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Surrogate/Spent Fuel Sabotage: Aerosol Ratio Test Program and Phase 2 Test Results

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Surrogate/Spent Fuel Sabotage: Aerosol Ratio Test Program and Phase 2 Test Results

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ABSTRACT

A multinational test program is in progress to quantify the aerosol particulates produced when a high energy density device, HEDD, impacts surrogate material and actual spent fuel test rodlets. This program provides needed data that are relevant to some sabotage scenarios in relation to spent fuel transport and storage casks, and associated risk assessments; the program also provides significant political benefits in international cooperation. We are quantifying the spent fuel ratio, SFR, the ratio of the aerosol particles released from HEDD-impacted actual spent fuel to the aerosol particles produced from surrogate materials, measured under closely matched test conditions. In addition, we are measuring the amounts, nuclide content, size distribution of the released aerosol materials, and enhanced sorption of volatile fission product nuclides onto specific aerosol particle size fractions. These data are crucial for predicting radiological impacts. This document includes a thorough description of the test program, including the current, detailed test plan, concept and design, plus a description of all test components, and requirements for future components and related nuclear facility needs. It also serves as a program status report as of the end of FY 2003. All available test results, observations, and analyses – primarily for surrogate material Phase 2 tests using cerium oxide sintered ceramic pellets are included. This spent fuel sabotage – aerosol test program is coordinated with the international Working Group for Sabotage Concerns of Transport and Storage Casks, WGSTSC, and supported by both the U.S. Department of Energy and Nuclear Regulatory Commission.

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The authors wish to acknowledge and express gratitude to the major contributions and support by multiple people to the ongoing definition and conduct of this surrogate/spent fuel sabotage and aerosol measurement test program. All of the participants of the international Working Group for Sabotage Concerns of Transport and Storage Casks are responsible for the continuing successes of this program. Most of the same people have also provided major technical inputs to the writing of this report, and the data within. For Sandia National Laboratories personnel in particular, we recognize Roy Dickey, for providing the designs and fabrication for most of the test components, and for the major conduct and guidance on all of the tests to date. Manny Vigil has provided guidance on the science of the explosive components and processes. Kevin Ewsuk and Chris DiAntonio have been responsible for the design and fabrication of the sintered ceramic, cerium oxide surrogate test pellets used in this program. Jeff Reich and James Hochrein have provided optical microscopy and elemental analysis services on the particulate materials collected from the tests. Richard Coats, Mike Gregson, and Susan Longley provide the major technical coordination and support for nuclear facilities related issues, and for guiding future radioactive, spent fuel test performance. Ken Reil, Sharon Walker, Don Berry, Paul Helmick, and Rob Naegeli also provide significant nuclear facilities guidance and support, as did John Guth and Jeff Philbin in the past. Richard Yoshimura, and Bob Luna - a consultant and retired Sandian, provided an excellent link with the technical definition and history of this test program. Mike Billone and Hanchung Tsai, Argonne National Laboratory, have provided most of the input for spent fuel characterization and related issues.

Of course, we also give special thanks to the Department of Energy, both the Office of Civilian Radioactive Waste Management and the National Nuclear Security Agency, and the Nuclear Regulatory Commission, Office of Nuclear Security and Incident Response, for providing major programmatic guidance and support for the successful, continuing conduct of this program.

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Surrogate/Spent Fuel Sabotage: Aerosol Ratio Test Program and Phase 2 Test Results

1. INTRODUCTION

Even in very severe accidents, casks used for spent nuclear fuel transport in the public domain are extremely resistant to releasing any of their contents. However, in some sabotage attack scenarios, aerosolized particles originating from disrupted fuel pellet materials could be released. A primary sabotage scenario considered is that of an attacker firing an armor-piercing weapon into a cask containing spent fuel assemblies, with the intent of trying to disperse some of the radioactive fuel contents, in order to cause harm to the public. Airborne aerosol materials have the potential to cause radiological consequences if released to the environment. Measurement of the actual amounts, nuclide content, and size distribution of the released materials from spent fuel is essential for predicting valid consequences of such radiological impacts. These factors have been measured in only a few tests involving high-energy impacts or sabotage events employing high energy density devices (HEDDs, e.g., armor-piercing weapons, conical shaped charges, etc.) that focus high explosive energy directly into the fuel materials. Large-scale tests to directly measure these parameters with transport casks containing actual spent fuel are prohibitive for multiple reasons, e.g., safety, costs, waste disposal, etc. A few tests have been conducted in the USA and in Germany with large-scale casks filled with depleted uranium fuel element simulants or surrogates. As a result of the lack of information concerning actual spent fuel behavior in such a sabotage event, conservative assumptions are used to develop source terms for consequence analyses and may significantly over-estimate that which would occur in a real sabotage attack.

The need for this spent fuel sabotage / aerosol test information has been strongly endorsed by both the U.S. and international program participants, as part of the international Working Group for Sabotage Concerns of Transport and Storage Casks, WGSTSC. The WGSTSC is coordinating this research to better understand the potential impacts of a sabotage attack on nuclear material transport and storage casks, to help quantify the risks, and to better protect people and the environment against radiological hazards arising from such sabotage events.

Table 1 provides a complete, current list of WGSTSC test program participants. In the U.S., Sandia National Laboratories has the lead role for conducting this test program, with support provided by the Department of Energy, DOE, the Nuclear Regulatory Commission, NRC, and Argonne National Laboratory, ANL. German participants, the Gesellschaft für Anlagen- und Reaktorsicherheit, GRS, and the Fraunhofer Institut für Toxikologie und Experimentelle Medizin, ITEM, are providing aerosol expertise, including design of the particle collectors and aerosol containment chambers, and analysis and evaluation of the aerosol particle data. They are also conducting related surrogate material aerosol particle formation and sampling, with non-HEDD testing. The Institut de Radioprotection et de Surete Nucleaire, IRSN, France, will provide depleted UO₂ surrogate fuel test rodlets and participates in the spent fuel test rodlet design. The Office of Civil Nuclear Security, OCNS, in the UK, participates in a consultative role. In addition, OCNS has also made available its substantial test facilities for conducting follow-on large scale cask plus payload testing, for a later stage to the current aerosol testing program.

There are significant benefits for the successful conduct of this program for all participants:

1. The cooperation of German, French, and British organizations and governmental entities provides a significant policy benefit, considering that the project not only can lead to improved safety of the environment from a postulated nuclear incident, but also will provide data useful to assess how to counter a terrorist threat. These benefits also project the fact that nuclear nations around the world are working together to mitigate terrorist threats to nuclear facilities and packages.
2. The spent fuel ratio data resulting from the tests and analyses will provide enhanced interpretations of both current and earlier data on surrogate test materials, as the data relate to actual spent fuel. Such analyses will provide improved safety assessments of the risks posed by a postulated nuclear incident, and allow improvements in the preparations for the response to such an incident.
3. The environmental benefit is that resultant test and programmatic data will allow safer and more secure transportation and storage of all the spent nuclear fuels, SNF, in the United States and the rest of the world. Through improved threat assessments and subsequent design, an additional margin of safety to the environment may be provided from the possible, albeit unlikely, sabotage scenario of SNF being struck by an HEDD.
4. The data will likely allow preparations against other related sabotage scenario events, which could save substantial resources that would otherwise have to be expended to address the wider scope of a potential incident resulting from the postulated event. Considering these results would be applied to transportation of large quantities of SNF over thousands of miles (kilometers) for tens of years, and then stored permanently, the cost savings could be considerable to the nuclear industry and the U.S. and foreign host governments.
5. As nations move toward permanent disposal of commercial SNF, the link between transportation and the repository surface facilities becomes ever more important. Transfer facilities and lag-storage areas on the repository surface need to address very similar questions relative to sabotage attacks. This SNF experiment work for storage and transportation can be directly applied to the surface facility operations, thereby facilitating the important process of evaluating sabotage risk to the entire back end of the fuel cycle system.

A major purpose of *this* document is to provide a thorough overview of the ongoing test program that supports the needs of the international Working Group for Sabotage Concerns of Transport and Storage Casks. Previous plans and detail from the draft “Master Plan for International WGSTSC Experiments to Determine Spent Fuel Ratio/Fractionation for Spent Nuclear Fuel” [Luna et al., 2002], and several recent conference presentations [Luna et al., 2001; Lake et al., 2001, Lake et al., 2002; Philbin et al., 2002; Molecke et al., 2003a; Molecke et al., 2003b; Molecke and Sorenson, 2003] have been incorporated herein, for completeness. This document provides a detailed description of the current test concept and design, a description of all current test components, requirements for future components, and a presentation of all available test results -- up through September 2003, observations, and aerosol particle analyses – primarily for surrogate material Phase 2 tests using cerium oxide sintered ceramic pellets. This document serves as a comprehensive program status report as of the end of FY 2003. Since Phase 2 of this test program is still in progress, only preliminary evaluations and interpretations of the aerosol particle data can be included. Thorough data interpretations and follow-on modeling will be documented separately, at a later time.

Table 1. WGSTSC Participants

International Working Group for Sabotage Concerns of Transport and Storage Casks	
Country:	Organization:
U.S.A.	Sandia National Laboratories (SNL) <ul style="list-style-type: none"> • Dept. 6141, Transportation Risk and Packaging • Dept. 2554, Explosive Components Facility (ECF) • Center 6800, Nuclear and Risk Technologies * ○ Dept. 1822, Materials Characterization, Analytical Chemistry ○ Dept. 1843, Ceramic Materials ○ Dept. 9117, Plasma, Aerosol, Non-continuum Processes
U.S.A.	U.S. Department of Energy (DOE) <ul style="list-style-type: none"> • Office of Civilian Radioactive Waste Management (OCRWM), Office of National Transportation, RW-30E • National Nuclear Security Agency (NNSA), Office of International Safeguards, NA-243
U.S.A.	U.S. Nuclear Regulatory Commission (NRC) <ul style="list-style-type: none"> • Nuclear Security and Incidence Response (NSIR) • Nuclear Regulatory Research (RES)
U.S.A.	Argonne National Laboratory (ANL), Energy Technology Division
Germany	<ul style="list-style-type: none"> • Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) • Bundesministerium für Umwelt Naturschutz und Reaktorsicherheit (BMU, Federal Ministry for the Environment, Nature Conservation and Nuclear Safety) • Fraunhofer Institut für Toxikologie und Experimentelle Medizin (ITEM)
France	Institut de Radioprotection et de Surete Nucleaire (IRSN)
UK	Office for Civil Nuclear Security (OCNS)

(* designated as Center 6400, in 2003)

2. GOALS AND OBJECTIVES

The primary goal of this surrogate/spent fuel aerosol ratio test program is to conduct experiments and supporting analyses to measure the aerosol, primarily respirable, particles produced from sabotage by a high-energy density device, HEDD, on spent fuel rods in a transport or storage casks. The anticipated product of this program is accurate data that verify and *significantly extend* prior broad estimates of the ratio between the amounts of aerosols generated from actual spent fuel versus surrogate materials when struck by an HEDD. Subsequent more accurate risk analyses could provide a better estimate of the hazards and potentially result in safer and significantly less expensive transportation and storage of the spent nuclear fuel.

Several different types of surrogate and actual spent fuel materials will be included in this test program and will be described in detail in this report. Experimental equipment and test measurement techniques described are being developed and optimized with the use of non-radioactive surrogate materials. A number of tests with actual, highly radioactive spent fuel must be conducted to obtain the required data; however, the number of these tests must be limited due to safety and expense constraints. The nuclear safety and facility requirements mandated for testing actual spent fuel and aerosolizing or “particulating” it with the explosive HEDD jet will be described later in this document.

The main objectives of the current test program include:

1. To provide reliable information for overall radiological consequence assessments for transportation sabotage scenarios, in support of the test and analysis activities of the WGSTSC members;
2. To provide technology transfers and support to NRC, IRSN, GRS, and OCNS vulnerability studies, by providing data and analyses for computer modeling of HEDD attacks against nuclear materials;
3. To support DOE and non-US participants National Authorities’ assessments of the physical protection requirements for nuclear materials in use, storage, and transport.

Information developed in this program may be used to guide development of future transportation security plans. This program also complements efforts to build and maintain strong collaborative relationships with our international partners, to counter nuclear terrorism activities. The data obtained will be shared with all participating WGSTSC partners. These objectives may be expanded in the future.

3. BACKGROUND

Significant prior work has been conducted in the U.S., Germany, and France to assess the potential impacts of hypothetical sabotage events on spent fuel casks, and to develop source terms for aerosol materials created as a result of HEDD impacts [Schmidt et al., 1981; Sandoval et al., 1983; Lange et al., 1994; Luna et al., 1999; U.S. DOE, 1999; Autrusson et al., 2003]. Philbin [Philbin et al., 2002a] has provided a good summarization of the various sabotage studies and aerosol experiments that have led up to the conduct of the current test program. Philbin [Philbin et al., 2002a] also provided a brief overview of the evaluations of the aerosol test results and documented the need for future confirmatory experiments, compared to earlier work, to be performed.

Early U.S. experiments on actual spent fuel and surrogate material [Sandoval et al., 1983; Schmidt et al., 1981] provided some data, though with significant variation, on the relation (ratio) between the amounts of aerosols produced from the actual spent fuel and surrogate materials. These experiments and subsequent analyses [Sandoval et al., 1983; Luna et al., 1999; U.S. DOE, 1999] predicted an aerosol spent fuel ratio, SFR, from HEDD impact events that fell within a range of about 0.5 to 12. This is quite a large spread in values for a parameter that has a direct influence on the predicted consequences of a successful sabotage attack.

In German experiments [Lange et al., 1994], a HEDD was fired into a full-scale, but 1/3-height, transport cask containing nine surrogate DUO₂ fuel assemblies. The total amount of airborne fuel particles with aerodynamic equivalent diameters, AED, < 100 μm, released through the breach formed by the HEDD jet was directly measured and classified aerodynamically in the size range between 0.01 μm and 100 μm AED. These full-scale experiments produced realistic source term data, but due to the use of surrogate DUO₂, significant uncertainties remained concerning proper application of the data to actual spent fuel. Further interpretations of the German surrogate aerosol results could be enhanced with a more precise measurement of the SFR.

It is clear [Luna et al., 2002] that the source term of released radioactive aerosol particles and, hence, any estimate of radiological consequences based on the data, suffer from unsatisfactory knowledge of the correlation of aerosol mass release data between the surrogate materials (unirradiated depleted UO₂) and actual spent UO₂ fuel. In addition, there is insufficient knowledge of the importance of enhanced release and sorption of volatile fission product elements; this is termed enrichment or fractionation. When fractionation occurs and the more volatile nuclides are found in the finest aerosol fractions (rather than equally dispersed), enrichment is said to have occurred. Enrichment was observed, but not well quantified, in both Battelle Columbus Laboratory and Idaho National Engineering and Environment Laboratory studies conducted in the early 1980s. [Alvarez et al., 1982; Schmidt et al., 1981]

The release of fine particles from a shipping cask after shaped charge impact is determined on the one hand by the initial “source term”, i.e. the “dust” generated inside the cask, and on the other hand by the “transport term,” characterizing the transport of airborne material from inside the cask to the outside environment. [Luna et al., 2001] Since the transport term is essentially independent of the type of fuel pellets used, the measurement of the source term for the surrogates and for the spent fuel, under the same transport conditions, would allow determination of the SFR as a function of particle diameter ranges.

4. EXPERIMENT DESCRIPTION AND DETAIL

4.1 Data Needs

The experimental program to be described is designed to measure two important features of the interaction of a HEDD (conical shaped charge, CSC) jet with spent fuel or surrogate material pellets contained within a Zircaloy™ cladding tube:

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

$$\text{SFR} = [\text{spent fuel aerosol particle masses}] / [\text{“surrogate” aerosol particle masses}]$$

2. The enrichment of volatile fission product nuclides like cesium and ruthenium, preferentially sorbed onto specific, respirable particle size fractions. Fission product “enrichment” has been referred to previously as “fractionation.”

The SFR determination involves, essentially, the comparison of the aerosol particle data from irradiated fuel to unirradiated fuel, obtained in paired experiments using the same apparatus, identical test conditions, and with the same HEDD. This is necessary in order to make appropriate correlations to prior experiments conducted primarily with depleted uranium oxide targets.

The aerosol testing requires sampling and measurement of the mass 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 and thoracic fractions, < 10 μm AED. The coarser aerosol particle range of 10 to 100 μm AED is of interest primarily for radiological ground-shine (dispersion, soil contamination) estimates. Multistage aerodynamic particle sizing devices are used to classify aerosolized particles according to their aerodynamic diameter. A corresponding concept has been developed by ITEM/GRS and was extensively used to characterize the formation of airborne particulates upon transient energy input into various types of brittle materials. Refer to further detail on aerosol apparatus and experimental set-up in Section 5.3, plus supporting data on brittle materials in Section 7.1. SFR values will be calculated for multiple, specific aerosol particle size ranges up to about 10 μm and for one or two specific size ranges between 10 and 100 μm AED.

A 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 fraction 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 test apparatus and test conditions. 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 AED is defined by means of the settling velocity of a unit density sphere, and, for spherical particles, is equivalent to: $(\text{particle geometric-diameter}) * (\text{particle density})^{1/2}$ [Hinds, 1999]. It is expected that the measure of particle diameter will be the AED rather than the particle physical or geometric size diameter. This acknowledges that most aerosol particle collection instruments expected to be used are calibrated for particles of unit density (water). In addition, it is simpler to use the AED parameter since it also accounts for particle shape as well as density [Luna et al., 2002].

The “surrogate” material for determining the SFR will be unirradiated, depleted uranium oxide pellets (very slightly radioactive, naturally). Other surrogate materials are also being used initially, specifically non-radioactive cerium oxide sintered ceramic pellets. It is much easier, safer, and far less expensive to conduct as many tests as possible using non-radioactive, then slightly radioactive surrogates.

4.2 Experimental Concept and Test Plan Design

The original spent fuel sabotage - aerosol ratio experiment concept was developed in earlier work by the WGSTSC and is contained in a document entitled “Joint GRS/SNL Proposal to Delineate the Ratio of Spent Fuel to Surrogate Aerosol Generation for More Accurate Prediction of Sabotage Consequences” [GRS/SNL, 2000]. The original, representative test matrix from this document is reproduced in Table 2, primarily to show how the current test program design has evolved and expanded.

Table 2. Original Representative Test Matrix [GRS/SNL, 2000]

Phase/ Test	Target Material	Number of Rod Targets	CSC* Used	Jet Tip Speed (10 ³ m/s)	Comments/Notes
1 / 1, 2,...	Glass / DUO ₂	1, 3 or 5	CSC1	≈ 9	Checkout and shakedown tests
2 / 1	DUO ₂	1	CSC1	≈ 9	
2 / 2	DUO ₂	1	CSC1	≈ 9	Duplicate for comparison to 2/1
2 / 3	DUO ₂	1	CSC2	≈ 9	Same tip speed as CSC1, but with $d_i / d_p = <0.2$ as goal
2 / 4	DUO ₂	5	CSC1	≈ 9	To look at aerosol from collateral effects on adjoining rods
3 / 1	Spent Fuel	1	CSC1	≈ 9	
3 / 2	Spent Fuel	1	CSC1	≈ 9	Duplicate for comparison to Experiment No. 3/1
3 / 3	Spent Fuel	1	CSC2	≈ 9	Analogously to Experiment No. 2 / 3 (If funding is available)

* Conical Shaped Charge - CSC1 is a specific CSC developed in early prior experiments – CSC2 has less explosive and smaller jet diameter. NOTE: CSC1 is the only HEDD that has been used in the following tests.

The current, overall program consists of four linked test phases, to be conducted in a sequential, cost-effective, and safe manner. Individual tests in each phase will use the same type of HEDD, CSC1 (in Table 2), but different test materials, with a similar geometry. Each individual test and test phase helps to “calibrate” or optimize the succeeding test phases, allowing us to fine-tune the test system and individual components, while providing an indication of anticipated system response and results. Successive phase testing allows us to add and evaluate multiple test variables and pellet response to HEDD jets.

Phase 1: The preliminary Phase 1 tests were conducted by Sandia National Laboratories, SNL, and Fraunhofer ITEM, using glass pellets and leaded glass plates as representative brittle materials. Six of these tests were performed in 2002, two with glass pellets contained in a Zircaloy cladding tube, four with leaded glass plates; leaded glass was selected because it has a higher density, closer to that of uranium oxide fuel. These glass brittle materials were impacted by a HEDD jet using the same test apparatus to be described for the Phase 2 tests. This test phase in-

cluded performance quantifications of the HEDD devices and refinement of the aerosol particle collection apparatus being used. Phase 1 test conduct was completed in 2002. Details of test Phase 1 design, equipment, and results will be documented separately [Molecke, Yoshimura, and Vigil, 2004].

Phase 2: The more extensive Phase 2 tests use nonradioactive cerium oxide, CeO_2 , in sintered ceramic pellets contained within a Zircaloy cladding tube assembly. CeO_2 is an excellent chemical “surrogate” material for both UO_2 fuel material and PuO_2 , and a representative ceramic for spent fuel UO_2 pellets. The cerium oxide has a density and melting point similar to those values of uranium dioxide, certainly more similar than those of glass. The Phase 2 test plan matrix is listed in Table 3, following. Comprehensive Phase 2 test component details will be discussed in Section 5. The Phase 2 surrogate tests allow us to evaluate multiple test variables, pellet and rod responses to HEDD jet impacts, and to fine-tune the experimental setup. The use of cerium oxide pellets is intended to bridge the program from the initial Phase 1 tests with glass, to a lesser number of advanced, slightly radioactive tests with DUO_2 pellets, Phase 3, to the fully radioactive tests with actual spent fuel pellets, Phase 4. The results and test experience gained in Phase 2 tests will be used to define and refine test apparatus used in later phases. Phase 2 test results should also allow us to anticipate or “calibrate” the results to be obtained from Phase 3 tests. The Phase 2 tests were initiated in September 2002 and are anticipated to be completed by July 2004.

As such, the goal of the overall test program with the surrogate materials is to compare and “calibrate” the subsequent Phase 3 DUO_2 tests and results with the more extensive data obtained from the Phase 2 cerium oxide pellet tests. Nonradioactive volatile fission product (species) enrichment measurements will also be performed. If the data correlations are judged adequate, we then can move on to the Phase 4 tests with actual spent fuel. If the correlation is not adequate, several more Phase 2 or Phase 3 tests can be added to the test matrix, to determine why the correlation is lacking, and to evaluate more tests variables, as needed.

Phase 3: The Phase 3 tests will use slightly radioactive, depleted uranium oxide, DUO_2 , pellets in comparable size, new Zircaloy cladding tube test rods; six of these tests are planned in 2004. Phase 3 test conduct will also allow us to complete the design, fabrication, and safety testing of the “total containment system” apparatus, primarily the explosive containment vessel and the aerosol collection chamber that will also be used for the Phase 4 tests with actual spent fuel. The Phase 3 test plan matrix is listed in Table 4, following; test component details will be discussed in Section 6. It is anticipated that Phase 3 tests will start in the July to September 2004 time period and should be completed within about six months.

Phase 4: The Phase 4 tests will use fully radioactive, actual spent fuel pellets in short test rodlets; eight of these tests are anticipated for conduct in 2005. Two types of spent fuel will be included in the test program. The first is high burnup, ~ 72 GWd/MTU (Gigawatt days per metric tonne of uranium) spent fuel, originating from the H.B. Robinson pressurized water reactor. The second is low(er) burnup, ~ 36 GWd/MTU spent fuel, originating from the Surry pressurized water reactor. Both fuels are being characterized and fabricated into test rodlets at Argonne National Laboratory, ANL, for HEDD-impact aerosol testing in the Gamma Irradiation Facility (GIF) at Sandia National Laboratories. Detailed plans for conduct of both the Phase 3 and Phase 4 spent fuel tests at SNL are in advanced stages of development.

Sandia National Laboratories [Philbin et al., 2002b] previously conducted and documented a finding that such an experimental program using highly radioactive spent fuel and explosives

(HEDD) could, potentially, be safely conducted within an available Sandia nuclear facility. The Phase 4 test plan matrix is listed in Table 5. Phase 4 test and component details are discussed in Section 6. It is anticipated that the Phase 4 tests will be performed during 2005.

Table 3. Phase 2 Tests: CeO₂ Surrogate Test Matrix

Test Phase 2: Cerium Oxide Surrogate Pellets/Rods				
Test #	Pressure	Dopants	Variables *	Date *
0	1 bar	no	top	10/2002
1A, 1B	1	no	top, center	10/2002
2A, 2B	1	no	French pellet & tube size	12/2002
3A, 3B	1	no	U.S. pellet & tube size, Respicon & Berner	7/2003
4A, 4B	1	yes	" + equipment design mods.	8/2003
5A – 5G	1	yes	" + vertical test chamber, instruments, Marple particle impactors.	9-12/2003
6A, 6B	40 blowdown	yes	" + equipment design modifications, Marple particle impactors	2004
7A, 7B	1	yes	German HLW glass rod, dopants (nonradioactive)	2/2004
8A – 8D	1	yes	particle impactors & sampling optim.	2004
9A – 9D ...	1	no	Total Containment System checkouts, N ₂ , @ SNL ECF, then GIF	2004

results included in this document
 ↑↑↑↑

(* HEDD used in all tests = CSC1. Dates are subject to future revisions)

Table 4. Phase 3 Tests: Advanced DUO₂ Surrogate Test Matrix

Test Phase 3: Depleted Uranium Oxide Pellets/Rods			
Test #	Pressure	Dopant	Variables *
3/1	1 bar	yes	Air (in aerosol chamber)
3/2	1	no	Air
3/3	1	yes	N ₂
3/4	40 (He)	yes	Air
3/5	40	no	Air
3/6	40	yes	N ₂
Conducted in SNL TA-V: GIF Cell 3, start < 9/2004 *			

(* subject to future revisions)

Table 5. Phase 4 Tests: Actual Spent Fuel Test Matrix

Test Phase 4: Actual Spent Fuel (PWR) Rodlets		
Test #	Pressure	Variables *
4/1	~ 40 bar	Robinson, high-burnup, ~72 GWd/MTU
4/2	~ 40 (He)	Air (in aerosol chamber)
4/3	~ 40	N ₂
4/4	~ 40	N ₂
4/5	~ 28 bar	Surry, low-burnup, ~38 GWd/MTU
4/6	~ 28 (He)	Air
4/7	~ 28	N ₂
4/8	~ 28	N ₂
Conducted in SNL TA V: GIF Cell 3, in 2005 *		

(* subject to future revisions)

5. PHASE 2 TEST COMPONENT 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); aerosol collection chamber or box; a conical shape charge, the HEDD; aerosol particle samplers (particle impactors, sampling tubes, pumps, etc.); explosive containment chamber (primarily for Phase 3 and 4 tests); 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. As such, detailed description of individual test components will be provided by test phase progression. This section describes experimental detail for test Phase 2 components, predominantly.

The overall test component setup for Tests 2/0, 2/1A and 2/1B, as well as many of the Phase 1 tests, is shown in Figure 1. These tests were conducted at the Sandia Explosive Components Facility, ECF, Building 905, in a contained explosive pad (room). The aerosol collection box, with a test rod and pellets inside, is shown at the center. The HEDD-jet stop block is shown mounted at the left, and the HEDD is located at right, mounted on a blast shield. The aerosol collection particle samplers (not visible) are mounted below the aerosol collection box. The four large tubes at front are flash X-ray diagnostic tubes; these were only used in the earlier tests, to observe the performance behavior of the HEDD jet.



Figure 1. Phase 2 Surrogate – HEDD Impact Aerosol Test Setup, 2/0, 2/1A & 2/1B

Figure 2 illustrates the overall test component setup for more recent Tests 2/4A and 2/4B. These tests were conducted at a remote, outdoor location of the Sandia Explosive Components Facility, at the Terminal Ballistics Facility or “Gun Site,” near Sandia Building 6750. The aerosol collection box, with a test rod and pellets inside, is shown at the center. The HEDD-jet stop block is shown mounted at the right, and the HEDD is located at left, mounted inside of a cylindrical blast shield. Several aerosol particle impactor/collector devices and associated vacuum hoses and pumps, are shown located below the aerosol collection box.



Figure 2. Phase 2 Surrogate – HEDD Impact Aerosol Test Setup, 2/4A & 2/4B, at Sandia ECF Gun Site

5.1 Surrogate Cerium Oxide Test Rods

The Phase 2 test rodlets consist of multiple cerium oxide surrogate, sintered ceramic test pellets contained within a Zircaloy-4 cladding tube. Cerium oxide was selected for use as a surrogate “spent fuel” pellet material because it has the following beneficial properties:

1. Primarily, it is chemically similar to, and representative of uranium oxide.
2. Cerium oxide is non-radioactive.
3. It can be fabricated into ceramic pellets by pressing and sintering, similarly to a uranium oxide fuel pellet.
4. Because of its representative brittle, ceramic (pellet) nature, it is expected to fracture under HEDD jet-impact conditions into aerosolized and respirable particles similarly to UO_2 and DUO_2 ceramic pellets.
5. Because of the chemical similarity of cerium oxide to UO_2 , it can be used for fission product enrichment/concentration testing. This was not possible for glass targets in Phase 1.

Cerium, a lanthanide element, is quite similar chemically, i.e., a good surrogate or homologue to the chemistry of uranium and plutonium, both actinide elements. Cerium has multiple oxidation states (+3, +4), similar to those of uranium and plutonium (+3, +4, +5, and +6). Cerium oxide, CeO_2 , (also termed ceria or ceric oxide) is also an oxide quite chemically similar to the UO_2 in nuclear fuel -- and the DUO_2 in the Phase 3 advanced surrogate tests; CeO_2 and UO_2 have the same ionic crystal, fluorite-type structure. For these reasons, CeO_2 has been used in multiple decontamination studies performed at Sandia National Laboratories [Molecke, 1999; Melgaard et al., 2003; Van Den Avyle et al., 2003].

Cerium oxide is commercially available in powder form, relatively inexpensive, and has a low hazards identification rating (Material Safety Data Sheet, MSDS). Cerium oxide has a theoretical density of about 7.13 g/cc compared to about 10.96 g/cc for UO_2 . For comparison, silica

glass (quartz) has a density of about 2.6 g/cc. Uranium oxide fuel pellets are commonly fabricated to about 95% of theoretical density, or 10.41 g/cc.

Cerium oxide is a refractory oxide with a very high melting point, approximately 2600 °C, compared to 2878 °C for UO₂. CeO₂ is an oxide ceramic like UO₂, and has similar physical properties (e.g., elastic moduli and Poisson's ratio, plus other thermal and mechanical properties), that have been comprehensively assessed and documented in the literature. From a shock physics viewpoint [Harper, 2004], the material properties important to shock aerosolization in our explosive, HEDD jet-impact tests, e.g., bulk modulus, bulk speed of sound, fracture toughness, and strength, compare reasonably well for both CeO₂ and UO₂. Therefore, we can conclude that cerium oxide is a good surrogate for uranium oxide (fuel pellets) from chemical, thermal, and physical or mechanical points of view, and an adequate surrogate from a shock physics perspective.

5.1.1 Cerium Oxide Pellet Fabrication

Cerium oxide powder has been pressed and sintered into ceramic pellets for our testing purposes, by the Ceramic Synthesis and Processing, Department 1843, at Sandia National Laboratories. The cerium oxide powder (99.9 % pure, about 2 μm grain size) was mixed with about 3 wt. % organic material binder, mechanically screened, then uniaxially dry pressed in a metal die (at ~200 MPa, ~29 kpsi, for the 7 mm-long pellets, Table 6) into “green” pellets, fired at about 600 °C for binder burnout, and then sintered at about 1600 °C [Ewsuk and Diantonio, 2002]. Measurements of apparent pellet porosity and Archimedes density are then made. The pellets were made to fit snugly (i.e., with minimal pellet-to-cladding gap) into Zircaloy 4 cladding tubes. Measured cerium oxide pellet specifications are presented in Table 6.

