SANDIA REPORT
SAND2002-1027
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Printed April 2002

The Need for Confirmatory Experiments on the Radioactive Source Term from Potential Sabotage of Spent Nuclear Fuel Casks

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Abstract

A technical review is presented of experiment activities and state of knowledge on air-borne, radiation source terms resulting from explosive sabotage attacks on spent reactor fuel subassemblies in shielded casks. Current assumptions about the behavior of irradiated fuel are largely based on a limited number of experimental results involving unirradiated, depleted uranium dioxide "surrogate" fuel. The behavior of irradiated nuclear fuel subjected to explosive conditions could be different from the behavior of the surrogate fuel, depending on the assumptions made by the evaluator. Available data indicate that these potential differences could result in errors, and possible orders-of-magnitude overestimates of aerosol dispersion and potential health effects from sabotage attacks. Furthermore, it is suggested that the current assumptions used in arriving at existing regulations for the transportation and storage of spent fuel in the U.S. are overly conservative. This, in turn, has led to potentially higher-than-needed operating expenses for those activities. A confirmatory experimental program is needed to develop a realistic correlation between source terms of irradiated fuel and unirradiated fuel. The motivations for performing the confirmatory experimental program are also presented.

ACKNOWLEDGMENTS

We have reviewed our position on the need for confirmatory experiments involving small samples of irradiated and nonirradiated fuel with other credible scientists and engineers who have also been active in the study of potential aerosol source terms from sabotage of spent fuel storage and transportation casks. We especially thank Dr. Robert Luna of Sandia National Laboratories and Dr. Gunter Pretzsch of the German Gesellschaft fur Anlagen-und Reactorsicherheit for their consultation and endorsement of this position. We also gratefully acknowledge the support of the U.S. Department of Energy and the U.S. Nuclear Regulatory Commission in the SNL/ITRI experiments referred to in this paper.

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The Need for Confirmatory Experiments on the Radioactive Source Term from Potential Sabotage of Spent Nuclear Fuel Casks

INTRODUCTION

Emergency planning for management of terrorist events involving radioactive materials requires a scientific basis for estimating potential aerosol dispersion. Current assumptions for release of radioactive materials from sabotage of spent nuclear fuel casks rely on an assumed, and possibly overly conservative correlation between the behaviors of unirradiated depleted uranium and irradiated nuclear fuel. Concerns have been raised that the current state of knowledge does not provide an adequate basis for cost-effective decision-making and regulatory requirements to protect public health (Newton et. al., 1996). This report reviews the studies and experiments that have been conducted to date and discusses their limitations. A confirmatory program of experiments with small samples of irradiated and unirradiated nuclear fuel should be undertaken to develop realistic source term estimates for use in radiation protection guidelines and regulations.

HISTORICAL BACKGROUND

Table 1 summarizes the studies and experiments that have been conducted over the past 30 years to estimate the potential radiological consequences of accidents and sabotage involving spent nuclear fuel and their transportation and/or storage casks.

Initial Source Term Estimates based on Engineering Judgment

During the early 1970s, concerns arose about the possible risks to the public from a potential terrorist attach on a civilian nuclear reactor spent fuel shipping cask. Several engineering analyses were published on the subject, including one by DuCharme *et al.* (1978) of Sandia National Laboratories, which became known as the "Urban Study". The Urban Study was based on engineering judgment and contained scant experimental data. The scenario was that the malevolent act would occur in downtown Manhattan on a Thursday afternoon. The postulated consequences of the Urban Study were that 1% of the cask contents would be dispersed as a respirable aerosol. These consequences were considered unacceptable and the U.S. Nuclear Regulatory Commission promulgated very restrictive interim regulations (draft 10CFR73.37) to protect the public from such a malevolent act.

