SANDIA REPORT
SAND2007-8070
Unlimited Release
Printed March 2008

Spent Fuel Sabotage Test Program, Characterization of Aerosol Dispersal: Interim Final Report

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Sandia is a multiprogram laboratory operated by Sandia Corporation,
a Lockheed Martin Company, for the United States Department of Energy’s
National Nuclear Security Administration under Contract DE-AC04-94AL85000.

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Spent Fuel Sabotage Test Program, Characterization of Aerosol Dispersal: Interim Final Report

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Abstract

This multinational, multi-phase spent fuel sabotage test program is quantifying the aerosol particles produced when the products of a high energy density device (HEDD) interact with and explosively particulate test rodlets that contain pellets of either surrogate materials or actual spent fuel. This program provides source-term data that are relevant to plausible sabotage scenarios in relation to spent fuel transport and storage casks and associated risk assessments. We present details and significant results obtained from this program from 2001 through 2007. Measured aerosol results include: respirable fractions produced; amounts, nuclide content, and produced particle size distributions and morphology; measurements of volatile fission product species enhanced sorption – enrichment factors onto respirable particles; and, status on determination of the spent fuel ratio, SFR, needed for scaling studies. Emphasis is provided on recent Phase 3 tests using depleted uranium oxide pellets plus non-radioactive fission product dopants in surrogate spent fuel test rodlets, plus the latest surrogate cerium oxide results and aerosol laboratory supporting calibration work. The DUO_2, CeO_2, plus fission product dopant aerosol particle results are compared with available historical data. We also provide a status review on continuing preparations for the final Phase 4 in this program, tests using individual short rodlets containing actual spent fuel from U.S. PWR reactors, with both high- and lower-burnup fuel. The source-term data, aerosol results, and program design have been tailored to support and guide follow-on computer modeling of aerosol dispersal hazards and radiological consequence assessments. This spent fuel sabotage, aerosol test program was performed primarily at Sandia National Laboratories, with support provided by both the U.S. Department of Energy and the Nuclear Regulatory Commission. This program has significant input from, and is cooperatively supported and coordinated by both the U.S. and international program participants in Germany, France, and others, as part of the International Working Group for Sabotage Concerns of Transport and Storage Casks (WGSTSC).

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* Sandia is a multi-program laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under contract DE-AC04-94-AL85000.

Acknowledgements

The authors wish to acknowledge and express gratitude to multiple people for their major contributions and support to this surrogate/spent fuel sabotage and aerosol measurement test program over the last five years. 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. Many of the same people have also provided major technical inputs to the writing of this report, analyses of the data within, plus designs and fabrication for many of the test components.

We want to acknowledge current and prior Sandia National Laboratories participants for their major contributory efforts. In particular, we recognize Ken Sorenson, the lead Sandia program manager and main interface with the International WGSTSC, plus Richard Yoshimura and Bob Luna (consultant, retired) who continue to provide an excellent link with the technical definitions, evaluations, and history of this test program. In the Explosive Projects/Diagnostics Department: Roy Dickey (retired), for providing the designs and fabrication for most of the test components, and for the major conduct and guidance on all of the tests through 2005; Manny Vigil (retired) provided guidance on the science of the explosive components and processes; Lloyd Bonzon (retired) explosive projects department manager; Mark Garrett, current manager; Marc Hagan, explosives technician, and Susan Bender, for ES&H support. In the Aerosol Sciences Department: Dan A. Lucero, Mai Luu, and Traci Durbin provided very valuable aerosol and testing support. In the Nuclear Facilities and Applied Technologies Center, Ken Reil, Richard Coats, Don Berry, Paul Helmick, and Rob Naegeli, for guiding future radioactive, spent fuel test performance plus significant nuclear facilities support, as did John Guth (retired) and Jeff Philbin (retired) in the past. In the Ceramic Processing and Inorganic Materials Department: Kevin Ewsuk, Denise Bencoe, and Chris DiAntonio provided key support on cerium oxide pellet and fission product-doped pellet fabrication, as well as technical support and guidance. In the Analytical Chemistry/Materials Characterization Department: Ted Borek, Jeff Reich, and James Hochrein provided test support and optical microscopy and elemental analysis services on the particulate materials collected from these tests, through 2005. In the High Consequence Assessment and Technology Department: Heather Gorenz, assisted in analyzing the data from the recent test 2/CSC. Appreciation is also expressed for Lorraine Herrera, who provided key support in shipping test samples to the GEL analytical chemistry laboratory, coordinating the analyses and results, and converting data received into the required formats and figures required for our full interpretations, plus Pam Puissant, for coordinating technical and contract negotiations with GEL Laboratory. Mike Billone, Tatiana Burtseva, and Hanchung Tsai (retired), Argonne National Laboratory, have provided most of the input for spent fuel characterization, rodlet fabrication, and related issues. We also express gratitude for the many contributions of our WGSTSC partners who have either retired or moved on: Florentin Lange, GRS, and Didier Brochard, IRSN. Thanks to all.

Finally, our appreciation is extended gratefully to those that have supported, funded, and/or managed this research program over the last five years at the U.S. Department of Energy: Alex Thrower, Nancy Slater Thompson, and Gary Lanthrum (DOE RW), Russell Hibbs (DOE NA, contractor), Diana D. Clark (DOE GC), and William Lake (DOE EM, retired); and, at the U.S. Nuclear Regulatory Commission: Francis (Skip) Young (NRC NSIR, now NRC OIP), Donald Helton (NRC RES), and Tin Mo (NRC RES, retired). Many of these individuals have also contributed objectives, suggestions, and informal documentation that are incorporated in part into this, and earlier SNL spent fuel sabotage program technical reports and presentations.

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1. Program Overview

The casks used for spent nuclear fuel transport are extremely resistant to releasing any significant fraction of their contents, even in very severe accident conditions. However, in recent years, concerns about the possibility of radiological sabotage against spent nuclear fuel shipping and storage casks within the United States and elsewhere have been rekindled. This increased interest is particularly due to the proposed increase in transport of spent fuel, e.g., to the planned Yucca Mountain Repository, increased number of proposed independent spent fuel storage installations, and the increased availability of explosives technology and materials. In some plausible, intentional sabotage scenarios, such as an attack employing a high energy density device (HEDD), i.e., explosive armor-piercing weapons, it is possible that a cask could be penetrated. Then, a small percentage of aerosolized particles produced within from disrupted fuel rod and pellet materials could be released as a radiological inhalation source hazard. If released to the environment in a significant quantity, the spent fuel respirable particles have the potential to cause radiological consequences.

Measurement of the actual amounts, nuclide content, and size distribution of the particles produced from spent fuel and related surrogate materials is essential for predicting the significance of aerosol releases from the cask and their radiological inhalation impacts. These aerosol source-term data are the input for follow-on modeling studies to quantify respirable hazards, associated radiological risk assessments, vulnerability assessments, and potential cask physical protection design or safeguards modifications. The need for accurately quantifying this information has been strongly supported by program participants in the U.S., Germany, France, and others, as part of the International Working Group for Sabotage Concerns of Transport and Storage Casks (WGSTSC). WGSTSC partners need, and are helping coordinate this research and subsequent modeling plus assessments, and to develop potential preventative measures for plausible radiological sabotage events, if necessary.

This comprehensive technical report is intended to provide a detailed summary of, and update to significant results as of the end of 2007 from the current, multinational test program that is measuring aerosol particle data for a spent fuel sabotage scenario relevant to spent fuel transport and storage casks. This technical report specifically summarizes test details, observations, aerosol results and comparisons, interpretations, and future plans documented over the last few years in previous Sandia technical reports [1-3]. We strive to minimize repetition of test details provided in these earlier reports. In addition, we will provide detailed results of the most recent tests, laboratory calibration information, programmatic progress and status, plus data interpretations and updates made since the last technical report in 2006 [3].

The current document is referred to as an “Interim Final” report because planned completion of depleted uranium oxide Phase 3 testing in 2007, and initiation of Phase 4 spent fuel sabotage testing activities at SNL were put into an indefinite state of suspension during the period of FY 2006 through 2008 due to insufficient or delayed allocations of DOE program funding. No further explosive-aerosol testing can be planned or performed at present. The option to restart the Phase 4 spent fuel test program has been left open, pending future adequate funding and program support. Future program strategies will be developed as appropriate. This Interim Final report provides data compilations and interpretations in a single document, most probably adequate to support follow-on modeling studies and consequence assessments, plus the potential restart of testing activities later, as warranted. A separate “Final Report,” following this Interim Final Report, is contemplated after the completion of Phase 4 spent fuel test activities plus analyses of the resultant data.

It is emphasized that this report provides the available results, interpretations, and technical details derived from the current spent fuel sabotage, aerosol measurement test program. It does not, nor was it intended to provide follow-on modeling evaluations or summaries of consequence assessments based on this, or other, earlier aerosol data. Such assessments and modeling are not currently supported parts of this test program. They must be conducted in the future for the full benefit and use of supporting organizations, regulators, and competent authorities. An excellent compilation of related assessments is documented separately [4], based on estimated source terms from earlier data on sabotage events related to spent fuel shipments.

1.1 Data Needs

The aerosol particle testing performed in this program requires sampling and quantification of the mass and chemical-physical characteristics of the aerosol particles produced from (spent fuel or surrogate material rod) target-HEDD jet impact, with particle aerodynamic equivalent diameters (AED) up to 100 µm (micrometers). The AED is defined by means of the settling velocity of a unit density sphere, and is equivalent to the particle geometric-diameter times the (particle density)$^{1/2}$. For evaluations of aerosol and radiological consequences, there has always been a special emphasis on *respirable particles*, commonly defined as 0 up to ~10 µm AED in size. Respirable particles also have been sub-categorized into the *respirable* portion (or fraction), 0 to ~4 µm, and the *thoracic* portion, ~4 to ~10 µm AED. Data from the coarser aerosol particles in the ~10 to 100 µm AED range, termed the *inhalable* portion, are of interest primarily for radiological "groundshine" (dispersion, soil contamination) or potential ingestion consequence estimates. Particles larger than 100 µm are not considered to be aerosols. Multistage aerodynamic particle sizing devices (impactor collectors) are used to classify aerosol particles according to their aerodynamic diameter. We initially used two 3-stage Respicon virtual impactors and one 9-stage Berner impactor [1]. The vast majority of aerosol data obtained since FY 2005 has used multiple 10-stage Marple impactors and custom designed large particle separators (LPS), to be detailed later.

Major measured aerosol source-term data needed, and results provided from our research include:

1. The **Respirable Fraction (RF)** of particles *produced* is defined as:

   $$\mathbf{RF} = \frac{\text{[mass of an element (i.e., U, Ce, Cs, etc.) in respirable particles]}}{\text{[mass of that element in the rod swept volume (particulated by the HEDD)]}}.$$

   This RF is expressed as a percentage, with special emphasis on U, Ce, Cs, other fission products, etc. RF values are particularly relevant to the far-field airborne dispersion and consequence modeling studies, e.g., for respirable particle releases from a sabotage damaged spent fuel cask.

2. The **Enrichment Factor (EF)** is the measurement of enhanced sorption of volatile fission product nuclides like cesium and, to a lesser extent, ruthenium, preferentially sorbed onto specific, respirable particle size fractions in the sub-µm to µm size range. This enhanced sorption relative to the bulk pellet material can be integrated over the total respirable size range of 0-10 µm AED, is expressed as the integrated Enrichment Factor (IEF) and defined as:

   $$\mathbf{IEF} = \mathbf{RF}\text{(fission product element)} \,/\, \mathbf{RF}\text{(uranium or surrogate cerium)}$$

   Differential EF values can also be measured as a function of individual particle size ranges, e.g., individually for 0 to 0.5 µm, 0.5-0.9 µm, 0.9-1.6 µm, 1.6-3.5 µm, etc., and are observed to vary quite significantly as a function of particle. Differential EF values are defined as:

EF = [fission product element fraction in a given size range] / [fission product element fraction in pellet].

3. The **Spent Fuel Ratio (SFR).** Measurement of a more accurate and precise value for the Spent Fuel Ratio for respirable particles is a major goal of this program. The SFR is defined as:

   SFR = [Spent Fuel respirable particle masses] / ["surrogate" DUO_2 respirable particle masses].

The integral SFR determination is, essentially, the comparison of the respirable, aerosol particle mass from irradiated, spent fuel (to be measured in Phase 4 of this test program) to the respirable, aerosol particle mass from unirradiated, surrogate DUO_2 fuel, as measured from Phase 3 tests and presented herein. These data are obtained in paired experiments using effectively the same apparatus, essentially identical test conditions, and using the same HEDD.

In addition to RF, EF, and SFR values, we also measure produced particle size distributions, elemental content, and morphology, in the respirable through aerosol range of 0 to 100 µm AED. In some cases, we have measured particle size distributions up to 1 mm in size, using mechanical sieving, and corresponding elemental content.