Table 6. Cerium Oxide Surrogate Pellet Specifications, as Fabricated

Test #	Ave. Weight	Average Theoretical Density	Average Diameter	Average Height	# Pellets per Rod	Comments
2/0, 2/1A, 2/1B	4.91 g	85.0 % 6.11 g/cc	8.65 mm	13.9 mm	6, 6, 5	initial batch for testing
2/2A, 2/2B	4.38 g	87.4 % 6.23 g/cc	8.20 mm	13.3 mm	5	sized to match French spent fuel
2/3A, 2/3B 2/4A, 2/4B	3.14 g	95.4 % 6.80 g/cc	9.17 mm	7.0 mm	9	sized to match H.B. Robinson U.S. spent fuel

5.1.2 Zircaloy Cladding Tubes

We have used Zircaloy 4 cladding tubes obtained from several sources, and of several different sizes. The dimensions and sources are summarized in Table 7. We purchased cladding tube material from Framatome ANP Richland, Inc. Richland, WA (both 9.55 mm and 10.6 mm outside diameter. This tubing was manufactured by ANF, Advanced Nuclear Fuels, GMBH, Duisburg, Germany. We anticipate using the 10.6 mm outside diameter tube for all future tests (in Phase 2 and Phase 3), because it is the closest in diameter currently available to both H.B. Robinson and Surry U.S. PWR, pressurized water reactor, spent fuels to be tested in Phase 4.

Figure 3 illustrates cerium oxide pellets contained within a Zircaloy-4 cladding tube; five 13.3 mm-long pellets are shown with a 10.6 mm outside diameter tube, held in position by copper rods. Figure 4 illustrates nine 7.0 mm-long pellets (shown standing at right angle to the tube) with a similar 10.6 mm outside diameter tube.

Table 7. Zircaloy-4 Cladding Tubes

Test #	Outside Diameter	Inside Diameter	Source	Comments
2/0	10.6 mm	9.32 mm	Not known/ Framatome ANP ?	Initial test
2/1A, 2/1B	10.0 mm	8.8 mm	Fraunhofer ITEM	Minimize pellet-clad gap
2/2A, 2/2B	9.55 mm	8.33 mm	Framatome ANP	Sized to match French PWR spent fuel
2/3A, 2/3B 2/4A, 2/4B & future 2/#	10.6 mm	9.32 mm	Framatome ANP	Sized to match H.B. Robinson U.S. spent fuel
Phase 3	10.6 mm	9.32 mm	Framatome ANP	
Phase 4	10.77 mm	9.25 mm	Westinghouse; ANL	H.B. Robinson spent fuel
Phase 4	10.72 mm	9.48 mm	Westinghouse; ANL	Surry U.S. spent fuel

Figure 3 illustrates cerium oxide pellets contained within a Zircaloy-4 cladding tube; five 13.3 mm-long pellets are shown with a 10.6 mm outside diameter tube, held in position by copper rods. Figure 4 illustrates nine 7.0 mm-long pellets (shown standing at right angle to the tube) with a similar 10.6 mm outside diameter tube.



Figure 3. Five CeO₂ pellets in tube



Figure 4. Nine CeO₂ pellets in tube

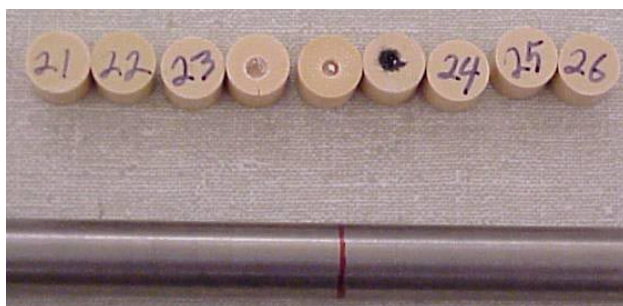


Figure 5. Fission product-doped cerium oxide pellets, Test 2/4A

dopants: ↑ ↑ ↑
 [SrO] [CsI] [RuO₂]

5.1.3 Fission Product Dopant Disks

One of the major goals of this overall experimental program is to quantify the potential enrichment of volatile fission product nuclides on respirable-size particulates produced from spent fuel–HEDD jet impact. Volatilized species of cesium (^{134}Cs , ^{137}Cs) and ruthenium (^{106}Ru) have been mentioned as most significant. Non-radioactive chemical forms of cesium and ruthenium have been added to the surrogate pellet test systems, starting with tests 2/4A and 2/4B. We have chosen cesium iodide, CsI, and ruthenium oxide, RuO_2 (anhydrous), for testing expediency, not because the fission product will be in these forms in actual spent fuel. The iodide species will also be representative of volatile fission product ^{129}I . Strontium is another major fission product species, ^{90}Sr , but it is not easily volatilized. It has also been added to this test system as strontium oxide, SrO, as a non-volatile “standard” fission product dopant, to be compared to the volatile Cs, Ru, and I species distribution for enrichment determinations. The boiling point, vaporization temperatures for CsI, RuO_2 , and SrO are 1280 °C, 1200 °C, and ~ 3000 °C, respectively. In the future, europium oxide, as Eu_2O_3 , will also be added and tested.

The initial fission product dopant samples were prepared by inserting the solid dopant chemicals into small “wells” pre-drilled in one end (prior to sintering) into the cerium oxide pellets, one chemical per pellet. Each doped pellet contain approximately 1000 ppm (0.1 wt %) of stable Cs, I, Ru, or Sr species, relative to the mass of the surrogate oxide pellet expected to be disrupted per test. The solid chemicals were held in place with a drop of super glue. Prior to HEDD impact, the dopant chemicals were not subjected to elevated temperatures so there would be no thermal volatilization. These doped pellets are illustrated in Figure 5, showing the pellets used in test 2/4A; the HEDD jet was centered on the center pellet, containing the CsI dopant. The actual weights of each fission product dopant chemical are listed in Table 8.

Table 8. Fission Product Dopant Chemicals

Phase 2 Test #	Cesium Iodide, CsI	Ruthenium Dioxide, RuO_2	Strontium Oxide, SrO
2/4A	32.6 mg	20.3 mg	18.7 mg
2/4B	30.4 mg	22.5 mg	19.2 mg

NOTE: A different technique for fabricating stand-alone fission product dopant disks, without the cerium oxide pellet “holders” is required for use in Phase 3 tests with DUO_2 pellets. Developmental testing of a suitable preparation technique (thin, resin base with solid chemicals embedded) is in process, and will be described at a later time. Sandia has agreed to prepare and provide these non-radioactive fission product dopant disks to the WGSTSC test partners.

All fission product dopant material in each test is expected to be 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 particulate materials. It is possible that the “cooled” fission product species will preferentially sorb onto the smaller surrogate/fuel aerosol particles, because the smaller particles have a higher surface area/mass ratio than larger particles. The aerosol particles will be collected selectively by size fraction using an aerosol particle sampler, then be chemically analyzed for each species by instrumental chemical techniques. Inductively coupled plasma/mass spectrometry, ICP-MS, analyses of the non-radioactive fission product dopant species are planned.

5.1.4 Other Phase 2 Surrogate Test Rods

With WGSTSC members agreement and cooperation, *non-radioactive* German high-level waste (HLW) glass test rods, containing multiple, non-radioactive fission product dopants, will be added to, and tested in Phase 2. Refer to the test matrix in Table 3. These tests with HLW glass can be considered as an extension of prior Phase 1 tests on brittle materials, with goals parallel to those of other Phase 2 tests. Several of these glass surrogate test rods, in stainless steel tubes, about 1.4 cm in diameter and 16 cm-long, will be supplied to Sandia by the GRS and Fraunhofer ITEM for aerosol-HEDD testing. This non-radioactive glass with fission product simulants originated at the German Karlsruhe Nuclear Research Center. Fraunhofer ITEM personnel will interactively participate in the HEDD-aerosol testing of the glass rods at Sandia. The post-test collected aerosol particle materials will be sent to Fraunhofer for detailed analyses. Fraunhofer will also conduct parallel, non-HEDD aerosol tests on this high-level waste glass material, in Germany. These surrogate HLW glass rod aerosol tests should be beneficial in providing additional data on volatile fission product enhanced sorption onto respirable particles.

5.2 Aerosol Collection Chamber and Associated Equipment

Figure 1 shows the non-ventilated, aerosol collection chamber box, 30 x 40 x 50 cm in size (width x length x height), 12 x 16 x 20 inch, used for all Phase 1 tests (with glass) and Phase 2 tests 2/0, 2/1A, 2/1B, 2/2A, and 2/2B. This box was fabricated out of welded aluminum plates, 1.2 cm-thick, and had two Lexan viewing windows, 1.6 cm-thick. These windows could be opened (un-bolted) for pretest rod-target insertion and post-test rod and residual particulate sampling. There is a small, open hole, about 1.9 cm-diameter, in both the front and back walls, to allow the HEDD jet to enter and exit (after penetrating through the surrogate pellet rod target), before being stopped in the adjacent HEDD jet stop block. At the bottom, mid-point of the box, there are two L-shaped sampling tubes leading to the two particle collectors below, to be described. These non-ventilated aerosol collection chambers were used for reasons of simplicity and durability. Because there is no laminar flow of gas through this chamber, only particles that remain airborne can be collected and classified aerodynamically. Thus, only particles generated by the test that have an AED of less than about 16 μm can be analyzed accurately. Aerosol particles larger than about 30 μm AED settle out within seconds of formation, and are only incompletely sampled.

The similar, but somewhat modified aerosol collection box shown in Figure 2, is 30 x 60 x 43 cm in size (width x length x height) and was used for Phase 2 tests 2/3A, 2/3B, 2/4A, and 2/4B. This box had larger, longer viewing windows, and three sampling tubes at the bottom, leading to three aerosol particle samplers. It also had closure valves, shown in Figure 6 for tests 2/3A and 2/3B, that sealed the HEDD-jet entrance and exit holes about 1 second after the HEDD explosion. This valve sealing was intended to minimize the loss of residual aerosols (remaining in the box) for post-test sampling. This box was slightly modified again for tests 2/4A and 2/4B, as shown in Figure 2; the exit hole closure valve was removed and the HEDD stop block was connected directly to the back side of the aerosol chamber. This was an additional measure to minimize loss of aerosol particulates formed by the HEDD jet impact from escaping through the exit hole.

The aerosol collection box design has evolved in order to provide improvements in testing and to bring it stepwise closer in design to the “total containment system” to be used and described for test Phases 3 and 4. Future Phase 2 aerosol collection chambers will be cylindrical in shape, as described for requirements in Section 6.4.1. Further aerosol chamber design details and testing results will be documented in the future.

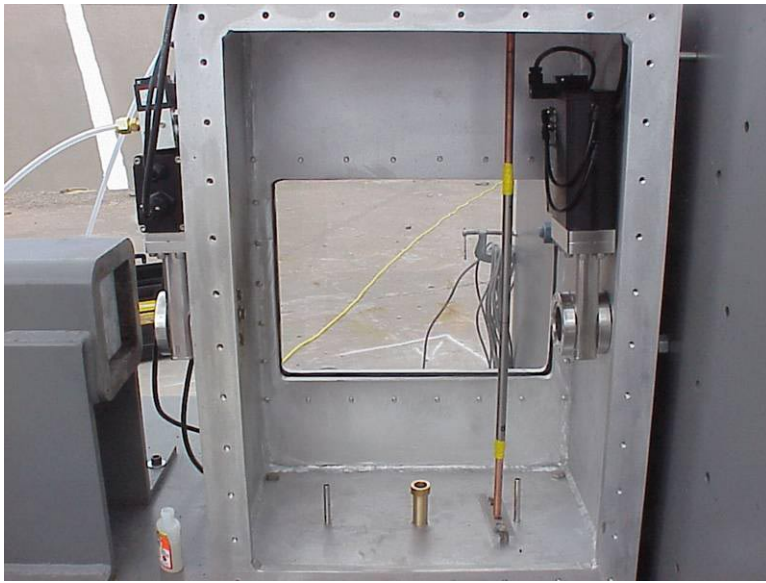


Figure 6.
Aerosol Collection Box,
Phase 2 Tests 2/3A and 2/3B
(showing entrance and exit
hole-closure valves)

5.3 Aerosol Particle Samplers

We have used two types of aerosol particle samplers to date, the Respicon™ 3-stage virtual particle impactor (two per test), and the Berner 9-stage particle impactor. The Respicon particle sampler was designed by, and recommended for use by Koch [Koch, et al., 1999]. It is commercially manufactured by TSI Incorporated, Shoreview, MN, and has three aerosol size particle collector stages: the top stage collects the *respirable* particle fraction of ~ 0 to $4 \mu\text{m}$ AED; the middle stage collects the *respirable/thoracic* sub-fraction of ~ 4 to $10 \mu\text{m}$ AED; and, the bottom stage collects the *inhalable* fraction of ~ 10 to about $100 \mu\text{m}$ AED. As described, the *inhalable* fraction can not be collected quantitatively in the non-ventilated, non-flow-through aerosol collection chamber. The Respicon particle samplers require the use of a vacuum pump equipped with a throttle valve that can draw 3.1 L/min through the samplers. The vacuum pumps are flow calibrated at each test, using a BIOS Dry-Cal DC-Lite Primary Flow Meter. The Respicon aerosol particle sampler is illustrated schematically in Figure 7 [TSI Incorporated, Shoreview, MN] and two are shown in Figure 8.

The Berner aerosol particle impactor, manufactured by Hauke G.m.b.h, Austria, was recommended by, and lent to Sandia, by Fraunhofer ITEM. It is the Berner AESAR™ Hauke model Type LPI 30/0,06, with a volume flow rate of 30 L/min , and a particle measuring range of $0.08 \mu\text{m}$ to $16 \mu\text{m}$, in nine stages. Associated equipment and supplies required to operate the Berner impactor include: a vacuum pump with flow rate of $16 \text{ m}^3/\text{h}$ minimum at $p=100 \text{ mbar}$; a make-up air supply input (“aquarium pump”) at an adjustable pressure ($0\text{-}3 \text{ bar}$) and pressure gauge, to limit (dilute) the flow to the Berner to approximately 5.6 L/min ; a 17 mm sampling probe (visible at bottom center of Figure 6) with calibrated critical orifice; assorted hoses, t-connector, and bushings; aluminum foil for sampling stages; and, silicone oil spray to prevent particle bounce on aluminum (catcher) foils. The flow rate through the Berner was checked with a BIOS (Butler, NJ) Dry-Cal DC-Lite Primary Flow Meter. The Berner impactor is illustrated schematically in Figure 9 and pictured in Figure 8; it is also visible, hooked up, below the aerosol collection box, in Figure 2.

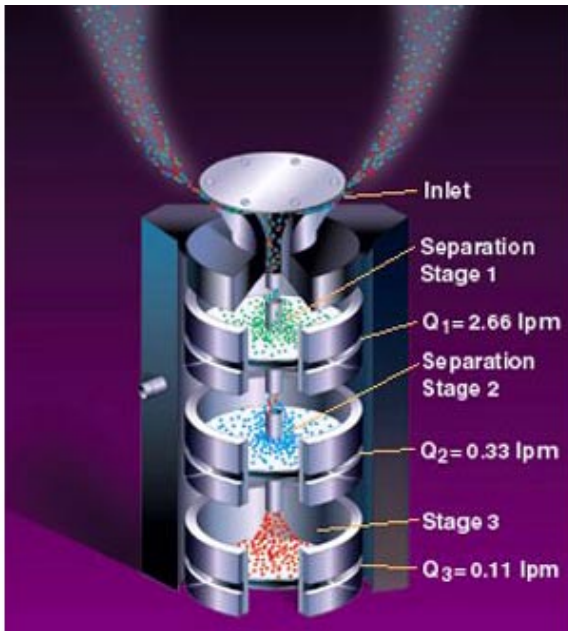


Figure 7. Respicon Virtual Particle Impactor, Schematic



Figure 8. Aerosol Particle Samplers: Respicon Virtual Impactors (left) Berner Particle Impactor (right)

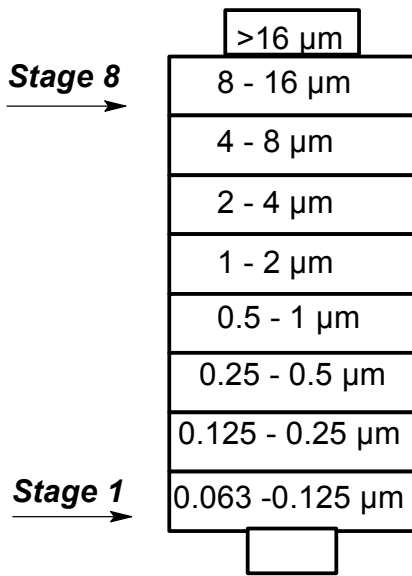


Figure 9. Berner Impactor Schematic and Particle Size Stages



Figure 10. Marple Cascade Impactor 8-stages, 0.4 – 21 μm AED

In the future conduct of Phase 2, 3, and 4 tests, a new, smaller aerosol particle impactor will be used, the multi-jet Marple Cascade Impactor (model 298), shown in Figure 10, to replace both the Respicons and the Berner particle samplers. Several commercially available aerosol particle collection devices were evaluated, based on recommendations from both Fraunhofer ITEM and Sandia National Laboratories aerosol and testing experts. We considered factors of sampler size and weight vs. remote manipulator ease of handling; means for attaching them to the aerosol col-

lection chamber, hardware issues, sampling and decontamination issues, etc.; desire for a low flow rate (Berner = 30 L/min, Respicon = 3 L/min, Marple = 2 L/min; lower is better); and, existing experience in use. Based on this evaluation process, the new aerosol sampler will be the Marple Cascade Impactor, with multiple (eight) stages providing complete and accurate aerodynamic particle size distributions from 0.4 to 21 micrometers AED. A final “back-up” filter collects all aerosol analytes smaller than 0.4 micrometers. We anticipate using about four Marple Cascade Impactors per test. Further evaluation tests will be conducted in the near future on the ease of use and sampling for the Marple devices.

5.4 HEDD Jet Stop Block

Two variations of the HEDD jet stop block used to date were illustrated in Figures 1 and 2. The internal components of this stop block consist of alternating plates of mild steel and polypropylene, each 1.2 cm-thick. The purpose of these plates is to stop the very energetic HEDD explosive jet, as well as the less energetic residual metallic slug or “carrot,” within a manageable distance; this distance is appreciably less than 30 cm. The polypropylene plates are critical for keeping this stopping distance to a minimum length.

6. TEST PHASES 3 and 4, COMPONENT PLANS and REQUIREMENTS

Since the spent fuel sabotage - aerosol test program Phases 3 and 4 have not been started, this section will describe test component planning, designs, and design concepts to date. The test components will be designed, fabricated, and performance tested as identically as possible for these two test phases, with the exception of the test rodlets. The Phase 3 test rodlets will contain unirradiated, depleted uranium oxide pellets, and Phase 4 test rodlets will be fabricated from actual spent fuel pellets plus original, irradiated cladding tube. The primary components and designs needed for the advanced surrogate and actual spent fuel tests are rods/rodlets, the aerosol collection chamber, the explosive containment vessel, the HEDD jet stop block, and the aerosol particle sampling devices, plus associated hardware.

A parallel list of test component design and performance requirements will also be provided for guidance to test participants. These component requirements are based on previous requirements documented in the Sandia feasibility study for performing this test program [Philbin et al., 2002b], plus modifications and updates based on recent test results and design concepts. Requirements for the test facility (safety, permitting, and related topics), and the post-test storage and disposal casks will also be described (separately).

6.1 Depleted Uranium Oxide Test Rodlets

Six test rodlets containing unirradiated, depleted uranium oxide pellets are required for Phase 3 testing, as listed in Table 4. The IRSN has agreed to fabricate and provide these depleted uranium oxide test rods to Sandia, as previously planned. These DUO₂ rods should have dimensions very similar to the U.S.-origin spent fuel, to allow the experimental determination of spent fuel ratio to be as valid as possible.

Issues associated with the post-test disposal of the French-origin, unirradiated materials have been addressed at Sandia and resolved [Blejwas, 2003]. "Post-test depleted uranium-contaminated hardware and samples generated during ... Phase 3 testing will be managed as low-level radioactive waste in accordance with the Sandia ES&H Manual. Representatives of the test program will prepare the appropriate documentation and submit the waste to the Radioactive Waste and Nuclear Material Disposition Department. This waste will then be eligible for disposal at the Nevada Test Site as part of Sandia waste stream ALSA000000011. The Radioactive Waste and Nuclear Material Disposition Department will be responsible for transportation to the Nevada Test Site for final disposal."

6.1.1 Requirements, Depleted Uranium Oxide Test Rodlets

- a) Six DUO₂ test rodlets need to be fabricated and supplied to Sandia National Laboratories by July 2004 (subject to revision). As listed in Table 4, three of these rodlets will require an internal pressurization of ~ 40 bar, with helium gas, to simulate the approximate pressures found within spent fuel rods. In addition, three of these test rodlets will be assembled with non-radioactive fission product dopant disks, one on each side of the center DUO₂ pellet. Sandia will supply these dopant disks to the IRSN.
- b) Zircaloy 4 cladding tube of 10.6 mm outside diameter, 9.32 mm inside diameter, shall be used; refer to Section 5.1.2. Sandia has agreed to supply this cladding tube to the IRSN.
- c) The DUO₂ pellets used should be as representative of uranium oxide fuel pellets as possible; e.g., theoretical density should be about 95%. There should be five ~ 13.8-mm long DUO₂ pellets per each test rodlet. The pellet diameter must fit within the Zircaloy tubing, e.g., they

should be about 9.1-9.2 mm. [NOTE: Only one DUO₂ pellet length, 13.8 mm, is required. This will be used as the comparison “advanced surrogate” material for both H.B. Robinson and Surry spent fuels. The pellet-length variable will be evaluated in Phase 2 tests only.

- d) The design of the DUO₂ test rodlets should be similar to and based on the spent fuel test rodlet design shown in Figure 11, following, and revisions thereto. The final DUO₂ test rodlet design will be jointly agreed upon by SNL, IRSN, and ANL.
- e) The post-test depleted uranium oxide particles remaining in the aerosol chamber should be manually collected, similar to the collection of post-test cerium oxide particles, weighed, mechanically sieved into size fractions (refer to Section 7.2, particle impact debris), and chemically analyzed for uranium and other metal components.

6.2 Spent Fuel Test Rodlets

The original spent fuel test rodlets were to be supplied to Sandia National Laboratories for testing by IRSN [GRS/SNL, 2000]. The spent fuel was to have been characterized and fabricated into test rodlets at the Commissariat à l’Energie Atomique, CEA, LECSTAR facility, in Cadarache, France. The fuel burnup was 50-58 GWd/MTU, with pellet size of about 8.3 mm diameter by 13.8 mm-long. Four of these test rods, internally pressurized to 40 bar with helium, were to be shipped to Sandia within a NAC LWT transport cask. IRSN provided an advanced preliminary design drawing of the LECSTAR spent fuel test rodlet to SNL in January 2003, for review and modifications appropriate for Sandia GIF handling.

Several outstanding Department of Energy issues were not able to be resolved in regard to the French spent fuel test rodlets. The DOE could not legally nor expeditiously take possession of non-U.S. origin fuel, primarily because no ultimate disposal path (location) was available for the foreign-origin spent fuel post-test waste materials, at a potential repository in the U.S.

6.2.1 High Burnup, H.B. Robinson Spent Fuel Test Rodlets

An alternate path forward was identified for acquiring domestic, U.S.-origin spent fuel, to substitute for the French LECSTAR-provided spent fuel test mock-ups. The new spent fuel material to be used in this test program originated at the H.B. Robinson PWR, Rod R01, and is currently at Argonne National Laboratory, near Chicago. It was supplied to ANL as part of a research program sponsored jointly by NRC, DOE, and EPRI. It is DOE-owned research material, not “commercial” SNF. This spent fuel was removed from the reactor in April 1995 and spent 5 years in wet storage. It has a peak high burnup of about 72 GWd/MTU, a ²³⁵U enrichment of about 2.90 wt. %, and is similar in dimensions to the French spent fuel. Argonne has already conducted some characterization tests on adjacent, sibling rods from this reactor [EPRI, 2001; Tsai and Billone, 2003]. ANL has agreed to fully characterize the spent fuel material (non-destructive and destructive characterization of fuel material and cladding) to be used in the Sandia tests, to fabricate the existing fuel into four test fuel rodlets, and to package and ship the test rodlets to Sandia within a DOE-owned T2, or other cask. This work will be performed under contract to Sandia, with DOE funding.

The test rodlet design proposed by IRSN/CEA has been slightly modified, and is the "starting" basis for the H.B. Robinson fuel test rod fabrication. There were four equivalent spent fuel pellets (~ ½ + 1 + 1 + 1 + ~ ½ pellet) in the French rodlet. There was no assurance that the fuel rod could be sectioned at the exact pellet-pellet interface; if the original rod is cut in the vicinity of a pellet-pellet interface, there is the risk of damage to the pellets due to the embrittlement of the

material. There was an equivalent total pellet (stack) length of ~ 55 mm (2.2 inch) for the French fuel. H.B. Robinson fuel pellets are only ~ 6.9 mm in length, half the length of the French pellets. Therefore, the spent fuel test rodlets made from H.B. Robinson fuel will require eight equivalent pellets (or ~½ + 7 whole + ~ ½ pellets) to achieve a comparable fuel length.

Argonne has provided a new engineering drawing derived from the original French design. This design has been modified as necessary, in coordination with Sandia nuclear facilities personnel, for applicable remote handling capabilities agreeable to Argonne, Sandia, and the IRSN. This spent fuel test rodlet design is illustrated in Figure 11. Specific program requirements for these spent fuel test rodlets are listed in Section 6.2.3, Requirements.

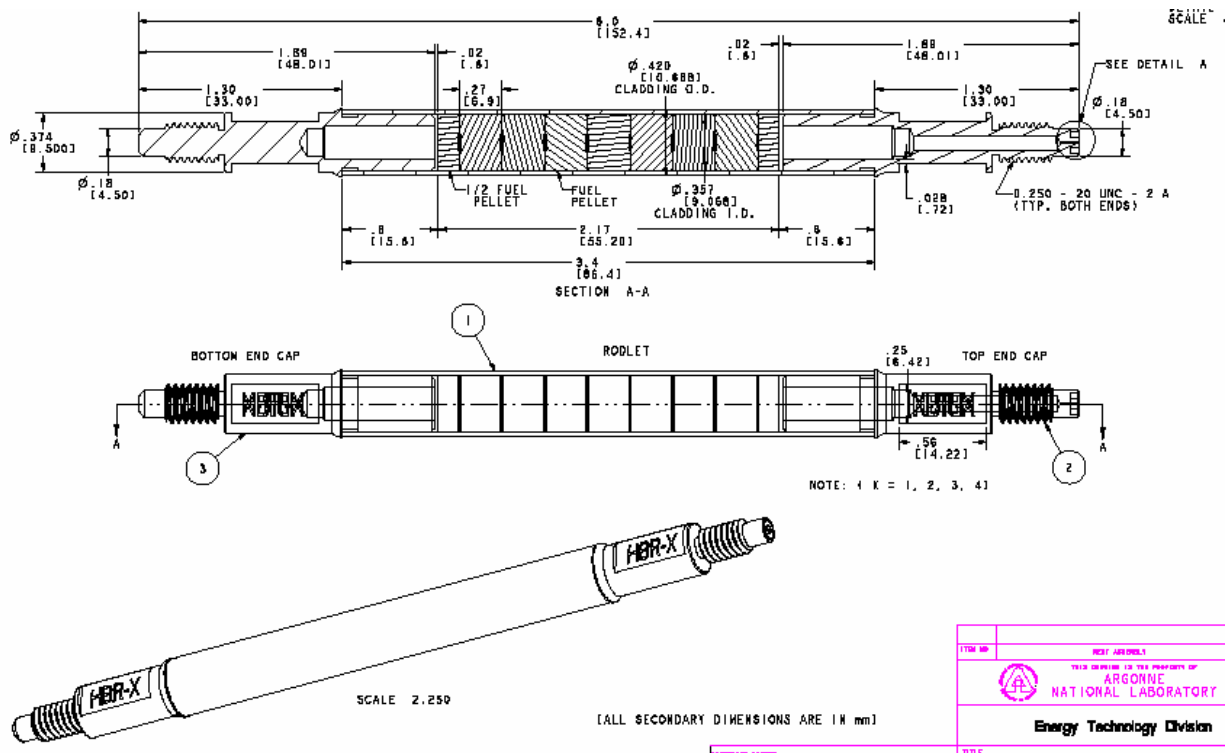


Figure 11. H.B. Robinson Spent Fuel Test Rodlet Design

6.2.2 Low Burnup, Surry Spent Fuel Test Rodlets

A hypothesis has been made that high burnup fuels, e.g., the H.B. Robinson spent fuel, may produce appreciably more aerosol and respirable particles than lower burnup spent fuels, primarily because of more extensive micro-fracturing from extended irradiation time and thermal stresses [Einziger, 2003]. However, there is an opposite hypothesis that the release of respirable particles from *unirradiated* fuel could be measurably *greater* than the release from irradiated fuel. “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.” [Philbin, 2002a]

To resolve these hypotheses with measured data, we have added a lower burnup spent fuel to the test program. This “low” burnup fuel was irradiated in the Surry PWR reactor, discharged in 1981, spent 3.8 years in wet storage, and then was stored in a He-filled Castor V/21 cask, as part of a 15-year dry storage test. The fuel burnup of the selected Surry fuel rod, H7, peaks at about 38 GWd/MTU, while the rod-average burnup is 36 GWd/MTU. Surry rod characterization results from sibling rods are documented in [Einziger et al., 2003]. This fuel rod currently resides at Argonne National Laboratory and will be characterized and fabricated into test rods for the ongoing test program, similarly to the H.B. Robinson high burnup spent fuel. The Surry fuel was supplied to ANL as part of a research program sponsored jointly by NRC, EPRI, and DOE RW.

The test rodlet design for the high-burnup Robinson rodlets, Figure 11, will be used as the starting basis for Surry test rodlet design. Modifications will be made based on differences between Surry and Robinson fuel pellet length and gas pressures. For example, Surry fuel pellet lengths are 15 mm, with a diameter of 9.3 mm, and a ^{235}U enrichment of 3.11 wt.%. Therefore, the spent fuel test rodlets made from Surry fuel will, tentatively, require four equivalent pellets (or $\sim\frac{1}{2} + 3$ whole + $\sim\frac{1}{2}$ pellets) to achieve a comparable fuel pellet length, about 60 mm (2.4 inch). ANL will perform the engineering design incorporating specifications stipulated by ANL and SNL nuclear facility personnel for applicable remote handling requirements at both sites.

6.2.3 Requirements, Spent Fuel Test Rodlets

The following requirements were defined by personnel in the Sandia Nuclear and Risk Technologies Center in order to partially satisfy nuclear facility operational and safety related issues and data needs.

- a) Rodlet design based on IRSN prior design, plus modifications by Sandia National Laboratories and Argonne National Laboratory, jointly, and in cooperation.
- b) Spent fuels (HB Robinson and Surry) are to be comprehensively characterized, and the test rodlets fabricated for Sandia by Argonne National Laboratory, under formal contract. There shall be no movable parts or springs within these assemblies.
- c) The test rodlets will be internally pressurized with He gas (refer to Table 5) at Argonne.
- d) Post-fabrication tests for each rodlet shall include: axial gamma scanning (for the integrity of fuel column and identification of mid-rodlet pellet position), leak checking (for rodlet weld leak-tightness) and visual inspection (for possible disturbance of cladding outside surface due to handling).
- e) Spent fuel test rodlets will be available for shipping from Argonne National Laboratory to Sandia by September 30, 2004. Actual shipment dates are to be determined. All eight test rodlets could be shipped within one transport cask.
- f) Rodlet end fitting extensions, for mounting the rodlets into the aerosol collection chamber, will be designed and fabricated by or for Sandia, with coordination by Explosive Component Facility and Nuclear Facilities personnel. The end cap extensions will be designed to make remote handling operations (insertion into the aerosol collection chamber and, if required, removal of intact, non-impacted rodlet) in the radiological facilities easier.
- g) The test rodlets, including end fitting extensions, and related mounting hardware, should be designed to be self-aligning within the test aerosol collection chamber. This self-alignment is defined to mean that the HEDD jet will be centered to impact on the central pellet in the rod.