Table 1
Brief History of Studies and Experiments to Estimate
the Potential Radiological Releases from Spent Fuel Accidents and Sabotage

Study or Experiment	Approach	Conclusion
First Urban Study (DuCharme, 1978)	Applied engineering judgment and limited engineering data	Estimated 1% release fraction for respirable aerosol for a transportation cask containing one PWR subassembly
Second Urban (Finley, 1980)	Refined methods and assumptions of First Urban Study	Reduced the estimated release fraction for respirable aerosol from a value of 1% to a value of 0.07%.
Waste Forms Response Investigation at Idaho (Alvarez, 1982)	Conducted miniature scale explosive attacks on unirradiated and irradiated fuel pellets to determine an irradiated-to-unirradiated "spent fuel ratio" (SFR).	Estimated that aerosol releases from irradiated fuel are 5.6 times greater than from unirradiated fuel (based on wet sieving results) but only 0.53 (based on a bulk aerosol filter results)
Cask Sabotage Experiment at Sandia (Sandoval, 1983)	Conducted subscale and full scale explosive attacks on fuel shipping casks with unirradiated fuel	Determined that 0.05% of the disrupted unirradiated fuel was released as a respirable aerosol. Multiplied the respirable aerosol release for unirradiated fuel by an SFR of 5.6 to estimate a release fraction of 0.0034% for respirable aerosol from one PWR subassembly of irradiated fuel.
Cask Sabotage Source Term Studies at Battelle (Schmidt, 1981, 1982, and Miller, 1986)	Conducted miniature scale explosive attacks on unirradiated and irradiated fuel pellets to determine an irradiated-to-unirradiated "spent fuel ratio" (SFR).	Estimated SFRs of 0.42 to 0.71, which were in agreement with the 0.53 SFR from the Idaho bulk aerosol filter result.
Dry Storage Cask Sabotage Study at Sandia (Philbin, 1988)	Conducted a full scale explosive attack on multiple fuel assemblies sections of unirradiated fuel in a mockup dry cask storage configuration	Measured the disrupted and ejected mass and estimated the respirable aerosol release from the damaged cask using the aerosol release fraction of Sandoval and a SFR of 1.0. Estimated release of respirable aerosol was 2.4 – 6.5 g, 0.05 % of swept mass.
Transportation Cask Sabotage Studies in Germany (Pretzsch, 1994)	Conducted a full-scale explosive attack on multiple fuel assembly sections of unirradiated fuel in a mockup nuclear fuel shipping cask configuration.	Measured release of respirable aerosol was 1 g each for 2 tests conducted at ambient pressure and 0.33 g for a third test conducted at cask pressure of 80% of atmospheric. A SFR of 1.0 was used to convert the data from unirradiated fuel to estimated releases for irradiated fuel.
SFR Studies (Luna, 2001 and Lange, 2001)	Reviewed experimental data to gain a better understanding of the SFR.	Noted that the uncertainty in the SFR still ranges from 0.7 to 12. Presented concepts for small-scale experiments to narrow the uncertainty in the SFR parameter.

A "Second Urban Study" was performed by Finley *et al.* in 1980 and resulted in a 14-fold reduction in the estimated release fraction for respirable aerosols (from 1% in the original Urban Study, to only 0.07%). Despite the improved analyses in the Second Urban Study, essentially no changes were made in the restrictive interim regulations on spent fuel shipments.

In the meantime engineering test programs were being established under the aegis of the U.S. Department of Energy and the U.S. Nuclear Regulatory Commission to develop an experimentally based risk estimate for the consequences of a potential sabotage attack on a spent fuel shipping cask.

Source Term Estimates based on Experiments at Sandia National Laboratories and the Idaho National Engineering Laboratory

The U.S. Department of Energy tasked Sandia National Laboratories with conducting an experimental program to develop the improved risk estimate for dispersion of radioactive aerosols from sabotage attacks on spent fuel storage and shipping casks. The DOE's Inhalation Toxicology Research Institute provided aerosol science support for the program. The experimental program involved a phased approach which included quarter scale attacks with conical shaped charges on simulated spent fuel casks. The simulated spent fuel assemblies were short sections of zircaloy-clad fuel pins containing depleted uranium dioxide (D-UO_2) pellets. A total of 25 short fuel pins (~13.5 kg D-UO_2) were placed in a quarter scale cask and subjected to shaped charge attack. The tests were conducted within a containment vessel to enable a total mass balance for the D-UO_2 involved in each test. After a total of 7 subscale tests, a confirmatory full-scale attack was conducted with the full-scale reference weapon, again in containment.