Release of fine particles from a shipping cask after a potential sabotage HEDD impact is determined both by the initial "source term", i.e. the "aerosol dust" generated inside the cask, and also by the "transport term," characterizing the transport of airborne material from inside the cask to the outside environment [13]. Since the transport term is essentially independent of type of fuel pellets used, 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 size ranges.

The measured SFR values (not yet available) provide a data bridge to previous large-scale surrogate (DUO_2) explosive-aerosol cask tests [5-6] performed in both the U.S. and Germany and to consequence assessments. The SFR values permit scaling to other geometries, from a simplified single, short fuel rodlet as tested herein, to rod bundles in casks, by means of supporting modeling studies. The primary test benefit of using the ratio of respirable, aerosol particles for the SFR determination is that it is not necessary to recover and analyze all of the aerosolized materials produced; only the identical portions of aerosol particles from both the spent fuel and surrogate fuel tests must be obtained, analyzed, and compared. This ratio drives the requirement for use of identical-as-possible test apparatus and test conditions for multiple test phases and materials. In addition, by focusing on the spent fuel ratio determination, we can use test rodlets containing only a few actual or surrogate fuel pellets for aerosol particle production. Entire fuel assemblies or casks full of fuel assemblies do not need to be tested.

1.2 Test Program Participants

Sandia National Laboratories (SNL), Materials Transportation Testing and Analysis Department (previous) / Fuel Cycle Experimentation and Analysis Department (current) has the lead role for managing and performing this research program. Other SNL Departments are currently providing major, required expertise, engineering, fabrication, testing, and facilities: Explosive Testing and Diagnostics, Aerosol Sciences, Radiation Sciences (Nuclear Facilities, Engineering, and Technologies, Ceramic Processing and Inorganic Materials, and, Radiation Protection. During the last five years, the SNL Ceramic Processing and Inorganic Materials and the Analytical Chemistry/Materials Characterization Departments also provided major technical support.

Overall sabotage and transportation program support is provided by both the U.S. Department of Energy (DOE) and the U.S. Nuclear Regulatory Commission (NRC). Currently, the DOE Office of Civilian Radioactive Waste Management (OCRWM), RW Office of Logistics Management, provides the major source of program funding. The DOE Sandia Site Office (SSO), National Nuclear Security Agency (NNSA), is responsible for the relevant nuclear facilities at SNL being used. The NRC, Office of Nuclear Regulatory Research (RES), is also a significant source of program support and funding, with NRC Nuclear Security and Incidence Response (NSIR), now the Office of International Programs (OIP), providing technical monitoring and oversight. Prior to 2003, the initial work for this SNL test program project was managed and funded by DOE Environmental Management (EM), National Transportation Program, and NRC Nuclear Material Safety and Safeguards (NMSS) -- included as a HEDD studies sub-set of a project entitled "Revalidation of NUREG-0170 Spent Fuel Shipment Risk Assessments." From 2003-2005, the DOE (NA), NNSA Office of International Safeguards, provided appreciable support and funding, primarily for Phase 4 spent fuel characterization and fabrication efforts. Argonne National Laboratory (ANL), Energy Technology Division, has provided the detailed characterization and fabrication work for all spent fuel test rodlets to be used in this program.

In addition to the U.S. participant organizations described, a group of other international organizations with the same concerns and data needs in regard to sabotage of nuclear materials joined together to collaboratively work on, support (with similar testing and modeling efforts), and coordinate the overall program effort. This collaborative group, together since 1999, is named the International Working Group for Sabotage Concerns of Transport and Storage Casks (WGSTSC), with major participation by German, French, and other organizations. The German participants include the Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), a non-profit, scientific-technical expert and research organization working for the German Federal Ministry for the Environment, Nature Conservation and Nuclear Safety (BMU); the Fraunhofer Institute of Toxicology and Experimental Medicine (ITEM), a research institute supporting GRS, that is providing supporting aerosol testing, expertise, and data analyses; and, also in support to GRS and ITEM, the Fraunhofer Ernst-Mach-Institut (EMI), Institut für Kurzzeitdynamik (Institute for High-Speed Dynamics), providing explosives test facilities and expertise. From France, the Institut de Radioprotection et de Surete Nucleaire (IRSN), is a research and analysis public establishment of an industrial and commercial nature, under the joint authority of the French Ministers of Defense, the Environment, Industry, Research and Health. IRSN has provided nuclear and materials expertise, unirradiated depleted UO_2 (surrogate, DUO_2) fuel test rodlets for Phase 3 testing, plus supporting modeling studies. Personnel from other organizations, the Japan Nuclear Energy Safety Organization (JNES) and Japan Atomic Energy Agency (JAEA), Japan, and the Office for Civil and Nuclear Security (OCNS), part of the Health & Safety Executive (HSE), Nuclear Directorate, in the UK, also have participated in WGSTSC cooperative roles.

1.3 Background History

To support the development of the regulations associated with transport and storage of spent nuclear fuel in the late 1970s [7], particularly assessments based on the NRC-supported "Urban Study" and its revision [8-9], the NRC determined that an experimental program was needed to validate the analyses used to develop the regulations. The NRC Urban Study [8] was probably the first study to formally address concerns for potential sabotage-related releases from spent fuel in transit. A limited number of small-scale explosive-aerosol tests with small depleted uranium oxide (DUO_2) and actual spent fuel targets were conducted at Battelle Columbus Laboratory (with NRC support) [10] and at the Idaho National Engineering Laboratory (INEL, now INL) (with SNL and

DOE support) [11]. These early tests were to assess the potential consequences of hypothetical sabotage events on spent fuel casks, to develop source terms for aerosol materials created as a result of HEDD impacts, and to measure the spent fuel ratio (SFR).

The studies performed at BCL [10] included seven tests with DUO_2 pellets in Zircaloy tube rods and another eight tests with short rodlets of actual spent fuel originating from the H.B. Robinson PWR (at 33 GWd/MTU burnup). A very small HEDD, used for oil well casing perforating, was used in this study. These experiments determined a SFR value of ~3.0 and also reported fission product enhanced sorption (Enrichment Factor values) in the "sub-µm" range; the reported EF for cesium was ~16, the EF for ruthenium was ~5.

The studies performed at INL [11] were similar to the BCL experiments. There were two HEDD-spent fuel tests using H.B. Robinson fuel (at 28 GWd/MTU burnup). A SFR value of ~5.6 was determined from wet sieve measurements of aerosols produced; particle impactor data was not available due to partial sample losses during analyses. A SFR value of ~ 0.53 was calculated based on available gravimetric data. Unfortunately, there was a large degree of uncertainty on these measurements. Enhanced fission product sorption EF values were also measured for "sub-µm" respirables < 0.2 µm; cesium EF = ~56, antimony EF = ~11, and ruthenium EF = ~16. Similarly, for respirables < 1 µm AED, cesium EF = ~35 and ruthenium EF = ~10. Measured Cs and Ru EF values determined in the current SNL-WGSTSC test program are reported in Section 4.2.

In addition, a small number of large, "cask-scale" tests were performed in both the U.S. and Germany using rod bundles with DUO_2. During the early 1980s, SNL performed [6] a series of 1/4- and full-scale tests of spent fuel casks containing a single rod bundle with DUO_2 in un-pressurized fuel pins. In later German experiments (1994), managed by GRS and performed at the Gramat facility in France [5], a HEDD was fired into a full-scale, but 1/3-height, Castor transport cask containing nine surrogate DUO_2 fuel assemblies with pressurized fuel pins. The total amount of airborne fuel particles 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. In both of the SNL and GRS experimental programs, the amount of respirable materials released from the penetrated casks to the surrounding volumes was measured and ranged from 7.7×10^{-4} to 4.6×10^{-3} for the SNL tests and 6.0×10^{-5} to 3.2×10^{-4} for the German tests [12]. Earlier tests performed with DUO_2 at Battelle Columbus Laboratory [10] yielded a respirable release value of about 3×10^{-5} for full-cask penetration. These respirable release values are different from, and can be compared with, the Respirable Fraction (RF) values produced, as defined and measured in this WGSTSC test program. Luna [12] compared the different measured respirable materials released from the earlier SNL and GRS tests and also provided a simple model to account for the effects of pin pressurization release and the ratio of pin plenum gas release to cask free volume. Luna also discussed the ranges of uncertainties associated with these release measurements. In a later evaluation, Luna et al. [13] derived RF values from the large-scale SNL and GRS DUO_2 test measured respirable release values; calculated surrogate DUO_2 RF values ranged from 1.7 to 5.3% for one-wall cask penetration, or 2.8 to 12% for two-wall cask penetration. These RF values will be compared to the smaller DUO_2 RF values measured in this SNL-WGSTSC test program, and presented in Section 4.1.1. These earlier SNL and GRS full-scale experiments produced realistic source term release data, but due to the use of surrogate DUO_2, significant uncertainties remained concerning proper application of the data to actual spent fuel.

These described experiments and subsequent analyses [4, 6, 14, 15] 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. The BCL SFR value of 5.6 was used in calculations that demonstrated that the NRC Urban Study [8] estimate for respirable release was orders of magnitude too high; however, authors of the Sandia study felt strongly that the best estimate of SFR, if there were more data, would more likely be of the order of 1 to 2 [15].

It is clear [12] 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 (DUO_2) and actual spent UO_2 fuel. Philbin [16] also provided a brief overview of the evaluations of these aerosol test results and documented the need for future confirmatory experiments, compared to earlier work, to be performed. These evaluations, plus concerns about the quality or reliability of earlier sabotage related test results led up to the conduct of the current SNL-WGSTSC test program. The accurate determination of the SFR value is the critical driving force or justification for the present experimental program.

1.4 WGSTSC History

The international working group, "Working Group for Sabotage Concerns of Transport and Storage Casks," (WGSTSC) was initiated in July 1999, at an information exchange meeting held at SNL, with participants from DOE, NRC, SNL, GRS, IPSN (now IRSN), and others. Participants concluded that more detailed analyses of the limited existing data, plus further HEDD-aerosol experimental studies were needed to better understand the possible impacts of potential terrorist acts on spent fuel shipments and other related radioactive materials. The small number of earlier experiments, as described, as well as non-similar experimental configurations, provided a general understanding of the relative behaviors of actual spent fuel and surrogate DUO_2 materials tested in the potential high energy environments that could exist in an attack. The lack of a detailed analysis has thus required estimates of potential consequences of an attack to be very conservative. The participants concluded that a common, or shared, experimental program could beneficially serve the data needs of all organizations.

By the second meeting of the WGSTSC, held in March 2000, near Paris, and hosted by IPSN, a decision was made to plan for further cooperative HEDD explosive-aerosol, small-scale testing with both surrogate and actual spent fuel materials. By the third meeting of the WGSTSC, held in July, 2000, in Berlin, and hosted by the GRS, the first spent fuel sabotage - aerosol ratio experiment concept was developed 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" [17]. The original, representative test matrix from this document is reproduced in Section 2, Table 2.1, primarily to show how the current test program design has evolved and expanded. Preliminary program testing was initiated in 2001 at both SNL, Albuquerque, NM, and at the Fraunhofer Institute (ITEM), in Hannover, Germany, and is described herein.

The International WGSTSC participants have continued to have formal, collaborative technical meetings approximately twice-a-year since 1999 to: discuss progress and future plans of the joint test program, evaluate the data generated, discuss modeling and assessment strategies, and discuss relevant safeguard-related items of common concern, all in an open, non-classified manner. A list of all WGSTSC meetings to date follows.

- 1st Meeting, July 1999, host: SNL, Albuquerque, NM.
- 2nd Meeting, March 2000, host: IPSN, Fontenay-Aux-Roses, France.
- 3rd Meeting, July 2000, host: GRS, Berlin, Germany.
- 1st Technical Meeting, December 2000, host: SNL, Albuquerque, NM.
- 4th Meeting, May 2001, host: OCNS, London, United Kingdom.
- 2nd Technical Meeting, November 2001, host: IRSN, Cadarache, France.
- 3rd Technical Meeting, July 2002, host: SNL, Albuquerque, NM.
- 4th Technical Meeting, November 2002, host: SNL, Albuquerque, NM.
- 5th Technical Meeting, May 2003, host: GRS, Cologne, Germany.
- 6th Technical Meeting, Nov. 2003, host: DOE, Washington, DC, and ANL, Chicago.
- 7th Technical Meeting, May 2004, host: OCNS, Edinburgh, Scotland.
- 8th Technical Meeting, November 2004, host: SNL, Albuquerque, NM.
- 9th Technical Meeting, May 2005, host: IRSN, Cadarache, France.
- 10th Technical Meeting, November 2005, host: SNL, Albuquerque, NM.
- 11th Technical Meeting, May 2006, host: GRS, Kandern and Garching, Germany.
- 12th Technical Meeting, December 2006, host: SNL, Las Vegas, NV.
- 13th Technical Meeting, May 2007, host: IRSN, Fontenay-Aux-Roses, Saclay, France.
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1.5 Objectives, Regulatory Uses, and Program Benefits

The primary objectives of this overall spent fuel sabotage aerosol program are to conduct experiments plus supporting analyses to obtain detailed data on the generation of aerosol, primarily respirable, particles resulting from intentional HEDD and other weapons attacks on actual spent fuel rods with various fuel characteristics, and similar surrogate materials.