- h) Prior to test usage, each spent fuel rodlet must be checked at Sandia to ensure cladding integrity and minimize potential contamination issues. Test rods shall be surface swiped to monitor for residual surface contamination.
- i) Radioactive rodlet and end fitting assembly requires insertion into aerosol chamber by remote operations at Sandia. This design is to include self-alignment, for HEDD jet impact on the center of the test rodlet, on the central pellet.
- j) The radioactive test rodlet will be the last component installed, prior to explosive testing.
- k) Post-test, fragmented spent fuel will be disposed of within the aerosol collection chamber. There will be no sampling of impact debris.
- l) Final Disposition: We anticipate temporary storage of post-test apparatus at Sandia for some time, possibly within a liner. The final disposition pathway/location must be agreed upon and approved by both Sandia (Nuclear and Risk Technologies Center) and the Department of Energy. The presumed disposition pathway is first, shipment to the Idaho National Engineering and Environmental Laboratory, then eventually to the U.S. Yucca Mountain Spent Fuel Repository. DOE (RW and EM) shall have prime responsibility of final disposition and associated expenses, once the post-test material is transported away from Sandia. The IRSN has agreed to cost-sharing for these post-test disposal expenses.
- m) Future research test reactor TRIGA rodlet (~ 3.8 mm diameter by 12.6 cm long) and possibly MTR plate (~ 10 x 10 cm) test targets, *if* incorporated into the test program, are to be designed similarly, to use the same or similar aerosol chambers and mounting hardware.
- n) The Sandia GIF facility nuclear safety assessment, authorization basis process shall be updated and modified, as appropriate, to include the H.B. Robinson high-burnup and Surry low-burnup spent fuel test rodlets, as well as the anticipated research test reactor fuels.
- o) Other requirements, TBD.

6.2.4 Test Program Expansion, Other Spent Fuel Forms

Future research reactor and MOX spent fuel tests have been discussed by the WGSTSC as a *potential* area of test program expansion. These types of fuel will be treated separately, later, *if and when* the need develops. It is critical that the fissile material in the MOX fuel be of U.S. origin. Subsequent fabrication of this material into rods and reactor irradiation in France, for example, would be acceptable. Such MOX spent fuel material could be accepted into the U.S., tested in this test program, then eventually disposed of in a U.S. nuclear waste facility.

- a) Research Test Reactor Fuel, for the NRC Office of Nuclear Reactor Regulation (NRR) or other organization, parallel to, but not part of the WGSTSC program, potentially including:
 - i) TRIGA rodlet
 - ii) MTR plate fuels (~ 10 x 10 cm) targets to be designed similarly, to use same aerosol chamber, similar mounting hardware.
- b) The Sandia GIF Documented Safety Assessment, DSA, may need updating/modification to incorporate TRIGA, MTR, or other research test reactor fuels.
- c) Further details will be documented at a later time, if and when research reactor fuels are formally incorporated into this test program.
- d) Other requirements, TBD.

6.3 Explosive Containment Vessel

The explosive containment chamber, or vessel, must safely isolate and protect personnel and facilities from the explosive detonation of the conical shape charge (HEDD) and resultant fragments and pressure pulses. It must direct the HEDD jet directly onto the test rod and must mate with the aerosol collection chamber and HEDD stop block. The design for this required test component has gone through several concept iterations over the past year; these concepts will be briefly described, then the current concept and requirements will be provided.

The initial explosive containment vessel, or “boom box,” shown in Figure 12, was presented at the 4th Technical Meeting of the Working Group for Sabotage Concerns for Transport and Storage Casks, in November 2002. This preliminary design, by the Sandia Explosive Components Facility [Dickey, 2002] consisted of a large, reusable explosive chamber, with a swing-open rear hatch, removable/cleanable liner for potential radioactive contamination, and an electrical cable feedthrough port. The conical shape charge is shown mounted in this vessel at its right side (at the center of the drawing), Figure 12. The aerosol containment chamber, containing the spent fuel test rodlet inside, plus the HEDD jet stop box, is shown at the right side of the figure, were designed to be removable post-test, for disposal. There was an explosive closure valve (not shown) between the explosive containment vessel and the aerosol containment chamber, intended to minimize potential HEDD post-detonation blowback of radioactive particles into the boom box. The aerosol containment chamber could be mounted either horizontally or vertically.

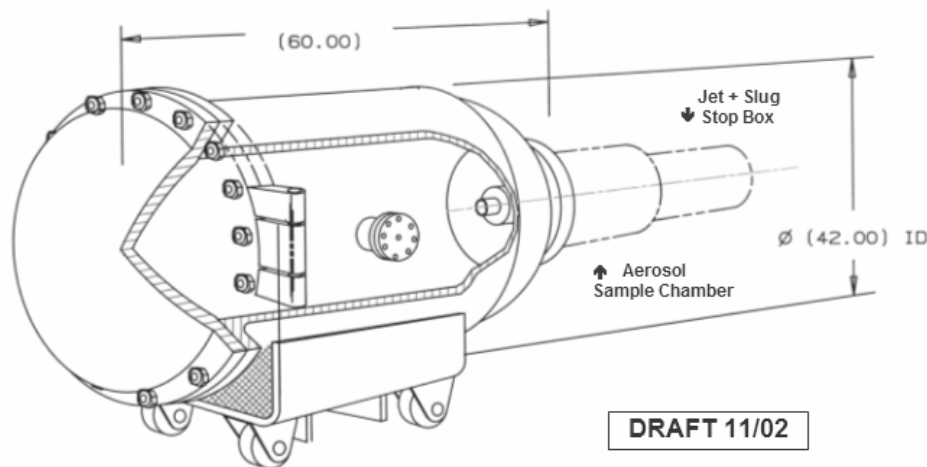


Figure 12. Explosive Containment Vessel, Preliminary Design (November 2002)

This explosive containment vessel design was modified and made approximately 33% shorter in length and 25% smaller in diameter, based on a series of BlastX modeling calculations [Dickey, 2003], and presented at the 5th Technical Meeting of the WGSTSC [Molecke and Sorenson, 2003], in May 2003. This revised, smaller version, is shown in Figure 13.

Significant, potential radiological contamination concerns were evaluated for the post-test explosive containment chamber, and to the efficacy and disposal of the large, removable polyethylene contamination liner within. There were also safety issues relating to the explosive valve in this design, and potential radiological contamination concerns for the GIF test facility when the aerosol chamber was disconnected from the boom box. A significant shift in design concept was required in order to satisfactorily resolve these important safety concerns, and to be agreeable to all participants involved.

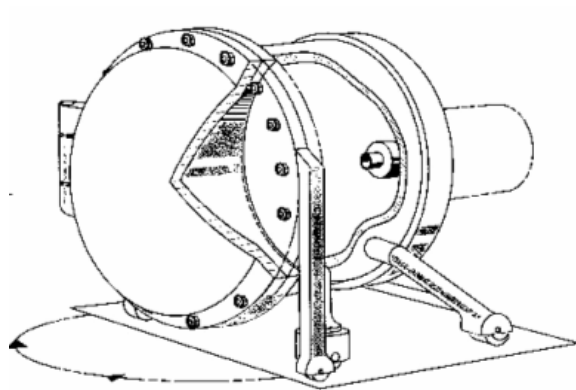


Figure 13. Explosive Containment Vessel, Revised Design (May 2003)

The current explosive containment chamber design *concept, still a work-in-progress*, is shown in Figure 14. Simplicity, safety, and total fabrication expense issues have been included in this design. Basically, the explosive containment vessel and the aerosol collection chamber are designed to be a single (non-separable), cylindrical chamber, with thick walls, designed to be used one time, then to be transportable and disposable within an available transportation cask. The overall design is, basically, shaped like a cannon barrel. This large cylinder is designed to be used in a vertical orientation, and allows for the use of a flow-through vertical elutriator design for the aerosol collection chamber. Design concept requirements are in the following section.

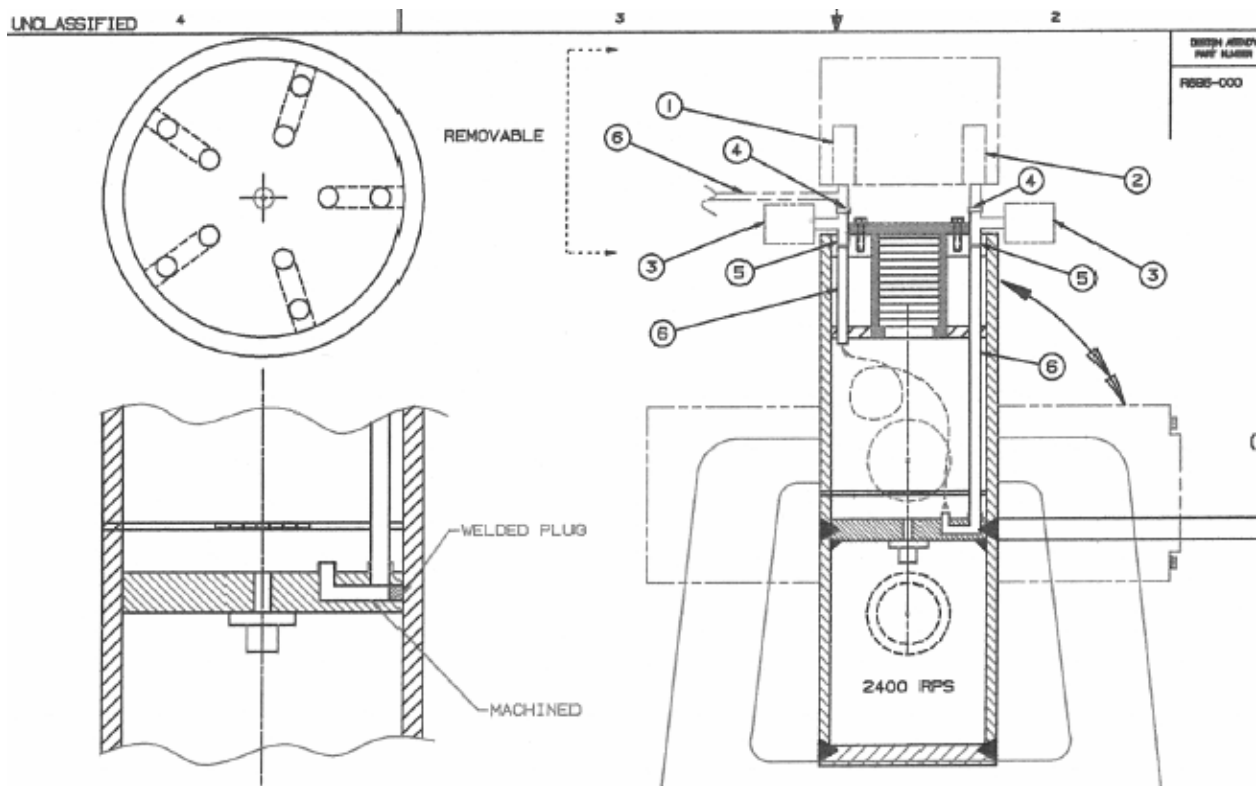


Figure 14. Design Concept, Explosive Containment/Aerosol Collection Test Chamber (10/03)

6.3.1 Requirements, Explosive Containment Vessel

The following requirements were defined by Sandia Explosive Components Facility personnel to help satisfy explosive safety and containment issues plus engineering qualification needs. In addition, further requirements were defined by personnel in the Sandia Nuclear and Risk Technologies Center, in order to partially satisfy corresponding nuclear facility operational and safety related issues.

- a) The explosive chamber must safely contain the HEDD up to (about 150 g); it must contain all explosive fragments and the transient explosive pressure pulse.
- b) It must satisfy requirements of the ASME code and the SNL Pressure Safety Manual.
- c) It must allow for the manual insertion of the HEDD and associated hardware and instrumentation, and for the accurate alignment of the HEDD jet onto the center of the target test rod-let. This requires a closable side port and electrical feed-throughs.
- d) The total explosive containment/aerosol collection chamber assembly must be designed with the goal of "total containment," i.e., the minimization of radioactive contaminant release.
- e) The external dimensions of the cylindrical, total explosive containment/aerosol collection chamber assembly must fit within the inner dimension of available transport casks, e.g., the GE 2000 cask. This requires a Certificate of Compliance, COC, for use of the GE 2000 cask at Sandia.
- f) The external diameter may also be limited by the available diameter of storage pit holes at INEEL, or at other locations. As such, it must also be capable of being enclosed in a thin, but leak-tight stainless steel disposal liner.
- g) The total explosive containment/aerosol collection chamber assembly must be oriented in the vertical position for testing purposes.
- h) There shall be no explosive valve or other shutter assembly between the explosive and the aerosol collection sub-chambers. Also, these two chambers will not be separable, i.e., they will be one piece.
- i) The explosive containment chamber will be capable of being used for, and withstanding multiple HEDD explosions. It is planned to reuse the test assembly for multiple Phase 3 DUO₂ tests. The Phase 4 spent fuel test chamber shall only be used one time only, then it will be disposable.
- j) Following each test, the explosive containment/aerosol collection chamber shall be checked for external surface contamination by remote swipe testing. Any contamination, most likely from separation of aerosol particle sampling apparatus, shall be decontaminated as required.
- k) Must fit into GIF Cell 3, and be movable, with appropriate handling fixtures.
- l) Requires electrical feed-throughs, for connection to the HEDD, and to strain gages and/or other instrumentation, needed to confirm the explosion process.
- m) Must satisfy all explosive design requirements of the Sandia Explosive Components Facility department.
- n) Must satisfy all nuclear and safety basis requirements for use in the Sandia GIF, and related Technical Area V requirements.

- o) Final component design, materials, and engineering detail for engineering and safety review, costing, and fabrication to be provided by R. Dickey (ECF), with major input and concurrence by Sandia Nuclear Facilities organizations (6800).
- p) Initial unit fabrication and availability for surrogate, non-radioactive testing and qualification, needed by April 2004.
- q) The total explosive containment/aerosol collection chamber assembly must be tested for leak tightness integrity prior to use, to two times the pressure design requirement.
- r) Other requirements, TBD.

6.4 Aerosol Collection Chamber

The primary purposes of the aerosol collection chamber for test phase 3 and 4 are to hold the radioactive test rodlet in the path of the HEDD jet, to provide for the adequate sampling of aerosol particles produced, and to be designed for “total containment,” i.e., to greatly minimize potential release of any radioactive contamination to the test facility, the Sandia GIF. Several aerosol collection chamber designs have been proposed, including a non-flow-through horizontal system capable of sampling particles in approximately the 0 to 16 μm size range, and a vertical flow-through or vertical elutriator design, capable of providing aerosol sampling in approximately the 0 to 100 μm size range. An earlier, now *obsolete*, design of the horizontal aerosol chamber and attached HEDD jet stop box is shown in Figure 15 [Molecke and Sorenson, 2003]. This “one-time use” aerosol collection chamber would be attached to the adjacent, reusable explosive containment vessel (Figures 12, 13), at left.

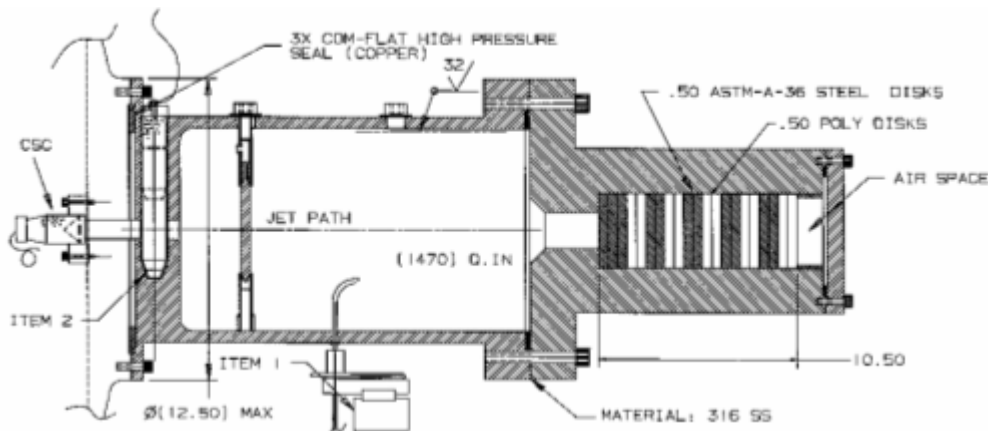


Figure 15. Horizontal Concept, Non-flow Aerosol Collection Chamber & HEDD Jet Stop Box (May 2003)

The vertical elutriator flow-through (or equivalent) aerosol collection chamber design concept is the only design being considered for further development, fabrication, and testing. As described in the previous section, this component will be cylindrical in shape and attached directly to the explosive containment vessel; refer to Figure 14 for a concept drawing. Specific aerosol design features are currently being developed. Current requirements for this component are as follows.

6.4.1 Requirements, Aerosol Collection Chamber

Many of the overall requirements for the aerosol collection chamber are identical to those for the attached, in-line explosive containment vessel. Major, joint requirements are re-listed, below.

- a) The total explosive containment/aerosol collection chamber assembly must be designed for “total containment,” i.e., to greatly minimize potential release of any radioactive contamination to the test facility. Any external chamber contamination that is detected, most likely from separation of aerosol particle sampling apparatus, shall be decontaminated as required.
- b) The external dimensions of the cylindrical, total explosive containment/aerosol collection chamber assembly must fit within the inner dimension of available transport casks, e.g., the GE 2000 cask. This requires a COC for GE 2000 cask use at Sandia.
- c) The external diameter may also be limited by the available diameter of storage pit holes at INEEL, or at other locations. As such, it may also be capable of being enclosed in a thin, disposal liner, if required in the future. [Note: Discussions on the issue of interim storage for post-test spent fuel wastes have been initiated with both INEEL and DOE; resolutions will be documented in the future.]
- d) The total explosive containment/aerosol collection chamber assembly must be oriented in the vertical position for testing purposes.
- e) The explosive containment chamber will be capable of being used for, and withstanding multiple HEDD explosions. It is planned to reuse the test assembly for the multiple Phase 3 DUO₂ tests only. The Phase 4 spent fuel test components shall only be used one time, then will be disposable. The “one-time only” use is based on the conclusion that it will be less radiologically safer, from a contamination control standpoint, as well as less expensive, to not open post-test chambers with particulated spent fuel and debris inside.
- f) It must satisfy requirements of the ASME code and the SNL Pressure Safety Manual.
- g) Must satisfy all explosive design requirements of the Sandia Explosive Components Facility department.
- h) Must satisfy all nuclear and safety basis requirements for use in the Sandia GIF, and related Technical Area V requirements.
- i) Final component design, materials, and engineering detail for engineering and safety review, costing, and fabrication to be provided by R. Dickey (ECF), with major input and concurrence by Sandia Nuclear Facilities organizations (6800).
- j) The total explosive containment/aerosol collection chamber assembly must be tested for leak tightness integrity prior to use, to two times the pressure design requirement.
- k) The initial unit fabrication and availability for surrogate, non-radioactive testing and qualification, needed before the initiation of Phase 3 tests. The total containment system must be operationally pre-tested w/ CeO₂ surrogate and HEDD, at the Sandia ECF in 2004, then re-tested and demonstrated at the GIF, before future use with DUO₂ and spent fuel.

Separate, additional aerosol collection chamber requirements:

- l) The stand-off distance, HEDD face to test rodlet center, must be 19 cm (7.5 inch).
- m) Must mate to, or incorporate the HEDD jet stop box in-line, not perpendicular, to minimize separation/contamination concerns.

- n) Must allow for remote handling insertion of radioactive test rodlet, with pre-alignment fixtures, and positive, secure attachment within the aerosol collection chamber.
- o) The test rodlet will be the last component installed prior to testing. A sealing cap device shall be installed over the rod opening after insertion, to complete the leak-tightness function.
- p) Must allow for remote or manual, intact rodlet removal, in the event of test termination.
- q) Internal chamber atmosphere must be capable of pretest gas-exchange flush. The internal chamber atmosphere shall be either air or nitrogen, at 1 bar.
- r) Must allow for external, top-side manual mounting and post-test dismounting (manual or remote) of multiple aerosol particle collectors (~ 3 or more particle impactors + 2 $\leq 100 \mu\text{m}$ filters, cyclone separators, or equivalent large-particle separators).
- s) Atmosphere sampling-volume capable of being pump, or vacuum bottle-drawn in < 1 minute. Volume flow rate requirements for all samplers estimated at ~ 10 L/min total.
- t) Must incorporate vertical elutriator design features, to keep larger aerosol particles (~ 20 to 100 μm) suspended or re-levitated long enough to allow effective particle sampling.
- u) An up-flow rate of 25 cm/sec is required, with pumps or pressure bottles for either air or nitrogen. A dispersal frit or other device at/near the chamber bottom should be implemented, to provide ~ upward laminar flow. Pump(s) exhaust, if used, must be HEPA filtered for contamination control.
- v) Vertical up-flow, aerosol sampling section length to be > 50 cm (20 inch), as available; Fraunhofer ITEM requested a length of 75 cm (29.5 inch).
- w) Must have atmosphere (air, N_2) inlet, with one-way valve (+ gas bottle?), or other device, to prevent internal vacuum during particle sampling.
- x) External connections for particle samplers and pumps, atmosphere change, electrical leads, etc. to be designed for contamination control for test use and post-test remote removal.
- y) Electrical on/off valves, controls for timing of external sampling pumps-to collectors are required.
- z) For Phase 3 DUO_2 testing and Phase 2 pre-testing with CeO_2 ONLY, the aerosol collection chamber must provide an open-able sampling port to collect post-test impact debris for sieving and analyses, and for clean-up. This version of the aerosol chamber should be reusable for multiple tests.
- aa) For Phase 4 tests with highly radioactive spent fuel samples, the post-test aerosol chamber with disrupted spent fuel, shall be disposed of without opening, with stop box attached, with disrupted fuel and rubble inside. Each such chamber will be used only one time.
- bb) Fiber-optic boroscope or similar optical device to be inserted into the post-test aerosol collection chamber, for viewing of the disrupted test rodlet. Device must be capable of being inserted without contamination release; may be disposed of with the test chamber. Other “viewing” design options may be considered.
- cc) Other requirements, TBD.

6.5 Requirements, Aerosol Particle Samplers

The following current aerosol sampling requirements were defined primarily by Sandia Plasma, Aerosol, & Noncontinuum Processes Department 9117 personnel, with significant input and consultation with aerosol experts and test partners at Fraunhofer ITEM. Explosive component and nuclear facility safety and engineering issues have also been incorporated.

- a) Aerosol particle samplers, e.g., Marple personal impactors or similar, must selectively collect particles in multiple size stages throughout the respirable aerosol size range, from 0 to 10 μm AED, and up to 20 μm AED if possible. Three of these particle impactors are required per each Phase 3 and 4 test, in order to provide multiple samples and data statistics.
- b) Additional aerosol particle samplers covering the range of $\sim 20 \mu\text{m}$ up to $\sim 100 \mu\text{m}$ AED are required. These can be small cyclone separators, metal foam or frit type filters (no organic material filters), or other similar large particle separator apparatus (to be defined). Three or more of these larger aerosol particle samplers are required per test.
- c) The size and weight of all particle samplers should be kept small, in order to facilitate remote handling operations, as needed, and to minimize the required flow rate through the sampler.
- d) All particle samplers and associated sampling tubes should be on the top end of the vertical aerosol collection chamber.
- e) All particle samplers must have manufacturer's calibrations.
- f) Particle sampling apparatus should be reusable, if at all possible, after removal of filtration stages and collected particulates. Procedures for apparatus decontamination shall be developed.
- g) Particle sampler apparatus must incorporate closure/separation valves, for post-test remote removal from the aerosol collection chamber. Particle release minimization, i.e., contamination control features must be used in these connections.
- h) All particle samplers must be designed to not release contaminants. External sampler housings may be used, as necessary. Post-test surface contamination checks are to be made. Surface decontamination shall be conducted, as required.
- i) The particle samplers and associated sampling tubes and valves must operate in a potential high-pressure environment (post-test internal explosive pressure of several atmospheres) while remaining accurately calibrated. Pressure mitigation efforts, e.g., the use of critical orifices and/or remote-control isolation valves, may be considered.
- j) The aerosol particle impactor samplers must be capable of being operated in a vertical orientation, right-side up.
- k) All associated airflow connections and pump connections for operating the particle samplers should be mounted on the top side of the aerosol collection chamber.
- l) Vertical tubing pathways through the housing of the adjacent HEDD jet stop box, if appropriately designed, are acceptable.
- m) Others requirements, TBD.

6.6 Requirements, HEDD Jet Stop Block and HEDD

As described earlier, the purpose of the HEDD jet stop block or box is to stop the very energetic HEDD explosive jet, as well as the less energetic residual metallic slug or "carrot," within a

manageable distance. The jet must not damage or penetrate the total containment aerosol collection system. The explosive engineering design requirements for this component follow.

- a) Must have adequate length and mass stop the HEDD jet and slug in less than 30 cm (12 inch) length; allow for a maximum of 8° jet misalignment.
- b) Will utilize multiple steel and polypropylene stop plates in series. The first plate, closest to the stop box/aerosol chamber interface, will be steel, in order to facilitate post-test surrogate particle impact debris sampling for surrogate materials only.
- c) Must align with the aerosol collection chamber, or be an integral part thereof.
- d) Design of the HEDD jet stop box and associated safety review qualifications are to be credibly based on CSC nominal performance and known discrepancies, Sandia Explosive Component Facility engineering experience, plus modeling, plus design requirements, plus demonstrations.
- e) The HEDD jet stop block housing may have allowable penetrations at its periphery for particle sampling tubes, air flow tube/pipes, or other penetrations, as long as the jet stop function is not impeded. For example, the air central jet stop area of the block may not incorporate an air-flow metal frit ring; destruction in this vicinity is anticipated.
- f) For Phase 3 DUO₂ testing and Phase 2 pre-testing with CeO₂ ONLY, the jet stop plates may be replaceable, to allow reuse of the stop block and other components.
- g) All conical shape charges (HEDD) used for the Phase 3 and 4 tests should be purchased at the same time, and specify that they all originate from the same manufacturing batch. This is desired in order to minimize differences in HEDD jet performance characteristics.
- h) Other requirements, TBD.

6.7 Pre-Test Transport Casks

- a) The transport cask must be acceptable for use with Argonne National Laboratory (West) and Sandia National Laboratories facilities.
- b) The transport cask must be certified by the Department of Transportation and NRC.
- c) The test rodlets, including interim packaging, must fit into the transportation cask inner cavity.
- d) Contamination between the transport cask and the shipped packages should not take place.

6.8 Post-Test Transport Casks

- a) Need availability of a transport cask of adequate dimensions to contain the explosive containment/aerosol collection chamber post-test components, presumably the GE 2000, or other, TBD. These transport casks must be available for use (rental, lease, etc.) following the termination of this test program. Test Phase 4 is currently scheduled to be complete at the end of 2005. There may be temporary, on-site storage of test containers at Sandia for several years, before off-site transport casks are needed.
- b) The transport cask must be acceptable for use and receipt at the post-test storage/disposal site, e.g., at the Idaho National Engineering and Environmental Laboratory.

- c) The transport cask, presumably the GE 2000 or other, TBD, requires a valid Certificate of Compliance, COC, be obtained (or modified) for use at Sandia.
- d) A transport *cask basket* must be fabricated to hold the explosive containment/aerosol collection chamber post-test components and liner in place securely, within the transport cask. This cask basket must have NRC certification.
- e) The scheduled availability or rental of an adequate number of transport casks, at least one and possibly more, is required.
- f) Sandia Nuclear Facilities personnel may require training in the use, handling, and loading operations for the selected transport cask.
- g) Adequate programmatic funds must be made available to Sandia National Laboratories for obtaining or revising the COC, and then for renting the use of the transport casks.
- h) Others, TBD.

6.9 Requirements, Sandia GIF Test Facility

The Sandia Gamma Irradiation Facility, GIF, is a Hazard Category 3 nuclear facility with an approved 10CFR830 DSA, Documented Safety Analysis.

The proposed Phase 4 test rodlet source term is well below Hazard Category 2 threshold limits, including the current GIF nuclear material inventory.

The current safety basis does not adequately address explosives (HEDD) or fissile materials (due to the lack of analysis in the current DSA), but will be modified in the required annual DSA update now underway.

The following nuclear facility considerations must be adequately addressed and resolved.

- a) Worker exposure to ionizing radiation:
 - Experiment will be conducted in an accessible shielded cell with remote handling capabilities.
 - Other than removal of the aerosol collection portion, the apparatus will be placed in a shipping container in its entirety with no attempt to dismantle. The apparatus may be contained first within an inner liner, leak tested, then placed within the shipping container/cask.
 - Minimal exposure to the spent fuel source term in the shielded GIF cell will be anticipated. Exposure to the test chamber outside of the GIF cell must also be minimized to limit worker exposure.
 - Work will be conducted according to SNL's radiation protection program which requires a radiation work permit and oversight by radiation protection personnel.
- b) Contamination control:
 - The experiment will be designed to confine the radioactive material.

- An independent, second level of confinement will be provided within the facility.
- Other than the aerosol collection portion, the spent fuel sample will not be removed from the apparatus.
- GIF pool access will be covered.
- During explosive testing, the GIF ventilation system will be secured and isolated.

c) Explosive Safety of HEDD

- The experiment chamber will be designed to contain all the products of combustion for twice the mass of explosive planned.
- The chamber design will be validated prior to use.
- Explosives handling will be performed by SNL's explosives experts trained in explosive safety.
- Explosive limits are governed by GIF and Sandia TA-V thresholds for explosive quantities.

d) Quality Programs

- Any item that is used in the GIF is subject to the SNL quality procurement process.
- A Project Experiment Quality Plan (PEQP) must be completed to encompass all testing.
- Prior to conduct, the experiment safety shall be reviewed by a committee of experts and approved by line management.

e) Testing and Disposal Path

- Clear and defined experiment requirements/goals must be set well before testing.
- Any modifications to experiment design or procedures will be reviewed by the Sandia National Laboratories safety committee.
- All aspects of the test program will be adequately funded by the experiment project management to encompass all portions of testing.
- A clear and defined disposal path is required and established before testing can begin.
- Test components can be stored at TA-V in the interim, yet all radioactive/hazardous components will eventually be removed from TA-V.

7. TEST CONDUCT AND AVAILABLE RESULTS

7.1 Summary of Phase 1 Tests with Glass

Three tests with glass pellets and four tests with leaded glass plates that were “particulated” by the impact with a HEDD jet were performed in June 2002. These preliminary tests were intended to help understand the physics of fragmentation of brittle materials impacted by the high-speed HEDD jet. They were also intended to check the validity of aerosol particle scaling laws established under the conditions of low-speed impact, in fragmentation experiments previously conducted at Fraunhofer ITEM with conventional rifle bullets. Flash X-ray diagnostic techniques and Imacon™ high-speed digital photography were used to observe the HEDD jet parameters and the target disintegration process. Dual Respicon™ particle samplers were used to collect and classify test aerosol particulates into three size fractions; other remaining fragments were collected and mechanically sieved. The results obtained with the leaded glass plates were reproducible, as shown in Figure 16.

