Initiation of Depleted Uranium Oxide and Spent Fuel Testing for the Spent Fuel Sabotage Aerosol Ratio Program

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ABSTRACT

We provide a detailed overview of an ongoing, multinational test program that is developing aerosol data for some spent fuel sabotage scenarios on spent fuel transport and storage casks. Experiments are being performed to quantify the aerosolized materials plus volatilized fission products generated from actual spent fuel and surrogate material test rods, due to impact by a high energy density device, HEDD. The program participants in the U.S. plus Germany, France, and the U.K., part of the international Working Group for Sabotage Concerns of Transport and Storage Casks, WGSTSC have strongly supported and coordinated this research program. Sandia National Laboratories, SNL, has the lead role for conducting this research program; test program support is provided by both the U.S. Department of Energy and Nuclear Regulatory Commission. WGSTSC partners need this research to better understand potential radiological impacts from sabotage of nuclear material shipments and storage casks, and to support subsequent risk assessments, modeling, and preventative measures. We provide a summary of the overall, multi-phase test design and a description of all explosive containment and aerosol collection test components used. We focus on the recently initiated tests on “surrogate” spent fuel, unirradiated depleted uranium oxide, and forthcoming actual spent fuel tests. The depleted uranium oxide test rodlets were prepared by the Institut de Radioprotection et de Surete Nucleaire, in France. These surrogate test rodlets closely match the diameter of the test rodlets of actual spent fuel from the H.B. Robinson reactor (high burnup PWR fuel) and the Surry reactor (lower, medium burnup PWR fuel), generated from U.S. reactors. The characterization of the spent fuels and fabrication into short, pressurized rodlets has been performed by Argonne National Laboratory, for testing at SNL. The ratio of the aerosol and respirable particles released from HEDD-impacted spent fuel to the aerosol particles produced from surrogate depleted uranium oxide, i.e., the spent fuel ratio, SFR, will be determined under closely matched test conditions. We shall briefly summarize similar results from completed, surrogate tests that used non-radioactive, sintered cerium oxide ceramic pellets in test rods, documented separately.

INTRODUCTION

We provide a detailed summary of recent technical accomplishments, results, and near-term plans from an ongoing, multinational test program that is measuring aerosol particle size distributions for some spent fuel sabotage scenarios relevant to spent fuel transport and storage casks. Casks used for spent nuclear fuel transport are extremely resistant to releasing any significant fraction of their contents, even in very severe accident conditions. However, in some credible sabotage scenarios for puncturing a cask, it is possible that a small percentage of aerosolized particles from disrupted fuel pellet materials could be released. The postulated scenario we evaluate is that of a saboteur attack on a nuclear cask using explosive, armor-piercing weapons, i.e., high energy density devices, HEDD(s). Any particulated spent fuel that is released from the cask as airborne respirable particles has the potential to cause radiological consequences, if released to the environment in a significant quantity. Measurement of the actual amounts, nuclide content, and size distribution of the released materials from spent fuel is essential for predicting the significance of the radiological impacts. The need for accurately quantifying this information has been strongly supported by program participants in the U.S. plus Germany, France, and the U.K., as part of the international Working Group for Sabotage Concerns of Transport and Storage Casks, WGSTSC. WGSTSC partners need this research to better understand potential radiological consequences, and to support subsequent risk assessments, detailed modeling, and to develop potential preventative measures.

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Sandia National Laboratories, SNL, Transportation Risk and Packaging plus other Departments, has the lead role for performing this research program. Overall sabotage and transportation program support is provided by both the U.S. Department of Energy (Office of Civilian Radioactive Waste Management/ Office of National Transportation, and National Nuclear Security Agency/ Office of International Safeguards) and the U.S. Nuclear Regulatory Commission (Offices of Nuclear Regulatory Research, and Nuclear Security and Incidence Response). Argonne National Laboratory, Energy Technology Division, has provided the detailed characterization and fabrication work for all spent fuel test rodlets to be used in this program. German participants, the Gesellschaft für Anlagen- und Reaktorsicherheit, GRS, and the Fraunhofer Institute of Toxicology and Experimental Medicine, ITEM, are providing supporting aerosol testing expertise and data analyses. The Institut de Radioprotection et de Surete Nucleaire, IRSN, France, has provided unirradiated depleted UO_2 (surrogate, DUO_2) fuel test rodlets. The Office for Civil Nuclear Security, OCNS, in the UK, participates in a consultatory role.