Specific program objectives include:

1. Provide *reliable*, measured source-term data and technology transfer for plausible sabotage radiological consequence modeling, and related security studies, relevant to transportation. The data obtained will be shared with all participating WGSTSC partners, with appropriate cooperative agreements.
2. Support evaluations to realistically estimate effects and consequences of sabotage attacks on SNF in particular, and hazardous materials, in general.
3. Provide a basis for National Authorities' assessments of appropriate levels of physical protection and safeguards requirements and strategies for nuclear materials in use, transport, and storage. Information developed in this program may be used to guide development of future transportation security plans.
4. Defensibly assess effectiveness of, and enhancements to, mitigation safeguards and preventative security strategies, implementation, if needed.
5. This program complements efforts to build and maintain strong, collaborative relationships with national and International WGSTSC partners on sabotage/security evaluations, to counter nuclear terrorism activities.
6. Support Yucca Mountain Repository (DOE OCRWM) transportation sabotage concerns and evaluations.

There are also multiple governmental, regulatory, and competent authorities' needs for results and findings of this experimental program:

7. Provide reliable source-term data and supporting analyses for the generation and release of respirable aerosol particles, plus atmospheric dispersal hazards, from a plausible sabotage attack on spent fuel in transport or storage casks. Separate modeling efforts and radiological consequence assessments are presumed to follow.
8. Provide reliable source-term data and technology transfer to help guide and validate technical bases for transport and storage regulations (i.e., 10 CFR Parts 71, 72, and 73).
9. Provide enhanced interpretations and clarifications to limited, 20+ yr-old transport cask aerosol data from Sandia, plus others, conducted in support of prior DOE transport and NRC Urban studies.
10. Provide further validation of NRC vulnerability studies … to enhance old, limited data.
11. Results and findings supplement NRC vulnerability assessments for nuclear facilities and operations, and also support the Office of Homeland Security, in response to terrorism activities.
12. In addition, measured data may reduce more speculative anti-nuclear statements about risks associated with radioactive and nuclear shipments.

There are significant benefits for the continuing, successful conduct and completion of this test program for all participants involved. The lack of adequate, detailed analyses based on prior, limited, and defensible data has required previous estimates of potential consequences of a sabotage attack to utilize very conservative assumptions in modeling assessments. This level of conservatism should be minimized by the data output and evaluations originating from the current test program. 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. In addition, the use of cooperatively shared expertise, supporting tests, analyses, and resources between organizations in the International Working Group for Sabotage Concerns of Transport and Storage Casks (WGSTSC) is being used to successfully accomplish these goals.

2. Experimental Program Summary

The original WGSTSC proposed explosive-aerosol test matrix from 2000 [1, 17] is shown in Table 2.1, primarily to show how the current test program design has evolved and expanded over time. This proposed test matrix consisted of three test phases. Phase 1, would test glass targets, as a "representative" brittle material, primarily as a "checkout or shakedown" series in support of other tests. Phase 2 was proposed to include tests with DUO_2 pellet targets and Phase 3 testing would consist of about three tests with single, short rodlets of actual spent fuel. Phase 2 and Phase 3 aerosol results would be used to determine the spent fuel ratio, SFR, value.

Table 2.1. Original WGSTSC Proposed 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_j / 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 overall test program plan and design was revised and approved by the WGSTSC members in 2002 and documented in Sandia Technical Report SAND2004-1832 [1]. A "new" Phase 2 was added to the previously defined test phases at this time. The new Phase 2 used non-radioactive surrogate target rodlets consisting of sintered pellets of cerium oxide (CeO_2, as a ceramic and chemically representative surrogate for UO_2 fuel pellets) within a Zircaloy cladding tube, both with or without added non-radioactive fission product chemical dopant materials. The multiple technical benefits of adding tests with surrogate CeO_2 pellets are discussed in Section 2.2.1.1. It was recognized that extensive nuclear safety procedures and approvals required to explosively particulate radioactive materials, e.g., DUO_2 and spent fuel in appropriate facilities at SNL could take several years. Non-radioactive Phase 2 testing with cerium oxide allowed testing to be performed in existing, approved facilities while the required explosive and nuclear approvals for radioactive Phase 3 tests with DUO_2 and Phase 4 Tests with actual spent fuel were in process (to be described). The technical report SAND2004-1832 [1] identified the number and sequence of tests for the total program. The current, overall program consisted of four, main, linked test phases, Phase 1 through Phase 4, to be conducted in a sequential, cost-effective, and safe manner. It also documented test component plans and requirements as of the end of FY 2003. It was recognized that the test complexities, expenses, and importance of data obtained would increase sequentially as we progressed from Phase 1 through Phase 4.

Changes to the test plan were made based on test observations and improvements plus programmatic decisions made during 2004 and 2005. During FY 2004, the extensive series of Phase 2 tests was extended to include a limited number of Phase 2/Phase 3 tests, intended as the initiation to, and practice for the following Phase 3 DUO_2 tests [2]. In addition, in FY 2005, Phase 2 was again extended to include a sub-set, Phase 2+ series of surrogate tests [3] that were intended to resolve several variables and their effects on fission product dopant respirable production and enhanced sorption onto other respirables produced. Both of these Phase 2 extensions will be described in detail in Sections 2.3 and 2.4.

Individual tests in each phase will use the identical type of HEDD (CSC1, in Table 2.1), but different test materials, with similar test apparatus geometries. 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 has allowed the addition and evaluation of multiple test variables and target material (pellet) response to HEDD jets, and consequent aerosol particle production.

2.1 Test Phase 1

Phase 1 testing was initiated in 2001 primarily at the Fraunhofer Institute (ITEM), Germany, and continued through the first half of 2002 at Sandia National Laboratories. There was significant testing cooperation between both organizations. This test phase included: performance quantifications of the HEDD devices; characterization of the HEDD jet properties with multiple tests; refinement of the aerosol particle collection apparatus being used; and, Phase 1 included HEDD jet-aerosol tests using leaded glass plates and glass pellets, serving as representative brittle materials. Phase 1 work, experimental results obtained, observations, and preliminary interpretations were comprehensively documented in Sandia Technical Report SAND2005-5873 [18] and will be summarized in this document. Phase 1 testing was quite important for the design and performance of the subsequent Phase 2 test program and test apparatus. The aerosol test chamber used for Phase 1 testing at SNL is very similar to the initial Phase 2 test chamber illustrated in Figure 2.2.1.

The Phase 1 tests, as originally defined [17] were to include pre-experiments with aerosol collection and classification units performed to:

1. Verify the aerosol scaling laws for the brittle material size distributions to be measured.
2. Study the energy transfer between projectiles having various speeds (bullet speeds, high-speed gas guns, up to and including a HEDD jet) and surrogate brittle material specimens with and without cladding.
3. Obtain information on the necessary size of the final aerosol test chamber suitable for subsequent Phase 3 and Phase 4 radioactive experiments in a suitable nuclear facility test cell.
4. Find out the necessary precautions to be taken for proper HEDD blast shielding of the aerosol instrumentation, and …
5. Optimize the basis design for the aerosol particle collection apparatus and test chamber.

2.1.1 Aerosol Scaling Law for Brittle Materials

In a meeting at the Fraunhofer Institute (ITEM) in Hannover, Germany, on May 25, 2001, personnel from the SNL, GRS, Fraunhofer, and NRC agreed that the linearity of the aerosol scaling law for brittle materials (including ceramic fuel pellets of uranium oxide, among others) must be confirmed for HEDD impact velocities (~ 8 km/sec), and that this validation should be conducted prior to initiation of the feasibility study for conduct of explosives and radioactive dispersion experiments at a nuclear facility (the final phases of this test program). Because the validity of the scaling law was untested for impact velocities greater than 1 km/sec (approximately bullet velocities), proposed high velocity HEDD tests would subject representative brittle surrogate materials, such as glass pellets, rods, and plates, to impacts from projectiles traveling at velocities very much greater than 1 km/sec [18].

If the linearity of the aerosol scaling law is shown to hold at HEDD jet impact velocities, at ~8 km/sec, then the test hardware to measure the particles released from surrogate and real spent fuel materials at HEDD velocities can be simplified. Specifically, the centrifugal classifier, vertical elutriator apparatus used by Fraunhofer ITEM to measure particles from 10 to 100 μm aerodynamic diameter (AED) could be replaced or modified for the Phase 4 tests that use highly radioactive spent fuel. This test sampling modification would greatly simplify the measurement process for the Phase 3 surrogate DUO_2 and Phase 4 spent fuel tests that require total particulate containment in a nuclear facility. The measurement of aerosols up to 10 μm AED could be performed with cascade impactor particle collection devices and the collection of aerosol particles up to 100 μm AED could be performed using a velocity and gravity separation technique, i.e., by vertical elutriation.

Earlier tests conducted at Fraunhofer Institut with a gas gun used rigid projectiles, and a scaling law for aerosols from brittle materials was developed for projectile impacts up to ½ km/sec. Refer to ([18] APPENDIX A, by W. Koch) for further detail. A graph developed from the gas gun impact tests that demonstrate the linearity of two scaling law parameters, specific energy input versus cumulative particle size for glass plate tests, is shown in Figure 2.1.1.

Fraunhofer [18] (W. Koch) also derived the universal power law function: $Q_3(x_{ae}) = a(x_{aed})^v$ describing the cumulative mass distribution of the airborne fragments, as shown for a few brittle materials examples in Figure 2.1.2. The exponent "v" takes values very close to one, irrespective of the material and the specific energy input. Thus the measurement of one point of the mass distribution allows for the prediction of the entire mass size distribution in the size range between 1 and 100 μm AED. In the sub-micron size range, the scatter becomes quite large for some ceramic materials (DUO_2, AlSi) due to the fact that intra-grain boundary cracks are required for the formation of particles in this size range. From these results, one can argue that for comparable modes of energy input, the spent fuel pellets, also being brittle ceramic material, behave very similar to the DUO_2, i.e., the SFR for the mass (< 100 μm, especially < 10 μm) being very close to 1. Further details are provided in [18], APPENDIX A, by W. Koch.

2.1.2 Phase 1 Test Aerosol Results

Aerosol particles produced from HEDD jet impact with four Phase 1 leaded glass plate targets were collected and sampled at SNL with two Respicon particle impactors (measuring respirables in the 0 to ~5 μm AED and ~5 to ~10 μm AED ranges, and aerosols in the ~10 to 100 μm AED range). The residual particles within the aerosol collection chamber were collected and removed for mechanical sieve fragment analyses. Laser diffraction spectroscopy (LDS) analysis was used for the smallest sized residual particles. Chemical analysis of the lead in the leaded glass plate particles was also used. Most materials collected from the aerosol box chamber originate from the glass plates. Further experimental details, particularly those relating to non-aerosol results and the evaluations of the HEDD and HEDD jet are presented in [18].

Particle size evaluations from the Respicon impactors, sieve measurements and LDS for all four tests are shown in Figure 2.1.3. The Respicon and LDS measurements agree surprisingly well [18]. (Respicon measurements from test 1 and 2 were disregarded due to losses of particles in the

Styrofoam layer in test 1 and 2). All four tests appear quite reproducible, in view of the large fragments formed. There is relatively large scatter in the small particle size data, < 10 µm AED; test 3 (with a smaller sized glass plate) results are significantly different, greater than respirable results from tests 1, 2, and 4. The combined particle size data from tests 1, 2, and 4 is shown as cumulative fraction vs. particle size in Figure 2.1.4. The size distribution fit to the data suggests a brittle material scaling law exponent of 1.2, close to 1. As a result of the large scatter in the respirable particle size range, extrapolation from 10 to 100 µm AED is not possible; further particle size measurements in this range would be desirable in the future.

