RESPIRABLE AEROSOLS RESULTING FROM HEDD INTERACTION WITH SURROGATE FUEL PELLETS

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ABSTRACT

An experimental program is underway to measure the aerosol source term for spent fuel and surrogate spent fuel subjected to the action of high energy density devices (HEDD). The program is supported and guided by an international working group composed of US, French, German, and United Kingdom agencies. Preliminary results as well as a description of the overall program are presented. Measurements made to date using cerium oxide pellets in Zircaloy cladding and a small precision HEDD indicate that about 1% to 3% of the disrupted surrogate pellet volume is converted to respirable aerosol. The completed experiment program will provide surrogate and spent fuel aerosol production in identical experiments which should permit an accurate estimate of the spent fuel ratio (SFR) that is needed to relate full scale attack release data on surrogate materials to expected releases from similar attack using HEDDs on spent fuel.

INTRODUCTION

Concern for the potential harm that could be caused by terrorist activity has entered many aspects of modern life as a result of the events of September 11, 2001. Because of the public’s extra sensitivity engendered by potential exposures to radiation and radioactive materials, the nuclear power industry is particularly concerned with understanding the potential risks and consequences of terrorist actions involving materials in the nuclear fuel cycle. One particular concern is nuclear materials in transportation because it is one aspect of the fuel cycle where material is outside the highly effective protection offered by security systems surrounding fixed facilities. One particular aspect of the fuel cycle, transportation of spent nuclear fuel, has been the focus of much of the effort because of the potential for future large scale shipments of spent fuel to the repository at Yucca Mountain, which has been approved for pursuit of a license from the NRC for storage of spent fuel and high level wastes. However, it should be noted that the concerns for potential terror or sabotage-related releases from spent fuel in transit actually date back to the mid-to-late 1970s when NRC funded the “Urban Study” (Finley, 1978), which looked at a variety of mechanisms for, and consequences resulting from, release of material from spent fuel casks and packages containing other radioactive materials.

This paper will briefly review the history of these analyses, discuss the progress on an ambitious experimental program underway under the aegis of an international working group, and provide recently derived parameters to support analyses that have been obtained from experiments completed at Sandia on behalf of the International Working Group for Sabotage Concerns of Transport and Storage Casks, WGSTSC.

Prior Work

Papers at previous Waste Management Symposia, PATRAMs and other technical conferences have explored various methods and data sources used for estimating the potential consequences of an optimally successful attack on a spent fuel cask using High Energy Density Devices (HEDDs). These papers have included full consequence analyses as well as re-analyses of older experimental data to glean new perspectives on the parameters used in analysis and data in hand or expected from new experiments. A list of relevant works is contained in the references; however, a brief review of selected prior work is needed to understand the basis for the ongoing work at Sandia under the aegis of the WGSTSC.

A report authored by Sandoval and others published by Sandia National Laboratories (SNL) in 1983 (Sandoval, 1983), contained an account of experiments at SNL and at INEL that were supported by the DOE in order to determine if the release fraction assumptions contained in the Urban Study (which led to very high potential doses) were at all valid. This was an important issue at the time because the NRC had placed new security requirements on spent fuel shipments in high population areas to minimize the likelihood of an attack using HEDDs. These requirements were of concern for the added unnecessary expense and complication to what had been a relatively straightforward activity. The SNL report also reviewed complementary experimental work conducted at Battelle Columbus Laboratory (BCL) for the NRC (Schmidt, 1982) as the basis of an analysis of the potential consequences of HEDD attack on various types of spent fuel casks. The results of that analysis suggested that the earlier analysis lead to consequences that were orders of magnitude too high.

One component of the SNL analysis was the SFR, the ratio of respirable aerosol mass from real spent fuel to that obtained for similar conditions of attack on surrogate spent fuel materials. Since it was prohibitively expensive to experiment with spent fuel in full-scale casks, spent fuel and surrogate material (depleted UO2) disruption by HEDDs was done in small scale by BCL and INEL (for SNL) with measurement of aerosols produced (Alvarez, 1982). A full-scale test using surrogate material in an obsolete truck cask provided the data on how much aerosol material might be released to the environment from an HEDD attack on a real cask. To extend the full scale release data to real transport situations required a value for SFR. For a variety of testing reasons, the small-scale data provided less than fully deterministic estimates of the SFR. Using various combinations of data, calculated estimate of SFR varied from 0.7 to almost 6. The largest value, 5.6 was used in calculations that demonstrated that the Urban Study estimate was orders of magnitude too high. However, the authors of the SNL study felt strongly that the best estimate of SFR, if there were more data, would more likely to be on the order of 1 to 2.