There are significant benefits for the successful conduct of this program for all participants involved:
(1) The cooperation of U.S., German, French, and British organizations and governmental entities provides a significant policy benefit, considering that this project can lead to improved safety of the environment from a postulated nuclear incident. The data generated from this program may be useful for determining how to counter or mitigate consequences of a terrorist threat with explosives and spent fuel.
(2) The spent fuel ratio - aerosol source term data and information to be derived from these tests, analyses, and subsequent modeling efforts will provide enhanced interpretations and clarifications to both current and earlier data on surrogate test materials [1,2,398 and very limited actual spent fuel results [4,5].
(3) Through improved radiological and safety assessments, quantifications of respirable hazards, vulnerability assessments, and potential physical protection design modifications, an additional margin of safety to the environment may be provided from a plausible, albeit unlikely, sabotage scenario.
(4) This test program's analysis of aerosol release relevant to transportation and storage scenarios may be directly applied to the spent nuclear fuel surface and repository facility operations, thereby facilitating the important process of evaluating sabotage risk to the entire back end of the nuclear fuel cycle.

A major purpose of this document is to provide an overview of the ongoing, multi-phase test program that supports the needs of the international WGSTSC. We provide summary descriptions of the overall test design, a description of all major test components, a brief review of test aerosol results and observations from surrogate material Phase 2 tests using sintered cerium oxide ceramic pellets. We present significant detail and test apparatus descriptions for the recently initiated Phase 3 tests which will include unirradiated, depleted uranium oxide and upcoming, Phase 4, actual spent fuel aerosol-explosive tests. We previously summarized the initiation of this test program [6] and documented the overall test design plus most of the earlier surrogate material aerosol test results [1].

DATA NEEDS

The experimental program has been designed [1] to measure two important features of the interaction of an explosive HEDD jet with spent fuel and surrogate material pellets contained within a Zircaloy cladding tube single rodlet:

1. The measurement of a more accurate and precise value for the Spent Fuel Ratio, SFR:

   SFR = [spent fuel aerosol particle masses] / ["surrogate" aerosol particle masses]

   The SFR determination essentially involves the comparison of the aerosol particle data from irradiated fuel to unirradiated surrogate fuel, obtained in paired experiments using the same apparatus, identical test conditions, and with the same HEDD. The SFR will be calculated from aerosol particles collected in multiple size ranges, from 0 up to about 20 μm aerosol equivalent diameter, AED. The AED is defined by means of the settling velocity of a unit density sphere, and is equivalent to the particle geometric-diameter times the $(\text{particle density})^{1/2}$. Multistage particle impactor samplers are used to classify aerosolized particles according to their aerodynamic diameter. The measured SFR values provide a data bridge to previous large-scale surrogate (DUO_2) cask tests [2,3] and consequence assessments. The SFR values permit scaling to other geometries, single fuel rod to rod bundles, by means of modeling.

2. The measurement of enrichment of volatile fission product nuclides like cesium and ruthenium preferentially sorbed onto specific, respirable particle size fractions in the sub-μm to μm size range.

The aerosol testing requires sampling and measurement of the mass and physical characteristics of the aerosol particles produced from HEDD jet impact, with aerodynamic equivalent diameters, AED, up to 100 μm (microme-

ters), and with special emphasis on the respirable and thoracic fractions, < 10 μm AED. Data from the coarser aerosol particles in the ~ 10 to 100 μm AED range are of interest primarily for radiological "ground-shine" (dispersion, soil contamination) consequence estimates.

TEST PROGRAM DESIGN

The overall program consists of four sequential test phases, Phase 1 through Phase 4. Individual tests in each phase use the identical type of HEDD, but different test materials, with a similar geometry. Successive phase testing has allowed us to add and evaluate multiple test variables and target material (pellet) response to HEDD jets, and consequent aerosol particle production.

The initial Phase 1 tests were conducted by SNL and Fraunhofer ITEM, using glass pellets and leaded glass plates as representative brittle materials. This test phase included performance quantifications of the HEDD devices and refinement of the aerosol particle collection apparatus being used. Four Phase 1 tests were performed in 2002; results were previously summarized [1].