For the purpose of inter-comparison of different test data from Phase 1 and 2 to previous experimental data from Fraunhofer ITEM, we have evaluated the cumulative size distribution, $Q(d)$, (y-axis, in Figure 16), defined as the material formed as particles and fragments with aerodynamic diameter smaller than (d) , divided by the mass of the HEDD jet-particulated specimen. The data points for $(d) > 100 \mu\text{m}$ AED are based on sieve analyses whereas the points at 10 and 5 μm were calculated from the Respicon™ particle distribution measurements. Some early Phase 2 cerium oxide aerosol particle Respicon data were analyzed in the same way as data from Phase 1 tests. The cerium oxide released mass smaller than 10 μm , respectively 5 μm , was normalized to the mass of the particulated pellets, and chosen to be 15 g in both tests. The results revealed $Q(5 \mu\text{m}) = 2.75 \times 10^{-3}$ and $Q(10 \mu\text{m}) = 3.2 \times 10^{-3}$, in remarkably good agreement to the glass test data. The data shown in Figure 16 for the leaded glass plates and preliminary cerium oxide tests 2/1A and 2/1B suggest the existence of a scaling law in the particle size range smaller than a few hundred μm . This is consistent with results found in earlier fragmentation experiments performed in Germany with multiple brittle materials. However, there is a relatively large scatter in the data for particles with sizes $< 10 \mu\text{m}$. The results suggest that for complete coverage of the relevant size range $< 100 \mu\text{m}$ in the spent fuel sabotage – aerosol measurement tests, one data point at 100 μm plus a detailed analysis in the range smaller than 10 μm should be adequate. Extrapolation from the small size data to larger sizes is inadequate.

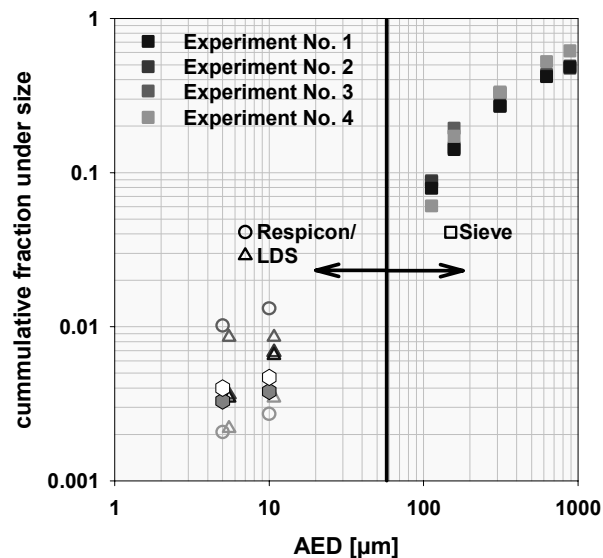


Figure 16. Glass vs. Cerium Oxide HEDD-Test Results for Validating Brittle Material Scaling Laws

(Fraunhofer ITEM)

□ ○ △ = Phase 1 Leaded Glass Plate data
● = Phase 2 CeO₂ – HEDD data

The extent of secondary glass particle fracturing, i.e., HEDD-disrupted particles crashing into the collection chamber walls and forming more respirable size particles, was determined to be small. This resulted in a conclusion that the horizontal dimension, or length, of the aerosol collection chamber could be limited to 20-30 cm. The aerosol collection chamber and test setup for the Phase 1 tests was illustrated in Figure 1; the horizontal dimension of this box was 40 cm. Further detailed results from Phase 1 tests will be documented separately [Molecke, Yoshimura, and Vigil, 2004].

A corresponding, supportive test concept has been developed by Fraunhofer ITEM/GRS and was extensively used to characterize the formation of airborne particulates upon transient energy input into various types of brittle materials, including CeO₂, glass, depleted UO₂, AlSi, and WZrO₂ [these detailed results will be documented in the future]. The aerosol test apparatus consists of a vertical elutriator to separate non-airborne fragments from airborne material with AED < 100 μm, and to further aerodynamically classify this fraction into a number of size intervals using a combination of a centrifugal classifier (20-100 μm) and a cascade impactor (0.1-20 μm). The possible effect of different modes of mechanical energy into the brittle material was studied: impact of objects against a hard target, interaction of the brittle material with bullets of different speed such as rifle bullets (< 1 km/s), and projectiles generated by a light gas cannon (2.9 km/s). This data [*to be documented*] revealed a striking universality in aerodynamic size distribution irrespective of the impact mode and the type of the brittle material. These results justify the calculation of the SFR from measurements for one or two specific size ranges between 10 and 100 μm and multiple specific size classes in the range smaller than 10 μm. This should accurately take into account the enrichment of volatile fission products in the respirable particle regime and thus provide useful input for the experimental design of the tests described herein.

7.2 Phase 2 Tests (2/0 through 2/2B)

By the end of September 2003, nine Phase 2 tests had been performed with cerium oxide pellets contained within Zircaloy cladding tubes, impacted by a HEDD explosive jet: tests 2/0, 2/1A, 2/1B, 2/2A and 2/2B (replicates), 2/3A and 2/3B (replicates), and 2/4A and 2/4B (replicates).

The first three tests, 2/0, 2/1A and 2/1B were intended as “system checkout and calibration tests,” to help fine tune the test setup, exercise all systems, and observe the extent of damage to the cerium oxide pellet stack – to determine how many of the sintered ceramic pellets were damaged and what length of Zircaloy tube was destroyed. For tests 2/0 and 2/1A, six pellets were stacked in the cladding tube; the HEDD jet was centered (using pretest laser alignment) on the center of the top cerium oxide pellet in the stack. The intent was to observe how many pellets are disrupted by the HEDD impact and shockwave transmission down through the pellet column. For test 2/1B, five pellets were used, with the HEDD jet aimed at the center of the middle pellet. Four diagnostic flash x-rays tubes were used to observe the explosive disruption process, and were set at four separate times of about 90 μsec to about 800 μsec. Figure 17 illustrates the X-ray results of the HEDD jet penetrating the test rod and particulating pellets at 90 μsec; the slower shape charge residual slug is seen in the views at 676 and 791 μsec, moving from right to left. Two Respicon particle samplers (replicates) were used for tests 2/1A and 2/1B; they were turned on at 10 seconds before the HEDD detonation and turned off at 60 seconds after.

Test 2/2A and 2/2B were similar to 2/1B. Five cerium oxide pellets were used, the HEDD jet was aimed at the center pellet, and two Respicon particle samplers were used. The Respicon pump was turned on at 10 seconds before the HEDD detonation and turned off at 30 seconds af-

ter. Use of the flash X-ray diagnostic system was discontinued. Refer to Tables 6 and 7 for detail on the cerium oxide pellets and Zircaloy cladding tubes used.

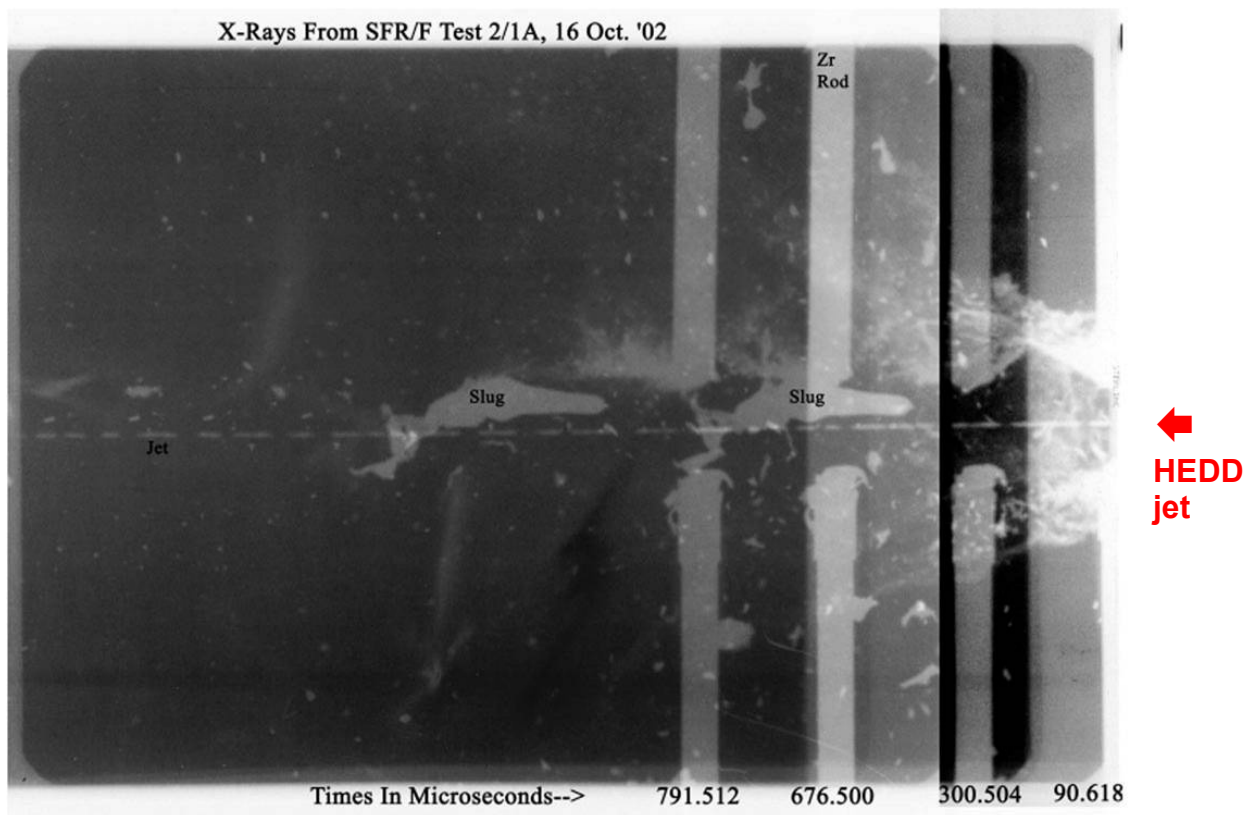


Figure 17. Flash X-Ray Pictures, Test 2/1A

Aerosol Particles: The aerosol particles collected on glass fiber filters in the Respicon particle samplers were initially observed to be loaded with grayish-brown, uniform particulates. This is the reason that the sampling time was decreased to 30 seconds for tests 2/2A and 2/2B.

We used gross weight analyses plus detailed chemical analyses, inductively coupled plasma/mass spectrometry, ICP-MS, for evaluation of major elements in all of the collected particles. Chemical analyses were necessary because much of the collected particle mass consisted of fine carbon soot -- a combustion byproduct produced by the HEDD explosive. The Respicon aerosol particulates were weighed, then chemically dissolved and analyzed. These collected particles weighed tens of mg per test. The major elements detected in the aerosol ranges were copper (from the HEDD jet), first, then cerium. Other significant elements detected were zirconium (from the cladding tube) plus aluminum and iron (from the HEDD housing). The majority of weight for the collected aerosol materials consisted of carbon soot (from the HEDD explosive) and oxygen, possibly with some minor nonmetallic elements; these elements cannot be analyzed by ICP-MS. Analytical results from the Respicon aerosol particle samples (respirable fraction: 0-4 μm ; thoracic fraction: 4-10 μm ; and inhalable fraction: 10 to < 100 μm) are listed in multiple tables, in **Appendix A**, Aerosol and Particle Analysis Results, Sections A.1.1A through A.1.2B.

Particle Impact Debris: The particulated test materials (aerosols plus non-aerosol particles) remaining in the aerosol collection box, i.e., the impact “debris” not collected by the aerosol particle samplers, were manually brushed from the inner aerosol box walls, Figure 18, and collected as well as possible, then mechanically sieved, and chemically evaluated using ICP-MS. Sieve meshes were: 1000, 500, 250, 125, and < 125 µm (residual) geometric size for the Phase 2 tests; additional sieves with meshes of 74, 44, 37, and 25 µm were used later. Several photos of the impact debris sieved materials are illustrated in Figure 19, for test 2/1B. The > 1000 µm sieve segment contained the majority of materials, predominantly fragments of Zircaloy tube and copper from the HEDD. The intermediate size ranges, from > 500 µm to > 125 µm, contained about 10 – 23 wt. % of the collected materials, and were fragments of non-aerosol cerium oxide, with carbon soot coatings. The sieved bottom residual segment, about 22 – 42 wt. % of the collected materials, was predominantly carbon soot, explosive residue, mixed with minor flecks of cerium oxide. Analytical results from the impact debris particle samples are listed in multiple tables in **Appendix A**, Aerosol and Particle Analysis Results. Figures of the distribution of metals (Ce, Zr, Cu, etc.) in the sieved fractions of the particle impact debris remaining in the aerosol collection box are also contained in **Appendix A**, Sections A.2.0 through A.2.2B.

Observations: Observed, post-test results were consistent for all tests, 2/0 through 2/2B: the top and bottom remaining segments of the pellet/Zircaloy tube remained essentially vertical; about 2 to 2 1/3 pellets (~ 13.9 mm/each pellet) and about 25-30 mm of Zircaloy tube were fragmented. The measured Zircaloy tubing gap varied primarily due to jagged flaps of Zircaloy of different lengths; refer to Figure 20. The amounts of post-test test rodlet tubing and pellets disrupted are listed in Table 9. In all tests, the CeO₂ pellets adjacent to the HEDD jet-impacted segment of Zircaloy tubing were firmly wedged into the tube, by “blowback” fine particles in the small tube-to-pellet gap (blown outward by the jet), and could not easily be removed from the cladding. These remaining, captive pellets were essentially whole, with some observable external fracturing, as shown in Figure 20. The end-most pellets, those adjacent to the copper holding rods were essentially undamaged.

Table 9. Observed Post-Test Rodlet Disruptions, Phase 2 Tests

Phase 2 Test #	Zircaloy Tube Gap mm (ave.)	# of Pellets * Particulated	Pellet Length Particulated	Pellet Weight Disrupted (particles + fragments)	“Blowback” ** Particle Weight
2/0	27-(30)-35				
2/1A	24-(26)-28				
2/1B	20 - 32				
2/2A	27-(27.3)-28	2.3 (long)	31 mm	10.24 g	
2/2B	26-(27)-28	2.3	31 mm	10.14 g	
2/3A	21-(25)-28	4.4 (short)	31 mm	~13.4 g	1.60 g
2/3B	22-(25)-29	4.8	34 mm	~15.2 g	0.85 g
2/4A	25-(29)-33	5.2	36 mm	~16.2 g	0.22 g
2/4B	22-(25)-30	4.7	33 mm	~15.0 g	2.28 g

(* refer to Table 6; ** refer to Section 7.3, observations)

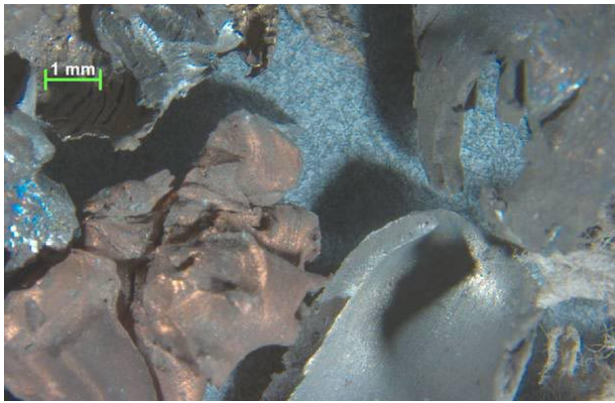


Figure 19a. Test 2/1B Impact Debris, 1.00 mm sieve



Figure 19b. Test 2/1B Impact Debris, 0.25 mm sieve



Figure 19c. Test 2/1B Impact Debris, 0.125 mm sieve

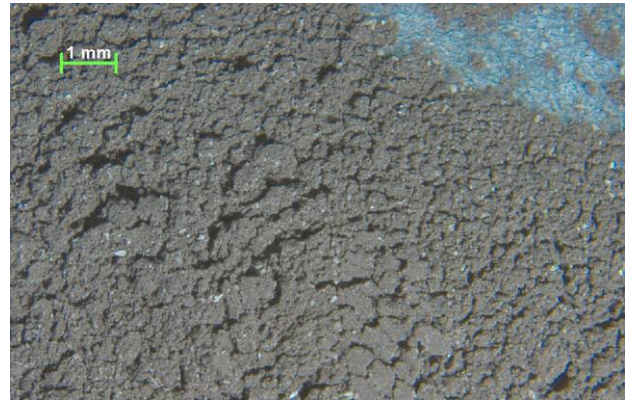


Figure 19d. Test 2/1B Impact Debris, < 0.125 mm sieve



Figure 18. Test 2/3A Impact Debris Particle Collection



Figure 20. Post-test 2/4A Rodlet Pieces

7.3 Phase 2 Tests (2/3A through 2/4B)

Tests 2/3A and 2/3B had several important differences from the preceding Phase 2 tests. The cerium oxide pellets used were shorter and slightly greater in diameter (than in tests 2/0 through 2/2B), refer to Table 6, and the Zircaloy cladding tube outside diameter was somewhat greater, refer to Table 7. These changes were made in order to closely match the respective spent fuel dimensions for H.B. Robinson spent fuel, to be tested in Phase 4. The earlier tests pellet and cladding dimensions were intended to simulate the dimensions of French spent fuel, originally to be obtained from IRSN [GRS/SNL, 2000; Molecke et al., 2003a]. The use of two Respicon aerosol particle samplers was supplemented with the addition, and independent operation of a Berner nine-stage aerosol particle impactor; refer to Section 5.3. The aerosol sampling time for all particle samplers was decreased to 15 seconds, starting at 5 seconds after the HEDD detonation; this change was recommended by Fraunhofer ITEM aerosol experts. Also, we used a new aerosol collection box with HEDD jet entrance and exit-hole closure valves, shown in Figure 6.

Tests 2/4A and 2/4B were very similar to 2/3A and 2/3B. In addition, non-radioactive fission product dopant pellets (chemicals in “wells” within the CeO₂ pellets) were added to the test system and particulated by the HEDD jet; these fission product dopant pellets are described in Section 5.1.3 and shown in Figure 5.

There also were some hardware modifications made to the overall test system, going from tests 2/3 to tests 2/4. As shown in Figure 6, the aerosol collection box used in tests 2/3A and 2/3B had two closure valves and it was not directly attached to the HEDD jet stop block. There also is a metal ring blast shield mounted on the vertical plate blast shield, as shown on the left side of Figure 2. This metal ring blast shield, with the HEDD/conical shaped charge mounted at its center, had not been used in previous tests. The purpose of the 0.91 m (36 inch) diameter metal ring used in tests 2/3A and 2/3B was to contain HEDD blast fragments, and to monitor the fragmentation pattern. The size and design of this ring was representative of a prior “total containment system” reusable boom box/explosive containment vessel design concept [Molecke and Sorenson, 2003]. For tests 2/4A and 2/4B, we used a smaller 0.58 m (23 inch) diameter metal ring blast shield ring, as shown in Figure 2. This smaller diameter blast shield is representative of a new design under development, of a one-use “cannon” boom box to be part of the total containment system design. One of the consequences of the blast shield ring use is to focus more of the conical shape charge explosive energy pressure forward, into the aerosol collection chamber. Test 2/4B utilized an electronic, monitored pressure transducer installed in the top corner of the aerosol collection chamber; a transient pressure pulse of about 5 bar, 75 psi, was measured. Further changes and tests of blast shield/explosive containment vessels, aerosol collection chambers, and other equipment are anticipated in future Phase 2 tests, tests 2/5A and others following.

Analytical results from the three-stage Respicon aerosol particle samples and the nine-stage Berner particle impactors are listed in multiple tables, separately, in **Appendix A**, Aerosol and Particle Analysis Results, Sections A.1.3A through A.1.4B. Results are presented for the major component species analyzed (as metals), cerium, zirconium, copper, as well as the minor components (HEDD-shell metals and alloying elements), such as aluminum, iron, tin, chromium, manganese, etc. Results for the major species distributed on each Berner stage are graphically shown in Figures A1.2 through A1.7.

Analytical results from the particle impact debris samples remaining in the aerosol collection chamber, collected and sieved into size fractions (1000 μm down through 25 μm, and residual) are also listed in multiple tables in **Appendix A**, Sections A.2.3A through A.2.4B.

Observations: We observed clean, not soot-contaminated, particles of cerium oxide rod debris or “blowback” material in the post-test Zircaloy tubes, adjacent to the copper rod supports, at the tube ends farthest from the HEDD-impact gap. This indicates that some of the jet-particulated cerium oxide was blasted at right angle to the HEDD jet, transporting through the available pellet-tube gap. These “blowback” cerium oxide particles were collected for weighing and sieving. Since there was no soot observed on these particles, no chemical analysis was necessary; the blowback material weights are listed in Table 9. Almost all of this blowback debris material was 250 μm in (geometric) size or smaller. A significant consequence of this observation is that the initial pellet-cladding tube gap is filled with this blowback debris, “cementing” the residual pellets in place. The post-test, remaining pellets could not move further, they could not be removed from the post-test cladding tube without significant force. In a sabotage scenario on actual spent fuel rods in a cask impacted by a HEDD device, we would not expect a significant number of non-destroyed pellets to pour out of the destroyed cladding tube ends. However, irradiated Zircaloy cladding will be more brittle than unirradiated cladding, due to hydrogen embrittlement. It is possible that additional fragmented material could fall out in this situation; this may be observed in the Phase 4 tests with actual (irradiated) spent fuel.

We also observed that the Zircaloy tube diameter expanded by about 0.2 mm closest to the HEDD-jet formed gap region, possibly from the HEDD-jet pressure pulse.

Based on information in Table 9: The HEDD-impact-produced gap lengths in the Zircaloy cladding tube/test rods were fairly consistent for all tests observed. This gap distance varied between approximately 25 to 30 mm. The consequent length of surrogate pellets particulated was always somewhat greater than the observed tubing gap; the pellet stack length particulated ranged from about 31 to 36 mm. The initial length of the surrogate CeO_2 pellets, varying from 13.3 to 7.0 mm, did not appear to effect the particulated length; the gaps between multiple pellets also did not seem to have an appreciable effect on this length.

The amount of impact debris and soot collected inside the post-test aerosol box for tests 2/4A and 2/4B appeared smaller in volume than in earlier tests; this was not anticipated. We had expected more impact debris, since the gap between the aerosol collection box and the HEDD jet stop block had been eliminated (refer to Figure 2), and the explosive momentum particle escape pathway was eliminated. For test 2/4A, we opened the HEDD jet stop block assembly post-test, separated out the stopping plates, and tried to sample debris particles by brush sweeping and collection. Particle collection was incomplete due to numerous ledges, nooks and crannies within the stop block.

Cerium distributions: From the cerium (element) distributions measured with the multistage Berner impactor in tests 2/3A & B and 2/4A & B, the vast majority by weight of collected cerium particles is within the range of 2-16 μm AED, with a much smaller amount in the 0.25-2 μm range; refer to Tables A1.24, A1.28, A1.37, and A1.45, and Figures A1.2, A1.3, A1.4, and A1.6, respectively (in the Appendix). The data from the 3-stage Respicon particle collectors is similar, but not as clearly defined. Particles collected on the largest size, “top” collection stage for each device, 10-100 μm for the Respicon and >16 μm for the Berner, cannot be accurately interpreted due to incomplete collection in a non-flowing aerosol collection chamber.

Volatile Fission Product Dopant Enhanced Sorption: Tests 2/4A and 2/4B were the first to incorporate the thermally volatile, non-radioactive dopant chemical fission product species cesium (as CsI), iodine (as CsI), ruthenium (as RuO_2), as well as non-volatile strontium (as SrO). There is clear evidence of enhanced sorption of the volatile species cesium and ruthenium onto the

smaller, respirable particles of surrogate cerium oxide. In Figures A1.5 and A1.7 and Tables A1.38 and A1.46, Berner particle analyses, the Cs and Ru dopants appear to maximize their concentrations in the 0.25 to 1 μm ranges, decreasing as the particles increase or decrease in size. The non-volatile strontium species does not show the same sorptive behavior, most of it was collected on the > 16 μm residual filter. Similar enhanced sorption of cesium and ruthenium was found on the smallest, respirable stage of the Respicon samplers, Tables A1.35 and A1.43. Again, the non-volatile strontium species was not enhanced on the smallest stage.

The volatile fission product dopant iodine was not detected on any of the Berner or Respicon particle collector stages. It is postulated that the volatilized iodine may have reacted with the bare steel inner wall of the aerosol collection test chamber. It is also possible that the analytical preparation methods selectively removed the iodine prior to analysis; this postulate shall be evaluated in the future.

Based on these observations of enhanced sorption of volatile fission product (surrogate) dopant species on the smallest, respirable particles, we would expect similar results from actual spent fuel aerosol tests, in Phase 4 of this test program.

8. SUMMARY AND DISCUSSION

In this technical report, we have documented a thorough overview of the ongoing test program that supports the needs of the international Working Group for Sabotage Concerns of Transport and Storage Casks. This surrogate and spent fuel sabotage – aerosol test program is being performed primarily at Sandia National Laboratories, with major input, participation, and supplemental testing from other U.S., German, French, and British partners in the WGSTSC. This program involves the testing of surrogate pellet materials and actual spent fuel test rod sections. The data being measured, and presented herein, support quantifications of aerosolized materials produced from actual spent fuel and surrogate material test rods, resulting from an impact by a high energy density device, HEDD.

In this document, we provide a detailed description of the test concept, overall design and programmatic details – this report serves as the formal test plan, at this point in time. We also supply detailed descriptions of all current experiment components, and provide requirements for future components. We present all available test results – up through September 2003, observations, and aerosol particle size and chemical analyses – primarily for surrogate material Phase 2 tests using cerium oxide sintered ceramic pellets. Since Phase 2 of this test program is still in progress, only preliminary evaluations and interpretations of the aerosol particle data can be included. This document is, therefore, a status report. Further detailed test results, interpretations, and comparisons to other available, relevant results will be documented in the future.

A major goal of this ongoing test program is to obtain an extensive, precise database on aerosol and respirable particle size distributions, and fission product enhancement on respirable particle sizes from both surrogate (cerium oxide pellet, unirradiated depleted uranium oxide) and actual spent fuel-HEDD interaction tests. This will allow us to calculate the spent fuel ratio and extend the test results to other nuclear fuel sabotage situations through follow-on modeling. Results and observations from the first nine Phase 2 surrogate cerium oxide tests documented herein 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 clear evidence of enhanced sorption of the volatile fission product species cesium and ruthenium onto the smaller, respirable particles of surrogate cerium oxide; the Cs and Ru dopants appear to maximize their concentrations in the 0.25 to 1 μm ranges, decreasing as the particles increase or decrease in size. The non-volatile strontium species does not show the same, enhanced sorptive behavior.

We have optimized the surrogate cerium oxide pellets to match the physical dimensions and theoretical density of actual high-burnup spent fuel pellets to be used in upcoming Phase 4 tests. Ten or more additional Phase 2 tests, including replicates, will be performed during 2003 and 2004. These tests will incorporate the variables of nonradioactive fission product dopants and test rod internal pressurization, both at 1 and 40 bar (of He), to simulate pressurization in spent fuel rods, and potential blow-down effects on HEDD-jet ruptured cladding tubes. Test procedures, apparatus and aerosol particle collection devices used will continue to be modified, optimized, and tested for performance. Phase 2 has been expanded to also incorporate non-radioactive, German surrogate high-level waste glass test rods doped with fission product simulants; these test rods will be similarly tested for aerosolization by HEDD jet, at Sandia, and by other, lower-energy techniques, in Germany. This is a cooperative testing and analysis effort with our WGSTSC partners, the GRS and Fraunhofer ITEM.

Current and future Phase 2 surrogate aerosol test results will be used for comparison to, and “calibration” of, the Phase 3 tests results with the “final surrogate,” unirradiated depleted UO_2 pellets. DUO_2 pellets contained within six fabricated test rods will be prepared by the Institut de Radioprotection et de Surete Nucleaire, France, and sent to SNL for testing in 2004. In 2005, we will compare and calibrate the surrogate test results with Phase 4 actual spent fuel test rodlets assemblies from PWR reactor fuel. This will yield a defensible SFR determination for multiple aerosol particle size ranges. There will be eight spent fuel HEDD-aerosol tests. Four tests, as originally planned, will be performed with high burnup (~ 72 GWd/MTU) H.B. Robinson spent fuel test rodlets. Four additional tests were added in mid-2003, with agreement of the Department of Energy and other WGSTSC partners. These additional tests use low(er) burnup (~ 38 GWd/MTU) Surry spent reactor fuel. All eight of these spent fuel test rodlets are being fully characterized and fabricated by Argonne National Laboratory, for HEDD-aerosol testing at SNL.

We have described the rapid, ongoing evolution of the test apparatus used for the HEDD explosive containment and aerosol collection chambers, and the HEDD jet stop box. The apparatus has progressed from three separate components to one, cannon-barrel shaped, integrated component, with attached explosive containment and aerosol collection chambers. We are currently testing and refining an interim, prototype vertical test apparatus for Phase 2 tests, starting with tests series 2/5. Details are provided on the engineering design and requirements for the prototype explosive containment and aerosol collection chamber apparatus for the Phase 3 and Phase 4 tests. The present design being designed and tested is the “vertical elutriator” design concept, as favored by many WGSTSC participants. In addition, we have progressed from using two 3-stage RespiconTM aerosol particle collection devices plus a 9-stage Berner AESARTM particle impactor collection device to the use of multiple 9-stage Marple particle impactors plus other aerosol collection devices. This will be described and reported on at a later time.

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 waste disposal concerns. These concerns were initially evaluated for safe testing feasibility [Philbin, et al., 2002b] within the nuclear facilities at SNL. Further detailed safety and radiological assessments are currently in progress, for test conduct in Hot Cell 3 of the Gamma Irradiation Facility, GIF, at SNL; these evaluations, as well as resultant nuclear facility test procedures, shall be documented separately. These same issues significantly increase testing expense and difficulty.

In summary, this ongoing test program is part of the collaborative, international Working Group for Sabotage Concerns of Transport and Storage Casks, WGSTSC, and is supported by organizations and experts in the United States, Germany, France, and Great Britain. 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 have provided a summary of the overall, multi-phase test design and a description of all explosive containment and aerosol collection test components used. The continuing, successful conduct of this program provides significant technical and policy benefits for all participants.

Beneficially, both the U.S. DOE and NRC are providing major financial support and technical guidance to SNL, in order to both resolve these issues and to perform the spent fuel sabotage aerosol measurement tests through completion, as anticipated in 2006. German organizations,

GRS and Fraunhofer ITEM, are providing aerosol expertise, including design of the particle collectors, aerosol containment chambers, and analysis and evaluation of the aerosol particle data. They are also conducting related surrogate material aerosol particle formation and sampling, without HEDD testing. The French IRSN is providing depleted UO₂ surrogate test assemblies and expertise on CSC – fuel rod interaction. Argonne National Laboratory is providing full characterization of the spent fuel elements to be tested, and they are fabricating the spent fuel into test rodlets for testing at SNL. The OCNS, UK, participates in a consultative role and has also made available its substantial test facilities for conducting follow-on, large-scale cask plus payload testing, for a later stage to the current aerosol testing program. The design, status, and results of the overall program, as well as posttest analyses, follow-on modeling, and interpretations of the aerosol data, will be shared by all WGSTSC participants, under a formal multilateral agreement, currently under development.