When the DOE decided to include an analysis of the consequences of a terrorist attack using an HEDD in the Yucca Mountain Project EIS, they contracted with SNL to extend the earlier SNL analysis to modern casks. That study (Luna, 1999), reviewed the data supporting the calculation of SFR and arrived at 3 as a central estimate for the value for SFR that was used in the analysis. However, it was noted that the actual value might be larger or smaller by as much as a factor of 3. A paper presented at WM2003 (Luna, 2003) based on data not available in 1999 also suggested that SFR is on the order of 2, but the aerosol data was for particles somewhat larger than those in the respirable size range.

The 1999 YMP study also included an estimate of the effect of cask blowdown caused by release of rod plenum gases in moving particles out of the cask after the original HEDD event. This was required because the surrogate rods used in the 1982 SNL experiment were not pressurized. That analysis required an estimate of the mass of respirable surrogate particles contained in the cask that might have been swept out by the plenum gas release. Through analysis of a variety of data on the impact breakup of brittle materials, a value of 5% respirable surrogate particle production (compared to the larger disrupted volume of fuel rods) was indicated, but the data from which it was obtained was for experiments with significantly smaller energy input per mass of solid material than would be typical of an HEDD interaction.

In 1994 GRS conducted a set of three full-scale tests in a 9-assembly storage cask with pressurized surrogate spent fuel rods (Pretszch, 1994). These tests produced from 1/3 to 1/9 of the release in the SNL test and significantly less release than would be predicted based on the YMP analysis. This suggests that internal trapping of aerosols is important in predicting their release.

Lacking full-scale tests of every cask containing spent fuel, or a full-scale test of every cask with realistic surrogate materials together with a good value of SFR, analytical models must be constructed to estimate the quantity of aerosols released. These models require information on:

- Aerosol production (the SFR and aerosol mass fraction as a function of particle diameter)
- How the resultant aerosol moves within the fuel rod bundles and between the compartments formed by the basket and ultimately to the environment.

To date, estimates of the potential consequences of HEDD attacks on spent fuel casks have been upper limit estimates as a result of using:

- Upper limit values for SFR,
- Aerosol production,
- Extrapolation from full-scale surrogate experiments of single element casks in which trapping of aerosols in internal compartments is non-existent.

That the latter factor is a concern is illustrated by comparison of results from the 1982 SNL test and that from the GRS-funded study in 1992-4.

Current Experimental Program

Since 2001, the need for accurately quantifying the information described above 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. The WGSTSC partners support the need for this research to better understand potential radiological consequences, and to support subsequent risk assessments, detailed modeling, and potential preventative measures.

Sandia National Laboratories has a leading role in performing the WGSTSC test program. Both the U.S. Department of Energy and Nuclear Regulatory Commission provide sabotage and transportation program support. 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 UO2 (surrogate) fuel test rodlets. The Office for Civil Nuclear Security, OCNS, in the UK, participates in a consultatory role.

The experimental program is designed to measure two important features of the interaction of a HEDD jet with spent fuel or surrogate material pellets contained within a Zircaloy cladding tube:

- Measurement of a more accurate and precise value for the Spent Fuel Ratio, SFR
- Enrichment of volatile fission product nuclides like cesium and ruthenium, preferentially sorbed onto specific, respirable particle size fractions, in the µm to sub-µm range

The aerosol testing requires sampling and measurement of the mass plus chemical and physical characteristics of the aerosol particles produced, with aerodynamic equivalent diameters, AED, up to 100 µm (micrometers), and with special emphasis on the respirable fraction (nominally < 10 µm AED). The coarser aerosol particle range of 10 to 100 µm AED is of interest primarily for evaluation of radiological effects of groundshine and soil contamination.

The overall program consists of four sequential test phases. Individual tests in each phase use the identical type of HEDD, but different test materials, with a similar geometry. SNL, and Fraunhofer ITEM, using glass pellets and leaded glass plates as representative brittle materials, conducted the initial Phase 1 tests. The more extensive Phase 2 tests use non-radioactive cerium oxide, CeO2, in sintered ceramic pellets contained within Zircaloy cladding tube assemblies, similar to spent fuel rods. CeO2 was selected as an excellent chemical "surrogate" and a representative ceramic material for UO2 fuel material for pressurized water reactor fuel rods. Twenty-four Phase 2 surrogate material tests conducted over the last two years have provided a large body of data and are the primary focus of this paper. They are the necessary precursors to calibrate and perform the more difficult Phase 3 and 4 tests.

Phase 3 tests will use slightly radioactive, unirradiated depleted uranium oxide, DUO2, pellets in comparable, new Zircaloy cladding tube test rods. Six of these tests will be performed in 2005. The DUO2 test rodlets have been designed and fabricated by our French test partner, IRSN, and their contractor, CERCA.