The more extensive Phase 2 tests use nonradioactive cerium oxide, CeO_2, as sintered ceramic pellets contained within Zircaloy cladding tube assemblies, similar to spent fuel rods. CeO_2 was selected [1,6] as an excellent chemical "surrogate" and a representative ceramic material for UO_2 fuel material for pressurized water reactor fuel rods. About 24 Phase 2 surrogate material tests were conducted in the 2002 - 2004 period, and have provided a large body of data [1]; Phase 2 test apparatus and results will be briefly reviewed herein. These Phase 2 tests were the necessary precursors to calibrate the equipment design and perform the more difficult Phase 3 and 4 tests; they allow us to anticipate the range of results from the latter tests.

Phase 3 tests, recently initiated, use slightly radioactive, unirradiated depleted uranium oxide, DUO_2, pellets in comparable, new Zircaloy cladding tube test rods. The overall Phase 3 aerosol-explosive test chamber is based on the similar, but less sophisticated Phase 2 chamber design(s). Six of the Phase 3 tests should be performed in late 2004 and early 2005. The DUO_2 test rodlets have been designed and fabricated by our French test partner, IRSN, and their contractor, CERCA (a subsidiary of Framatome-ANP), and will be described in detail.

Phase 4 tests will use radioactive spent fuel pellets in short test rodlets. Four of the Phase 4 tests will use high burnup (~ 72 GWd/MTU) spent fuel originating from the H.B. Robinson pressurized water reactor. Another four Phase 4 tests will use a low-medium burnup (~ 38 GWd/MTU) spent fuel originating from the Surry pressurized water reactor. All of the spent fuel is being characterized in detail and fabricated into test rodlets at Argonne National Laboratory, for HEDD-impact aerosol testing in the Gamma Irradiation Facility, GIF, at SNL during 2005. The final calculation of the spent fuel ratio, SFR, as a function of aerosol particle size ranges, will be based on a comparison of the aerosol particle results from the Phase 4, actual spent fuel data, to the Phase 3, "surrogate" DUO_2 data. These data will be obtained from paired sets of experiments using identical test conditions and apparatus.

EXPERIMENTAL DETAILS

The major components required for conduct of these surrogate and spent fuel sabotage, HEDD impact, and aerosol measurement tests consists of: test rods and target pellets (Zircaloy-4 cladding tubes, ceramic pellets of cerium oxide, non-radioactive fission product dopant disks, depleted uranium oxide, or spent fuel; support rods and hardware); a test chamber consisting of an aerosol collection chamber and an explosive containment vessel; a conical shape charge, the HEDD; aerosol particle samplers (particle impactors, sampling tubes, pumps, etc.); a HEDD-jet stop box; and, a test facility to perform the tests in. Test components, while specific to individual test phases, are very similar and will be described in detail, starting with the test chamber. All nonradioactive, surrogate material tests to date have been performed at the SNL Explosive Components Facility. All Phase 3, DUO_2 (slightly radioactive), and Phase 4, spent fuel (highly radioactive) aerosol-explosive tests will be performed at the Sandia Gamma Irradiation Facility, GIF, Test Cell 3, using closely controlled radiological and explosive safety conditions, under both SNL and DOE-Sandia Site Office authorizations.

Test Chamber: The vertical test chamber used for Phase 2 tests is shown in Figure 1. The (open) aerosol collection chamber, with a horizontal surrogate material test rodlet inserted and visible, is located in the top "aerosol collection chamber." The "explosive containment chamber" is on the bottom. When the explosive HEDD installed in the bottom chamber is remotely detonated, a HEDD jet shoots upward through a small-diameter hole in the thick steel plate between the two chambers, penetrates the test rodlet, and is stopped in the thick HEDD jet-stop block

on the top of the test chamber, not visible. The entire test chamber, approximately 0.6 m-diameter by 1.3 m-high, is fabricated out of thick steel to contain the explosive blast and all aerosol particles produced. It is a durable and demonstrated leak-tight system that has been used for more than 20 tests. It has been instrumented to measure multiple temperatures and pressures in the top aerosol collection chamber and in the aerosol equipment shown at the top of Figure 1.