APPENDIX A, Aerosol and Particle Analysis Results

A.1 Aerosol Particle Measurements, Phase 2 Tests

Table A1.1 General Test and RESPICON Particle Sampler Information

Test #	Notes: Test Modifications	Sampling Time	Respicon ID (replicates)	Flow Rate L/min	Total Filter Stages Loading
2/0	system checkout, calibration	--	(none)	--	--
2/1A	checkout, w/ 2 Respicons, 6 pellets in rodlet	60 sec	A	3.162	26.665 mg
			B	3.064	33.540 mg
2/1B	replicate of 2/1A, but w/ 5 pellets	30 sec	C	3.154	29.389 mg
			D	3.058	24.675 mg
2/2A	aerosol replicate tests, w/ 5 pellets & cladding sized to match French PWR fuel rods	30 sec	E	3.090	30.248 mg
F			3.105	26.652 mg	
2/2B		30 sec	G	3.090	24.834 mg
			H	3.090	26.303 mg
2/3A	aerosol replicate tests, 2 Respicons + Berner, w/ 9 pellets & cladding sized to match: H.B. Robinson U.S. fuel rods; new aerosol box w/ 2 valves	15 sec	I	3.111	9.165 mg
(dislodged)		J	none	no sample	
		K	3.195	16.141 mg	
2/3B		15 sec	L	3.095	21.204 mg
2/4A	replicates of 2/3A & 3B, but w/ fission product dopants added. 1 closure-hole valve	15 sec	M	3.137	4.343 mg
			N	2.992	9.863 mg
2/4B		15 sec	O	3.111	6.925 mg
			P	3.262	6.703 mg

A.1.1A Aerosol Fraction Analyses and Results, Test 2/1A

The filters were sectioned for analysis, with 3/4th of a filter digested using microwave assisted digestion using nitric acid and hydrogen peroxide. The digestate was then analyzed by ICP/MS. The total of the metals found and the weighed filter loading is provided. It may be seen that the metals detected do not exceed 50% of the material on the filter; the remainder may be oxygen, soot (carbon), and other nonmetallic species not detectable by the analytical techniques selected.

Table A1.2 Test 2/1A Respicon Respirable Fraction (0-4 μm stage) Analyses

	A, top	B, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	A	B	A	B	A	B
Cerium	1.813	0.929	28.9	19.4	12.2	5.8	3.3	3.7
Copper	3.278	3.028	52.3	63.3	22.1	18.9		
Zirconium	0.545	0.254	8.7	5.3	3.7	1.6		
Aluminum	0.354	0.335	5.7	7.0	2.4	2.1		
Iron	0.165	0.158	2.6	3.3	1.1	1.0		
Tin	0.061	0.055	1.0	1.1	0.4	0.3		
Barium	0.011	0.001	0.2	0.0	0.1	0.0		
Lead	0.036	0.024	0.6	0.5	0.2	0.2		
Metals Detected	6.263	4.784	100.0	100.0	42.2	29.9	% sums	
Filter Loading	14.847	15.999						

Table A1.3 Test 2/1A Respicon Thoracic Fraction (4-10 µm stage) Analyses

	A, middle	B, middle	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	A	B	A	B	A	B
Cerium	0.521	0.366	31.7	22.1	9.1	5.7	5.4	5.4
Copper	0.845	0.949	51.4	57.4	14.8	14.7		
Zirconium	0.097	0.068	5.9	4.1	1.7	1.1		
Aluminum	0.087	0.115	5.3	7.0	1.5	1.8		
Iron	0.063	0.086	3.8	5.2	1.1	1.3		
Tin	0.013	0.015	0.8	0.9	0.2	0.2		
Barium	0	0.043	0.0	2.6	0.0	0.7		
Lead	0.017	0.011	1.0	0.7	0.3	0.2		
Metals Detected	1.643	1.653	100.0	100.0	28.7	25.5		
Filter Loading	5.72	6.476						

Table A1.4 Test 2/1A Respicon Inhalable Fraction (10-100 µm stage) Analyses

	A, bottom	B, bottom	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	A	B	A	B	A	B
Cerium	0.398	0.429	32.0	25.6	6.5	3.9	8.5	7.9
Copper	0.542	0.901	43.5	53.9	8.9	8.1		
Zirconium	0.047	0.054	3.8	3.2	0.8	0.5		
Aluminum	0.045	0.112	3.6	6.7	0.7	1.0		
Iron	0.057	0.098	4.6	5.9	0.9	0.9		
Tin	0.006	0.012	0.5	0.7	0.1	0.1		
Barium	0.023	0.011	1.8	0.7	0.4	0.1		
Lead	0.127	0.056	10.2	3.3	2.1	0.5		
Metals Detected	1.245	1.673	100.0	100.0	20.4	15.1		
Filter Loading	6.098	11.065						

Table A1.5 Test 2/1A Distribution of Cerium on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams			
A	1.813	0.521	0.398	2.732
B	0.929	0.366	0.429	1.724
	wt%			
A	66.4	19.1	14.6	
B	53.9	21.2	24.9	

A.1.1B Aerosol Fraction Analyses and Results, Test 2/1B

The filters were sectioned for analysis, with 3/4th of a filter digested using microwave assisted digestion using nitric acid and hydrogen peroxide. The digestate was then analyzed by ICP/MS.

Table A1.6 Test 2/1B Respicon Respirable Fraction (0-4 µm stage) Analyses

	C, top	D, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	C	D	C	D	C	D
Cerium	1.006	0.694	31.2	27.5	9.1	9.5	5.4	4.3
Copper	1.678	1.377	52.1	54.6	15.1	18.9		
Zirconium	0.185	0.161	5.7	6.4	1.7	2.2		
Aluminum	0.201	0.17	6.2	6.7	1.8	2.3		
Iron	0.087	0.071	2.7	2.8	0.8	1.0		
Tin	0.031	0.026	1.0	1.0	0.3	0.4		
Barium	0.014	0.001	0.4	0.0	0.1	0.0		
Lead	0.018	0.021	0.6	0.8	0.2	0.3		
Metals Detected	3.22	2.521	100.0	100.0	29.0	34.7	% sums	
Filter Loading	11.107	7.268						

Table A1.7 Test 2/1B Respicon Thoracic Fraction (4-10 µm stage) Analyses

	C, middle	D, middle	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	C	D	C	D	C	D
Cerium	0.284	0.235	24.9	31.0	7.4	6.6	7.9	6.5
Copper	0.409	0.384	35.9	50.7	10.6	10.8		
Zirconium	0.036	0.036	3.2	4.8	0.9	1.0		
Aluminum	0.048	0.044	4.2	5.8	1.2	1.2		
Iron	0.027	0.023	2.4	3.0	0.7	0.6		
Tin	0.007	0.005	0.6	0.7	0.2	0.1		
Barium	0.32	0.024	28.1	3.2	8.3	0.7		
Lead	0.008	0.006	0.7	0.8	0.2	0.2		
Metals Detected	1.139	0.757	100.0	100.0	29.6	21.2	% sums	
Filter Loading	3.848	3.568						

Table A1.8 Test 2/1B Respicon Inhalable Fraction (10-100 µm stage) Analyses

	C, bottom	D, bottom	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	C	D	C	D	C	D
Cerium	0.75	0.507	37.7	32.4	5.2	3.7	11.2	9.4
Copper	0.924	0.786	46.4	50.2	6.4	5.7		
Zirconium	0.067	0.054	3.4	3.5	0.5	0.4		
Aluminum	0.127	0.098	6.4	6.3	0.9	0.7		
Iron	0.097	0.084	4.9	5.4	0.7	0.6		
Tin	0.01	0.009	0.5	0.6	0.1	0.1		
Barium	0	0.001	0.0	0.1	0.0	0.0		
Lead	0.017	0.026	0.9	1.7	0.1	0.2		
Metals Detected	1.992	1.565	100.0	100.0	13.8	11.3	% sums	
Filter Loading	14.434	13.839						

Table A1.9 Test 2/1B Distribution of Cerium on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams			
C	1.006	0.284	0.750	2.040
D	0.694	0.235	0.507	1.436
	wt%			
C	49.3	13.9	36.8	
D	48.3	16.4	35.3	

A.1.2A Aerosol Fraction Analyses and Results, Test 2/2A

The filters were sectioned for analysis, with ½ of a filter digested using microwave assisted digestion using nitric acid and hydrogen peroxide. The digestate was then analyzed by ICP/MS.

Table A1.10 Test 2/2A Distribution of Cerium on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams			
E	5.095	3.081	8.319	16.495
F	4.937	1.741	8.013	14.691
	wt%			
E	30.9	18.7	50.4	
F	33.6	11.9	54.5	

Table A1.11 Test 2/2A Respicon Respirable Fraction (0-4 µm stage) Analyses

	E, top	F, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	E	F	E	F	E	F
Cerium	5.095	4.937	66.6	64.9	49.1	43.2	6.4	6.3
Copper	1.484	1.575	19.4	20.7	14.3	13.8		
Zirconium	0.791	0.784	10.3	10.3	7.6	6.9		
Iron	0.150	0.172	2.0	2.3	1.4	1.5		
Magnesium	0.074	0.088	1.0	1.2	0.7	0.8		
Tin	0.029	0.036	0.4	0.5	0.3	0.3		
Lead	0.017	0.009	0.2	0.1	0.2	0.1		
Chromium	0.006	0.006	0.1	0.1	0.1	0.1		
Manganese	0.002	0.002	0.0	0.0	0.0	0.0		
Hafnium	0.001	0.001	0.0	0.0	0.0	0.0		
Terbium	0.001	0.001	0.0	0.0	0.0	0.0		
Metals Detected	7.65	7.611	100.0	100.0	73.7	66.6		
Filter Loading	10.374	11.422						

Table A1.12 Test 2/2A Respicon Thoracic Fraction (4-10 µm stage) Analyses

	E, middle	F, middle	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	E	F	E	F	E	F
Cerium	3.081	1.741	76.2	75.7	67.0	69.1	9.4	8.9
Copper	0.447	0.216	11.1	9.4	9.7	8.6		
Zirconium	0.329	0.195	8.1	8.5	7.2	7.7		
Iron	0.089	0.059	2.2	2.6	1.9	2.3		
Magnesium	0.081	0.073	2.0	3.2	1.8	2.9		
Tin	0.009	0.005	0.2	0.2	0.2	0.2		
Lead	0.004	0.006	0.1	0.3	0.1	0.2		
Chromium	0.003	0.003	0.1	0.1	0.1	0.1		
Manganese	0.001	0.001	0.0	0.0	0.0	0.0		
Hafnium	0.001	0.001	0.0	0.0	0.0	0.0		
Terbium	0	0	0.0	0.0	0.0	0.0		
Metals Detected	4.045	2.3	100.0	100.0	88.0	91.3		
Filter Loading	4.597	2.518						

Table A1.13 Test 2/2A Respicon Inhalable Fraction (10-100 µm stage) Analyses

	E, bottom	F, bottom	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	E	F	E	F	E	F
Cerium	8.319	8.013	80.7	81.0	54.5	63.0	10.2	10.5
Copper	0.815	0.833	7.9	8.4	5.3	6.6		
Zirconium	0.817	0.766	7.9	7.7	5.3	6.0		
Iron	0.203	0.162	2.0	1.6	1.3	1.3		
Magnesium	0.099	0.086	1.0	0.9	0.6	0.7		
Tin	0.01	0.01	0.1	0.1	0.1	0.1		
Lead	0.033	0.016	0.3	0.2	0.2	0.1		
Chromium	0.006	0.005	0.1	0.1	0.0	0.0		
Manganese	0.003	0.003	0.0	0.0	0.0	0.0		
Hafnium	0.001	0.001	0.0	0.0	0.0	0.0		
Terbium	0.001	0.001	0.0	0.0	0.0	0.0		
Metals Detected	10.307	9.896	100.0	100.0	67.5	77.8	%sums	
Filter Loading	15.277	12.712						

A.1.2B Aerosol Fraction Analyses and Results, Test 2/2B**Table A1.14 Test 2/2B Respicon Respirable Fraction (0-4 µm stage) Analyses**

	G, top	H, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	G	H	G	H	G	H
Cerium	3.996	4.505	62.6	62.2	42.0	44.3	5.1	5.0
Copper	1.260	1.471	19.7	20.3	13.2	14.5		
Zirconium	0.785	0.895	12.3	12.4	8.3	8.8		
Iron	0.214	0.233	3.4	3.2	2.2	2.3		
Magnesium	0.088	0.089	1.4	1.2	0.9	0.9		
Tin	0.028	0.035	0.4	0.5	0.3	0.3		
Lead	0.007	0.006	0.1	0.1	0.1	0.1		
Chromium	0.006	0.005	0.1	0.1	0.1	0.0		
Manganese	0.002	0.003	0.0	0.0	0.0	0.0		
Hafnium	0.001	0.000	0.0	0.0	0.0	0.0		
Terbium	0.001	0.001	0.0	0.0	0.0	0.0		
Metals Detected	6.388	7.243	100.0	100.0	67.2	71.2	%sums	
Filter Loading	9.512	10.179						

Table A1.15 Test 2/2B Respicon Thoracic Fraction (4-10 µm stage) Analyses

	G, middle	H, middle	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	G	H	G	H	G	H
Cerium	0.674	1.663	63.8	70.7	36.0	66.4	6.6	7.0
Copper	0.126	0.261	11.9	11.1	6.7	10.4		
Zirconium	0.102	0.238	9.7	10.1	5.4	9.5		
Iron	0.059	0.090	5.6	3.8	3.2	3.6		
Magnesium	0.085	0.086	8.0	3.7	4.5	3.4		
Tin	0.003	0.005	0.3	0.2	0.2	0.2		
Lead	0.002	0.003	0.2	0.1	0.1	0.1		
Chromium	0.003	0.003	0.3	0.1	0.2	0.1		
Manganese	0.001	0.001	0.1	0.0	0.1	0.0		
Hafnium	0.001	0.001	0.1	0.0	0.1	0.0		
Terbium	0.000	0.000	0.0	0.0	0.0	0.0		
Metals Detected	1.056	2.351	100.0	100.0	56.4	93.9	%sums	
Filter Loading	1.873	2.505						

Table A1.16 Test 2/2B Respicon Inhalable Fraction (10-100 µm stage) Analyses

	G, bottom	H, bottom	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	G	H	G	H	G	H
Cerium	8.015	8.372	79.1	80.1	59.6	61.5	9.2	10.1
Copper	0.929	0.896	9.2	8.6	6.9	6.6		
Zirconium	0.869	0.832	8.6	8.0	6.5	6.1		
Iron	0.199	0.223	2.0	2.1	1.5	1.6		
Magnesium	0.093	0.098	0.9	0.9	0.7	0.7		
Tin	0.010	0.010	0.1	0.1	0.1	0.1		
Lead	0.007	0.008	0.1	0.1	0.1	0.1		
Chromium	0.005	0.005	0.0	0.0	0.0	0.0		
Manganese	0.002	0.003	0.0	0.0	0.0	0.0		
Hafnium	0.001	0.001	0.0	0.0	0.0	0.0		
Terbium	0.001	0.001	0.0	0.0	0.0	0.0		
Metals Detected	10.131	10.449	100.0	100.0	75.3	76.7	%sums	
Filter Loading	13.449	13.619						

Table A1.17 Test 2/2B Distribution of Cerium on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams			
G	3.996	0.674	8.015	12.685
H	4.505	1.663	8.372	14.540
	wt%			
G	31.5	5.3	63.2	
H	31.0	11.4	57.6	

A.1.3A Aerosol Fraction Analyses and Results, Test 2/3A

Table A1.18 Test 2/3A Distribution of Cerium on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams			
I	1.417	1.602	2.300	5.319
J				
	wt%			
I	26.6	30.1	43.2	
J				

Table A1.19 Test 2/3A Respicon Respirable Fraction (0-4 µm stage) Analyses

	I, top	J, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	I	J	I	J	I	J
Cerium	1.417		53.0		44.5		3.3	
Copper	0.619		23.1		19.5			
Zirconium	0.430		16.1		13.5			
Aluminum	0.164		6.1		5.2			
Iron	0.027		1.0		0.8			
Tin	0.010		0.4		0.3			
Chromium	0.006		0.2		0.2			
Manganese	0.001		0.0		0.0			
Metals Detected, mg	2.674		100.0		84.0		%sums	
Filter Loading	3.182							

Table A1.20 Test 2/3A Respicon Thoracic Fraction (4-10 µm stage) Analyses

	I, middle	J, middle	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	I	J	I	J	I	J
Cerium	1.602		79.1		64.5		8.5	
Copper	0.190		9.4		7.7			
Zirconium	0.188		9.3		7.6			
Aluminum	0.026		1.3		1.0			
Iron	0.015		0.7		0.6			
Tin	0.002		0.1		0.1			
Chromium	0.003		0.1		0.1			
Manganese	0.000		0.0		0.0			
Metals Detected, mg	2.026		100.0		81.6		%sums	
Filter Loading	2.483							

Table A1.21 Test 2/3A Respicon Inhalable Fraction (10-100 µm stage) Analyses

	I, bottom	J, bottom	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	I	J	I	J	I	J
Cerium	2.300		79.6		65.7		11.7	
Copper	0.306		10.6		8.7			
Zirconium	0.196		6.8		5.6			
Aluminum	0.028		1.0		0.8			
Iron	0.045		1.6		1.3			
Tin	0.002		0.1		0.1			
Chromium	0.010		0.3		0.3			
Manganese	0.002		0.1		0.1			
Metals Detected, mg	2.889		100.0		82.5		%sums	
Filter Loading	3.500							

Berner Aerosol Particle Sampling and Results, Test 2/3A

The Berner impactor is a 9-stage aerosol collection device; refer to the schematic of this device shown in Figure 9 and A1.1. Stages 1 through 8 are loaded with 10 µm thick aluminum discs that had been treated with a silicone spray, to form an adhesive surface for the particles. Stage 9 had a small glass fiber filter that was soaked with paraffin oil to collect very large particles. The Berner impactor is shown schematically in Figure A1.1, along with the particle sizes collected. After the HEDD impact and subsequent sampling period, the Berner was disassembled and the aluminum filters and particles weighed, refer to Table A1.22, then the filters plus particles were submitted for elemental chemical analysis.

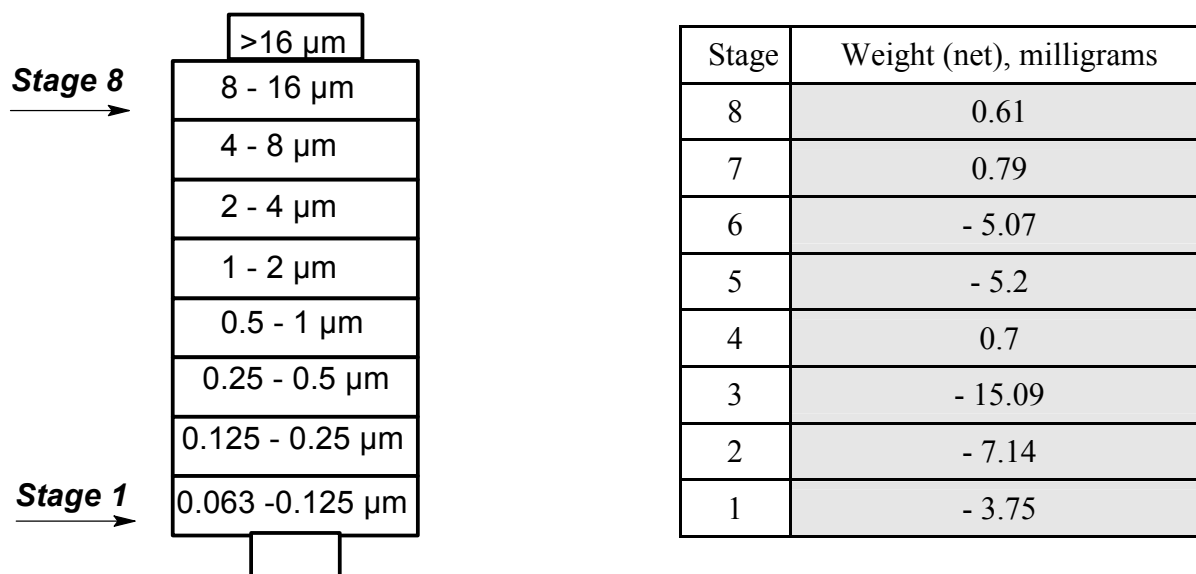


Table A1.22 Test 2/3A Berner Particle Collection

Figure A1.1 Berner Impactor Schematic and Particle Size Stages

Half of the Berner stages have measured “negative” masses.” This measurement is a test artifact, caused by excessive amounts of silicone oil spray applied to the aluminum disc, then attaching itself to adjacent hardware. Particulate material was observed and collected on each stage of the Berner; the “data” in Table A1.22 is not useful. All of the collected particulate on the Berner filters were analyzed in total using the digestion method previously described for the glass fiber filters, and were analyzed by ICP/MS for all elements except aluminum and silicon. The metals analysis from each Berner stage, in milligrams, are shown in Table A1.23.

Table A1.23 Test 2/3A Berner Particulate Metal Analysis Results, milligrams

	Stage 9	Stage 8	Stage 7	Stage 6	Stage 5	Stage 4	Stage 3	Stage 2	Stage 1
2/3A	>16μm	8-16μm	4-8μm	2-4μm	1-2μm	0.5-1μm	0.25-0.5μm	0.125-0.25μm	0.063-0.125μm
Cerium	14.698	3.923	5.663	3.704	0.500	0.079	0.084	0.031	
Copper	1.185	0.419	0.701	0.744	0.317	0.348	0.658	0.200	0.006
Zirconium	0.889	0.459	0.901	1.024	0.352	0.208	0.249	0.078	0.004
Barium	1.101								
Tin	0.008	0.005	0.007	0.007	0.006	0.010	0.022	0.007	
Chromium	0.007	0.004	0.009	0.008	0.003	0.002	0.006	0.002	
Manganese	0.005	0.006							
Magnesium	0.029		0.003						
Hafnium						0.002	0.002		0.003
Lead	0.009	0.001							
Metals Found, mg	17.931	4.817	7.284	5.487	1.178	0.649	1.021	0.318	0.013

A graph of the Berner particle metal results for Stage 8 through Stage 1 is shown in Figure A1.2. The major species are visible. There is no cerium present in the Stage 1 filter.

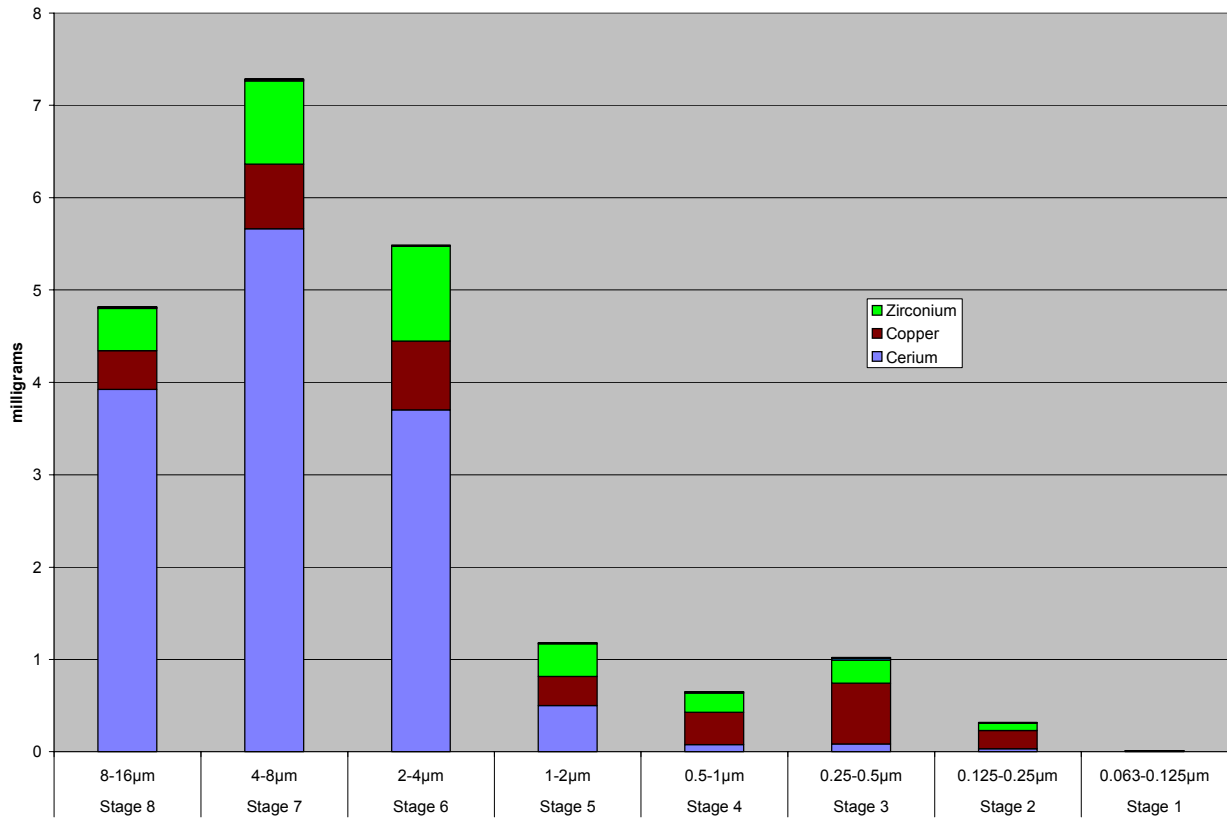


Figure A1.2 Test 2/3A Berner Particle Metal Analysis, milligrams

Table A1.24 Test 2/3A Berner Particulate Cerium Distribution

Stage	Particle Size	Ce, milligrams	wt%
9	>16µm	14.698	51.2
8	8-16µm	3.923	13.7
7	4-8µm	5.663	19.7
6	2-4µm	3.704	12.9
5	1-2µm	0.500	1.7
4	0.5-1µm	0.079	0.3
3	0.25-0.5µm	0.084	0.3
2	0.125-0.25µm	0.031	0.1
1	0.063-0.125µm	0.000	0.0
Total		28.682	

A.1.3B Aerosol Fraction Analyses and Results, Test 2/3B

Table A1.25 Test 2/3B Respicon Respirable Fraction (0-4 µm stage) Analyses

	K, top	L, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	K	L	K	L	K	L
Cerium	2.436	3.011	66.1	69.0	52.4	56.6	5.4	5.9
Copper	0.699	0.764	19.0	17.5	15.0	14.4		
Zirconium	0.455	0.508	12.3	11.6	9.8	9.5		
Iron	0.039	0.040	1.1	0.9	0.8	0.8		
Aluminum	0.032	0.019	0.9	0.4	0.7	0.4		
Tin	0.013	0.013	0.4	0.3	0.3	0.2		
Chromium	0.010	0.010	0.3	0.2	0.2	0.2		
Manganese	0.001	0.001	0.0	0.0	0.0	0.0		
Metals Detected, mg	3.685	4.366	100.0	100.0	79.2	82.0		
Filter Loading	4.651	5.322						

Table A1.26 Test 2/3B Respicon Thoracic Fraction (4-10 µm stage) Analyses

	K, middle	L, middle	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	K	L	K	L	K	L
Cerium	3.364	3.840	83.6	83.9	70.0	69.5	11.4	11.3
Copper	0.333	0.362	8.3	7.9	6.9	6.5		
Zirconium	0.294	0.340	7.3	7.4	6.1	6.2		
Iron	0.025	0.025	0.6	0.5	0.5	0.5		
Aluminum								
Tin	0.004	0.004	0.1	0.1	0.1	0.1		
Chromium	0.005	0.006	0.1	0.1	0.1	0.1		
Manganese								
Metals Detected, mg	4.025	4.577	100.0	100.0	83.7	82.8		
Filter Loading	4.809	5.528						

Table A1.27 Test 2/3B Respicon Inhalable Fraction (10-100 µm stage) Analyses

	K, bottom	L, bottom	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	K	L	K	L	K	L
Cerium	4.608	7.026	86.3	86.7	69.0	67.9	14.2	14.1
Copper	0.328	0.515	6.1	6.4	4.9	5.0		
Zirconium	0.324	0.499	6.1	6.2	4.8	4.8		
Iron	0.058	0.048	1.1	0.6	0.9	0.5		
Aluminum								
Tin	0.003	0.005	0.1	0.1	0.0	0.0		
Chromium	0.015	0.013	0.3	0.2	0.2	0.1		
Manganese	0.002	0.002	0.0	0.0	0.0	0.0		
Metals Detected, mg	5.338	8.108	100.0	100.0	79.9	78.3		
Filter Loading	6.681	10.354						

Table A1.28 Test 2/3B Distribution of Cerium on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams			
K	2.436	3.364	4.608	10.408
L	3.011	3.84	7.026	13.877
	wt%			
K	23.4	32.3	44.3	
L	21.7	27.7	50.6	

Berner Aerosol Particle Sampling and Results, Test 2/3B

Analyses of the Berner impactor particle stages was as described for Test 2/3A. Again, a majority of the Berner stages have “negative” mass, so the data is not presented; this is due to excessive amounts of silicone applied to the aluminum discs. Material was collected on each stage of the Berner and analyzed.

The analyzed metal results for Berner stages 9 through 1 are tabulated in Table A1.29. A graph of the metal results of Stage 8 through Stage 1 is shown in Figure A1.3. The major species are visible. There is no cerium present in the Stage 2 and Stage 1 filters.

Table A1.29 Test 2/3B Berner Collector Metal Analysis Results, milligrams

	Stage 9	Stage 8	Stage 7	Stage 6	Stage 5	Stage 4	Stage 3	Stage 2	Stage 1
2/3B	>16µm	8-16µm	4-8µm	2-4µm	1-2µm	0.5-1µm	0.25-0.5µm	0.125-0.25µm	0.063-0.125µm
Cerium	15.062	3.344	4.780	3.656	0.437	0.106	0.056		
Copper	0.893	0.271	0.410	0.471	0.213	0.216	0.326	0.191	0.022
Zirconium	0.964	0.307	0.561	0.683	0.221	0.101	0.127	0.056	0.005
Iron	1.083								
Barium	0.993								
Tin	0.008	0.004	0.005	0.005	0.004	0.004	0.013	0.005	
Chromium	0.423	0.004	0.006	0.006	0.003	0.001	0.003	0.002	
Manganese	0.024								
Magnesium	0.026								
Terbium	0.002								
Lead		0.001							
Metals Found	19.478	3.931	5.762	4.821	0.878	0.428	0.525	0.254	0.027

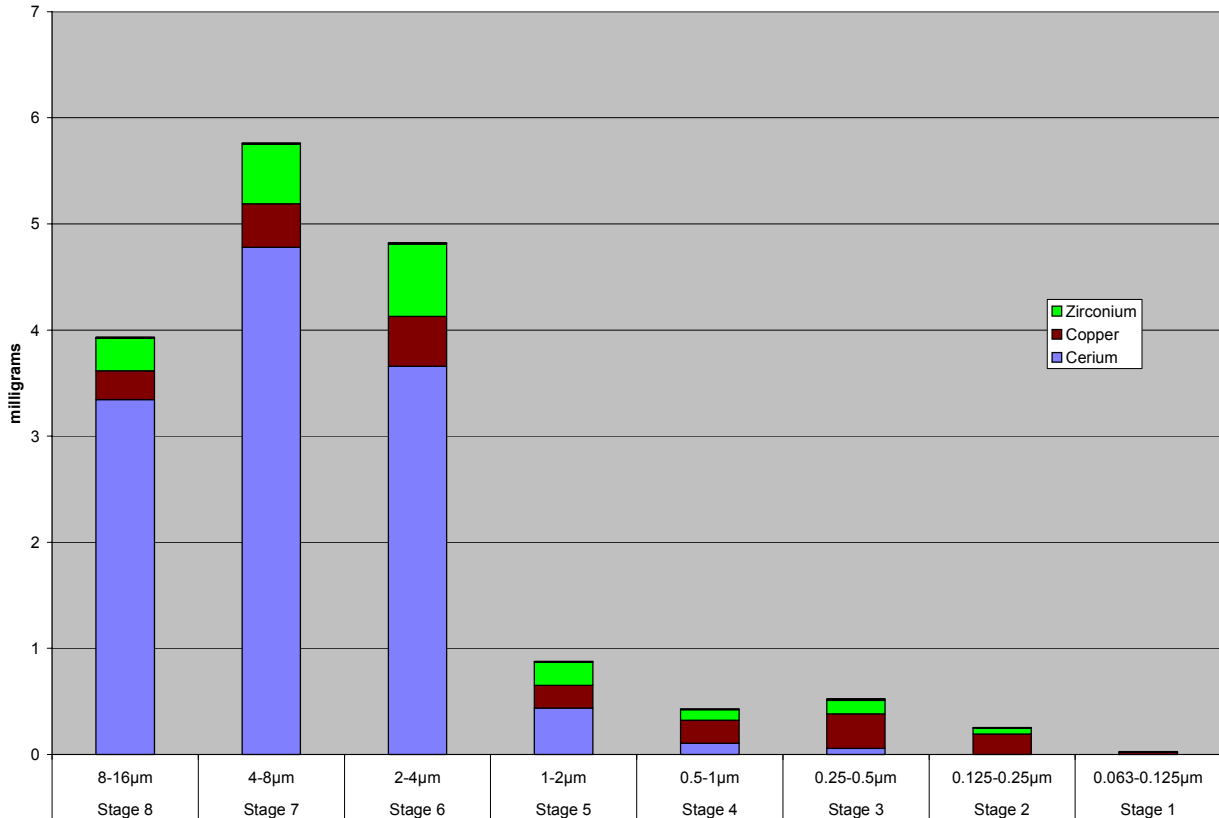


Figure A1.3 Test 2/3B Berner Particle Metal Analyses, milligrams

The distribution of cerium through the Berner is shown in Table A1.30.