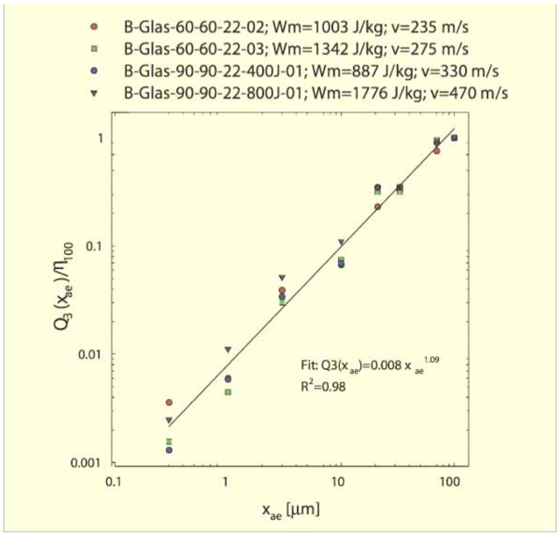


Figure 2.1.1. Cumulative Particle Size vs. Particle Size (µm), Glass Slabs

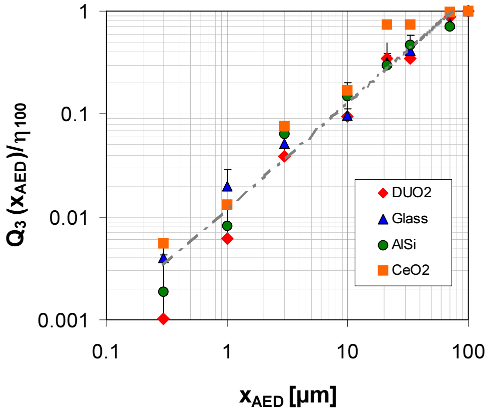


Figure 2.1.2. Cumulative mass size distribution in the relevant size range normalized to its value at x_{AED}=100 µm: low speed pellet impact test

Where $Q_3(x_{ae})$ = the cumulative airborne mass fraction as a function of the aerodynamic diameter, $m_{airborne}(x_{ae})$ = the cumulative airborne mass as a function of the aerodynamic diameter, and $Q_3(x_{ae=100\mu m})$ = the release fraction smaller than 100 µm AED, η100

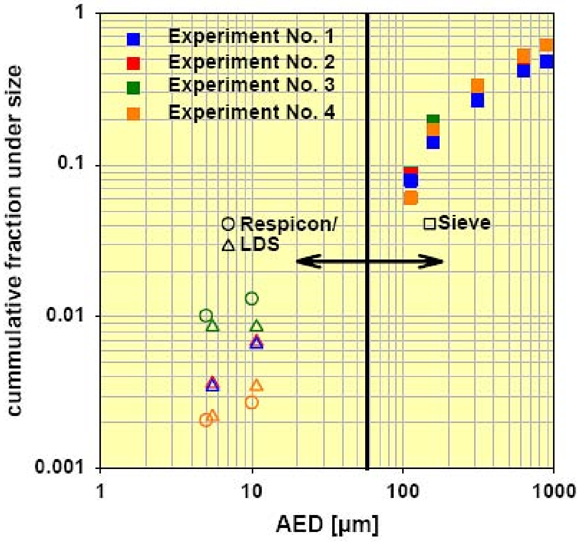


Figure 2.1.3. Phase 1 Glass Plate Test Cumulative Particle Size Distribution

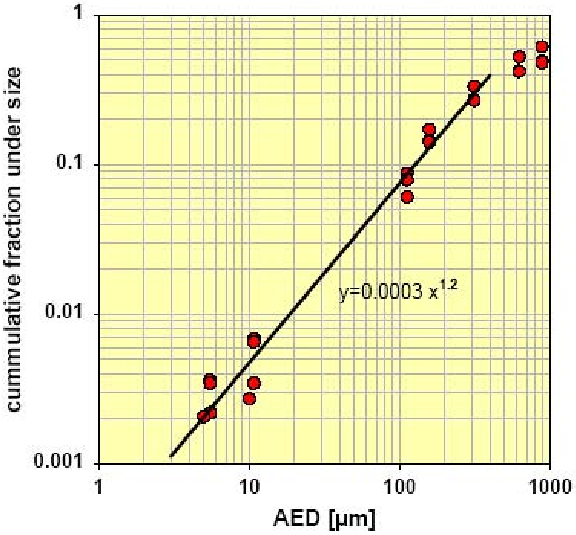


Figure 2.1.4. Phase 1 Glass Plate Test Cumulative Fraction Under Size

Some of the preliminary results and conclusions from these Phase 1 glass plate-HEDD jet impact tests, and how they led to modifications in the subsequent Phase 2 test program, are as follows:

1. Under the experimental conditions used, the fragmentation process of the glass plates is reproducible and gives fairly consistent results. However, uncertainties remain (to be addressed in future Phase 2 testing). These experiments did not allow us to exactly discriminate between primary (HEDD jet/ projectile impact related) and secondary (chamber wall impact related) fragmentation processes. The secondary processes, however, appear to be small. The amount of fragmented material escaping the aerosol collection box through the HEDD jet entrance and exit holes could not be quantified and characterized.
2. An aerosol collection chamber with uncovered walls (i.e., with no Styrofoam sheets) can be used in the future. Collection chamber dimensions in the HEDD jet direction should be of the order of 30 cm (12 inch). It would be desirable to close the CSC jet entrance and exit holes as soon as possible after CSC detonation. These suggestions were incorporated in subsequent Phase 2 surrogate testing [1].
3. The experimental set-up used in Phase 1 enables a suitable simulation of the HEDD jet effect on the representative brittle glass material for the purposes of performing future comparison experiments with DUO_2 surrogate and spent fuel targets for the determination of the Spent Fuel Ratio as a function of aerosol particle size.
4. The contribution of the explosive soot particles must be accounted for, primarily in the particles size range below 10 µm AED. This conclusion led to the use of elemental chemical analyses for collected particles, and was initiated in the following Phase 2 test program [1].

The representative surrogate, leaded plate glass particle size distribution appears to follow universal power law behavior for brittle materials with, however, some uncertainties (observed scatter) in the respirable size range, < 10 µm AED. More detailed size and nuclide specific analyses in the respirable size range are required, and were implemented in test Phase 2.

2.2 Test Phase 2

The extensive Phase 2 series of tests used nonradioactive, sintered ceramic pellets of cerium oxide (CeO_2) contained within Zircaloy-4 cladding tube assemblies, similar to spent fuel rods. Twenty four Phase 2 surrogate material tests were conducted from October 2002 through April 2004. The complete Phase 2 test matrix is provided in Table 2.2.1, and lists major test variables and dates. The Phase 2 surrogate tests allowed us to evaluate multiple test variables, pellet and rod responses to HEDD jet impacts, and to fine-tune the experimental setup. The primary compilation of Phase 2 tests details and results was documented in both SAND2004-1832 and SAND2005-4446 [1-2]. This section describes experimental detail for test Phase 2 test components, predominantly. We shall summarize the most important of these details herein. A significant extension of Phase 2 was added in 2005 and is termed test Phase 2+; these tests are described in detail in Section 2.3.

2.2.1 Test Component Details

The major components required for conduct of the Phase 2 surrogate spent fuel sabotage, HEDD impact, and aerosol measurement tests include: the test rodlet (pellets of cerium oxide, in many cases, fission product dopant chemicals, and Zircaloy-4 cladding tubes), the test chamber, aerosol particle samplers, multiple temperature and pressure measuring instrumentation, a conical shape charge (the HEDD), an a facility to perform the explosive-aerosol testing within.

Table 2.2.1. Phase 2 Tests: CeO_2 Surrogate Test Matrix

Test #	Pressure	Dopants	Variables	Date
0	1 bar	no	top; system checkout	10/2002
1A, 1B	1	no	top, center; Respicon samplers	10/2002
2A, 2B	1	no	optimized for French pellet & tube size	12/2002
3A, 3B	1	no	optimized for U.S. pellet & tube size, Respicon & Berner samplers	7/2003
4A, 4B	1	yes	" + rev. equipment design, FP dopants	8/2003
5A – 5G	1	yes	" + vertical test chamber, instruments, Marple particle impactors.	9/03-1/04
6A, 6B	28-38 (blow-down)	yes	" + equipment design modifications, Marple impactors, Large Particle Sep.	4-5/2004
7A, 7B	1	yes	**German HLW glass rod, dopants** (nonradioactive)	2/2004
8A – 8D	1	yes	particle impactors & sampling optim.	2-4/2004

FY 2003 ⬆⬆⬆⬆

FY 2004 ⬆⬆⬆⬆

Successive tests in the Phase 2 (2/0 through 2/8) series allowed us to add and evaluate effects of multiple variables on target aerosolization response to HEDD jet impact, including: internal rodlet pressurization; different fission product dopant additions and form/distribution factors; several types of aerosol particle samplers; and, target rodlet materials (CeO_2 pellets of various lengths, German surrogate high-level waste glass rodlets or pellets).

2.2.1.1 Surrogate Target Pellets: Most Phase 2 tests incorporated sintered ceramic pellets of cerium oxide (CeO_2, ceria, cerium dioxide, or ceric oxide). CeO_2 was selected for use as a surrogate "spent fuel" pellet material because it has the following beneficial, technical properties:
(a) primarily, it is chemically similar to, and representative of unirradiated uranium oxide; (b) it is non-radioactive; (c) it can be fabricated into ceramic pellets by pressing and sintering, similarly to a new uranium oxide fuel pellet; (d) 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 fuel (both fresh and spent) and DUO_2 ceramic pellets.

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). CeO_2 is also an oxide quite chemically similar to the UO_2 and PuO_2 in nuclear fuel -- and the DUO_2 in the Phase 3 surrogate tests. CeO_2 and UO_2 have the same ionic crystal, fluorite-type structure. For these reasons, CeO_2 was used in multiple decontamination studies performed at Sandia National Laboratories [19-21]. CeO_2 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.

Cerium oxide is a refractory oxide with a very high melting point, approximately 2600 °C, compared to 2878 °C for UO_2. CeO_2 is an oxide ceramic like UO_2, 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, 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 CeO_2 and UO_2. Therefore, the participants in the WGSTSC concluded that CeO_2 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.

Cerium oxide powder was pressed and sintered into ceramic pellets for our testing purposes by the Ceramic Synthesis and Processing Department at Sandia National Laboratories. The cerium oxide powder (99.9 % pure, about 5 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 7 mm-long pellets) into "green" pellets, fired at about 600 °C for binder burnout, and then sintered at about 1600 °C. Measurements of apparent pellet porosity and Archimedes density were then made. The pellets were made to fit snugly (i.e., with minimal pellet-to-cladding gap) into Zircaloy-4 cladding tubes. The average theoretical density of sintered pellets used ranged from 85% - 95%, or 6.11-6.80 g/cc. The complete measured specifications for cerium oxide pellets used were presented in Table 6 in [1] and in Table 4.1 in [2].

With WGSTSC members cooperation, non-radioactive German high-level waste (HLW) glass test rods, containing multiple, non-radioactive fission product dopants, were added to, and tested in Phase 2 for tests 2/7A and 2/7B [2]. 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. The glass test rods were fabricated at the Karlsruhe Nuclear Research Center and provided to SNL for joint, cooperative testing by Fraunhofer ITEM and GRS. These glass test rods were 14 mm in diameter and 163 mm long, contained within a stainless steel cladding tube with about a 1 mm wall thickness. The glass matrix was composed of about 84 wt. % SiO_2, MgO, MnO_2, CaO, Na_2O and other glass frit material. The remaining 16 wt. % contained dopants, primarily thermally volatile Cs_2O (0.44 wt. %) and MnO_2 (0.30 wt. %), plus nonvolatile La_2O_3 (1.82 wt. %) and Nd_2O_3 (1.04 wt. %). These surrogate HLW glass rod aerosol tests were beneficial in providing additional data on volatile fission product enhanced sorption onto respirable particles.

2.2.1.2 Fission Product Dopants: One of the major goals of this overall experimental program is to quantify the potential enrichment of volatile fission product nuclides sorbing onto respirable-size particulates produced from a spent fuel–HEDD jet impact. Volatilized species of cesium (^{134}Cs, ^{137}Cs) and ruthenium (^{106}Ru) have been mentioned as most significant because of concerns about inhalation radiotoxicity. Cs exists in several forms in spent nuclear fuel (SNF) [22-23], primarily as Cs vapor, plus complex oxides (e.g., cesium uranate, Cs_2UO_4, and Cs molybdenate, Cs_2MoO_4). In colder rod regions (e.g., at fuel cladding bond, especially at crack tips), CsI vapor can condense. In general, the fission product yield of Cs is much higher than that of I, so all the iodine may be tied up as CsI, but certainly not all of the Cs. A really crude estimate for Cs based on fission yield is to assume 3% ^{235}U enrichment and all fissions coming from ^{235}U. If the fissions yield about 0.22 Cs/^{235}U fissioned, about 0.3 wt.% Cs would be generated. Based on isotopic measurements of ^{137}Cs in PWR fuel operated to about 50 GWd/MTU, the ^{137}Cs content is 0.16 wt.%. Ru exists in SNF mostly in metallic form with low vapor pressure. Therefore, it is not a volatile fission product. Ru is of radiological importance because of its high radiotoxicity. RuO_2 is not stable above 1350 ºC. If sufficient oxygen is present (e.g., if UO_2 transforms to U_3O_8), Ru will start to oxidize to RuO_3 and RuO_4, both with very high vapor pressures (boiling point is only 108 ºC for RuO_4). Given the very short-time/high-temperature of a HEDD jet, we may assume that Ru comes off in metallic form. If we dope the test rodlet/pellets with RuO_2, the produced aerosol may contain Ru.