The new, more refined aerosol-explosive, vertical test chamber for all Phase 3 tests is shown in Figure 2, is based on the Phase 2 test chamber. It incorporates recessed, removable flange covers for the top aerosol and bottom explosive chambers. This chamber has been explosively over-tested successfully to twice the HEDD-produced pressures expected in planned usage. This chamber has also been modeled for stress analyses, welds have been X-rayed and dye-penetrant tested, and it has been hydrostatically and leak tested.

The aerosol-explosive, vertical test chambers for Phase 4, spent fuel tests, have been designed but not yet fabricated. These are very similar to the Phase 3 test chamber, except that there is no flanged access port to the top aerosol collection chamber. A transparent drawing of a Phase 4 explosive-aerosol test chamber is shown in Figure 3. The internal HEDD jet stop block, the horizontal spent fuel test rodlet, and four vertical aerosol sampling tubes are visible. Once the remotely inserted spent fuel rod has been explosively disrupted by the HEDD jet, the post-test chamber will NOT be opened (it will be examined using fiber optics equipment), in order to prevent escape of radioactive particulates. The only particle sampling is via the top-mounted aerosol impactor sampling devices, to be described. Each Phase 4 test chamber will be used one time only, temporarily stored at Sandia with the particulated spent fuel contained within, then shipped off-site to an approved, limited-term radioactive material facility prior to final disposal. The total volume of the Phase 4 aerosol chamber is identical to the Phase 3 aerosol chamber; compensation has been made for the lack of the internal flange support in the Phase 4 top chamber.

Fig. 1 Phase 2 Test Chamber (open), aerosol chamber (top), with test rod, and explosive containment chamber (bottom)

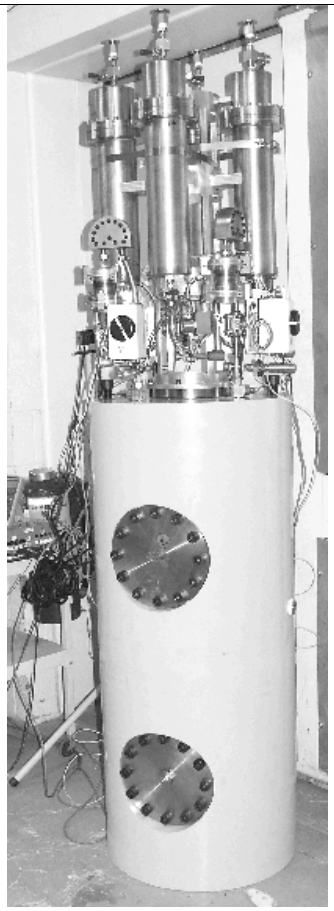

Fig. 2 Phase 3 Test Chamber and Aerosol Sampling System

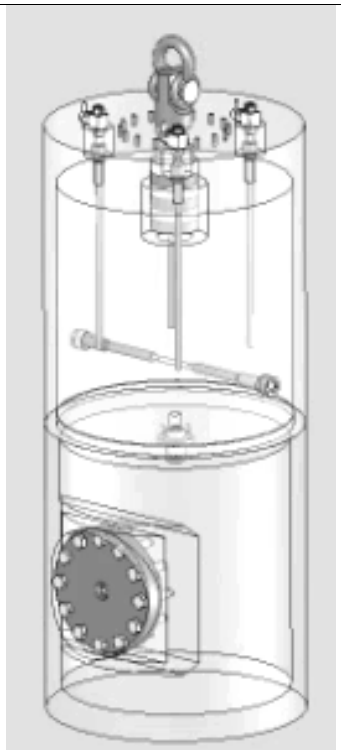

Fig. 3 Phase 4 Test Chamber, transparent drawing

Target Pellets and Test Rodlets: All test rodlets have been fabricated with ceramic pellets (either the surrogate cerium oxide, DUO_2, or spent fuel/UO_2) inserted with a minimal pellet-to-cladding gap into Zircaloy-4 cladding tubes, about 10.6 mm O.D. Pellet sizes and cladding tubing were sized to match H.B. Robinson fuel rods. Phase 2 tests used nine cerium oxide pellets fabricated by the Ceramic Synthesis and Processing Department at SNL. The pellets were fabricated by uniaxially dry powder pressing CeO_2 powder in a steel die followed by sintering for 2 hours in air at ~ 1600°C. The sintered pellets have an average density of ~ 6.7 g/cc; their ~ 95 % theoretical density is similar to that of UO_2 fuel pellets. Pellets are nominally 9.2 mm in diameter and 7.0 mm long. In many tests, two thin fission product dopant disks were placed on either side of the central pellet. These disks contained non-radioactive cesium iodide and ruthenium oxide (thermally volatile fission product species) and non-volatile strontium oxide, contained within a resin-base.