Table A1.30 Test 2/3B Berner Particulate Cerium Distribution

Stage	Particle Size	milligrams	wt%
9	>16µm	15.062	54.9
8	8-16µm	3.344	12.2
7	4-8µm	4.780	17.4
6	2-4µm	3.656	13.3
5	1-2µm	0.437	1.6
4	0.5-1µm	0.106	0.4
3	0.25-0.5µm	0.056	0.2
2	0.125-0.25µm	0.000	0.0
1	0.063-0.125µm	0.000	0.0
Total		27.441	

A.1.4A Aerosol Fraction Analyses and Results, Test 2/4A

Table A1.31 Test 2/4A Respicon Respirable Fraction (0-4 µm stage) Analyses

	M, top	N, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	M	N	M	N	M	N
Cerium	1.1257	1.5012	51.4	54.6	40.2	35.8	4.1	4.5
Copper	0.6240	0.7071	28.5	25.7	22.3	16.9		
Zirconium	0.2726	0.3318	12.4	12.1	9.7	7.9		
Iron	0.0922	0.1181	4.2	4.3	3.3	2.8		
Cesium	0.0388	0.0464	1.8	1.7	1.4	1.1		
Strontium	0.0129	0.0115	0.6	0.4	0.5	0.3		
Tin	0.0120	0.0139	0.5	0.5	0.4	0.3		
Chromium	0.0086	0.0113	0.4	0.4	0.3	0.3		
Nickel	0.0018	0.0028	0.1	0.1	0.1	0.1		
Ruthenium	0.0013	0.0008	0.1	0.0	0.0	0.0		
Hafnium	0.0004	0.0004	0.0	0.0	0.0	0.0		
Molybdenum	0.0003	0.0004	0.0	0.0	0.0	0.0		
Terbium	0.0002	0.0002	0.0	0.0	0.0	0.0		
Manganese	0.0001	0.0011	0.0	0.0	0.0	0.0		
Metals Detected, mg	2.1913	2.7470	100.0	100.0	78.2	65.5	%sums	
Filter Loading	2.803	4.193						

Table A1.32 Test 2/4A Respicon Thoracic Fraction (4-10 µm stage) Analyses

	M, top	N, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	M	N	M	N	M	N
Cerium	0.2228	0.4859	64.1	65.0	53.0	45.9	4.6	5.1
Copper	0.0564	0.1355	16.2	18.1	13.4	12.8		
Zirconium	0.0489	0.0956	14.1	12.8	11.6	9.0		
Iron	0.0000	0.0012	0.0	0.2	0.0	0.1		
Cesium	0.0023	0.0058	0.7	0.8	0.5	0.5		
Strontium	0.0132	0.0130	3.8	1.7	3.1	1.2		
Tin	0.0011	0.0023	0.3	0.3	0.3	0.2		
Chromium	0.0024	0.0058	0.7	0.8	0.6	0.5		
Nickel	0.0000	0.0010	0.0	0.1	0.0	0.1		
Ruthenium	0.0001	0.0004	0.0	0.1	0.0	0.0		
Hafnium	0.0005	0.0004	0.1	0.1	0.1	0.0		
Molybdenum								
Terbium	0.0000	0.0001	0.0	0.0	0.0	0.0		
Manganese								
Metals Detected, mg	0.3477	0.747	100.0	100.0	82.8	70.5	%sums	
Filter Loading	0.4200	1.059						

Table A1.33 Test 2/4A Respicon Inhalable Fraction (10-100 µm stage) Analyses

	M, top	N, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	M	N	M	N	M	N
Cerium	0.6290	1.2481	73.1	72.6	56.2	27.1	6.6	9.2
Copper	0.0883	0.1852	10.3	10.8	7.9	4.0		
Zirconium	0.0948	0.1357	11.0	7.9	8.5	2.9		
Iron	0.0246	0.0804	2.9	4.7	2.2	1.7		
Cesium	0.0020	0.0036	0.2	0.2	0.2	0.1		
Strontium	0.0119	0.0096	1.4	0.6	1.1	0.2		
Tin	0.0015	0.0020	0.2	0.1	0.1	0.0		
Chromium	0.0062	0.0357	0.7	2.1	0.6	0.8		
Nickel	0.0013	0.0121	0.2	0.7	0.1	0.3		
Ruthenium	0.0002	0.0007	0.0	0.0	0.0	0.0		
Hafnium	0.0006	0.0005	0.1	0.0	0.1	0.0		
Molybdenum	0.0000	0.0002	0.0	0.0	0.0	0.0		
Terbium								
Manganese								
Metals Detected, mg	0.8605	1.7191	100.0	100.0	76.8	37.3	%sums	
Filter Loading	1.1200	4.6110						

Table A1.34 Test 2/4A Distribution of Cerium on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams			
M	1.1257	0.2228	0.629	1.9775
N	1.5012	0.4859	1.2481	3.2350
	wt%			
M	56.9	11.3	31.8	
N	46.4	15.0	38.6	

Table A1.35 Test 2/4A Distribution of Fission Product Dopants on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams	CESIUM		
M	0.0388	0.0023	0.0020	0.0431
N	0.0464	0.0058	0.0036	0.0560
	wt%			
M	90.0	5.3	4.6	
N	83.2	10.4	6.5	
	milligrams	RUTHENIUM		
M	0.0013	0.0001	0.0002	0.0016
N	0.0008	0.0004	0.0007	0.0020
	wt%			
M	81.3	6.3	12.5	
N	42.1	21.1	36.8	
	milligrams	STRONTIUM		
M	0.0129	0.0132	0.0119	0.0380
N	0.0115	0.0130	0.0096	0.0340
	wt%			
M	33.9	34.7	31.3	
N	33.7	38.1	28.2	
	milligrams	IODINE		
M	(not detected)			
N	(not detected)			
	wt%			
M	(not detected)			
N	(not detected)			

Berner Aerosol Particle Sampling and Results, Test 2/4A

Some of the Berner stages have ‘negative’ mass; this is due to excessive amounts of silicone applied to the aluminum discs. Material was collected on each stage of the Berner. All of the Berner filters were analyzed in total using the digestion method previously described for the glass fiber filters, and were analyzed by ICP/MS for all elements except aluminum and silicon. The metals analysis from each Berner stage, in milligrams, are shown in Table A1.36.

Table A1.36 Test 2/4A Berner Particulate Metal Analysis Results, milligrams

	Stage 9	Stage 8	Stage 7	Stage 6	Stage 5	Stage 4	Stage 3	Stage 2	Stage 1
2/4A	>16µm	8-16µm	4-8µm	2-4µm	1-2µm	0.5-1µm	0.25-0.5µm	0.125-0.25µm	0.063-0.125µm
Cerium	2.2801	1.3641	2.3977	1.8960	0.1665	0.0983	0.0504	0.0081	0.0008
Copper	0.1796	0.2348	0.4178	0.4692	0.2607	0.2625	0.6353	0.1065	0.0124
Zirconium	0.1680	0.1943	0.3923	0.4213	0.1429	0.0814	0.1275	0.0221	0.0032
Iron			0.1973	0.1509		0.0110	0.0759	0.1400	0.1299
Magnesium	0.1906		0.0048	0.0000					0.0012
Chromium		0.0016	0.0046	0.0030	0.0012	0.0015	0.0038		
Nickel	0.0010	0.0006	0.0016	0.0010			0.0005		
Manganese			0.0014	0.0008		0.0026			0.0004
Tin	0.0002	0.0004	0.0013	0.0013	0.0013		0.0078		
Terbium		0.0002	0.0006	0.0003					
Molybdenum							0.0004		
Lead									0.0004
Cesium	0.0011	0.0018	0.0028	0.0054	0.0020	0.0067	0.0105	0.0031	0.0007
Strontium	0.0119	0.0008	0.0017	0.0020	0.0000	0.0002	0.0001	0.0000	0.0000
Ruthenium	0.0005	0.0010	0.0024	0.0014	0.0021	0.0021	0.0033	0.0007	0.0001
Metals Found, mg	2.8330	1.7996	3.4263	2.9526	0.5767	0.4663	0.9155	0.2805	0.1491

A graph of the Berner particle metal results for Stage 9 through Stage 1 is shown in Figure A1.4. The major species cerium, copper (from the HEDD), and zirconium (cladding tube) are visible, as are iron, magnesium, and other lesser species.

The distribution of cerium measured throughout the Berner particle stages is shown in Table A1.37. Similarly, the analyzed distribution of the fission product dopants cesium, ruthenium, and strontium are shown in Table A1.38. The fission product dopant iodine was not detected. A graph of the Berner particle fission product dopant results for Stage 9 through Stage 1 is shown in Figure A1.5.

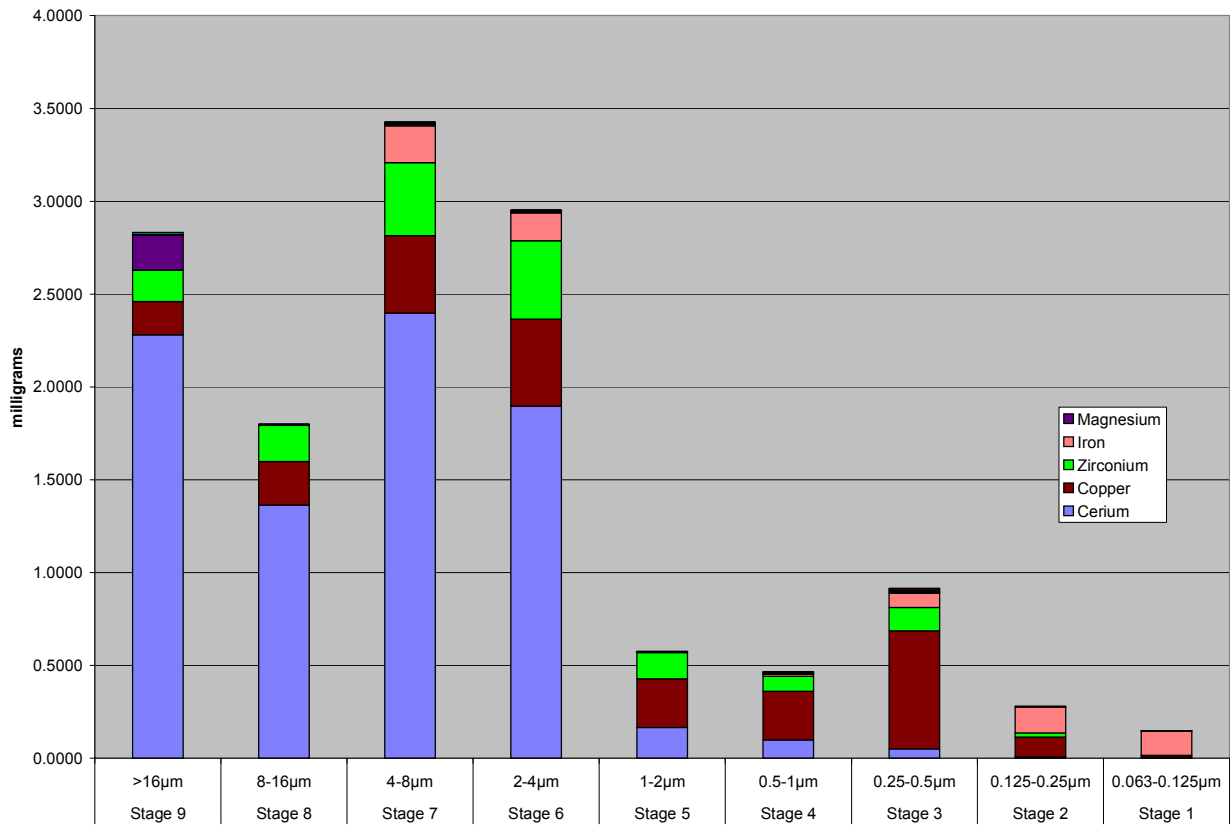


Figure A1.4 Test 2/4A Berner Particle Metal Analysis, milligrams

Table A1.37 Test 2/4A Berner Particulate Cerium Distribution

Stage	Particle Size	Ce, milligrams	wt%
9	>16µm	2.2801	27.6
8	8-16µm	1.3641	16.5
7	4-8µm	2.3977	29.0
6	2-4µm	1.8960	22.9
5	1-2µm	0.1665	2.0
4	0.5-1µm	0.0983	1.2
3	0.25-0.5µm	0.0504	0.6
2	0.125-0.25µm	0.0081	0.1
1	0.063-0.125µm	0.0008	0.0
Total		8.2620	99.9

Table A1.38 Test 2/4A Berner Particulate Fission Product Dopant Distribution

Stage	Particle Size	milligrams	wt%	milligrams	wt%	milligrams	wt%
		CESIUM		RUTHENIUM		STRONTIUM	
9	>16µm	0.0011	3.2	0.0005	3.7	0.0119	71.3
8	8-16µm	0.0018	5.3	0.0010	7.4	0.0008	4.8
7	4-8µm	0.0028	8.2	0.0024	17.6	0.0017	10.2
6	2-4µm	0.0054	15.8	0.0014	10.3	0.0020	12.0
5	1-2µm	0.0020	5.9	0.0021	15.4	0.0000	0.0
4	0.5-1µm	0.0067	19.6	0.0021	15.4	0.0002	1.2
3	0.25-0.5µm	0.0105	30.8	0.0033	24.3	0.0001	0.6
2	0.125-0.25µm	0.0031	9.1	0.0007	5.1	0.0000	0.0
1	0.063-0.125µm	0.0007	2.1	0.0001	0.7	0.0000	0.0
Totals		0.0341	100.0	0.0136	99.9	0.0167	100.1

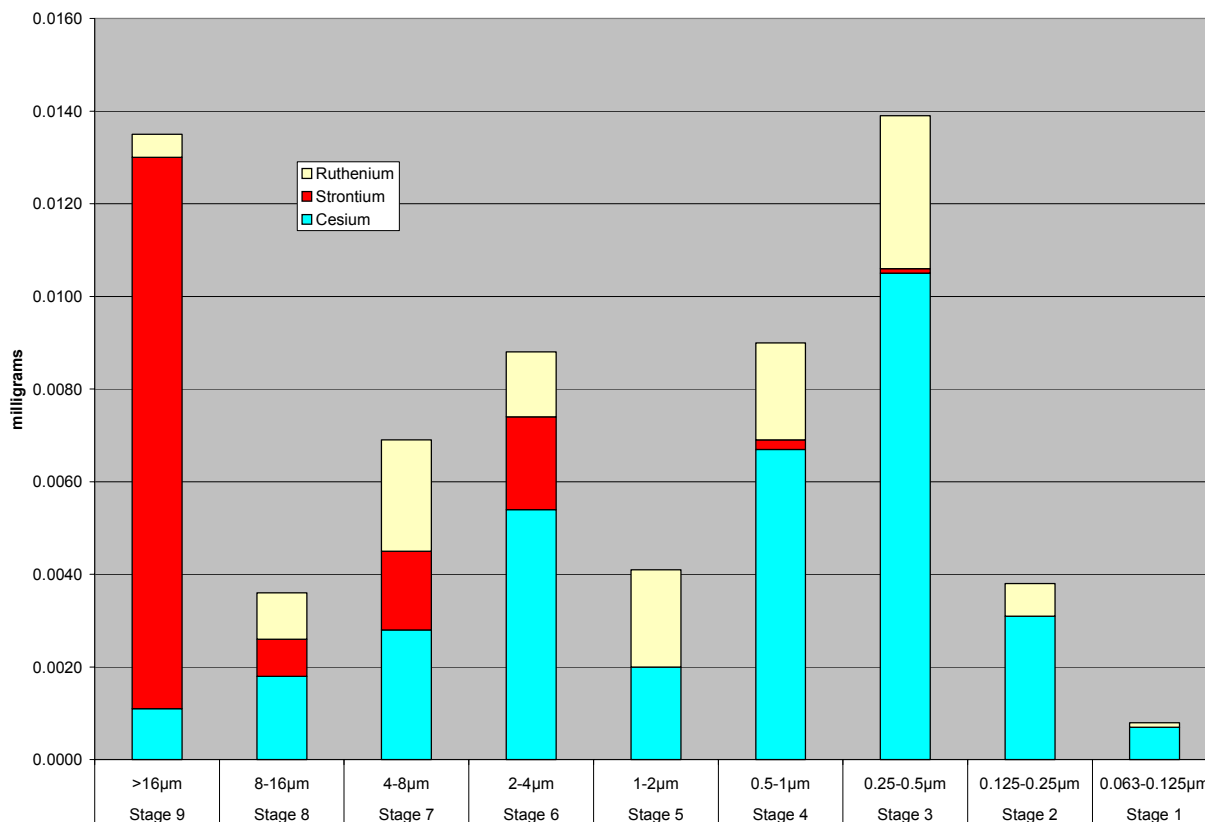


Figure A1.5 Test 2/4A Berner Particle Fission Product Dopant Analyses, milligrams

A.1.4B Aerosol Fraction Analyses and Results, Test 2/4B

Table A1.39 Test 2/4B Respicon Respirable Fraction (0-4 µm stage) Analyses

	O, top	P, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	O	P	O	P	O	P
Cerium	1.0748	0.8632	37.7	38.5	24.7	24.8	3.9	3.9
Copper	1.1713	0.9011	41.1	40.2	26.9	25.9		
Zirconium	0.2742	0.2195	9.6	9.8	6.3	6.3		
Iron	0.246	0.2021	8.6	9.0	5.7	5.8		
Cesium	0.0329	0.0236	1.2	1.1	0.8	0.7		
Strontium	0.0113	0.0085	0.4	0.4	0.3	0.2		
Tin	0.0144	0.0096	0.5	0.4	0.3	0.3		
Chromium	0.0125	0.0092	0.4	0.4	0.3	0.3		
Nickel	0.0028	0.002	0.1	0.1	0.1	0.1		
Ruthenium	0.0004	0.0006	0.0	0.0	0.0	0.0		
Hafnium	0.0004	0.0003	0.0	0.0	0.0	0.0		
Molybdenum	0.0008	0.0005	0.0	0.0	0.0	0.0		
Terbium	0.0002	0.0002	0.0	0.0	0.0	0.0		
Manganese	0.0056	0.0034	0.2	0.2	0.1	0.1		
Metals Detected, mg	2.8476	2.2438	100.0	100.0	65.4	64.5	%sums	
Filter Loading	4.351	3.477						

Table A1.40 Test 2/4B Respicon Thoracic Fraction (4-10 µm stage) Analyses

	O, top	P, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	O	P	O		mg	mg
Cerium	0.3331	0.3654	52.3	51.5	36.3	36.7	4.0	4.2
Copper	0.1583	0.1866	24.8	26.3	17.2	18.8		
Zirconium	0.0836	0.0870	13.1	12.3	9.1	8.7		
Iron	0.0440	0.0509	6.9	7.2	4.8	5.1		
Cesium	0.0033	0.0041	0.5	0.6	0.4	0.4		
Strontium	0.0084	0.0092	1.3	1.3	0.9	0.9		
Tin	0.0014	0.0014	0.2	0.2	0.2	0.1		
Chromium	0.0042	0.0039	0.7	0.5	0.5	0.4		
Nickel	0.0006	0.0005	0.1	0.1	0.1	0.1		
Ruthenium	0.0001	0.0002	0.0	0.0	0.0	0.0		
Hafnium	0.0003	0.0003	0.0	0.0	0.0	0.0		
Molybdenum								
Terbium	0.0001	0.0001	0.0	0.0	0.0	0.0		
Manganese								
Metals Detected, mg	0.6374	0.7096	100.0	100.0	69.4	71.3	%sums	
Filter Loading	0.9180	0.9950						

Table A1.41 Test 2/4B Respicon Inhalable Fraction (10-100 µm stage) Analyses

	O, top	P, top	wt% detected		wt% on filter		Ce/Zr	
	mg	mg	O	P	O		mg	mg
Cerium	0.8071	0.9236	67.3	71.7	48.7	40.7	6.0	8.2
Copper	0.1676	0.1494	14.0	11.6	10.1	6.6		
Zirconium	0.1345	0.1127	11.2	8.7	8.1	5.0		
Iron	0.0612	0.0757	5.1	5.9	3.7	3.3		
Cesium	0.0019	0.0016	0.2	0.1	0.1	0.1		
Strontium	0.0099	0.0119	0.8	0.9	0.6	0.5		
Tin	0.0016	0.0027	0.1	0.2	0.1	0.1		
Chromium	0.0113	0.0077	0.9	0.6	0.7	0.3		
Nickel	0.0027	0.0013	0.2	0.1	0.2	0.1		
Ruthenium	0.0001	0.0002	0.0	0.0	0.0	0.0		
Hafnium	0.0000	0.0000	0.0	0.0	0.0	0.0		
Molybdenum	0.0005	0.0005	0.0	0.0	0.0	0.0		
Terbium	0.0002	0.0002	0.0	0.0	0.0	0.0		
Manganese	0.0013	0.0009	0.1	0.1	0.1	0.0		
Metals Detected, mg	1.1999	1.2884	100.0	100.0	72.5	56.7	%sums	
Filter Loading	1.6560	2.2710						

Table A1.42 Test 2/4B Distribution of Cerium on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams			
O	1.0748	0.3331	0.8071	2.215
P	0.8632	0.3654	0.9236	2.152
	wt%			
O	48.5	15.0	36.4	
P	40.1	17.0	42.9	

Table A1.43 Test 2/4B Distribution of Fission Product Dopants on Respicon Filters

	Respirable	Thoracic	Inhalable	
	<i>Top</i>	<i>Middle</i>	<i>Bottom</i>	Total
	milligrams	CESIUM		
O	0.0329	0.0033	0.0019	0.0381
P	0.0236	0.0041	0.0016	0.029
	wt%			
O	86.4	8.7	5.0	
P	80.5	14.0	5.5	
	milligrams	RUTHENIUM		
O	0.0004	0.0001	0.0001	0.0006
P	0.0006	0.0002	0.0002	0.001
	wt%			
O	66.7	16.7	16.7	
P	60.0	20.0	20.0	
	milligrams	STRONTIUM		
O	0.0113	0.0084	0.0099	0.0296
P	0.0085	0.0092	0.0119	0.030
	wt%			
O	38.2	28.4	33.4	
P	28.7	31.1	40.2	
	milligrams	IODINE		
O	(not detected)			
P	(not detected)			
	wt%			
O	(not detected)			
P	(not detected)			

Berner Aerosol Particle Sampling and Results, Test 2/4B

Table A1.44 Test 2/4B Berner Particulate Metal Analysis Results, milligrams

	Stage 9	Stage 8	Stage 7	Stage 6	Stage 5	Stage 4	Stage 3	Stage 2	Stage 1
2/4B	>16μm	8-16μm	4-8μm	2-4μm	1-2μm	0.5-1μm	0.25-0.5μm	0.125-0.25μm	0.063-0.125μm
Cerium	8.8112	0.3422	1.5828	1.0483	0.2329	0.4914	0.2608	0.0126	
Copper	0.9292	0.2368	0.4428	0.5138	0.4255	1.3242	1.0743	0.0444	
Zirconium	0.6618	0.1484	0.3476	0.3990	0.1703	0.1347	0.0870	0.0062	0.0001
Iron		0.0488	0.1214	0.0194	0.0489	0.0000	0.0683		
Magnesium	0.0324	0.0047	0.0020	0.0016	0.0095	0.0083	0.0111	0.0063	
Chromium	0.0077	0.0030	0.0039	0.0043	0.0035	0.0084	0.0071	0.0008	
Nickel	0.0027	0.0012	0.0014	0.0015	0.0011	0.0018	0.0014	0.0004	
Manganese		0.0029	0.0018	0.0016	0.0028	0.0052	0.0048	0.0014	
Tin	0.0044		0.0025	0.0035	0.0029	0.0114	0.0100		
Terbium	0.0011	0.0001		0.0002	0.0001	0.0002			
Molybdenum				0.0002	0.0003	0.0010	0.0008	0.0001	
Titanium		0.0026		0.0005				0.0008	0.0005
Hafnium	0.0001								
Barium		0.0002				0.0001			
Lead	0.0016	0.0005	0.0002	0.0002	0.0001	0.0002	0.0002	0.0001	
Cesium	0.0040	0.0005	0.0025	0.0040	0.0043	0.0125	0.0083	0.0010	0.0001
Strontium	0.0225	0.0002	0.0009	0.0006	0.0002	0.0003	0.0002	0.0000	0.0000
Ruthenium	0.0042	0.0008	0.0005	0.0009	0.0012	0.0023	0.0016	0.0002	0.0000
Metals Found, mg	10.4829	0.7929	2.5103	1.9996	0.9036	2.0020	1.5359	0.0743	0.0007

A graph of the Berner particle metal results for Stage 9 through Stage 1 is shown in Figure A1.6. The major species cerium, copper, and zirconium are visible, as are iron and magnesium.

The distribution of cerium measured throughout the Berner particle stages is shown in Table A1.45. Similarly, the analyzed distribution of the fission product dopants cesium, ruthenium, and strontium are shown in Table A1.46. The fission product dopant iodine was again not detected. A graph of the Berner particle fission product results for Stage 9 through Stage 1 is shown in Figure A1.7.

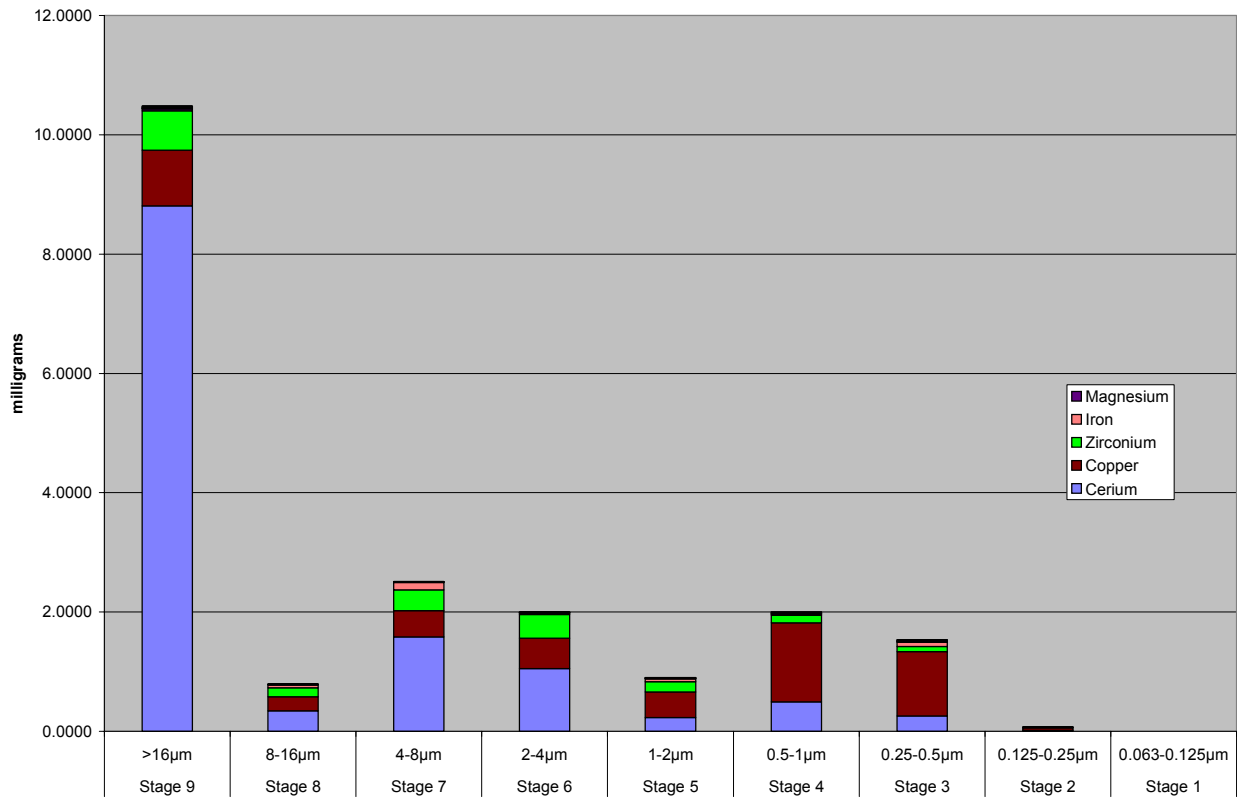


Figure A1.6 Test 2/4B Berner Particle Metal Analysis, milligrams

Table A1.45 Test 2/4B Berner Particulate Cerium Distribution

Stage	Particle Size	Ce, milligrams	wt%
9	>16µm	8.8112	68.9
8	8-16µm	0.3422	2.7
7	4-8µm	1.5828	12.4
6	2-4µm	1.0483	8.2
5	1-2µm	0.2329	1.8
4	0.5-1µm	0.4914	3.8
3	0.25-0.5µm	0.2608	2.0
2	0.125-0.25µm	0.0126	0.1
1	0.063-0.125µm	0.0000	0.0
Total		12.7822	99.9

Table A1.46 Test 2/4B Berner Particulate Fission Product Dopant Distribution

Stage	Particle Size	milligrams	wt%	milligrams	wt%	milligrams	wt%
		CESIUM		RUTHENIUM		STRONTIUM	
9	>16µm	0.0040	10.8	0.0042	35.9	0.0225	90.4
8	8-16µm	0.0005	1.3	0.0008	6.8	0.0002	0.8
7	4-8µm	0.0025	6.7	0.0005	4.3	0.0009	3.6
6	2-4µm	0.0040	10.8	0.0009	7.7	0.0006	2.4
5	1-2µm	0.0043	11.6	0.0012	10.3	0.0002	0.8
4	0.5-1µm	0.0125	33.6	0.0023	19.7	0.0003	1.2
3	0.25-0.5µm	0.0083	22.3	0.0016	13.7	0.0002	0.8
2	0.125-0.25µm	0.0010	2.7	0.0002	1.7	0.0000	0.0
1	0.063-0.125µm	0.0001	0.3	0.0000	0.0	0.0000	0.0
Totals		0.0372	100.1	0.0117	100.1	0.0249	100.02

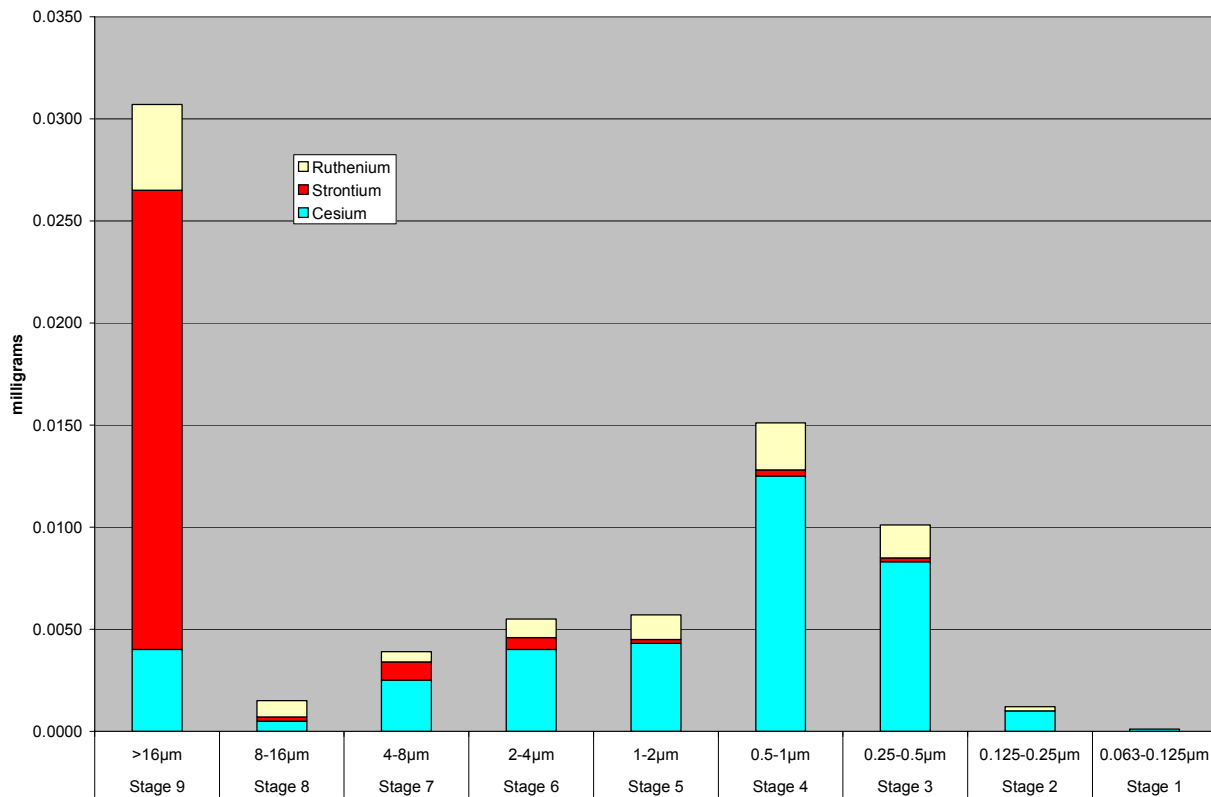


Figure A1.7 Test 2/4B Berner Particle Fission Product Dopant Analyses, milligrams

A.2 Impact Debris Particle Measurements, Phase 2 Tests

The particulate materials remaining in the aerosol collection box after each test were collected, mechanically sieved, weighed, and chemically analyzed by ICP/MS, in order to determine how much material was fragmented, particulated, and aerosolized. This impact debris contains all of this material minus the aerosols collected in the aerosol particle impactors minus any material which either escaped through the HEDD jet entrance and exit holes in the box or were incompletely sampled and lost.