Phase 4 tests will use highly radioactive, actual 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. Four additional Phase 4 tests will use a lower 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 2005-6.

The final calculation of the spent fuel ratio, SFR, as a function of aerosol particle size range, will be based on a comparison of the aerosol particle results from the Phase 4, actual spent fuel data, to the Phase 3, "surrogate" DUO2 data. These data will be obtained from paired sets of experiments using identical test conditions and apparatus (Molecke 2003a, 2003b, 2004)

Experimental Detail, Phase 2

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 are specific to individual test phase and have been modified as a function of time. The following details focus on the most recent Phase 2 surrogate explosive-aerosol tests, all performed at the Explosives Component Facility at SNL (Molecke, 2004).

In many tests, two thin fission product dopant disks are placed on either side of the central CeO2 pellet. These disks contained non-radioactive chemicals, cesium iodide, ruthenium oxide (thermally volatile fission product species), and non-volatile strontium oxide, contained within a plastic resin-base material. All fission product dopant materials were totally aerosolized and possibly vaporized by the shock wave and thermal pulse from the HEDD jet. As the temperature cools after the jet impact, aerosolized and/or volatilized species can sorb onto nearby cerium oxide particulate materials that are size segregated by the particle impactors and become available for chemical analysis. We then can evaluate if preferential enhancement of these species occurred.

The vertical test chamber used in Phase 2 tests is shown in Figure 1. The open aerosol collection chamber, with a horizontal test rodlet inserted and visible, is located in the top "aerosol collection chamber." The "explosive containment chamber" is on the bottom. When the 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 aerosols produced. It is a durable and demonstrated leak-tight system.

Target Rod Disruptions

The observed effects of HEDD explosive jet impact on the CeO2 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, yielding a 21-30 mm average gap in the Zircaloy cladding tube. The total Zircaloy tubing gaps observed varied from 16-35 mm, primarily due to jagged flaps of Zircaloy of different lengths; refer to Figure 2. A total of 3.7 to 6 of the original 9 CeO2 ceramic pellets, each 7 mm long, were fragmented/aerosolized, about a 26-42 mm length. In most tests, the CeO2 pellets adjacent to the destroyed segment of Zircaloy tubing were firmly wedged into the tube, by "blowback" fine particles 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. Observed results for the surrogate German high-level glass target rods (also with non-radioactive fission product dopants), impacted by the same HEDD jet in the same test chamber, were similar. About 29-44 mm of (stainless steel) cladding tube was destroyed; there was about 37 mm of disrupted glass length-- the rest was contained

While there have been no actual spent fuel results obtained at the present that allow estimation of the SFR, the results from tests completed to date are relevant to the problem of estimating the potential impact of HEDD attacks on spent fuel casks. Table I contains some of the preliminary results of the program.

Table I. Respirable Fraction Produced, Derived from Phase 2 Surrogate Experiments

	CeO_2 Disrupted		Cerium Oxide Respirable Fraction (%)					
Test	**CeO_2 (mg)**	**Ce (mg)**	**Respicon A**	**Respicon B**	**Berner**	**Marple A**	**Marple B**	**Test Avg.**
2/1A	11304	9203	0.48%	0.28%				0.38%
2/1B	11304	9203	0.53%	0.40%				0.46%
2/2A	10209	8311	4.93%	4.00%				4.46%
2/2B	10209	8311	2.81%	3.81%				3.31%
2/3A	13906	11321	2.65%		5.10%			3.88%
2/3B	15251	12416	4.53%	5.52%	4.17%			4.74%
2/4A	16149	13147	1.02%	1.56%	2.32%			1.63%
2/4B	14803	12051	1.16%	0.97%	1.72%			1.28%
2/5A	13286	10816	0.40%	0.40%				0.40%
2/5E	18869	15361				0.93%		0.93%
2/5G	16720	13612				0.54%		0.54%
2/8C	16017	13039				0.55%	0.70%	0.62%
2/8D	11286	9188				0.74%	0.89%	0.81%
Avg. all				**2.09%**	**3.33%**		**0.72%**	**1.97%**

The data presented relate to measurements done with differing aerosol collection samplers and in somewhat different conditions. The tests from 2/1A to 2/4B were done in an unsealed, "square-box" chamber with relatively simple sampling geometry (Molecke 2003b). Experiments from 2/5A to 2/8D were performed by sampling from the closed system shown in Figure 1. There may be appreciable particle loss in the sampling stream from thermophoresis and other effects; this is being quantified. The results show relatively good agreement in spite of the variations among the experiments. As a result of the sampling complexity, data from the 9-stage Marple samplers (tests 2/5E-2/8D) appear to be somewhat at odds with some of the earlier measurements, but no corrections have been applied yet to account for losses in the sampling system, which for the experiments (2/5E-2/8D), could increase values by about a factor of 2. From the non-Marple data, the average respirable fraction is 2% +/- 0.9% and for the Marple data 0.7% +/- 0.2%. As indicated earlier, the Marple data is likely to become as high as 1.5% when all corrections are made. A value of around 1.5% to 2% for the respirable fraction is somewhat below, but not inconsistent with, the 5% value used in the YMP analysis (Luna, 1999) and suggests that the estimated release predicted in that analysis is likely to be conservative.