The Phase 3 target rodlets were fabricated by CERCA, in Romans-Sur-Isère, France, for IRSN, for testing at SNL. Each test rod contains five 13.8 mm-long pellets of ~ 95% theoretical density DUO_2, with dished ends, as shown in Figure 5. The test rodlet design, shown in Figure 4, was a collaborative effort by IRSN, SNL, and Argonne National Laboratory. Three of the rodlets contain two of the non-radioactive fission product dopant disks (as used in the Phase 2 tests, provided by SNL) surrounding the DUO_2 pellet in the center, shown in Figure 4 and Figure 5. Three of the rodlets are filled with air at atmospheric pressure; the other three are internally pressurized with He at 4 MPa, similar to PWR fuel rods, within the end plenum regions of the rodlet, shown in Figure 4. Laser end-cap and seal welding was used to fabricate the rodlets. The completed rodlets were He-leak tested, all welds were X-ray tested, and then the rodlets were shipped to SNL.

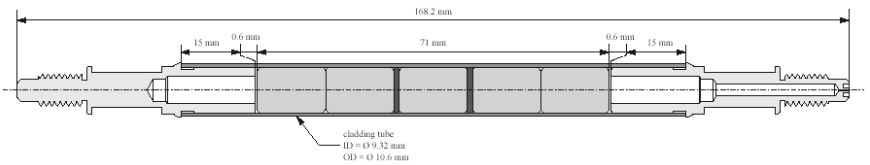

Fig. 4 Phase 3 DUO_2 Test Rodlet DUR-4, with dopant disks, He-pressurized

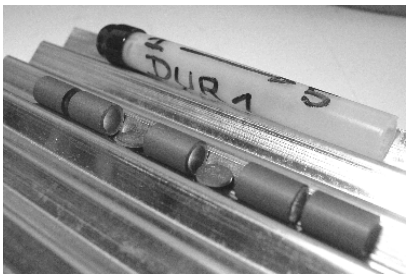

Fig. 5 DUO_2 Pellets and Dopant Disks, for Test Rodlet DUR-1 ➡

The Phase 4 spent fuel pellets, both from the high-burnup H.B. Robinson reactor and the lower-burnup Surry reactor, have been fully characterized in the Argonne National Laboratory Alpha Gamma Hot Cell (AGHC) facility. Characterizations include: visual exams, axial gamma scanning, optical metallography, cladding hydrogen content, and isotopic analyses – for following aerosol and radiological source term material behaviour evaluations. The spent fuel pellets will be contained within their original, irradiated Zircaloy cladding tubes; new Zircaloy end fittings, similar to the design shown in Figure 4, will be added, then circumferentially sealed with rotary tungsten inert gas (TIG) welding. Each of the four H.B. Robinson test rodlets will contain about eight 6.9 mm-long fuel pellets. The test rodlets will be internally pressurized with He to 4.4 MPa. The Surry test rodlets will each contain four 15.5 mm-long pellets, with an internal He pressurization of 3.3 MPa. Laser welding will be used to seal the pressurization hole in the end fitting. Argonne will complete the spent fuel rodlet fabrication effort soon, including post-welding leak testing and external contamination control, and then transport the rodlets to SNL for testing in 2005.

