
Total		2470

Based on this preliminary review of the DOE National Spent Fuel Program database, the SMS verification method is applicable for most of the predicted DOE inventory fuel types. The strong candidates include fuels from EBR-II, Fort St. Vraine, Shippingport, High Flux Isotope Reactor, Advanced Test Reactor, Fast Flux Test Facility and Fermi. In fact for some of these fuels, the National Spent Nuclear Fuel Program has already performed modeling and isotopic inventory calculations for other purposes (e.g., Total System Performance Assessment, bounding calculations and criticality safety considerations). Future discharges are also considered strong candidates because it is likely that these fuels will have adequate process knowledge due to today's reactor operation and record-keeping requirements. Figure 4 is a graphical representation of the relative mass of each fuel type in the strong candidate category.

Fuels from N-reactor, the Transient Reactor Test Facility, the High Flux Beam Reactor, U.S. universities and foreign fuels are examples of weak candidates for the SMS verification method. For some of these, uncertainty in the operating records of facilities not regulated by the DOE means that adequate process knowledge may not be available. However, it is recognized that suitable records may exist on a case-by-case basis and that additional analysis is required to determine which of these fuel types may have sufficient process knowledge for verification using the SMS method.

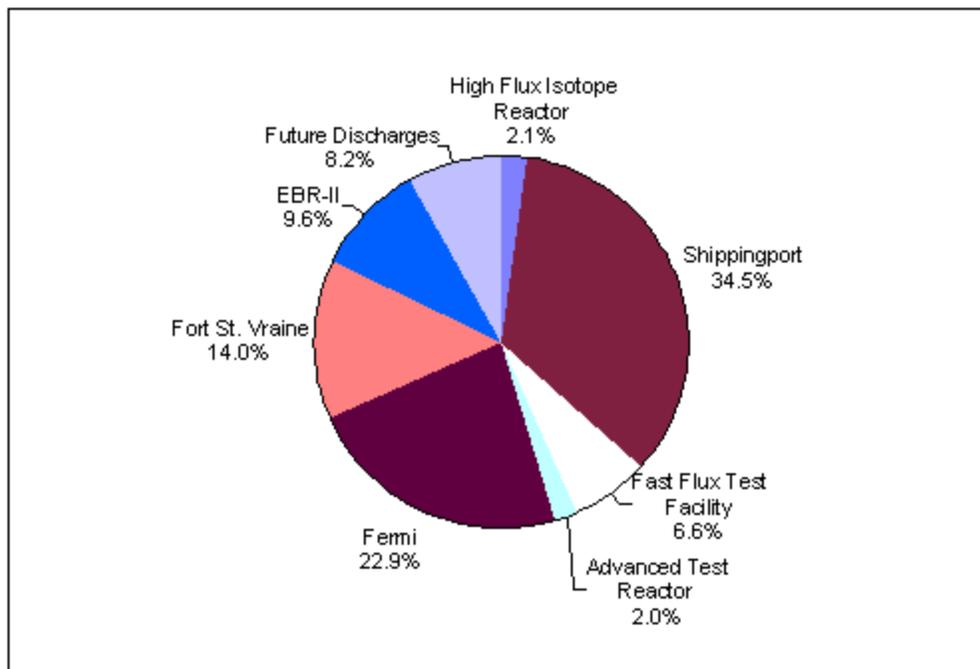


Fig. 4. Relative fraction (by mass) of each fuel type in the strong candidate category.

Finally, a small portion of the fuels listed in the National Spent Fuel Program database are unsuitable candidates for the SMS verification method. It is unlikely that the required process knowledge pedigrees are available for damaged fuels and scrap. For these materials, a verification approach that directly measures a fissile material signature is more suitable.

It is important to note that the spent fuel inventory analysis described above does not consider radiation signature measurement uncertainty in the categorization of each fuel type. The existence of adequate process knowledge only makes the fuel a candidate, it does not guarantee that the SMS verification method will provide adequate confirmatory information for that fuel. As an example, the coincident neutron signal used in the BN-350 application to verify plutonium content may not be useful for light-water reactor fuels with significant quantities of the Cm-242 and Cm-244. The uncertainties associated with isotopic inventory prediction, radiation signature prediction and radiation signature measurement must be carefully analyzed and considered for each specific fuel type.

CONCLUSION

The SMS spent fuel verification method is based on reasonable process knowledge, computational prediction of isotopic inventory and confirmatory measurements to verify declared values of important fuel parameters such as fissile mass and burnup. The SMS itself is a versatile experimental platform capable of mating to both hot cell and underground dry storage facilities. Although initial testing of the SMS verification method will take place at the ANL-W

RSWF using EBR-II fuels, it is expected that the SMS verification method can be used for many different fuel types in the DOE inventory.

ACKNOWLEDGEMENTS

The Shielded Measurement System project is funded through the Department of Energy Office of Safeguards and Security Technology Development Program.

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