Aerosol Measurement Summary: We have documented a compilation of aerosol data 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 major aerosolized elements detected were copper (from the HEDD jet), first, then cerium, followed by zirconium (from the cladding tube). Less than 1% of the total disrupted CeO_2 pellet mass was in the respirable size range. The cerium aerosol particles peak in concentration in about the 2-8 µm AED range, the copper aerosols concentrate in the < 2 µm and smaller sizes.

For the most recent Phase 2 tests, we used 2 or 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. These Marple aerosol results are similar to the earlier data. The cerium concentrations are maximized in the 1.5-15 µm AED range. Preliminary results indicate a secondary concentration peak in the ~70-100 µm size range, as collected on the Large Particle Separator (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.

A substantial degree of progress and quantity of data have been obtained from this test program over the past year. We have:

- performed about 19 additional explosive-aerosol measurement tests, essentially completing Phase 2 of the test program. Results and observations from all Phase 2 surrogate cerium oxide tests performed have been quite consistent.
- 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.
- observed a clear indication of enhanced sorption of the volatile fission product species cesium and ruthenium onto the smaller, respirable particles of surrogate cerium oxide.
- optimized the design and operation of the test chamber, joint vertical aerosol collection chamber and explosive containment vessel, so that it is leak-tight, durable, and safe for repeated use.
- optimized, designed, and assembled an aerosol sampling system that satisfies the needs of both U.S. and German aerosol experts involved with the program.

- used these test equipment optimizations to design and fabricate of the next test chambers to be used for Phase 3 and Phase 4 testing of nuclear materials.
- completed fabrication for six depleted uranium oxide Phase 3 test rodlets and are nearing completion for 8 high- and low-burnup spent fuel test rodlets to be used for Phase 4 testing.
- made progress, in detailing Phase 3 and 4 testing details, and in documenting additional relevant data and analyses.

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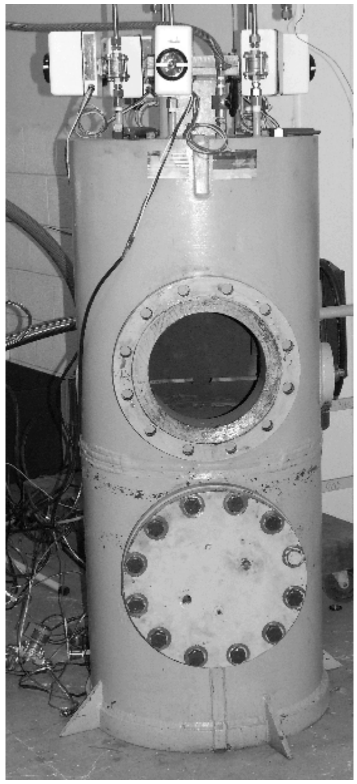

Fig. 1. Sandia phase 2 surrogate explosive-aerosol test chamber, and disrupted test rodlet

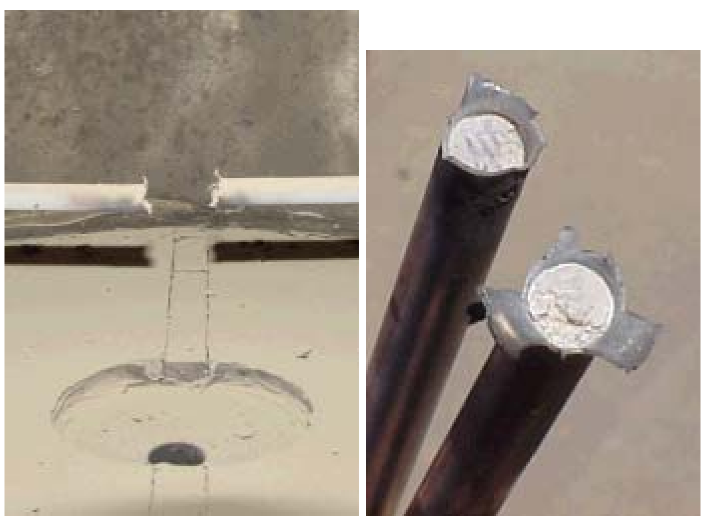

Fig. 2. Disrupted CeO2 test rodlet