Particle Analyses: We have used several types of multistage aerosol particle samplers to date (Respicon™ 3-stage virtual particle impactors, Berner 9-stage particle impactor [1]), but now use four 8-stage multi-jet Marple cascade impactors per test. These impactors are designed to measure aerosol particle size distributions from about 0.4 – 21 µm AED, with an additional pre-filter stage (larger particles) and final stage (~ all particles < 0.4 µm). We also use separate, in-line, large-particle separators, LPS, for collecting ~30-100 µm AED particles; refer to Figure 2, top area. The LPS collectors were jointly designed by SNL and ITEM aerosol experts; the design is based on work published previously [7]. Each Marple and LPS sampling sub-system requires a vacuum bottle to draw a calibrated, nominal 2 L/min flow rate through the samplers; a critical orifice and small HEPA filter are also used. One second after the HEDD detonation, explosion proof valves on top of the chamber (visible in Figures 1 and 2) are opened, so that aerosol produced can be collected in the top-mounted particles collectors for a period of 10 seconds. This proceedure effectively samples a representative, reproducible portion of the still-suspended aerosol particles. Aerosol and larger surrogate Cerium oxide or DUO_2 particle materials (but not spent fuel) that were not collected in the Marple and LPS samplers are collected post-test, after the aerosol collection chamber is opened. These

particles are then mechanically sieved to determine size distributions from 25 to 1000 μm (geometric). The mass of all collected particles, on the impactor stages or as sieved materials, are determined by weight gain measurements, photographed, then chemically dissolved; detailed chemical analyses of major elements and fission product species in the particles were obtained using inductively coupled plasma-mass spectrometry, ICP/MS. Chemical analyses were necessary because much of the collected particle mass consists of fine carbon soot, a combustion byproduct of the HEDD explosive detonation.

Test Variables: To date, 24 Phase 2 surrogate material aerosol-explosive tests have been performed plus two Phase 2/Phase 3 cross-over surrogate tests. We previously documented test details and summary results [1] for the first nine tests. Multiple test variables incorporated in this test system included: (a) use of fission product dopant disks in some tests; (b) "blank" tests, with no CeO_2 pellets or no target rod at all, in order to evaluate behavior of soot particles and test contaminants; (c) two tests with non-radioactive German high-level waste glass with fission product surrogates, in a cooperative effort with our German test partners; (d) two tests with the test rodlet internally pressurized to 2.8 to 3.8 MPa with helium, in comparison to most tests at atmospheric pressure, in air; (e) multiple modifications and optimizations to the test chamber aerosol containment efficiency and aerosol particle sampling apparatus; and, (f) temperature and pressure sensors were installed in the aerosol collection chamber and in aerosol particle sampling lines to monitor conditions from pre-detonation to post-sampling, to aid analyses. One additional test variable is used for the Phase 3 tests, with DUO_2, and the Phase 4 tests, with irradiated UO_2 spent fuel; the internal test chamber initial atmosphere is either air or nitrogen. This variable allows us to evaluate any changes in aerosol particle formation if some of the uranium is converted to higher oxidation states (from +4 to higher) by the high energy and temperature HEDD jet.

Test Facility Requirements and Controls: The combination of an explosive, high energy density device and highly radioactive spent fuel test rods in Phase 4 of this program (as well as slightly radioactive DUO_2 pellets, in Phase 3) gives rise to *significant* radiological safety testing concerns. These concerns necessitate extensive facility environmental and safety assessment evaluations, contamination and radiation controls, plus remote handling and posttest disposal concerns. These same issues significantly increase testing expense and difficulty. The SNL Gamma Irradiation Facility, GIF, Test Cell 3, where the radioactive Phase 3 and Phase 4 tests will be performed, has been operated as a clean facility for gamma irradiation testing, with no contamination. The GIF is a Hazard Category 3 nuclear facility with a DOE approved Documented Safety Analysis, DSA. However, the existing safety basis documentation did not adequately address the use of explosives (the HEDD) and fissile materials, specifically spent fuel, due to the lack of analysis in the original DSA. Because of the unique needs and requirements of this spent fuel sabotage explosive-aerosol test program (with all test component and surrogate plus spent fuel materials requirements documented in detail [1]) , the current DSA has been substantially modified by Sandia and submitted to the DOE Sandia Site Office for review and approval (in process). The following nuclear facility safety and radiological concerns have been addressed and (tentatively) resolved, permitting us to proceed with Phase 3 and Phase 4 testing: worker exposure to ionizing radiation, contamination control (inside a leak-tight test chamber plus external secondary confinement control), explosives safety and handling, testing, and identification of post-test spent fuel material disposal path. A full NEPA determination for the entire test program has also been performed and approved by SNL. Identification of the required test facility supporting equipment has been identified and will be tested in Phase 3 tests prior to their necessitated use in Phase 4 tests.