After sieving, the different fractions were examined using an optical microscope. The pieces collected from the 1.00 mm sieve are mainly Zircaloy tube fragments from the impact region and residual copper pieces from the conical shape charge. There were no obviously visible pieces from the cerium oxide pellet that in this size region; these were later quantified with ICP-MS.

Starting with Test 2/2A and continuing onward, the collected impact debris material was sieved first using a set of 48mm-diameter metal sieves with progressively smaller mesh openings; 1.000 mm, 0.500 mm, 0.250 mm, and 0.125 mm with a final catch pan, i.e., < 0.125 mm. These ‘fines’ were then sieved further with disposable, smaller-screen mesh at 0.149 mm, 0.074 mm, 0.044 mm, 0.037 mm, and <0.037 mm to further differentiate the debris. There is usually a slight amount of particles from the < 0.125 mm fines that was retained on the ostensibly larger pore-size 0.149 mm disposable sieve.

The mechanical sieves used separate fine materials based on geometric particle size. Table A2.1 provides a conversion for geometric diameter (size) to aerodynamic equivalent diameter, AED, for both cerium oxide ceramic pellets and uranium oxide fuel pellets.

Table A2.1 Conversion Chart, Geometric to Aerosol Equivalent Diameters

Particle Density		Particle Density (95%)	Particle Density (95%)
1.00		6.78	10.41
Geometric Diameter		AED CeO₂	AED UO₂
(μm)	Screen Size	(μm)	(μm)
1000	1.000 mm	2600	3230
500	0.500 mm	1300	1610
250	0.250 mm	650	807
125	0.125 mm	325	403
149	100 mesh	388	481
74	200 mesh	193	239
44	325 mesh	115	142
37	400 mesh	96	119
25	600 mesh	65	81
AED = (Geometric Diam.) X (density) ^{1/2}			

A.2.0 Impact Debris in Aerosol Box, Analyses and Results, Test 2/0

Each sieved fraction, with the exception of the material collected in the 1.000 mm sieve, was homogenized by grinding with a mortar and pestel. An approximately 0.05 gram portion of the ground fraction was digested in a Teflon beaker using 8mL of concentrated nitric acid (HNO₃) and 5mL of DI water and refluxing for 30 minutes. After cooling, 2 mL of 30% hydrogen peroxide (H₂O₂) was added, and the mixture heated for an additional 15 minutes. After cooling the second time, 0.5mL of conc. hydrofluoric acid (HF) was added, and this mixture was heated for 15 minutes. This digestate was then diluted to 100 grams with DI water and analyzed by ICP/MS. In all cases, an amount of solid material in the digestate was observed. This solid material was filtered, washed, dried, and weighed. It is reported as “solids,” and is considered to be cerium oxide. Analysis of a portion of this material by energy dispersive spectroscopy shows that it is composed of cerium and some fluoride. While suspending this material for weighing, it all became a uniform brown color, which is the color of cerium oxide. Therefore, the undissolved solids residue is considered to be cerium oxide.

The elemental analysis results and residue weighings as weight percent of metal are summarized in the following tables. The “solids” have been converted to cerium metal from cerium oxide. The amount of cerium increases with decreasing particle size. There is a large, unknown weight percentage in the 0.125 mm fraction; it is assumed to be carbonaceous soot from the shaped charge.

Table A2.2 Test 2/0, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%	Cerium as CeO ₂	Undissolved Solids as CeO ₂	Total CeO ₂
1.000 mm	3.7745 g	44.6	0 wt %	0 wt %	0 wt %
0.500 mm	0.5987 g	7.1	37.5 wt %	7.7 wt %	45.2 wt %
0.250 mm	0.6774 g	8.0	47.5 wt %	24.9 wt %	72.4 wt %
0.125 mm	0.7092 g	8.4	34.4 wt %	28.0 wt %	62.4 wt %
< 0.125 mm	2.7006 g	31.9	23.7 wt %	70.2 wt %	93.9 wt %
Total	8.4604 g				

Table A2.3 Test 2/0, Elemental Analysis wt% of Sieved Impact Debris

Sieve Fraction	0.500 mm	0.250 mm	0.125 mm	<0.125 mm
Cerium	30.55	38.64	28.03	19.32
Solids as Cerium	6.26	20.30	22.76	57.14
Copper	10.19	7.513	2.159	3.649
Zirconium	37.8	5.812	1.224	4.536
Aluminum	1.353	4.257	1.765	1.141
Iron	3.62	4.091	0.094	0.452
Tin	0.471	3.293	0	0.041
Barium	0.036	0.073	0.049	0
Lead	0.142	0.056	0.11	0.005
Total	90.4	84.0	56.2	86.3

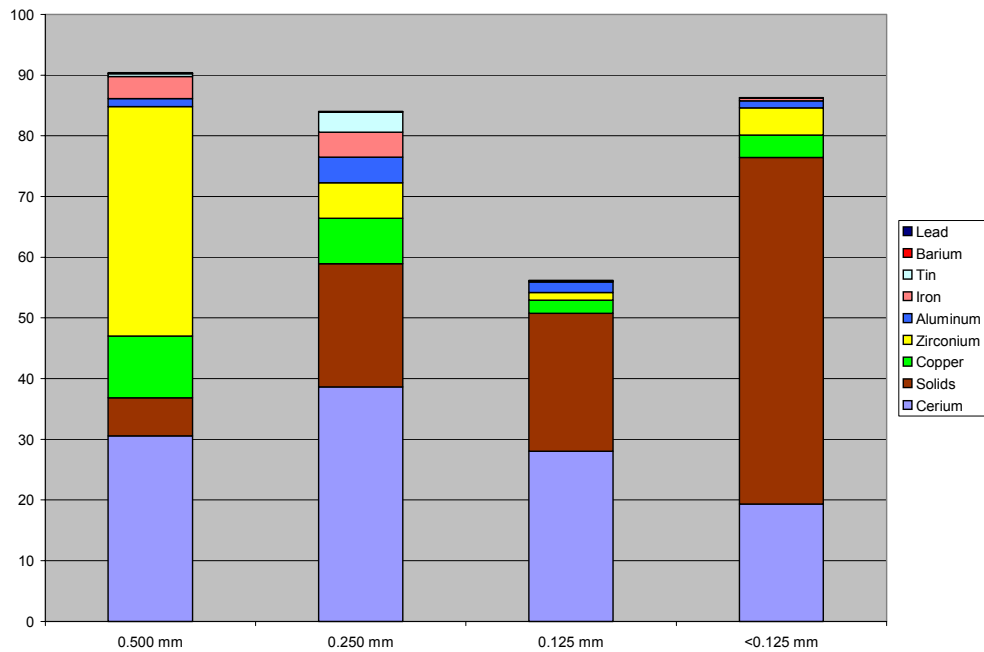


Figure A2.1. Test 2/0, Distribution of Metals in Sieved Fractions

A.2.1A Impact Debris in Aerosol Box, Analyses and Results, Test 2/1A

Table A2.4 Test 2/1A, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%	Cerium as CeO ₂	Undissolved Solids as CeO ₂	Total CeO ₂
1.000 mm	9.7224 g	67.8	0 wt %	0 wt %	0 wt %
0.500 mm	0.6271 g	4.4	8.7 wt %	2.4 wt %	11.2 wt %
0.250 mm	0.4536 g	3.2	36.7 wt %	21.8 wt %	58.5 wt %
0.125 mm	0.4117 g	2.9	47.4 wt %	16.1 wt %	63.5 wt %
< 0.125 mm	3.1216 g	21.8	56.0 wt %	30.0 wt %	86.0 wt %
Total	14.3364 g				

Table A2.5 Test 2/1A, Elemental Analysis wt% of Sieved Impact Debris

Sieve Fraction	0.500 mm	0.250 mm	0.125 mm	<0.125 mm
Cerium	7.12	29.89	38.57	45.58
Solids as Cerium	1.98	17.75	13.14	24.41
Copper	49.20	25.11	6.10	5.33
Zirconium	39.71	8.60	3.62	5.47
Aluminum	3.13	8.17	3.37	1.23
Iron	0.25	3.50	0.63	0.72
Tin	0.56	0.09	0.02	0.07
Barium	0.00	0.73	0.22	0.00
Lead	0.00	0.08	0.06	0.09
Total	101.9	93.9	65.7	82.9

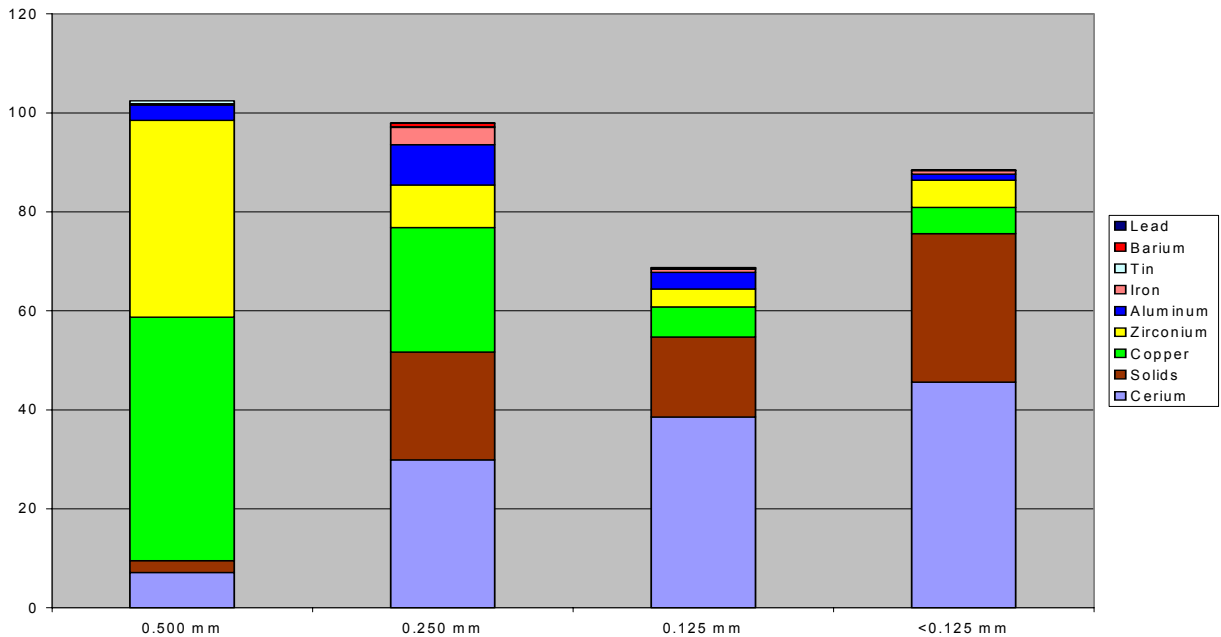


Figure A2.2. Test 2/1A, Distribution of Metals in Sieved Fractions

A.2.1B Impact Debris in Aerosol Box, Analyses and Results, Test 2/1B

Table A2.6 Test 2/1B, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%	Cerium as CeO ₂	Undissolved Solids as CeO ₂	Total CeO ₂
1.000 mm	4.5073 g	35.7	0 wt %	0 wt %	0 wt %
0.500 mm	0.753 g	6.0	31.8 wt %	13.5 wt %	45.3 wt %
0.250 mm	1.1124 g	8.8	50.0 wt %	18.7 wt %	68.6 wt %
0.125 mm	0.9307 g	7.4	32.0 wt %	20.9 wt %	53.0 wt %
< 0.125 mm	5.3149 g	42.1	19.4 wt %	48.9 wt %	68.3 wt %
Total	12.6183 g				

Table A2.7 Test 2/1B, Elemental Analysis wt% of Sieved Impact Debris

Sieve Fraction	0.500 mm	0.250 mm	0.125 mm	<0.125 mm
Cerium	31.82	49.97	32.04	19.43
Solids as Cerium	13.47	18.67	20.92	48.86
Copper	15.48	6.225	1.756	3.433
Zirconium	29.38	5.436	1.342	4.388
Aluminum	1.299	4.257	1.672	0.807
Iron	0.181	2.949	1.138	0.795
Tin	0.375	0.914	0	0.041
Barium	0	0	0	0
Lead	0.002	0	0.016	0.009
Total	92.0	88.4	58.9	77.8

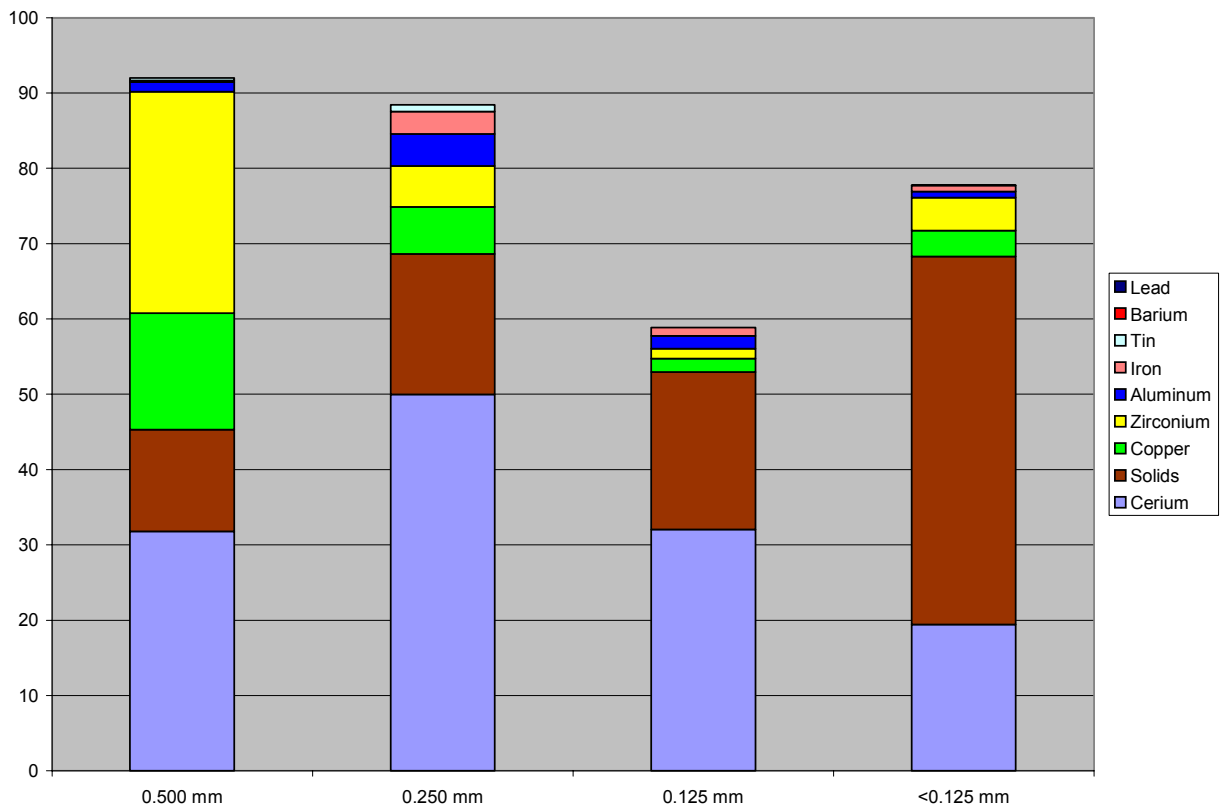


Figure A2.3 Test 2/1B, Distribution of Metals in Sieved Fractions

A.2.2A Impact Debris in Aerosol Box, Analyses and Results, Test 2/2A

Table A2.8 Test 2/2A, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%
1.000 mm	11.4013 g	62.5
0.500 mm	0.6842 g	3.8
0.250 mm	1.0416 g	5.7
0.125 mm	1.1424 g	6.3
0.149 mm	0.0369 g	0.2
0.074 mm	1.2987 g	7.1
0.044 mm	1.8231 g	10.0
0.037 mm	0.142 g	0.8
<0.037 mm	0.6341 g	3.5
Total	18.2043 g	

Table A2.9 Test 2/2A, Elemental Analysis wt% of Sieved Impact Debris

Test 2/2A Sieve Fraction	0.500 mm	0.250 mm	0.125 mm	0.149 mm	0.074 mm	0.044 mm	0.037 mm	<0.037 mm
Cerium	26.850	62.330	75.990	73.290	76.300	71.920	71.600	71.270
Copper	21.220	9.500	2.274	2.326	1.726	3.761	2.983	4.438
Zirconium	39.330	6.472	1.287	2.356	1.488	3.355	2.921	4.328
Aluminum	5.606	4.993	1.018	1.162	0.954	1.165	0.837	0.992
Iron	0.100	0.740	0.277	0.000	0.237	0.444	0.438	0.475
Tin	0.388	0.052	0.005	0.000	0.005	0.016	0.012	0.016
Chromium	0.046	0.010	0.002	0.000	0.002	0.003	0.004	0.004
Magnesium	0.042	0.032	0.010	0.000	0.018	0.025	0.021	0.014
Barium	0.029	0.120	0.137	0.000	0.119	0.111	0.103	0.097
Manganese	0.002	0.010	0.003	0.000	0.003	0.005	0.005	0.005
Terbium	0.002	0.004	0.004	0.000	0.004	0.004	0.004	0.003
Hafnium	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Lead	0.001	0.002	0.005	0.000	0.008	0.011	0.013	0.009
Total	93.6	84.3	81.0	79.1	80.9	80.8	78.9	81.7

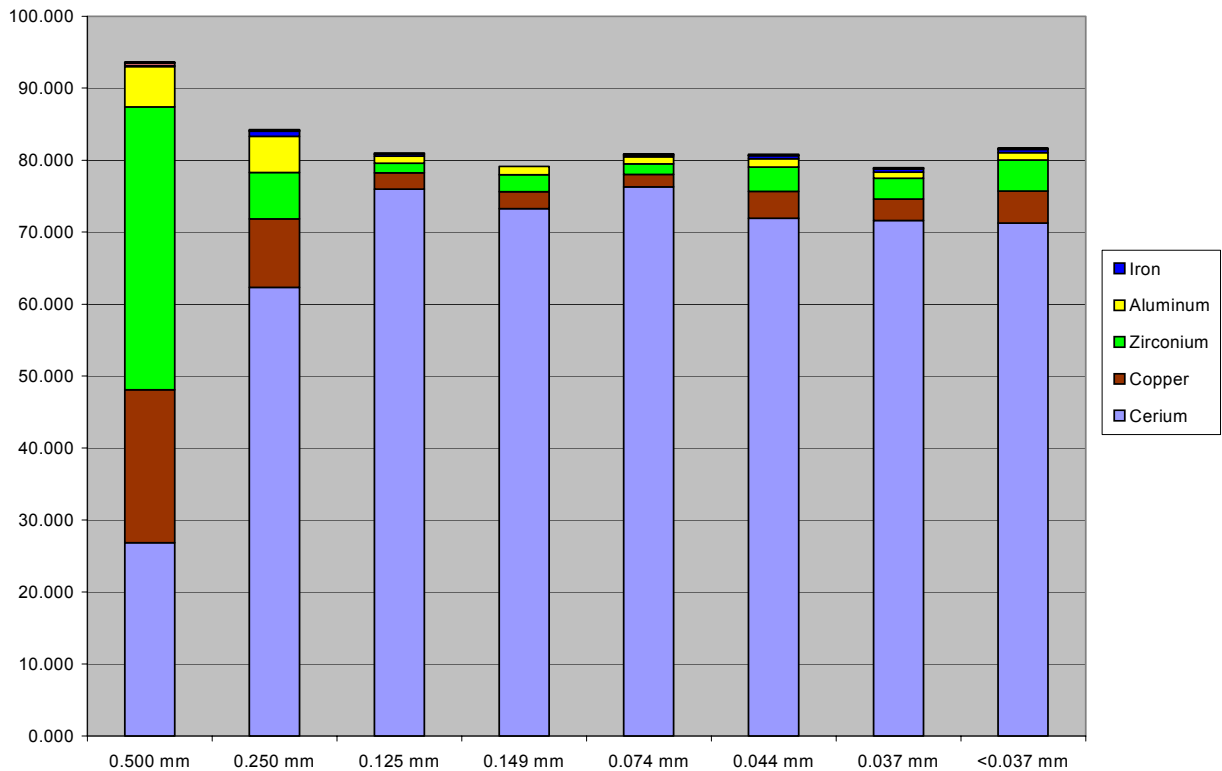


Figure A2.4. Test 2/2A, Distribution of Metals in Sieved Fractions

A.2.2B Impact Debris in Aerosol Box, Analyses and Results, Test 2/2B

Table A2.10 Test 2/2B, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%
1.000 mm	3.5807 g	41.1
0.500 mm	0.6014 g	6.9
0.250 mm	0.7548 g	8.7
0.125 mm	0.8554 g	9.8
0.149 mm	0.1229 g	1.4
0.074 mm	0.7893 g	9.1
0.044 mm	1.0308 g	11.8
0.037 mm	0.1306 g	1.5
<0.037 mm	0.8357 g	9.6
Total	8.7016 g	100.0

Table A2.11 Test 2/2B, Elemental Analysis wt% of Sieved Impact Debris

Test 2/2B Sieve Fraction	0.500 mm	0.250 mm	0.125 mm	0.149 mm	0.074 mm	0.044 mm	0.037 mm	<0.037 mm
Cerium	24.080	66.460	81.110	75.350	83.460	79.970	69.630	73.930
Copper	8.976	4.755	1.278	0.826	0.940	1.785	2.079	3.484
Zirconium	50.600	5.099	1.599	1.772	1.293	2.557	2.772	4.760
Aluminum	6.039	5.183	0.926	0.808	0.581	0.750	0.732	0.961
Iron	3.419	2.094	0.456	0.681	0.374	0.451	0.590	0.551
Tin	0.526	0.042	0.006	0.014	0.004	0.010	0.016	0.020
Chromium	0.059	0.010	0.002	0.003	0.001	0.003	0.000	0.004
Magnesium	0.043	0.035	0.007	0.011	0.005	0.007	0.008	0.007
Barium	0.017	0.006	0.003	0.293	0.010	0.020	0.000	0.040
Manganese	0.042	0.029	0.006	0.006	0.005	0.006	0.007	0.007
Terbium	0.002	0.006	0.000	0.010	0.007	0.006	0.007	0.005
Hafnium	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Lead	0.001	0.014	0.006	0.026	0.013	0.011	0.012	0.008
Total	93.8	83.7	85.4	79.8	86.7	85.6	75.9	83.8

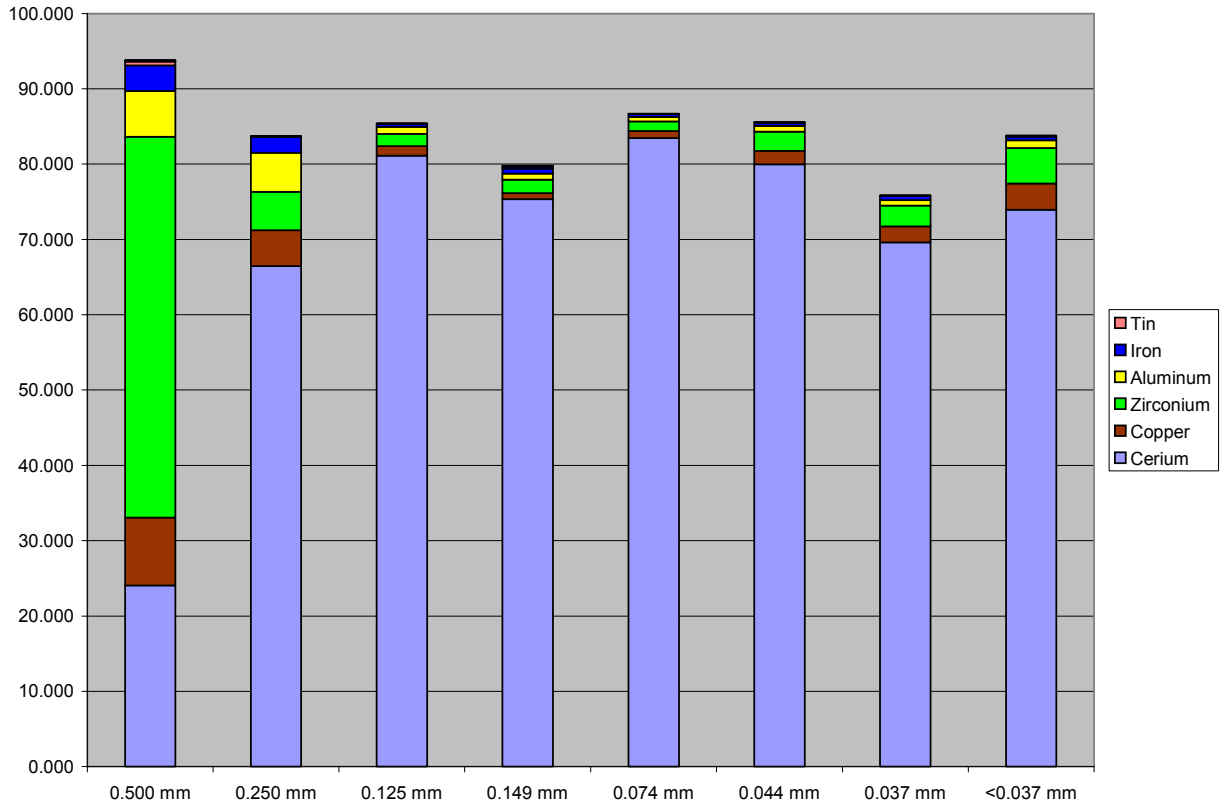


Figure A2.5. Test 2/2B, Distribution of Metals in Sieved Fractions

A.2.3A Impact Debris in Aerosol Box, Analyses and Results, Test 2/3A

Table A2.12 Test 2/3A, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%
1000 µm	8.00335 g	53.0
500 µm	0.76382 g	5.1
250 µm	0.94716 g	6.3
125 µm	1.19101 g	7.9
100 µm	0.47247 g	3.1
74 µm	0.29420 g	1.9
37 µm	1.08110 g	7.2
25 µm	1.00500 g	6.7
<25 µm	1.34820 g	8.9
Total	15.10631 g	100.0

Table A2.13 Test 2/3A, Elemental Analysis wt% of Sieved Impact Debris

Test 2/3A Sieve Fraction	125 µm	100 µm	74 µm	37 µm	25 µm	<25 µm
Cerium	75.510	79.590	79.970	77.240	77.250	72.500
Copper	3.084	2.200	2.485	1.844	3.337	4.583
Zirconium	0.849	0.610	0.982	1.058	2.917	4.461
Aluminum	1.231	0.579	0.583	0.437	0.799	0.749
Iron	1.226	1.171	0.668	0.704	0.520	0.444
Tin	0.003	0.000	0.001	0.003	0.016	0.032
Chromium	0.096	0.020	0.023	0.016	0.036	0.059
Magnesium	0.014	0.007	0.008	0.007	0.014	0.017
Barium	0.000	0.000	0.000	0.000	0.059	0.020
Manganese	0.018	0.013	0.008	0.010	0.009	0.010
Terbium	0.007	0.007	0.007	0.007	0.006	0.006
Total	82.0	84.2	84.7	81.3	85.0	82.9

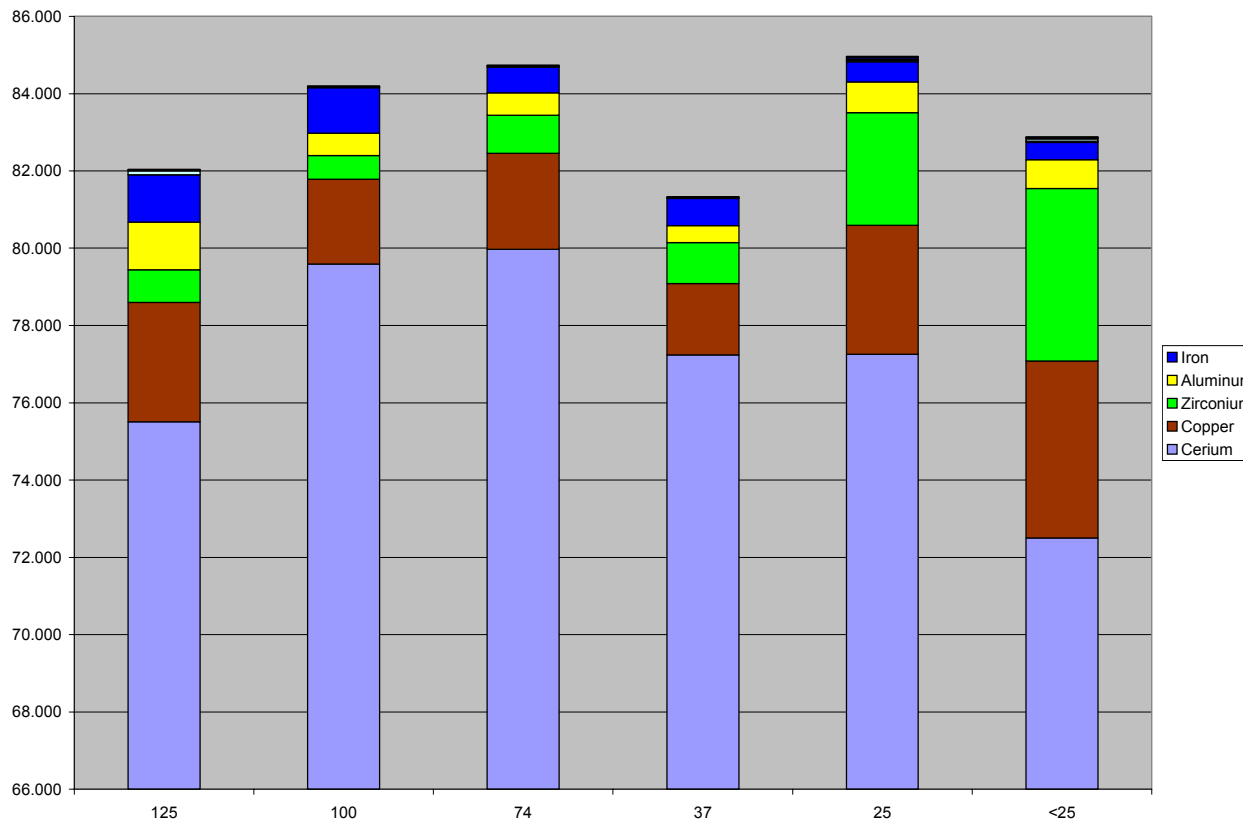


Figure A2.6 Test 2/3A, Distribution of Metals in Sieved Fractions

A.2.3B Impact Debris in Aerosol Box, Analyses and Results, Test 2/3B

Table A2.14 Test 2/3B, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%
1000 µm	2.21456 g	23.0
500 µm	0.78298 g	8.1
250 µm	0.95636 g	9.9
125 µm	0.81005 g	8.4
100 µm	0.18184 g	1.9
74 µm	0.03470 g	0.4
37 µm	1.44280 g	15.0
25 µm	0.74310 g	7.7
<25 µm	2.46170 g	25.6
Total	9.62809 g	100