Non-radioactive chemical forms of cesium (as CsI) and ruthenium (as RuO_2) have been added to the surrogate pellet test systems, starting with tests 2/4A and 2/4B [1]. 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. We also added europium oxide, as Eu_2O_3, as another non-volatile "standard" fission product dopant. For Phase 2 tests, we incorporated approximately 1000 ppm (0.1 wt %) of stable Cs, I, Ru, or Sr species per test rodlet, relative to the mass of the surrogate oxide pellet expected to be disrupted per test. Refer to Table 4.2 in [2]. These dopants were incorporated by inserting the solid dopant chemicals into small "wells" pre-drilled into one end (prior to sintering) of the cerium oxide pellets, one chemical per pellet, for tests2/4A, 2/4B, and 2/5A; 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.

We used a different technique for fabricating *stand-alone* fission product dopant disks, without the cerium oxide pellet "holders" starting with test 2/5E. These dopant disks allowed us to incorporate all fission product dopants into a minimal space, without the need to pre-drill holes or wells into pellets; this was particularly important for dopant disk use in Phase 3 test DUO_2 pellets; refer to Section 2.5. SNL personnel fabricated plastic resin-based dopant disks to slip-fit within the Zircaloy-4 tubing. These dopant disks were approximately 9.1 mm-diameter and 1 mm- thick, each, with a minimum amount of resin used. All of the solid fission product chemicals were mixed into the liquid resin, then solidified (cured) into a rod-shape, then cut with a diamond-blade saw into individual disks. Finally, individual disks were lapped to a thickness of 1 mm. Two resin-based disks containing dopant chemicals were used per test, one on each side of the central cerium oxide pellet in the target rodlet. Refer to Section 4.1.3 in [2] for further specifications and photographs of the fission product dopant additives. All fission product dopant materials were within the swept volume and were totally aerosolized and possibly vaporized by the shock wave and thermal pulse from the action of the HEDD jet.

2.2.1.3 Zircaloy Cladding Tubes: The Zircaloy-4 cladding tubes used were purchased primarily from Framatome ANP Richland, Inc. Richland, WA. This tubing was manufactured by Advanced Nuclear Fuels (ANF), GMBH, Duisburg, Germany, and was 10.6 mm outside diameter and 9.32 mm inside diameter. (NOTE: In tests 2/1A through 2/2B, we used Zircaloy tubes of 9.55 or 10.0 mm-diameter O.D). This tubing is the closest in diameter available (at the time) to U.S. PWR, pressurized water reactor fuel rods. All Zircaloy cladding tubes for test series 2/5 and 2/8 were 305 mm (12.0 inches) long. Test rodlet tubes for 2/6A and 2/6B were 254 mm (10.0 inches) long and had special end caps designed to contain internal pressurization (to be described). Zircaloy-4 consists of zirconium, with ~ 1.45 wt. % Sn, 0.11 % Cr, 0.23 % Fe, 0.34 % Fe + Cr, and 0.12 % oxygen. Some of the more significant trace impurities, at the ppm level, include: Al, C, Hf, Nb, Ni, Pb, Si, Ta, and W. Refer to Section 4.1 in [2] for further specifications and photographs of the assembled Phase 2 surrogate test rodlets.

2.2.1.4 Aerosol Collection Chambers: Three separate aerosol collection chambers were used during the Phase 2 series of tests. They are illustrated in Figures 2.2.1, 2.2.2, and 2.2.3. Figure 2.2.1 shows the first non-ventilated (non-flow), 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. This non-ventilated aerosol collection chamber was used for reasons of simplicity and durability. The similar, but somewhat modified second aerosol collection box shown in Figure 2.2.2, 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 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 escaping from the aerosol box for post-test sampling [1]. Neither version of the "square box" aerosol collection chambers can be considered as "leak-tight" or total containment, particles can and do escape. Figures 2.2.1 and 2.2.2 show a vertical, installed rodlet.

The vertical, aerosol collection/ explosive containment test chamber used in Phase 2 tests 2/5A through 2/8D (named "Grandma") is shown in Figure 2.2.3 (with steel flange cover-plates removed, not shown). The open aerosol collection chamber, with a horizontal test rodlet inserted and visible, is located in the top "aerosol collection chamber." The "explosive containment chamber" is on the bottom. When the HEDD installed in the bottom chamber is remotely detonated, a HEDD jet shoots upward through a small-diameter (2.5 cm) hole in the thick steel plate between the two chambers, penetrates the test rodlet (inserted horizontally; self aligning insertion), and is stopped in the thick HEDD jet-stop block on the top of the test chamber, not visible. A schematic of the explosive-aerosol test process and apparatus is illustrated in Figure 2.2.4. The entire test chamber (body, exclusive of top-mounted aerosol apparatus and valves) is approximately 0.6 m-diameter by 1.3 m-high, and is fabricated out of thick steel to contain the explosive blast and all aerosols produced. This vertical test chamber is a durable and demonstrated particulate leak-tight, total containment system [2], as differentiated from the earlier "square box" aerosol chamber. This test chamber was instrumented to measure multiple temperatures and pressures in both the top aerosol collection chamber and in the aerosol sampling apparatus lines shown at the top of Figure 2.2.3.

Figure 2.2.1. Initial Phase 2 Test Apparatus

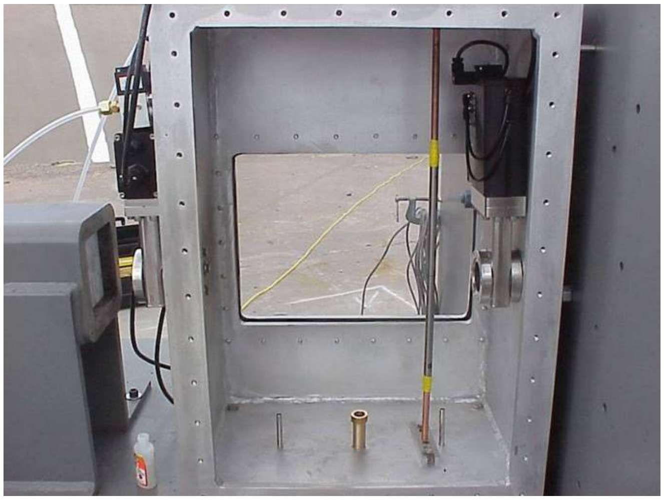
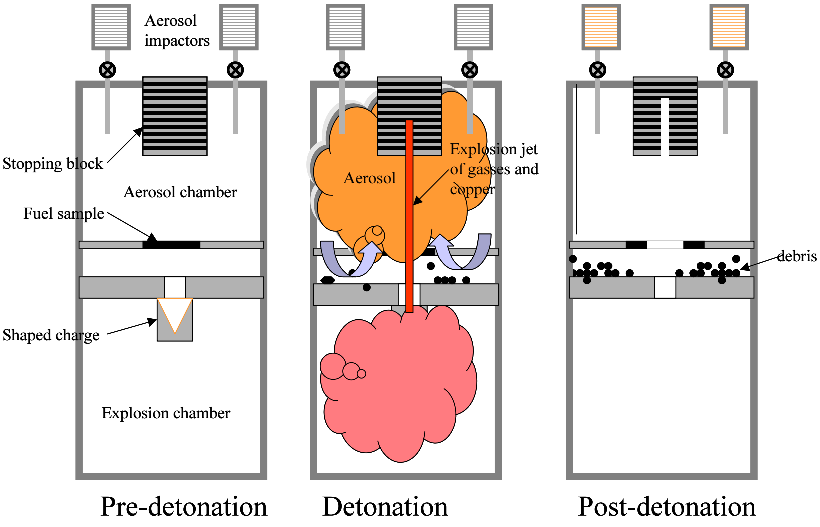


Figure 2.2.2. Second Phase 2 Test Apparatus

Figure 2.2.3. Third Phase 2 Vertical Explosive-aerosol Test Chamber



Pre-detonation Detonation Post-detonation

Figure 2.2.4. Explosive-Aerosol Test Apparatus Schematic

2.2.1.5 Aerosol Particle Sampling System: Several types of aerosol particle samplers were used during conduct of the Phase 2 tests. The Respicon™ 3-stage virtual particle impactor (two/ test), and a Berner 9-stage particle impactor were used through test 2/5A, then replaced; both of these impactors have been described in detail earlier [1]. Starting with test 2/5B, multiple 9-stage, multi-jet Marple cascade impactors (model 298) were used in each test, operating in an independent and replicate manner. These impactors measure aerosol particle size distributions from about 0.4 - 21 µm AED, including a final (stage 9), base stage (~ all particles < 0.4 µm), plus a pre-filter stage (stage #0, for larger particles up to about 35 µm AED). The impactor is constructed of aluminum with fiberglass substrate collection media (modified for Phase 2+ tests [3]). In addition to the Marple impactors, a separate, in-line, large-particle separator (LPS) was used for collecting the larger ~30-100 µm AED particles, starting with test 2/5E. The LPS is the semi-cylindrical component visible at the top of Figure 2.5.5. The LPS collectors were jointly designed by SNL and Fraunhofer ITEM aerosol experts and fabricated by SNL; the design is based on work published previously [24]. Each Marple and LPS sampling sub-system requires a vacuum bottle and a critical orifice to draw a calibrated, nominal 2 L/min flow rate through the samplers; a small HEPA filter is also used, before the vacuum bottle.

For tests 2/6A, 2/6B, and 2/8A-D, a separate, additional line of six sequential Gelman filter samples were incorporated, to monitor particle stratification and settling over about the 2.5 - 60 second period after HEDD detonation. The Gelman filter samples are shown near the top of Figure 2.2.3. These are in addition to the Marple impactors and LPS. Two sampling levels in the aerosol chamber (near top and lower/at rod-target level) were used for the impactor samples with two impactors at each level. The Gelman filters effectively collected all aerosol particles which pass through their glass fiber media. Further details on the Marple impactors, the LPS, and the Gelman filters plus supporting sampling apparatus were described in Section 4.3 of [2].

2.2.1.6 Conical Shape Charge (CSC): Referred to as the HEDD in this document. Individual tests in each phase of this program used identical precision CSCs, containing PBX-N5 explosive (95% HMX and 5% VITON A). The CSC incorporates a copper (Cu) cone and produces a copper jet. Within the test chamber, the CSC is held in place with a polyvinyl chloride (PVC) fixture-holder, and has a CSC-to-rodlet stand-off distance of 7.5 inches (19 cm). Further characterization and performance parameters and details of the CSC were provided in Appendix B of [18].

2.2.1.7 Test Facilities: Phase 2 (plus Phase 2+ and Phase 2/Phase 3) tests were all performed at various locations of the Sandia Explosive Technology Group (ETG), predominantly in SNL building 905 (previously referred to as the Explosive Components Facility, ECF; for tests 2/0, 2/1A-1B, 2/2A-2B, 2/6A-6B, 2/7A-7B, 2/8A-8D, and 2/9A-9B). Other Phase 2 tests (2/3A-3B, 2/4A-4B, and 2/5A-5G) were performed outside of the SNL ETG remote-site building 6750, the Gun Site/Terminal Ballistics Facility, in SNL Tech Area 3.

2.2.2 Phase 2 Test Major Results

2.2.2.1 Particle Collection and Analyses: The aerosol particles produced by HEDD jet impact with the Phase 2 target rodlets were collected on the various stages of the aerosol particle devices used, with the Respicon, Berner, Marple, or LPS collectors [1-2]. The particulated test materials (aerosols plus non-aerosol particles) remaining in the aerosol collection box or chamber , i.e., the impact "debris" not collected by the aerosol particle samplers, were manually brushed from the inner aerosol box walls, collected as well as possible, then mechanically sieved. This impact debris consists of heterogeneous fragments and particles of cerium oxide pellets, plus some plastic (detritus from the HEDD holding fixture), copper from the conical shape charge cone and jet, Zircaloy metal pieces from the cladding tube, plus aluminum or iron from the inner walls of the test chamber and/or the HEDD-jet stop block, etc. Some particulate material was “blown out” of the “square box” aerosol chamber in tests 2/1A - 2/4B, through HEDD jet entrance and/or exit holes. Sieve meshes used were: 1000, 500, 250, 125, and < 125 µm (residual) geometric size; additional sieves with meshes of 74, 44, 37, and 25 µm geometric were used later.

We used gravimetric weight analyses plus detailed chemical instrumental analyses, inductively coupled plasma/ mass spectrometry (ICP/MS), for evaluation of major elements in all of the collected aerosol particles and for impact debris up to 125 µm geometric size (equivalent to 325 µm AED for CeO_2 particles). Chemical analyses were necessary because much (about half) of the collected particle mass consisted of fine carbon soot -- a combustion byproduct produced by the HEDD explosive. The major elements detected in the aerosol ranges were copper (from the HEDD jet), first, then cerium. The collected aerosol particles were predominantly carbon soot (explosive residue) with significant copper content coincident with the mass distribution peak in the 1– 3 µm AED range. Other significant elements detected were zirconium, plus aluminum and iron. All data obtained for the collected aerosol particles and impact debris, including weights, weight percents, and elemental analyses for each particle size range, were listed and plotted in Appendix A of in both technical reports [1-2]. Representative photographs of the impact debris as a function of sieved size were also presented. Representative plots of analyzed major elements and fission product dopant elements as a function of size, from 0 to ~ 35 µm AED, for tests 2/5G are shown in Figures 2.2.5 and 2.2.6 from [2].