At the same time that the subscale and full scale tests were taking place at Sandia, a second (and extremely essential) part of the program was being conducted at the Idaho National Engineering Laboratory. That part of the test program involved the direct experimental comparison of miniature scale attacks on single fuel pellets of both D-UO_2 and irradiated spent nuclear fuel, in standard zircaloy cladding to determine a spent fuel ratio (SFR). SFR is defined as the ratio between the spent fuel respirable aerosol mass and the surrogate fuel respirable aerosol mass when all other experiment parameters and conditions are essentially constant. The explosive devices were small Type Y-1005 conical shaped charges, which are frequently used to fracture rock in oil field production enhancements. The irradiated spent fuel pellets were from the H.B. Robinson Unit 2 civilian power reactor and had been aged about 6.5 years after removal from the reactor. This direct experimental comparison of D-UO_2 pellets and irradiated spent fuel pellets was to provide the critical damage ratio for relative aerosolization rates that would allow the results of the Sandia tests with D-UO_2 to be extrapolated to realistic situations involving actual irradiated nuclear fuel.

Unfortunately, after the miniature explosive tests were conducted on the D-UO_2 and spent fuel pellets in Idaho, a mishap occurred during preparation of some of the cascade impactor samples for analysis; a tray containing many of the critical samples was dropped and the identity of the individual samples was lost. This precluded the correlation of the relative release rates of aerosol from D-UO_2 and irradiated spent fuel pellets based on the actual cascade impactor aerosol measurements in the Idaho experiments.

Because neither time nor funds were available to repeat the expensive test series, personnel at the Idaho National Engineering Laboratory re-examined the wet sieving results for the bulk particle debris that remained after the explosive attacks on the D-UO_2 and irradiated fuel pellets. This was an indirect attempt to obtain the needed correlation between the behaviors of D-UO_2 and irradiated spent fuel.

Use of the wet sieving data from the Idaho study provided a correlation value for Spent Fuel–to–DUO_2 aerosol release fraction of 5.6:1. This result was substantially higher than the ratio of 0.53:1 obtained from analyses of bulk aerosol filter samples taken during the tests. Note that the wet sieving data were considered of limited reliability because they were not based on aerosol samples and because such wet sieving results have not been accurate in other studies. The wet sieving method does not account for the fact that particles can agglomerate to form larger particles in settling, thus distorting the size distribution. The aerosol portion (with aerodynamic diameters < 10 micrometers, μm) cannot be examined separately by this method nor can the results be weighted versus aerodynamic diameter, as is possible with the resolved cascade impactor data. Nevertheless, in the absence of reliable aerosol data, the decision was made to be extremely conservative in selecting the correlation coefficient for extrapolating the Sandia results (with D-UO_2) to potential scenarios involving irradiated spent fuel. Therefore, the higher value of 5.6:1 (from the wet sieve data) was selected and used in officially revising (for regulatory purposes) the potential health effects expected from this type of sabotage. Nevertheless, even with this conservative value for spent fuel–to–DUO_2 ratio (5.6:1), the small respirable aerosol mass release, 2.94 g, obtained from the experiments led to markedly lower respirable release fractions and health effects than had previously been assumed in either of the earlier Urban Studies.

These new results lead to some relaxation of regulatory requirements (10 CFR 73.37), including the elimination or altering of certain physical protection requirements and easing of restrictions for driving spent fuel casks through densely populated urban areas.

Source Term Estimates based on Experiments at Battelle Columbus Laboratory

Meanwhile, the U.S. Nuclear Regulatory Commission asked Battelle Columbus Laboratory to investigate the respirable aerosol release rate from a hypothetical terrorist attack on a civilian spent fuel shipping cask. Their approach was to use a simulated, subscale cask containing a five-pin array of short sections of spent fuel; attack the simulated cask and contents with an explosive device; and characterize the resultant aerosol release (Schmidt *et al.*, 1981). Because of the high radiation field from the irradiated fuel pellets, the study was conducted in a lead-lined glove box. Unfortunately, rather than using conventional filtration and cascade impaction techniques to characterize the aerosol source term, the Battelle approach used electron microscopy of witness plate samples to obtain a size distribution. Such an approach is not technically justified because it is not possible to distinguish individual particles that piled up sequentially on the collection plate from aggregate aerosols that agglomerated during the explosive process. For particles larger than 1 µmdiameter, this generally leads to an underestimation of the particle size distribution in the respirable size range. For particles with diameters on the order of 0.3 µm, this may lead to an over estimate of the material in the size range associated with deposition in the pulmonary region of the lung. In addition, a simple microscopic examination of witness plates cannot distinguish uranium particles from particles of zircaloy, explosives residue, or any other materials in the test system. This can result in further improper assumptions about the true particle size distribution of the fuel material. In comparison, collection of the airborne debris by cascade impaction results in an appropriate measurement of the actual distribution of radioactivity as a function of aerodynamic particle size.