Table A2.15 Test 2/3B, Elemental Analysis wt% of Sieved Impact Debris

Test 2/3B Sieve Fraction		125 µm	100 µm	74 µm	37 µm	25 µm	<25 µm
Cerium		74.260	77.170	74.480	79.800	77.090	73.720
Copper		3.184	2.311	2.478	2.172	2.734	3.934
Zirconium		0.817	0.842	0.909	1.340	2.197	3.840
Aluminum		1.292	0.598	0.622	0.280	0.392	0.436
Iron		1.045	0.820	2.234	0.466	0.302	0.292
Tin		0.002	0.002	0.003	0.003	0.010	0.022
Chromium		0.095	0.037	0.026	0.020	0.032	0.055
Magnesium		0.014	0.008	0.009	0.006	0.008	0.010
Barium		0.023	0.007	0.024	0.000	0.002	0.000
Manganese		0.013	0.009	0.021	0.006	0.005	0.007
Terbium		0.007	0.007	0.000	0.008	0.007	0.007
Total		80.8	81.8	80.8	84.1	82.8	82.3

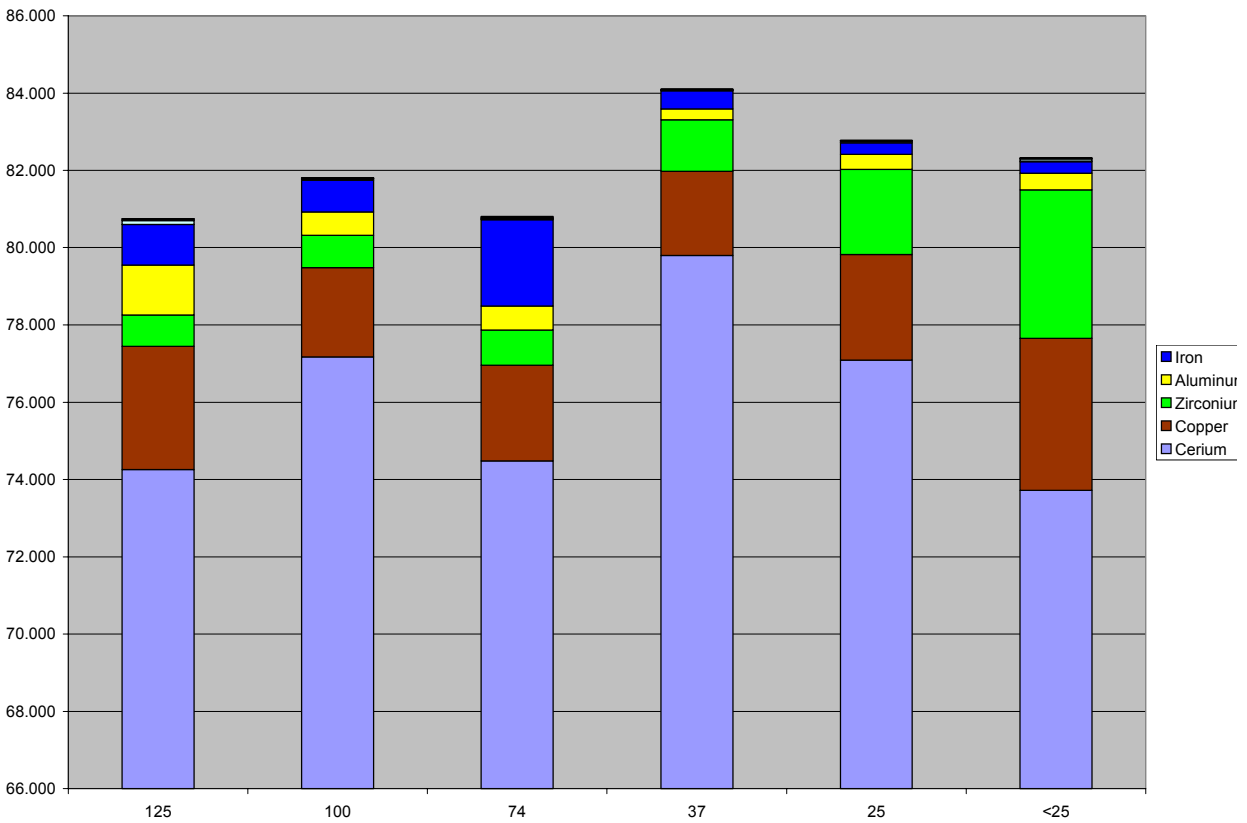


Figure A2.7 Test 2/3B, Distribution of Metals in Sieved Fractions

A.2.4A Impact Debris in Aerosol Box, Analyses and Results, Test 2/4A

Table A2.16 Test 2/4A, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%
1000 µm	1.7580	25.3
500 µm	0.6056	8.7
250 µm	0.6621	9.5
125 µm	0.6226	9.0
100 µm	0.1422	2.0
74 µm	0.3512	5.0
37 µm	0.7817	11.2
25 µm	0.1359	2.0
<25 µm	1.8965	27.3
Total	6.9558	100.0

Table A2.17 Test 2/4A, Elemental Analysis wt% of Sieved Impact Debris

Test 2/4A Sieve Fraction	125 µm	100 µm	74 µm	37 µm	25 µm	<25 µm
Cerium	36.780	38.640	46.190	60.180	70.260	69.230
Iron	35.420	31.600	27.420	14.550	5.900	2.854
Copper	11.020	8.928	7.669	5.093	4.026	4.492
Zirconium	0.631	0.557	0.726	1.415	2.044	3.272
Aluminum	2.113	0.907	0.696	0.680	0.721	0.806
Manganese	0.272	0.231	0.178	0.093	0.044	0.021
Tin	0.011	0.007	0.006	0.009	0.017	0.024
Chromium	0.100	0.044	0.037	0.026	0.032	0.040
Magnesium	0.029	0.017	0.013	0.021	0.022	0.017
Nickel	0.049	0.025	0.020	0.012	0.013	0.015
Titanium	0.037	0.030	0.025	0.026	0.025	0.016
Molybdenum	0.008	0.005	0.004	0.002	0.001	0.001
Strontium	0.007	0.013	0.014	0.046	0.077	0.072
Cesium	0.005	0.005	0.006	0.010	0.018	0.032
Ruthenium	0.001	0.007	0.001	0.002	0.003	0.001
Terbium	0.004	0.004	0.005	0.006	0.007	0.007
Lead	0.000	0.002	0.000	0.000	0.000	0.000
Hafnium	0.000	0.000	0.000	0.004	0.000	0.000
Total	86.5	81.0	83.0	82.2	83.2	80.9

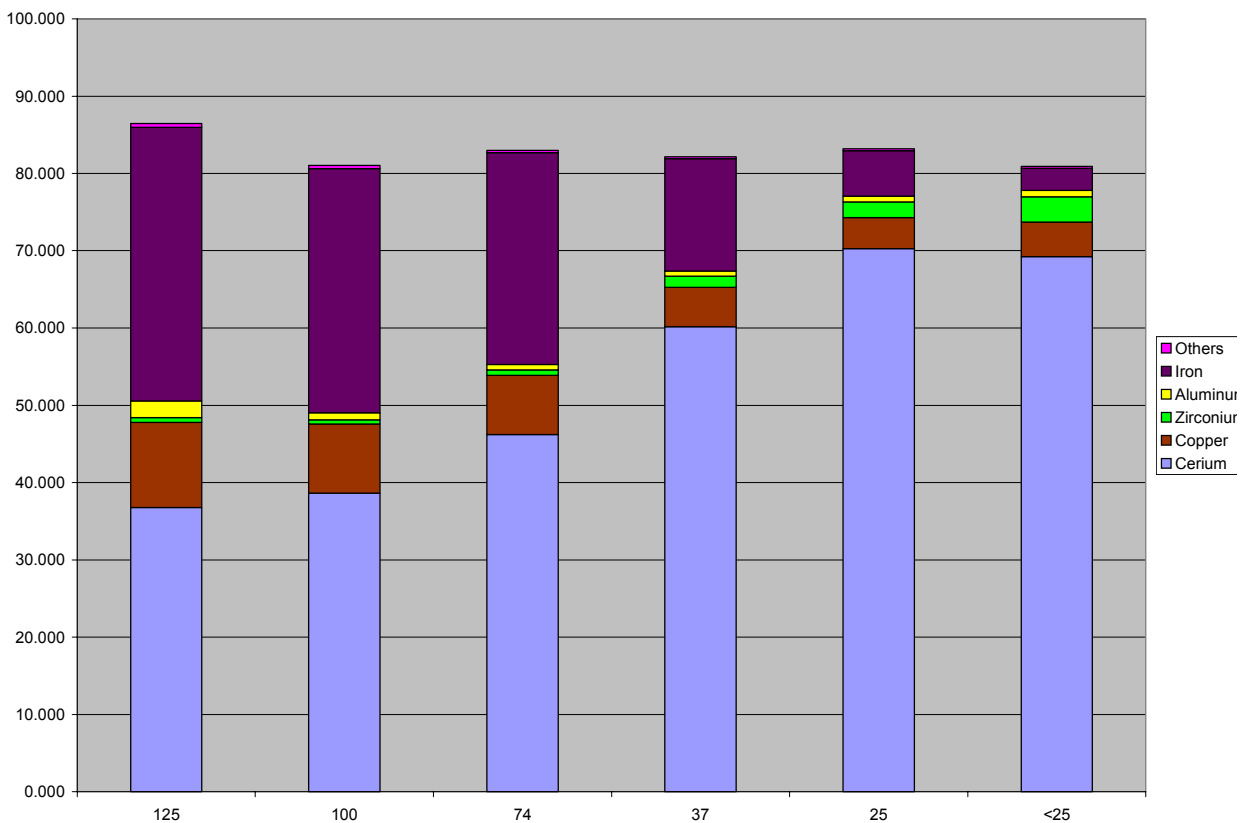


Figure A2.8 Test 2/4A, Distribution of Metals in Sieved Fractions

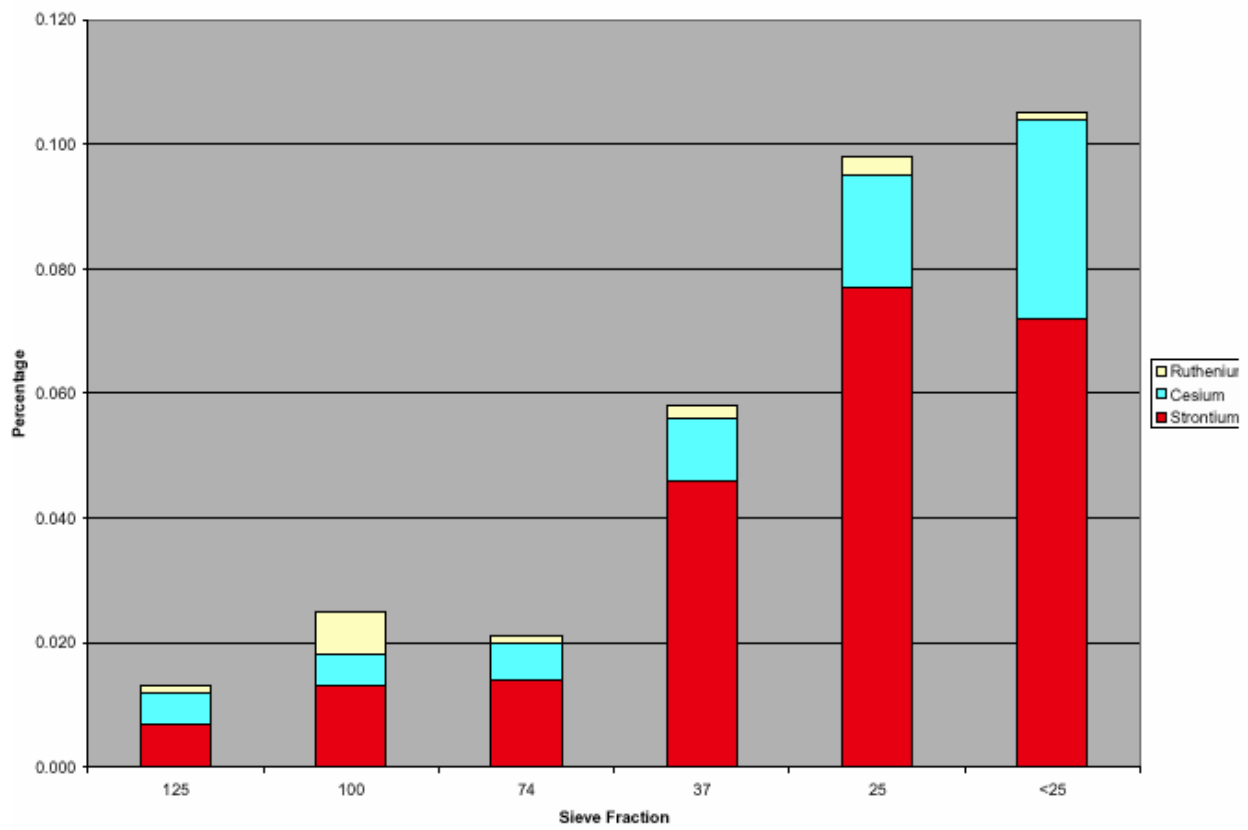


Figure A2.9 Test 2/4A, Weight Percent Distribution of Fission Product Dopants in Sieved Fractions

A.2.4B Impact Debris in Aerosol Box, Analyses and Results, Test 2/4B

Table A2.18 Test 2/4B, Weight Distribution of Impact Debris

Sieve Fraction	Weight	%
1000 µm	5.3469	47.8
500 µm	0.4764	4.3
250 µm	0.4826	4.3
125 µm	0.5790	5.2
100 µm	0.2329	2.1
74 µm	0.4338	3.9
37 µm	1.6150	14.4
25 µm	0.6760	6.0
<25 µm	1.3420	12.0
Total	11.1846	100.0

Table A2.19 Test 2/4B, Elemental Analysis wt% of Sieved Impact Debris

Test 2/4B Sieve Fraction		125 µm	100 µm	74 µm	37 µm	25 µm	<25 µm
Cerium		69.780	66.050	66.550	62.270	58.890	57.300
Copper		4.778	4.427	5.062	5.598	6.399	7.213
Zirconium		1.748	1.408	1.851	2.615	3.544	4.137
Aluminum		2.419	1.756	2.273	2.352	2.488	2.912
Iron		4.270	5.117	5.270	5.159	4.864	5.264
Tin		0.005	0.004	0.006	0.012	0.020	0.024
Chromium		0.073	0.030	0.030	0.034	0.044	0.050
Magnesium		0.014	0.012	0.018	0.019	0.024	0.027
Nickel		0.028	0.011	0.011	0.013	0.017	0.019
Titanium		0.008	0.012	0.014	0.011	0.010	0.010
Molybdenum		0.003	0.001	0.002	0.001	0.001	0.002
Strontium		0.040	0.036	0.053	0.056	0.049	0.041
Cesium		0.004	0.004	0.007	0.014	0.024	0.030
Ruthenium		0.000	0.000	0.002	0.001	0.001	0.003
Terbium		0.006	0.006	0.006	0.006	0.006	0.005
Barium		0.032	0.010	0.003	0.000	0.000	0.001
Manganese		0.023	0.025	0.027	0.027	0.028	0.031
Terbium		0.006	0.006	0.006	0.006	0.006	0.005
Total		83.231	78.909	81.185	78.188	76.409	77.069

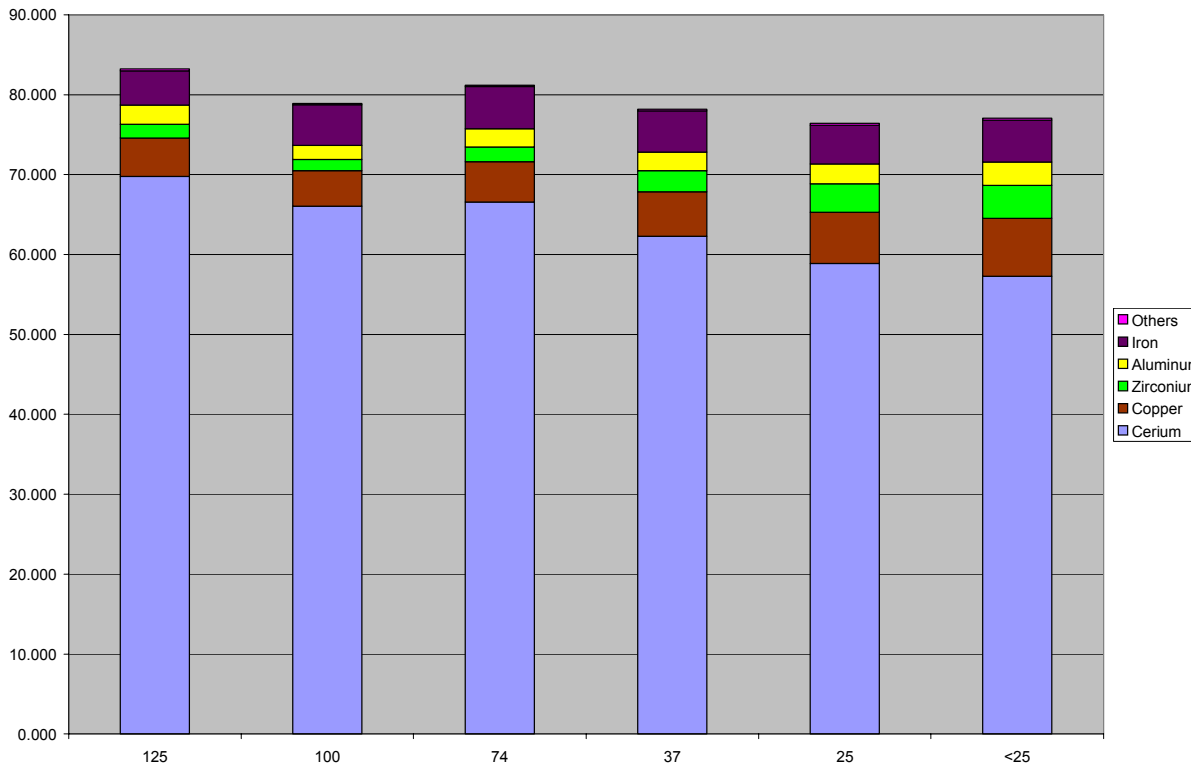


Figure A2.10 Test 2/4B, Distribution of Metals in Sieved Fractions

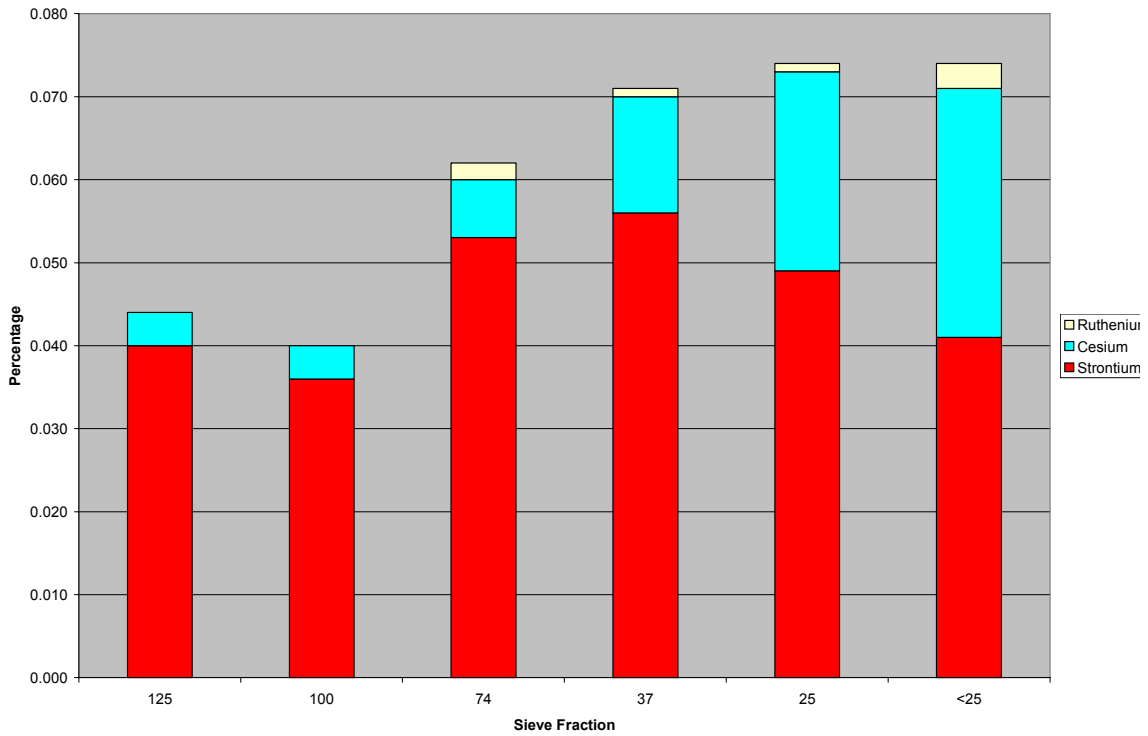


Figure A.11 Test 2/4B, Weight Percent Distribution of Fission Product Dopants in Sieved Fractions

REFERENCES

- Alvarez et al., 1982. Alvarez, J.L., et al. *Waste Form Response Project Correlation Testing*. EGG-PR-5590. Idaho National Engineering Laboratory. Idaho Falls, ID. September 1982.
- Autrusson et al., 2003. Autrusson, B., O. Loiseau, K. Cheval, D. Brochard, J.M. Sibeaud, and A. Delmas, *France Shipment Casks Subjected To Shaped Charge*. Presented at the 44th Annual Meeting, Institute of Nuclear Materials Management, INMM, July 13-17, 2003, Phoenix, AZ.
- Blejwas, 2003. Blejwas, T.E., Sandia National Laboratories. Letter "Management of HEDD Project DU Waste," to N. Slater Thompson, Department of Energy. May 15, 2003.
- Dickey, 2002, 2003. Dickey, R.R., Sandia National Laboratories, personal communications.
- Einzigler et al., 2003. Einziger, R.E., H. Tsai, M. C. Billone and B.A. Hilton. *Examination of Spent PWR Fuel Rods After 15 Years in Dry Storage*, NUREG/CR-6831. To be published in September 2003.
- Einzigler, 2003. Einziger, R.E., Consultant, personal communication. 2003.
- EPRI, 2001. *Design, Operation, and Performance Data for High Burnup PWR Fuel from the H. B. Robinson Plant for Use in the NRC Experimental Program at Argonne National Laboratory*. EPRI, Palo Alto, CA. 2001. 1001558.
- Ewsuk and Diantonio, 2002. Ewsuk, K. and C. DiAntonio, memorandum to M.A. Molecke, Sandia National Laboratories, "Cerium Oxide (CeO₂) Pellet Fabrication," September 24, 2002.
- GRS/SNL, 2000. Gesellschaft für Anlagen- und Reaktorsicherheit and Sandia National Laboratories, "Joint GRS/SNL Proposal to Delineate the Ratio of Spent Fuel to Surrogate Aerosol Generation for More Accurate Prediction of Sabotage Consequences." Unpublished. July 2000.
- Harper, 2004. Harper, F.T., Sandia National Laboratories, personal communications. 2004.
- Hinds, 1999. Hinds, W.C., *Aerosol Technology*. John Wiley & Sons, 1999.
- Koch et al., 1999. Koch, W., W. Dunkhorst, and H. Lödding, *Design and Performance of a New Personal Aerosol Monitor*, *Aerosol Sci. & Tech.*, Vol. (31), pg.231-246, 1999.
- Lake et al., 2001. Lake, W.H., Kapoor, A.K., Davis, J., Young, F.I., Yoshimura, R.H., Vigil, M.G., Luna, R., Lange, F., Pretzsch, Autusson, B., and Brochard, D. *International Initiative in Transportation Sabotage Investigations*, SAND2001-2858C. Presented at PATRAM 2001, September 30-October 5, 2001, Chicago, IL.
- Lake et al., 2002. Lake, W.H., Kapoor, A.K., Davis, J., Young, F.I., Murphy, A., Yoshimura, R.H., Philbin, J.S., Sprung, J.L., Vigil, M.G., Lange, F., Pretzsch, Koch, W., Nolte, O. Lodding, H, Autusson, B., Brochard, D., and Luna, R., *International Cooperation in Measuring Spent Fuel Response in High Energy Environments*, SAND2002-0628C. Presented at Waste Management (WM) 2002 Conference, February 24-28, 2002, Tucson, AZ.
- Lange et al., 1994. Lange, F., G., Pretzsch, J. Döhler, E. Hörmann, H. Busch, W. Koch. *Experimental Determination for UO₂-Release from a Spent Fuel Transport Cask after Shaped Charge Attack*. Proc. INMM 35th Meeting, Naples, Florida. July 17-20, 1994.
- Luna et al., 1999. Luna, R. E., K. S. Neuhauser, and M. G. Vigil. *Projected Source Terms for Potential Sabotage Events Related to Spent Fuel Shipments*, SAND99-0963. Sandia National Laboratories. June 1999.

Luna et al., 2001. Luna, R.E., H.R. Yoshimura, M.G. Vigil, J.S. Philbin, F. Lange, G. Pretzsch, W. Koch, and Y.S. Cheng. *Perspectives on Spent Fuel Sabotage*, SAND2001-0603C. Presented at Waste Management 2001 Conference, February 25-March 2, 2001, Tucson, AZ.

Luna et al., 2002. Luna, R.E., H.R. Yoshimura, M.A. Molecke. DRAFT "Master Plan for International WGSTSC Experiments to Determine Spent Fuel Ratio/Fractionation for Spent Nuclear Fuel." Unpublished, internal draft. September 2002.

Melgaard et al., 2003. Melgaard, D.K, G.J. Shelmidine, J.A. Van Den Avyle, and M.A. Molecke. *Radionuclide Surrogate Decontamination of Metal Using Electroslag Remelting*, SAND2002-3379C. Presented at TMMS 2003 132st Annual Meeting of the Minerals, Metals & Materials Society, San Diego, CA, March 2-6, 2003.

Molecke, 1999. Molecke, M.A., memorandum to J.A. Van Den Avyle, Sandia National Laboratories, "Surrogate Chemistry Recommendations for Future ESR Melt Studies," Feb. 17, 1999.

Molecke et al., 2003a. Molecke, M.A., H.R. Yoshimura, M.G. Vigil, F. Lange, G. Pretzsch, W. Koch, O. Nolte, B. Autrusson, D. Brochard, W. Lake, A. Kapoor, F.I. Young, and R.E. Luna. *Spent Fuel Response in High Energy Environments: Initial Results, International Program*, SAND2003-0779C. Presented at the 2003 International High-Level Radioactive Waste Management Conference, American Nuclear Society, March 30-April 2, 2003, Las Vegas, NV.

Molecke et al., 2003b. Molecke, M.A., H.R. Yoshimura, S.W. Longley, F. Lange, G. Pretzsch, W. Koch, O. Nolte, B. Autrusson, D. Brochard, N. Slater Thompson, and F.I. Young. *Spent Fuel/Surrogate Aerosol Ratio Test Program and Preliminary Results*, SAND2003-2706C. Presented at the 44th Annual Meeting, Institute of Nuclear Materials Management, July 13-17, 2003, Phoenix, AZ

Molecke and Sorenson, 2003. Molecke, M.A., and K.B. Sorenson, Spent Fuel Sabotage, *Spent Fuel Ratio-HEDD: International Test Program Status Update*, SAND2003-2263P. Presented at the 5th Technical Meeting of the International Working Group for Sabotage Concerns of Transport and Storage Casks, WGSTSC, May 21-22, 2003, Cologne, Germany.

Molecke, Yoshimura, and Vigil, 2004. Molecke, M.A., R.H. Yoshimura, and M.G. Vigil, Spent Fuel/Surrogate Aerosol Ratio Tests, Phase 1 Result. SAND2004-xxxx. Sandia National Laboratories. In preparation (rough draft, December 2002).

Philbin et al., 2002a. Philbin, J.S., Hoover, M.D., Newton, G.J. *The Need for Confirmatory Experiments on the Radioactive Source Term from Potential Sabotage of Spent Nuclear Fuel Casks*, SAND2002-1027. Sandia National Laboratories. April 2002.

Philbin et al., 2002b. Philbin, J. S., R. E. Naegeli, R. L. Coats, P. H. Helmick, M. J. McDonald, S. W. Longley, S. A. Walker, and D. T. Berry, *Performing Spent Fuel Ratio Experiments at Sandia National Laboratories: A Feasibility Study*, SAND2002-2509. Sandia National Laboratories. August 2002.

Sandoval et al., 1983. Sandoval, R.P., J.P. Weber, H.S. Levine, A.D. Romig, J.D. Johnson, R.E. Luna, G.J. Newton, B.A. Wong, R.W. Marshall, Jr., J.L. Alvarez, and F. Gelbard. *An Assessment of the Safety of Spent Fuel Transportation in Urban Environs*, SAND82-2365. Sandia National Laboratories. June 1983.

Schmidt et al., 1981. Schmidt, E.W., M.A. Walters, and B.D. Trott. *Shipping Cask Sabotage Source Term Investigation*, BMI-2089, NUREG/CR-2472. Battelle Columbus Laboratory, Columbus, OH. December 1981.

Tsai and Billone, 2003. Tsai, H. and M. C. Billone. *Characterization of High-Burnup PWR and BWR Rods, and PWR Rods after Extended Dry-Cask Storage*. Proceedings of the 2002 Nuclear Safety Research Conference, NUREG/CP-0180, March 2003.

U.S. DOE, 1999. U.S. Department of Energy. *Draft Environmental Impact Statement for a Geologic Repository for the Disposal of Spent Nuclear Fuel and High-Level Radioactive Waste at Yucca Mountain*. Office of Civilian Radioactive Waste Management, DOE/EIS-0250D. Las Vegas, NV. July 1999.

Van Den Avyle et al., 2003. Van Den Avyle, J.A., D. Melgaard, M.A. Molecke, G.J. Shelmidine, U. Pal, and S.I. Bychkov, *Final Report- Optimization of Thermochemical, Kinetic, and Electrochemical Factors Governing Partitioning of Radionuclides During Melt Decontamination of Radioactively Contaminated Stainless Steel*, SAND2003-0054P. Sandia National Laboratories. January 2003.

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