2.2.2.2 Aerosol Particle Results: The Phase 2 test aerosol particle results are presented in Section 4, combined in a tabular and graphical manner with similar Phase 2+ and Phase 3 results, to provide an overall data compilation and to allow full comparisons between tests with different variables. The measured Phase 2 test Respirable Fraction (RF) values for respirable aerosol particles of cerium oxide, zirconium, and the fission product elements of cesium, ruthenium, strontium, and europium, are presented in Section 4.1. Similarly, the measured Enrichment Factor (EF) values for cesium, ruthenium, strontium, and europium are presented in Section 4.2.

The calculated CeO_2 cumulative distributions (cumulative mass fraction vs. particle size) for tests 2/4A through 2/8D, for particles from ~0.25 to ~650 µm AED were presented in Figures 6.28 through 6.36 in [2]. The cerium oxide particle data collected with particle impactors matches up well with the larger impact debris particle data obtained from mechanical particle sieving. The particle sizes from the sieve data were adjusted to match up with the aerodynamic size from the impactor and all the data were normalized by the total mass of CeO_2 disrupted and dispersed into the chamber. The data from the tests 2/4A, 2/4B, 2/5G, 2/8C, and 2/8D indicate 10% to 30% of dispersed CeO_2 material is less than 250 micrometer sieved size (650 µm AED) and the general trend is along a single, nearly straight line on a log-log plot. The data for tests 2/6A and 2/6B, with the internally pressurized test rodlets, show a slightly higher fraction (>30%) of debris less than 250 micrometers sieve size, lower fractions of mass in the aerosol samples, and a steeper slope in the debris size distribution than the other tests [2]. The cerium oxide cumulative fraction distributions suggest that the HEDD impact produces mechanical fragmentation without phase change; the cerium oxide surrogate material behaves as a representative brittle material.

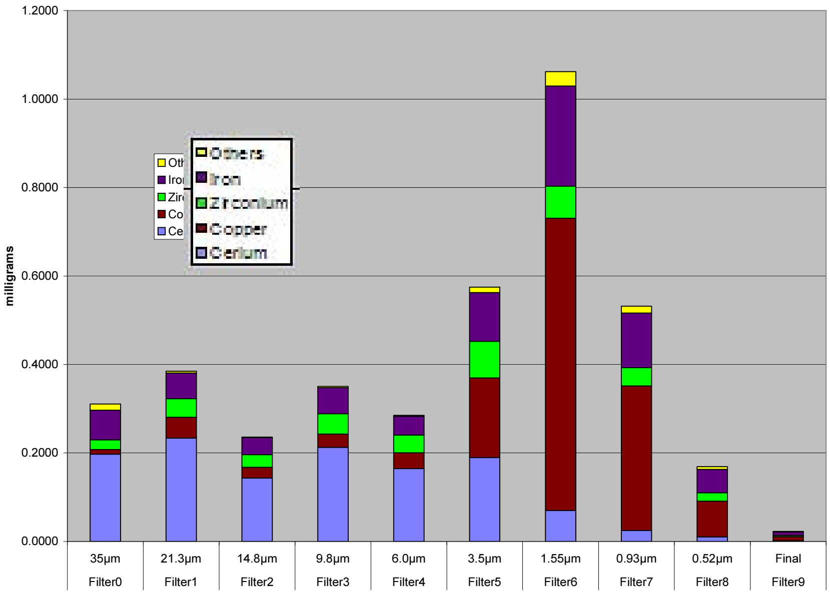


Figure 2.2.5. Test 2/5G Marple Metals Analysis Distribution, milligrams

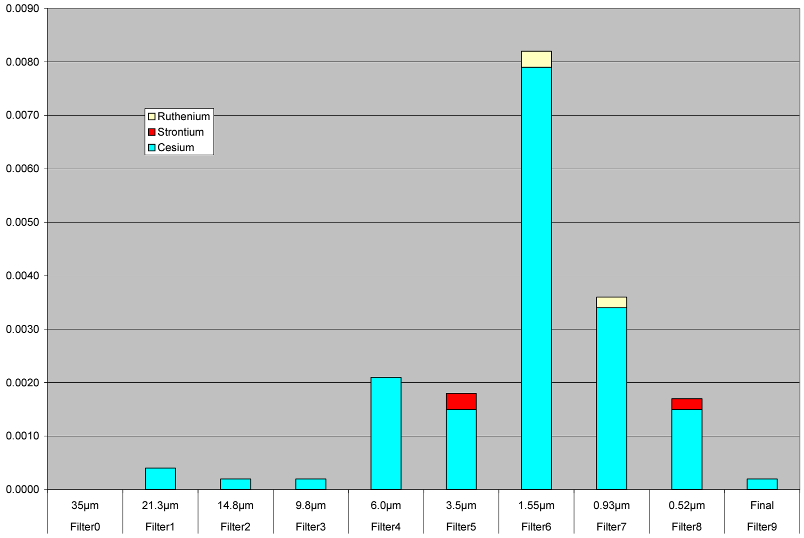


Figure 2.2.6. Test 2/5G Marple Fission Product Dopant Analysis Distribution, mg

Note: The CeO_2 cumulative mass fraction distribution results do not extend up to 100% because large, non-aerosol-size particles were not analyzed, even though the total CeO_2 disrupted mass was used for subsequent calculations.

2.2.2.3 Particle Stratification Within Aerosol Sampling Chamber: Tests in the 2/6 and 2/8 series used six sequential Gelman filter bank samples and four simultaneous, replicate Marple impactor samplers. A major purpose of these tests was to characterize the particle mass loading and distribution within the aerosol chamber over the 2.5 to 12.5 second sampling period (after HEDD detonation) for the impactors, as well as the 2.5 to 47.5 second sampling periods for the Gelman filter bank. Two sampling levels within the aerosol test chamber were used for the impactor samples, high and low (near rodlet height), with two impactors at each level. No significant stratification between high and low sampled particulate material concentrations was observed, based on results from these six tests. Taken test by test, the means for both levels in each test were within the 95% confidence interval of each other and, taken level by level, the means for both tests at each level were generally within the 95% confidence interval. No significant stratification of particles below about 20 µm AED occurred and no appreciable aerosol particle settling was seen over the size range of particles sampled within the aerosol chamber during the 2.5 to 12.5 second sampling period after HEDD detonation. The time-sequential sampling from the Gelman filters appear to indicate that the soot aerosols remain suspended over the 2.5 to 47.5 second sampling period. Settling is a function of particle size and may present a greater depletion for larger particles. All subsequent tests, specifically for Phase 3 and Phase 4, will be sampled at only one internal aerosol chamber location level, selected as the "lower," rodlet height level.

2.2.2.4 Aerosol Particle Morphology: Electron microscopy and energy dispersion x-ray spectroscopy (EDS) of respirable particulate material collected from stage 6 of the Marple impactor (~ 1.5 to 3.5 µm AED) from test 2/6A provided a good indicator of the aerosol chemical composition and morphology. The collected aerosol particles were predominantly carbon soot with significant copper content coincident with the mass distribution peak in the 1– 3 µm AED range; there was a similar distribution with the volatile fission product cesium. The high soot content acts as sites for vapor condensation. Four types of particle morphology were observed in electron micrographs presented in [3] and may assist in future modeling evaluations:

1. Typical agglomerated particles that range from obvious chain aggregates to more compacted densely packed material. The major constituents are copper, iron, and aluminum, with some zirconium present. The agglomerations are likely these materials condensed onto soot particles.
2. Discrete particles and spheres (copper melted material that formed spheres), with major constituents of Cu and Ce, alone and in combination.
3. Large particles that are 10s of µm in size, both monolithic and cracked. These may have formed in situ or in the gas phase. They are possibly compact agglomerates with a very high void fraction, and are likely to be a form of soot. And,
4. Most interestingly, long, thin ribbons/wires of copper were observed, present as discrete particles and as constituents of agglomerates. The copper was present as extrusions, i.e., as solid material that never melted.

The small particle mode arising from soot formation provides multiple condensation sites for vapor and/or nucleation particles. Soot and Cu and Cs size distributions are correlated. Copper is present everywhere and appears to enter the aerosol chamber as solid, liquid, and vapor. Cerium oxide is present as individual particles, with some that may have melted. Cesium was not seen in the EDS analysis. Iodine was seen in one micrograph, Figure 7.18 in [3]. Other dopant materials have not been seen so far. More microscopy is needed.

2.2.2.5 Rodlet Pressurization Effects: It was initially presumed that HEDD jet impact onto internally pressurized target rodlets may lead to an increase in the amount of pellet particulates being "blown out," with a larger resultant measured respirable fraction. Phase 2 CeO_2 explosive-aerosol tests 2/6A and 2/6B were specifically performed with internally pressurized (helium gas) rodlets at 27.6 bar and 40 bar, respectively, to evaluate the effects of pressurization. Conclusions from these evaluations are presented in Section 4.1.1 because these evaluations require comparisons to respirable fraction results from other Phase 2, Phase 2+, and Phase 3 tests.

2.2.2.6 Target Rodlet Disruption: The observed effects of HEDD explosive jet impact on the Phase 2 test rodlets, both for CeO_2 pellet-containing rodlets and the empty Zircaloy cladding tube used in test 2/5B, were fairly consistent. The HEDD jet impacts the center-point of each target rodlet, yielding a 21-30 mm *average* gap in the Zircaloy cladding tube. The total Zircaloy tubing gaps observed in Phase 2 tests varied from 16-35 mm, primarily due to jagged flaps of Zircaloy of different lengths; refer to Table 6.2 and Figures 6.5 through 6.15 (photographs) in [2]. 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.

In all tests, the CeO_2 pellets adjacent to the HEDD jet-impacted segment of Zircaloy tubing were firmly wedged into the tube by "blowback" fine particles of CeO_2 in the small tube-to-pellet gap (blown outward, perpendicular to and by the jet); pellets could not easily be removed from the cladding. The remaining, captive pellets were essentially whole, with some occasional observable external fracturing. Almost all of the blowback debris material was 250 µm (geometric) in size or smaller. A significant consequence of this blowback debris observation is that 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 fragmented cladding tube ends.

2.2.2.7 Instrumentation Results, Temperatures and Pressures: Temperature measurements from thermocouples installed in these tests were quite dependent on their locations, either within the aerosol chamber itself or within the top-mounted aerosol sampling tubing. Temperature readings in the 2/5 series of tests were taken primarily as indicators of HEDD detonation. Consequently, thermocouples were located within the aerosol chamber with survival from fragmentation as a major concern. As such, these obtained test 2/5 temperature readings may not reflect overall aerosol chamber conditions. For example, for test 2/5D, the observed temperature rise from a thermocouple at the top of the aerosol chamber increased about 20 ºC above initial ambient conditions at 20 seconds after detonation, and peaked 5 minutes after detonation at 31 ºC above initial ambient temperature.

Efforts to make more representative aerosol chamber temperature measurements were instituted in the series 2/8 and 2/6 tests. The thermocouples were generally shielded within sampling tubes, and may have inadvertently touched adjacent surfaces or were contacted by hot fragments. The aerosol chamber thermocouples were generally shielded within internal sampling tubes, and may have inadvertently touched adjacent surfaces or were contacted by hot fragments. Peak temperatures measured, as shown in Table 2.2.2, were in the 180 to 840 °C (453-1113 K) range about 6 to 11 seconds after HEDD detonation, then decreasing to < 50 °C after a few minutes [2-3]. Above the aerosol test chamber, within the Marple aerosol sampling system, the measured temperatures never exceeded ~ 40 ºC, generally with measurements of ~ +2 to +13 °C, decreasing to essentially ambient temperature within a few minutes. The significantly higher temperatures within the aerosol test chamber were reduced significantly during the aerosol transit through the sampling tube.

Pressure transducers installed within the square aerosol collection chamber of test 2/4B, measured [1] a transient pressure pulse of about 5 bar, 75 psi. For test 2/8D, measured peak pressures within the aerosol sampling chamber were ~85 psig (5.9 bar) at ~200 msec. after HEDD detonation, decreasing to < 1 psig (< 0.07 bar) after 5 minutes [2]. Within the top Marple aerosol sampling systems, the pressure transducers were isolated from the aerosol chamber by ball valves until about 2 seconds after detonation when the aerosol sampling was initiated. These measured pressures in tests 2/8A and 8B were in the range of +0 to + 28 psig (+1.9 bars). Measured upstream pressures for tests 2/8D, 2/6A, and 2/6B were quite similar, peaking at +24 psig (+1.7 bars) [2].