Because of these and other problems that arose in the experimental program, the first draft assessment from the Battelle-Columbus studies indicated a very high respirable aerosol release fraction. The U.S. Nuclear Regulatory Commission called personnel from Sandia National Laboratories, the Inhalation Toxicology Research Institute, Battelle-Columbus, and the U.S. Department of Transportation to a meeting in Washington, DC, to discuss the results and their interpretation. Based on recommendations from the Sandia/ITRI team, the U.S. Nuclear Regulatory Commission provided additional funding to Battelle-Columbus to repeat some of their studies using the aerosol sampling devices and approaches developed at ITRI. Schmidt *et al.* (1982) reported the results of those tests and Sandoval *et al.* presented a thorough statistical analysis of the results relevant to the correlation ratio in the 1983 report.

Using scaling theories and statistical analysis, Sandoval *et al.* compared the Battelle spent fuel filter data to their D-UO_2 cascade impactor data and obtained release fraction ratios for spent fuel-to-D-UO_2 in the range of 0.42 to 0.71. The lower value (0.42) is the average value when two outlier data points (which failed statistical tests for a Gaussian distribution) are ignored. The lower value also agrees better with results from the INEL bulk aerosol filter results. Thus, there is evidence that the irradiated-to-unirradiated aerosol release ratio is less than 1. Quoting from the Sandoval report, “There are potential errors in all of these correlation ratios. While estimates for the Battelle results are based on actual spent fuel aerosolized mass, <u>there were no comparable results</u> for D-UO_2 in the same test set-up. Given this uncertainty the most definitive statement that can be made is that the value is likely to be on the order of unity.” However, for conservatism in the health risk assessment, the highest experimental value for the correlation, 5.6:1, (based on the INEL wet sieve data), was the value chosen for regulatory purposes. Miller *et al.* (1986) at Battelle performed subsequent tests with nonirradiated surrogates of high-temperature gas-cooled reactor (HTGR) fuel, nonpower reactor (research reactor) fuel, and vitrified high-level waste, *not* with power reactor fuel. Miller *et al.* (1986) concluded that the sabotage of shipments of these fuels should have no greater consequences than those predicted for shipments of PWR spent fuel. This last test campaign did not involve any tests with irradiated materials, however, so no new correlation ratios were developed for irradiated-to-nonirradiated samples.

Thus, in spite of all the disparity and ultimate conservatism used with regard to the spent fuel to D-UO_2 correlation, the experiment programs demonstrated substantially lower consequences than the earlier Urban Study reports. The reduced health consequences for a sabotage attack on spent fuel allowed the U.S. Nuclear Regulatory Commission to revise its requirements for the physical protection of irradiated nuclear fuel in transit and drop several of the more restrictive requirements on handling of civilian spent fuel. This provided some relief to the civilian nuclear reactor industry in the United States.

Subsequent Experiments at Sandia National Laboratories on the Extent of Fuel Damage during Sabotage Attack on a Simulated Dry Storage Cask

In 1986, the Nuclear Regulatory Commission/Nuclear Material Safety and Safeguards Division asked Sandia/ITRI to measure the release of fuel material from a simulated, dry storage cask subjected to a high-energy, directed-explosive device. See Philbin *et al.* (1988). The purpose of this experiment was to estimate the source term and potential health effects from a sabotage attack upon an on-site spent fuel storage cask. (Note: the on-site casks are much larger than typical transportation casks and contain considerably more spent fuel.) The spent fuel was simulated by unirradiated mechanical mockups of 15-inch long subassemblies of boiling water reactor rods containing D-UO_2. The D-UO_2 was the only practical target for a large scale, open-air test. In the experiment, instrumented basket plates separated nine mockup D-UO_2 fuel subassemblies. These were mounted in individual compartments, which were lined up behind 11.5 inches of steel representing a section of a large storage cask wall. The explosive device penetrated five of the nine subassemblies, 12.9 kg of D-UO_2 were displaced, and 2 kg were ejected from the test article (i.e., released from the cask). The previously well-documented, measured respirable release fraction (0.0005, Sandoval *et al.*, 1983) for D-UO_2 fuel (under similar conditions) was used to derive a respirable, released mass of 6.5 g. The respirable released mass is a product of D-UO_2 displaced (12.9 kg) and the release fraction (0.0005). The uncertainty in the respirable released mass was ± 9%.