RESULTS AND OBSERVATIONS

Target Rod Disruptions: Observed effects of HEDD explosive jet impact on the Phase 2 CeO_2 pellet-containing test rodlets were fairly consistent for all tests performed. The HEDD jet hits the center-point of the target rod in < 90 μsec, "particulating" a portion of the test rodlet and included pellets, leaving the remainder intact. The total Zircaloy cladding tube gaps observed averaged 21-30 mm, but varied from 16-35 mm, primarily due to jagged flaps of Zircaloy of different lengths. A total of 3.7 to 6 of the original 9 CeO_2 ceramic pellets, each 7 mm-long, were fragmented/aerosolized, about a 26-42 mm length. In most tests, the CeO_2 pellets adjacent to the destroyed segment of Zircaloy tubing were firmly wedged into the tube, by fine particle "blow-back" of material in the small tube-to-pellet gap, and could not easily be removed from the cladding. These remaining, captive pellets were essentially whole, with some observable external fracturing. The end-most pellets were essentially undamaged. All fission product dopant (disk) materials, if included, were totally particulated and possibly vaporized by the shock wave and thermal pulse from the HEDD jet. As the temperature cools after the jet impact, some aerosolized and/or volatilized species were sorbed onto nearby cerium oxide and explosive residue soot (carbon) particulate materials, and then were segregated by size by the particle impactors. Observed results were similar for the surrogate German high-level glass target rods (with cesium, lanthanum and neodymium oxide dopants), impacted by the same HEDD jet in the

same test chamber. About 29-44 mm of (stainless steel) cladding tube was destroyed; there was about 37 mm of disrupted glass length -- the rest was contained.

Aerosol Summary: We have fully documented a compilation of all aerosol data [1] from the first nine Phase 2 tests, collected with Respicon and Berner particle collectors, in the range of 0 to about 16 µm AED. The full results and interpretations of aerosol data from the remainder of the 24 Phase 2 tests will be documented in the near future. The major aerosolized elements detected were copper (from the HEDD), first, then cerium, followed by zirconium (from the cladding tube). Less than 1-2 % of the total disrupted CeO_2 pellet mass was in the aerosol size range [1,6]. The cerium aerosol particles peak in concentration in about the 2-21 µm AED range, the copper and zirconium aerosols are maximized in the 2 +/- 1 µm range. For the most recent Phase 2 tests, we used 4 Marple impactors, a large particle separator, plus a separate line of six sequential Gelman filter samples, to monitor particle stratification and settling over about the 2.5-60 second period after HEDD detonation. Two sampling levels in the aerosol chamber (near top and lower/at rod-target level) were used for the impactor samples with two impactors at each level. No significant particle stratification, high vs. low level, or over the total sampling time, was observed. Preliminary normalized total (target plus soot) particle sampling data for these tests indicate a mean of 16-24 mg/liter at the 95% confidence interval; about 10 mg/liter of particulates were from the target material. Most of the aerosol particles collected in the Marple impactors have been chemically analyzed; interpretations and supporting laboratory calibrations are still in process and will be documented later. Preliminary results indicate a secondary particle concentration peak in the ~70-100 µm size range, as collected on the LPS. There is also a significant quantity of aerosolized iron, from the steel test chamber and jet stop-block interaction with the HEDD jet, concentrated in the 0.5 to 4 µm AED range.

Fission Product Enhancement: We have incorporated dopant disks of thermally volatile, non-radioactive chemical fission product species cesium, iodine, ruthenium, as well as non-volatile strontium in multiple surrogate cerium oxide pellet aerosol-explosive tests. The possibility exists that the dopant disk design may cause results to be biased in a more conservative direction compared to actual spent fuel, due to enhanced mixing of the aerosolized dopant species. There is a clear indication of enhanced sorption of the volatile cesium and ruthenium onto the smaller, respirable size particles of surrogate cerium oxide. The peak cesium concentrations were maximized in the respirable range of 0.5-1.5 µm AED. The preliminary, calculated cesium enhancement factor ranges from X1 at 1 µm AED up to about a factor of X20 at the smaller 0.4 µm AED. The calculated enhancement factor for cesium dopant in the German HLW glass tests similarly ranged from X1 to X10 (at 0.2 µm). Data on ruthenium were not as distinct; it was found over the 1-16 µm size range. The non-volatile strontium species does not show the same sorptive behavior as volatilized cesium. Most of the strontium was collected along with larger sized particles, although a small amount was observed in the aerosol range. It is postulated that the enhanced volatile fission product sorption on the smallest particle sizes is due to two factors: (1) the smallest aerosol particles have a larger surface area for sorption compared to the larger particles, and (2) the explosive residue carbon soot particle sizes are maximized in concentration at < 4 µm AED, also providing a large surface area for enhanced volatilized species sorption.