Table 2.2.2. Peak Measured Temperatures In Phase 2 Aerosol Chambers

Phase 2+ Test	Peak Temperature
2/6A	840 ºC
2/6B	840 ºC
2/8A	220 ºC
2/8B	200 ºC
2/8C	180 ºC
2/8D	340 ºC

2.3 Test Phase 2+

The predominant explosive-aerosol testing performed during FY 2005-06 included nine new Phase 2+ tests, tests 2/10A-2/10F, performed at SNL, outside of the remote-site SNL ECF building 6750, located at the Gun Site/ Terminal Ballistics Facility. Tests 2/10G – 2/10I were performed in Germany by Fraunhofer Institut (ITEM and EMI) in cooperation with GRS. These Phase 2+ tests were added to the original Phase 2 program matrix following discussions at the 8th Technical Meeting of the WGSTSC, in November 2004. Both the SNL and German test partners reported significant, but differing levels of cesium fission product dopant enrichment (EF values) in the smaller respirable particle sizes that were coincident with the copper (from the HEDD jet) and explosive soot respirable particles. This information plus results were originally documented at a following 10th Technical Meeting of the WGSTSC in [25]. Consequently, the Phase 2+ tests were specifically added to the original Phase 2 matrix, and used various fission product-doped surrogate cerium oxide or German surrogate HLW glass pellets to evaluate the release and enrichment (Enrichment Factor, EF) behavior of cesium, primarily, plus ruthenium, strontium, and europium dopants. Furthermore, some tests with German inactive HLW glass were added to compare results with former glass experiments. The expanded Phase 2+ test matrix is presented in Table 2.3.1. One additional Phase 2+ test, 2/CSC, was added in 2007 and is described separately in Section 3.1.

The Phase 2+ series of surrogate tests were intended to resolve several variables and their effects on fission product dopant respirable production and enhanced sorption onto other respirables produced. The primary variables to be evaluated are:

1. The effect of the position of the HEDD, either detonated outside of a semi-open aerosol chamber (similar to the 2/0 through 2/4 series of Phase 2 tests [1], refer to Figures 2.2.1 and 2.2.2, or similar tests performed at the Fraunhofer Institut using the separate vertical elutriation chamber [18], shown in Figure 2.3.3, or detonated inside the sealed and co-joined Phase 2 aerosol collection chamber-explosive containment chamber, Figure 2.2.3, on the enrichment of cesium and other fission products. It was postulated that when the HEDD is detonated within an enclosed test system, the presence of more explosive byproduct soot and an associated high internal temperature rise may increase the extent of fission product enrichment onto the more plentiful respirable material soot present.
2. The distribution homogeneity of the fission product dopant within the surrogate material pellet, whether distributed throughout the pellet (as in an actual spent fuel pellet, or within the doped, vitrified German glass rodlet in tests 2/7A and 7B [2] or glass pellets in tests 2/10E and 2/10F,

or dispersed within individual pellets, as in tests 2/10E and 10F, or located at discrete localized positions within fission product dopant disks adjacent to cerium oxide pellets, as in many other Phase 2 tests.

Table 2.3.1. Phase 2+ Surrogate Test Matrix

Test #	Pressure	Dopants	Variables	Date
2/10A, 10B	1	yes, 6 FP disks	9 CeO_2 pellets, new, ½ size test chamber, check-out & replicate	4-6/2005
2/10C, 10D	4 internally doped CeO_2 CsI, Ru, SrO, Eu_2O_3		+ 6 CeO_2 pellets, semi-open chamber, w/valved entrance port	7/2005
2/10E, 10F	1	yes, in glass	**9 German HLW glass pellets** joint w/ Fraunhofer & GRS (nonrad.)	8/2005
2/10G, 10H	4 internally doped CeO_2 CsI, Ru, SrO, Eu_2O_3		+ 6 CeO_2 pellets, testing in Germany, to cross-calibrate vertical elutriator, and Berner impactor set-up	6/2006
2/10I	1	yes, in glass	**12 German HLW glass pellets** in Germany (cross-calibration, nonrad.)	6/2006
2/CSC	1	8 FP disks	performed in a 50 m^3 test chamber (* details and results in Section 3.1)	3/2007

2.3.1 Phase 2+ Test Components

The major components required for conduct of the Phase 2+ tests 2/10A - 2/10F are very similar to the preceding Phase 2 and Phase 2/Phase 3 tests (Section 2.4), and the following Phase 3 tests (Section 2.5). The notable differences are in the Phase 2+ test explosive-aerosol test chamber used and in the means for including fission product dopants either adjacent to or within the pellets. The German test setup also has a different configuration of aerosol sampling devices.

NOTE: Details for Phase 2+ test 2/CSC will be described separately, in Section 3.1.

2.3.1.1 Test Rodlets, Surrogate Pellets, and Fission Product Dopants: All Phase 2+ surrogate test rodlets consist of non-pressurized Zircaloy-4 cladding tubes containing either multiple pellets of CeO_2 ceramic pellets plus multiple fission product dopant disks (tests 2/10A and 2/10B), or pellets of fission product doped (internally) CeO_2 pellets (tests 2/10C, 10D, 10G, and 10H), or pellets of German non-radioactive high-level waste (HLW) glass pellets with internal dopants (tests 2/10E, 2/10F, and 2/10I). Photographs of these test rodlets, pellets, and end holder rods during assembly, pellet weights in grams plus densities and dimensions, and other details were presented in Section 4 and Table 4.1 in Sandia Technical Report SAND2006-5674 [3].

Fraunhofer ITEM and GRS provided Sandia with the surrogate HLW glass pellets for cooperative testing, in tests 2/10E and 2/10F. These glass pellets were core-drilled out of a bulk glass sample originally fabricated at the Karlsruhe Nuclear Research Center in Germany. The glass composition is essentially identical to the HLW glass rodlets used in earlier tests 2/7A and 2/7B, described in Section 2.2.1 [1]. The vitrified glass matrix was composed of about 84 wt. % SiO_2, MgO, MnO_2, CaO, Na_2O and other glass frit material. The remaining 16 wt. % contained dopants, primarily thermally volatile Cs_2O (0.47 wt. %, melting point 490°C) and MnO_2 (0.30 wt. %, melting point 535°C), plus nonvolatile La_2O_3 (1.82 wt. %) and Nd_2O_3 (1.03 wt. %), plus copper and iron. The cesium dopant was presumably retained and not volatilized in the (molten, during fabrication) glass material in silicate compounds, and is homogeneously distributed in each glass pellet.

For tests 2/10A and 2/10B, we used discrete, plastic resin-based fission product dopant disks, as described in Section 2.2.1. However, the measured concentrations of fission product species sorbed onto respirable particles was very low in the past Phase 2 tests (2/5E, 2/5G, 2/6A, 2/6B, 2/8C, and 2/8D) that incorporated two fission product disks [2]. Therefore, we incorporated six fission product dopant disks for these Phase 2+ tests. Three fission product dopant disks were located on either side of the central (of nine total) cerium oxide pellets. The HEDD jet-disrupted (particulated) region in these test rodlets includes all of the central pellet, all six dopant disks, and essentially all of the adjacent two cerium oxide pellets. The total concentrations of each fission product chemical species in each Phase 2+ test rodlet are listed in Table 2.3.2.

Table 2.3.2. Fission Product Dopant Chemicals, per Phase 2+ Test (revised* 2007)

Phase 2+ Test #	**Cesium Iodide, CsI**	**Ruthenium, as RuO_2**	**Strontium Oxide, SrO**	**Europium Oxide Eu_2O_3**	**Shape Factor & Weight**
	Actual (calculated) dopant weights, post-sintering				
2/10A	101 mg	18.8 mg	14.6 mg	15.2 mg	6 disks (0.534 g)
2/10B	98.7 mg	18.5 mg	14.3 mg	14.9 mg	6 disks (0.524 g)
2/10C	204 mg	1.48 mg	8.0 mg	29.3 mg	4 pellets (FP internal)
2/10D	260 mg	1.49 mg	8.0 mg	29.5 mg	4 pellets(FP internal)
2/10E	49 mg (as Cs_2O)	31 mg (MnO_2)	188 mg (La_2O_3)	107 mg (Nd_2O_3)	9 **glass** (10.35 g)
2/10F	49 mg (as Cs_2O)	32 mg (MnO_2)	192 mg (La_2O_3)	108 mg (Nd_2O_3)	9 **glass** (10.53 g)
2/10G	261 mg	1.54 mg	8.3 mg	30.5 mg	4 pellets (FP internal)
2/10H	270 mg	1.49 mg	8.1 mg	29.5 mg	4 pellets (FP internal)
2/10I	65 mg (as Cs_2O)	43 mg (MnO_2)	256 mg (La_2O_3)	144 mg (Nd_2O_3)	12 **glass** (14.04 g)
2/CSC	118 mg	22.3 mg	17.1 mg	18.1 mg	8 disks (0.628 g)

Note: Concentrations estimated as ± 10% for CsI; ± 20% for SrO and Eu_2O_3; ± 50% for RuO_2.
Note *: The fission product dopant concentations in Table 2.3.2 are revised and corrected from the similar values presented in Table 4.2 in [3]. These revisions resulted from additional chemical analyses of internally doped pellets performed in 2007.

In order to obtain cerium oxide ceramic pellets with the fission product dopants internally incorporated within the pellet matrix in an near-homogeneous manner, Sandia Ceramics personnel developed new doped pellet fabrication techniques. The fission product dopants of Ru metal powder (instead of previously used ruthenium oxide) plus granular non-volatile SrO and Eu_2O_3 were mixed with CeO_2 powder, then sintered to a theoretical density of ~75-80%. The resultant pellets were subsequently melt-infiltrated with CsI in solution in a secondary pellet heat treatment. The actual cesium iodide content was measured directly by weight gain after the melt-infiltration process. Further doped pellet fabrication techniques are described in [3].

2.3.1.2 Phase 2+ Aerosol Collection/ Explosive Test Chamber Systems: A new, semi-open aerosol collection test chamber was specifically designed and fabricated for use in the Phase 2+, 2/10A-10F series of tests, and is shown in Figure 2.3.1. It is, basically, intermediate in design between the semi-open, “square box” aerosol collection chamber used in the earlier Phase 2/4 series of tests, Figure 2.2.2, and the top, cylindrical half (only) of the Phase 3 vertical explosive-aerosol test apparatus, shown in Figure 2.5.1. The Phase 2+ test chamber is similar in construction to, and has the same internal volume as the Phase 3 aerosol top chamber, with the same four internal aerosol sampling tubes (sampling from the test rodlet height/area) and the same four independent, replicate sets of Marple impactor and Large Particle Separator systems. The test rodlet is also positioned and installed horizontally, as shown in Figure 2.3.2. The HEDD is located externally, on the side of the test chamber (in Figure 2.3.1, below the fragment deflector, but not installed yet), and fires horizontally through a 1 inch-diameter (2.5 cm) hole. The HEDD-jet stop block assembly is also horizontal and located on the outside of the cylinder, shown at right side in Figure 2.3.1, but connected to it; internal components are cleaned after each test and replaced as required. The test chamber is "open" for only about one second after detonation and then is sealed by an internal, fast-acting gate isolation valve, visible in Figure 2.3.2. This internal valve is opened at -4 seconds prior to HEDD detonation, and then closed at +1 seconds following detonation. We expected somewhat lower internal temperatures and somewhat less soot contamination in the “semi-open” Phase 2+ chamber compared with the totally sealed system in the Phase 3 explosive-aerosol system. The Phase 2+ test chamber has a removable, bolted-on top lid that allows for access to the inside for post-test particulate debris removal and decontamination. Three of the Phase 2+ tests, 2/10G, 2/10H, and 2/10I, were performed in Germany by Fraunhofer Institut für Toxikologie und Experimentelle Medizin (ITEM) and the Fraunhofer Ernst-Mach-Institut (EMI) in cooperation with GRS. The vertical elutriator aerosol collection chamber used, pictured in Figure 2.3.3, is similar to earlier tests performed in Phase 1 [18]; refer to Section 2.1. The HEDD was mounted in the blue, fragment-control cylinder at right which prevents the aerosol chamber from damage. In addition, a fast-closing valve separates the aerosol chamber from the detonation area after the CSC jet has entered the vertical elutriator chamber. The jet is caught by a stop block attached to the back chamber wall, behind the vertically oriented target. This vertical elutriator test setup uses a direct particle measurement technique incorporating a vented aerosol chamber with an upward homogeneous airflow of 25 cm/s separating the aerosol range below 100 µm AED from larger particles collected on a porous plate at the chamber floor. Particles transported with the airflow are led to further sampling devices at the top of the setup (in Figure 2.3.3). The two test target rodlets for tests 2/10G and 2/10H were supplied by SNL, and were very similar to target rodlets for the preceding tests 2/10C and 2/10D. In test 2/10I, the rodlet was filled with inactive HLW glass pellets. The latter test was added to the matrix to check the reproducibility of former glass tests with this setup. As the aerosol backup filter was damaged in test 2/10I, there are no reliable absolute data (RF) available from this test, but relative results (IEF) are expected to be reasonable.