In the absence of a definitive ratio for spent fuel-to-unirradiated fuel, a ratio of 1:1 was assumed for the solids release fission-product inventory. In other words, it was assumed that an equal amount (6.5 g) of 5 yr old spent fuel would become respirable particles (less than 10 μm Activity Median Aerodynamic Diameter), if spent fuel, rather than D-UO_2, had been the target in the explosive attack test. The 6.5 g of respirable spent fuel particles (i.e., UO_2 and solid fission products) plus 50% of the Kr-85 (gas) from a single, full-length spent fuel subassembly were taken as the source term for the scenario dose calculations. This source resulted in estimated whole body and lung doses of 1 and 2 rem (at 100 m downwind) and 0.1 and 0.2 rem (at 1000 m downwind). These doses would not be exceeded in 95% of the potential weather conditions. Almost the entire dose was attributable to the respirable solids release; the dose accrued from the Kr-85 gas was very small and, hence, not important.

The source term and dose results for this scenario could be renormalized if a better correlation of respirable release data existed between irradiated and unirradiated UO_2 fuel subjected to these conditions. Additional experiments are needed to develop a reliable correlation for the respirable release fractions (of UO_2 and solid as well as gaseous fission products) in explosive attack sabotage scenarios for spent fuel and unirradiated UO_2.

German Experiments

In the early 1990s, scientists at Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) conducted experiments to evaluate the release of radioactive debris, primarily the fraction of respirable particles, from an explosive sabotage attack on a large nuclear fuel shipping cask. See Pretzsch and Lange (1994a). In a separate report Pretzsch and Lange (1994b) evaluated the radiological consequences (dose commitment to individuals) of radioactivity release from the sabotage attack test. The cask and D-UO_2 subassemblies were about one third of normal length but other dimensional features were full size. Three fuel subassemblies were arranged one behind the other. A directed high-energy device penetrated the cask wall and then hit the three fuel elements as it bored its way through the material in its path. Three main experiments were performed, the last of which was configured with a slightly negative pressure (0.8 bar) inside the test cask (to simulate conditions prototypic of an interim storage facility).

The airborne debris was ejected from the test cask (through the entrance shot hole) into a sampling chamber that was connected to the test cask. The debris was forced by blowers through an opening in the side of the sampling chamber and directed into various aerosol-physical measurement devices, where it was divided into fractions (from coarse to fine). Respirable fine debris having aerodynamic equivalent diameter of < 12.5 μm was divided into five particle size groups (or fractions). The first two tests (with ambient pressure in the cask) yielded 1 g of respirable fine debris and the third test (at 0.8 bar pressure) yielded about 0.33 g. Most of the debris was released immediately after the impact of the charge and mainly from the first in the line of three elements impacted. Using deterministic calculations and allowing for a slightly higher release fraction for volatile fission products, Pretzsch and Lange calculated an effective dose from inhalation (lung) of 18 mSv (1.8 rem) at 50 m. Based on the penetration achieved and the amount of respirable debris released, one can assume that the explosive device used in the German tests had lower specifications for penetrating power and deposited less total energy in the fuel subassemblies than the explosive device used in the Sandia/ITRI tests. If so, then these results are consistent with the Sandia results. Once again, however, the "mass fractions" of the released aerosol particles measured as a function of the aerodynamic diameter were assumed to be the same (1:1) for irradiated fuel as for unirradiated fuel. Thus, in the absence of more definitive correlation data, no correction could be applied to allow for potentially different mechanical response that may occur between the irradiated (fragmented) and unirradiated (solid) fuel resulting from the explosive forces and associated shock wave. A definitive correlation experiment would enable the Germans to reexamine the consequences of spent fuel sabotage scenarios based on their D-UO_2 test results.