DISCUSSION & SUMMARY

We provide a concise summary of an ongoing, multi-phase, spent fuel sabotage explosive-aerosol measurement test program, focusing on a description of the test components for the recently initiated, surrogate cerium oxide and radioactive DUO_2 Phase 3 and upcoming spent fuel Phase 4 of the program. We also review some of the more important aerosol results and observations from the recently completed Phase 2 surrogate material tests using cerium oxide ceramic pellets in test rodlets. This test program, performed primarily at SNL, has had major design input, participation, and supplemental testing from other U.S., German, French, and British partners, as part of a collaborative, international Working Group for Sabotage Concerns of Transport and Storage Casks, WGSTSC. This testing began in 2002.

A substantial degree of progress and quantity of data has been generated from this test program over the past two years. We have performed 24 Phase 2 explosive-aerosol measurement tests, essentially completing Phase 2 of the test program [1,6] and, with the additional two most recent tests, starting the cross-over Phase 2 / Phase 3 tests. Results and observations from the all surrogate cerium oxide explosive-aerosol tests performed, as well as supplemental tests using non-radioactive German high-level waste glass rod targets, have been quite consistent. We have characterized and chemically analyzed both the aerosol particles collected by multi-staged particle impactor collection devices, plus the residual impact debris remaining in the post-test aerosol collection chamber. We have observed a clear indication of enhanced sorption of the volatile fission product species cesium and ruthenium

onto the smaller, respirable particles. We have optimized the test chamber (joint vertical aerosol collection chamber and explosive containment vessel) through multiple designs and improvements so that it is leak-tight, durable, and safe for repeated use. We have optimized, designed, and assembled an aerosol sampling system that satisfies the needs of both U.S. and German aerosol experts involved with the program.

These data allow us to calculate the spent fuel ratio (calculated for individual particle size distributions) and extend the test results to other nuclear fuel sabotage situations through follow-on radiological consequence, vulnerability modeling and assessments. The primary test benefit of using the ratio of aerosol particles for the SFR determination is that it is not necessary to recover and analyze all of the aerosolized materials produced, only that the identical portion of aerosol particles from both the spent fuel and surrogate fuel tests be obtained, analyzed, and compared. This ratio drives the requirement for use of identical-as-possible test apparatus and test conditions for multiple test phases and materials. In addition, by focusing on the spent fuel ratio determination, we can use test rodlets containing only a few actual or surrogate fuel pellets for aerosol particle production – we do not need to test entire fuel assemblies nor entire casks full of fuel assemblies. The explosive detonation/impact of spent fuel by a high energy density device, HEDD, plus subsequent aerosolization and potential release to the environment, is relevant to a credible terrorist attack scenario on nuclear fuel transport or storage casks.

This test program, and the radiological consequence and aerosol dispersion modeling efforts that will follow, support data and policy needs of the U.S. Department of Energy, Nuclear Regulatory Commission, and the European WGSTSC partners. Both the U.S. DOE and NRC are providing substantial financial support and technical guidance to SNL, in order to both resolve nuclear programmatic issues and to perform the spent fuel sabotage aerosol measurement tests through completion, as anticipated in 2005. Final post-test clean up and off-site spent fuel material transport will be conducted in 2006. All the WGSTSC partners need information and results from this research to better understand potential radiological impacts from sabotage of nuclear material shipments and storage casks, and to support subsequent risk and safety assessments, modeling, and preventative measures. The current results will also be compared to, and extend, previous limited spent fuel ratio testing and dispersion release modeling performed in the past [2-5]. The continuing, successful conduct of this program provides significant technical and policy benefits for all participants.

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