Inadequacy of the Current Ratio for Respirable Aerosol Release from D-UO_2 and Irradiated Spent Fuel

In the large-scale test programs that have been described above, it was practical to use unirradiated UO_2. It was assumed that the irradiated and unirradiated UO_2 behaved the same or nearly the same. However, there is insufficient evidence to make a definitive correlation between the aerosol release fractions for irradiated UO_2 and unirradiated UO_2. If a correlation could be developed, based on a small scale test with both types of fuels, the correlation could be used to reinterpret the large scale test data with a correction factor to account for any real differences between unirradiated and irradiated UO_2. The motivation for this is that the release of respirable particles from unirradiated fuel is suspected to be measurably greater than the release from irradiated fuel, which is typically fragmented from previous exposure to extreme radiation and heat stress. Specifically, the propagation of a shock wave through the highly fragmented pellets of irradiated fuel could lead to a substantially lower fraction of respirable material than might be expected from the shattering of solid pellets by a high explosive shock wave. If proven, this could lead to relaxed regulatory guidelines on the shipment, storage and handling of spent fuel.

As noted above, the commonly accepted ratio of 5.6:1 for the release fraction of respirable aerosol from irradiated spent fuel and depleted uranium is likely to be overly conservative by an order of magnitude. The limited data from bulk filter samples in the Idaho study indicate that the ratio is not greater than 0.53:1. Using "early time" uranium dioxide aerosols collected on filters from the Battelle-Columbus spent fuel tests (after careful screening to reduce variability in the data) and relying on "scaling" theories in order to make a comparison to the Sandia full scale D-UO_2 test data, Sandoval *et al.* (1983) developed a 0.42:1 ratio. When two outlier data points were included in the correlation calculations, the ratio is still only 0.71:1 (spent fuel:D-UO_2). The *same test set-up* was not used at Battelle and Sandia, however, so there is still considerable uncertainty in these correlations.

Taking into account the best information currently available, it is likely that the correlation of respirable aerosol release fraction from irradiated spent fuel and depleted uranium is on the order of 0.42-0.53, and in any case, not larger than 1:1. However, the limitations of the previous test programs have prevented the widespread acceptance of either of these lower ratios for regulatory purposes.

A recent paper by Luna et. al. (2001) focused on the importance of gaining better understanding of the spent fuel ratio (SFR). This reference reports that the SRF has a range of 0.7 to 12 between the lowest and highest estimates. Furthermore, this recent paper has presented concepts for small-scale experiments to narrow the uncertainty in the SFR parameter.

RECOMMENDATION

A confirmatory experimental program should be undertaken to determine an appropriate correlation coefficient for the relative aerosol release fraction between D-UO_2 and irradiated spent fuel. An appropriate experiment would involve a repeat of a portion of the failed tests at Idaho in which individual pellets of zircaloy-clay D-UO_2 and irradiated spent fuel were attacked by a Y-1005 type penetrator.

A joint German/U.S. team has proposed a well-instrumented set of tests with carefully scaled explosives impacting single pellets of irradiated and unirradiated fuel. An air stream moving perpendicular to the path of the explosive would sweep the debris from the explosive/fuel interaction out of the test chamber into analysis instruments. Completion of these proposed tests would provide the necessary information to fully correlate the extensive aerosol data from the previous experimental programs. The proposed tests and analysis would provide a more realistic value of the aerosol source term for use in estimating risks to human health from sabotage of spent fuel storage and shipping casks.

Motivations for the performing the confirmatory experimental program are: 1) to develop a strong technical basis and a definitive correlation for a spent fuel–to–DUO_2 release fraction, 2) to remove ultra conservatism in existing evaluations of health risk assessments for cask sabotage scenarios, if this can be justified, and 3) to reevaluate the appropriateness of regulations on the transportation and storage of spent fuel. Realistic estimates of aerosol dispersion parameters will provide a stronger scientific basis for emergency planning and preparedness for terrorist attacks. If the proposed work shows the current assumptions to be overly conservative, there could be a relaxation of existing regulations for transportation and storage of spent nuclear fuel. This would result in cost savings in the form of reduced security requirements (e.g., number of personnel, the need to obtain advance approval of shipments and routes with local authorities), fewer numbers of shipments (with an increase in the number of spent fuel subassemblies per shipment), etc. The cost savings could be on the order of several tens of millions of dollars per year and could preclude unnecessary diversions of resources from other critical areas of emergency preparedness.

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