2.3.1.3 Aerosol Particle Sampling Systems: We used four, replicate aerosol particle sampling systems for the Phase 2+ tests at SNL, essentially identical to those used and described in Section 4.3 of [2] for the later Phase 2 tests and also used in Phase 3 tests (Section 2.5). Each independent sampling system includes a: Marple particle impactor (Model 298), with an external containment enclosure that was designed and fabricated by SNL, and installed onto the impactor housing to provide a leak-proof double containment; a large particle samplers (LPS); plus, associated sampling tubes, a primary ball-type valve (with a 3/8 inch, 0.95 cm inner diameter) which provide isolation from the initial pressure pulse from the explosive charge, a secondary normally closed solenoid valve, critical orifice that is connected to the outlet of the impactor, an in-line HEPA filter, and vacuum bottle, all visible in Figure 2.3.1. Each aerosol particle sampling system is independently removable from the top of the test chamber, while the aerosol test chamber (below) remains sealed with use of individual manual-closure valves.

In the German aerosol test apparatus, the vertical airflow passes through a foam layer that retains fragments in the size range between 10 and 100 µm AED, to limit the aerosol loading of following sampling devices. A coarse-particle classifier collects particles in three size intervals covering the range between 100 and 21 µm. In this unit, the airflow performs a 180° bend, forcing aerosol deposition in different regions of the inner surface of the outer bend wall depending on the AED. Only particles with aerodynamic diameter larger than 21 µm are able to penetrate the bend and are collected on a backup filter. A side stream of this fraction is taken and is further classified by two conventional Berner cascade impactors (5 and 9 stages) covering the particle size range between 0.1 and 20 µm AED.

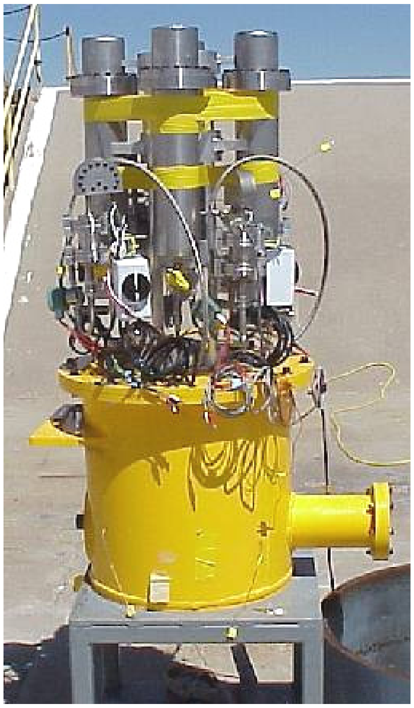


Figure 2.3.2. Phase 2+ Chamber Interior, showing Rodlet, Closure Valve, Thermocouples

⬅ **Figure 2.3.1. Phase 2+ Test Chamber and Aerosol Apparatus**

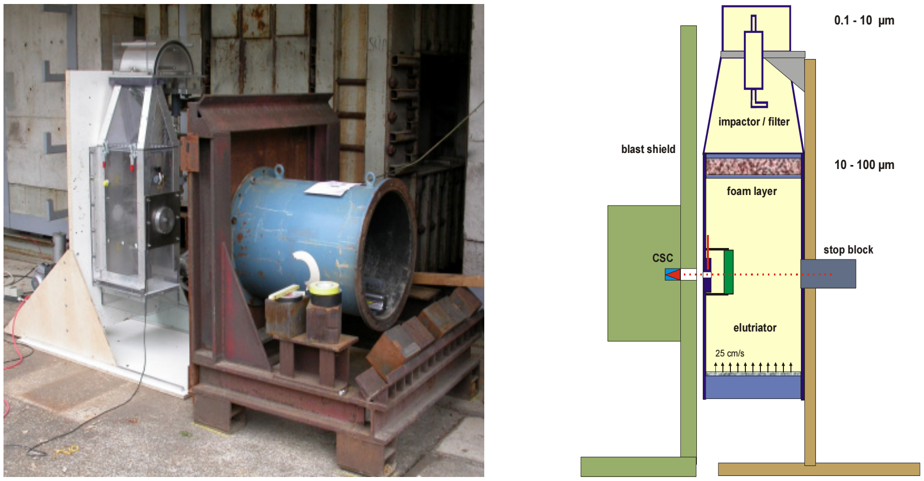


Figure 2.3.3. Fraunhofer Vertical Elutriator Test System

2.3.1.4 Aerosol Particle Post-test Chemistry Analyses: Previously, the SNL Analytical Chemistry Dept. 1822 had provided all chemical analyses for the surrogate Phase 2 aerosol samples through test 2/9B, and provided some peripheral support for the Phase 2+ surrogate tests. SNL Dept. 1822 major contributions to this overall program have been the development of the digestion scheme for analysis of the particle debris and aerosol samples collected from these tests, the analysis of samples from field tests and laboratory preparations, and the presentation of the data in a format in which the important details from the analysis may be readily found. Because SNL Analytical Chemistry could not accept the upcoming Phase 3 DUO_2 radioactive materials for analysis, we contracted the General Engineering Laboratory, GEL, in Charleston, South Carolina, to perform future DUO_2 particulate mechanical sieving, dissolution, and following chemical elemental analyses using Inductively Coupled Plasma-Mass Spectroscopy, ICP/MS. For consistency in results, GEL was also tasked with performing analyses of the surrogate material aerosol particles, starting with Phase 2+ test 2/10A. Post-test aerosol samples for tests 2/10E and replicate 2/10F, with surrogate HLW glass pellets, were shipped to the Fraunhofer Institute for post-test chemical analyses; particles from tests 2/10G to 2/10I, performed in Germany, were also analyzed by Fraunhofer using ICP/MS.

2.3.1.5 Instrumentation: Multiple pressure and temperature instrumentation were used in these Phase 2+ tests, including fourteen thermocouples and five pressure transducers. Figure 2.3.4 provides a schematic of the Phase 2+ test chamber instrumentation locations and the aerosol system layout. Table 2.3.3 lists the Phase 2+ thermocouple locations. Twelve of these thermocouples are installed within the test aerosol chamber. Four of these were installed through holes drilled in the lower wall portion of the test chamber and located at the entrance of each of the internal sample tubes; refer to Figure 2.3.2. Eight more thermocouples were installed through holes drilled in the top of the chamber lid and located about 6 inch (15 cm) down from the inner wall of the chamber lid; two thermocouples each were run down the impactor sample tubes, at 180 degrees from each other. Two more thermocouples were installed above the aerosol chamber, within the particle sampling tubes on the downstream side of the Large Particle Separators. The pressure transducers are located one each on the downstream side of the Large Particle Separators and one on the inside lower part of the test chamber. There were two thermocouples and one pressure gauge within the German aerosol test chamber.

2.3.2 Test Phase 2+ Major Results

2.3.2.1 Aerosol Particle Results: The Phase 2+ test aerosol particle results including measured Respirable Fraction (RF) values and fission product dopant measured Enrichment Factor (EF) values are presented in Sections 4.1 and 4.2, combined in a tabular and graphical manner with similar Phase 2, Phase 2/Phase 3, and Phase 3 results. In addition, interpretations and inter-comparisons of these data will be discussed in Section 4. EF values were measured and will be presented both integrated over the 0 to 10 μm AED respirable range and in differential form as a function of particle size range through the aerosol size range. In a comparison of Phase 2+ results for measured RF values for both CeO_2 and cesium, there did not appear to be an obvious, nor conclusive difference seen in RF values between target rodlets with discrete, adjacent dopant disks and those with internally doped pellets. In addition, there did not appear to be a major difference in measured RF value based on pellet material, whether surrogate CeO_2 or HLW glass; observed differences were significantly less than a factor of two. Measured Cs EF values will be discussed separately.

2.3.2.2 Target Rodlet Disruption: The observed effects of HEDD explosive jet impact on the Phase 2+ test rodlets were very consistent to those measured in Phase 2 tests. For Phase 2+ tests and Phase 2/Phase 3 tests, there was a measured gap of 24-(27 ave.)-32 mm; the total Zircaloy-4 tubing gap varied from 15-40 mm, due primarily to jagged flaps of Zircaloy of different lengths.

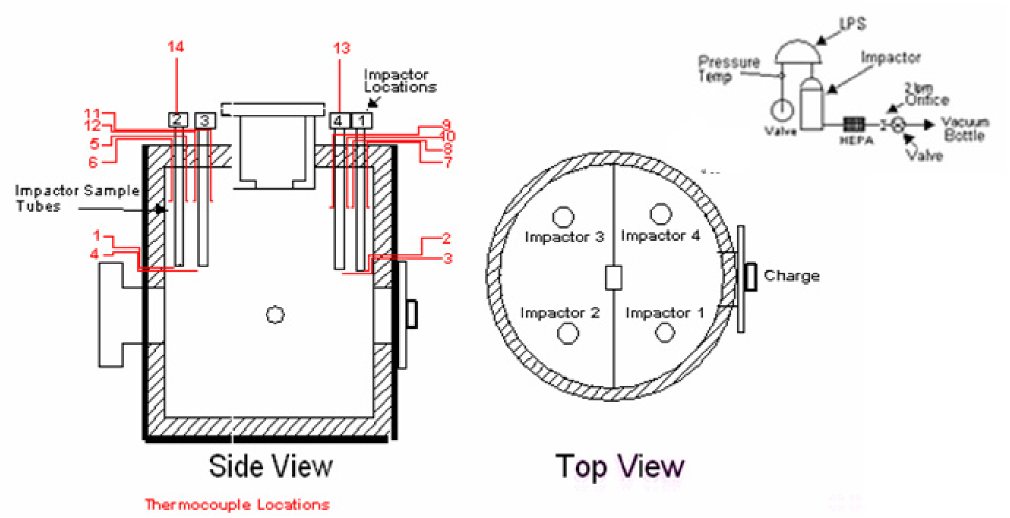


Figure 2.3.4. Schematic of Phase 2+ Test Chamber Instrumentation and Aerosol System

Table 2.3.3. Phase 2+ Thermocouple Sample Locations

1. Located at Impactor # 2 sample tube inlet
2. Located at Impactor # 1 sample tube inlet
3. Located at Impactor # 4 sample tube inlet
4. Located at Impactor # 3 sample tube inlet
5. Located on Impactor # 2 sample tube, 6" (15 cm) from bottom of chamber lid
6. Located on Impactor # 2 sample tube180 from 5, 6" from bottom of chamber lid
7. Located on Impactor # 1 sample tube, 6" from bottom of chamber lid
8. Located on Impactor # 1 sample tube180 from 5, 6" from bottom of chamber lid
9. Located on Impactor # 4 sample tube, 6" from bottom of chamber lid
10. Located on Impactor # 4 sample tube180 from 5, 6" from bottom of chamber lid
11. Located on Impactor # 3 sample tube, 6" from bottom of chamber lid
12. Located on Impactor # 3 sample tube180 from 5, 6" from bottom of chamber lid
13. Located on Impactor # 4 LPS co-located with pressure transducer
14. Located on Impactor # 2 LPS co-located with pressure transducer

2.3.2.3 Measured Temperature and Pressure Results: The peak temperatures measured in Phase 2+ tests are listed in Table 2.3.4. As mentioned for Phase 2 tests, temperature measurements were quite dependent on the thermocouple locations; they varied appreciably and reached some very elevated values. As an example, Figure 2.3.5 presents measured temperatures in test 2/10D as a function of time (seconds). It is obvious that temperatures within the aerosol chamber are not homogeneous, but vary significantly as a function of location. More details on individual temperature measurements were presented in Section 6.4 in [3]. The maximum measured temperatures measured in the Fraunhofer vertical elutriator test chamber were appreciably lower. Measured peak pressures in the SNL semi-open test chamber were measured immediately after HEDD detonation, and then decreased to about 1.9 bar at 0.04 seconds and 1 bar after 0.5 seconds.
** The much higher peak temperature measured in test 2/10A was possibly the result of the thermocouples inadvertently touching or being located very near the metal surfaces of the chamber wall or sampling tubes, as described in Section 2.2.2.7. Thermocouples were located more appropriately in subsequent tests.

Table 2.3.4. Peak Measured Temperatures and Pressures In Phase 2+ Aerosol Chambers

Phase 2+ Test	Peak Temperature	Peak Pressure	Comments
2/10A	560 ºC (833 K)**	- -	SNL semi-open chamber 1 bar @ 0.5 seconds
2/10B	230 ºC (503 K)	7.4 bar	
2/10C	280 ºC (553 K)	7.4	
2/10D	330 ºC (603 K)	5.1	
2/10E	260 ºC (533 K)	5.5	
2/10F	330 ºC (603K)	6.5	
2/10G	220 ºC (493K)	1.3	Fraunhofer vertical elutriator
2/10H	195 ºC (468K)	1.6	
2/10I	140 °C (413K)	1.3	

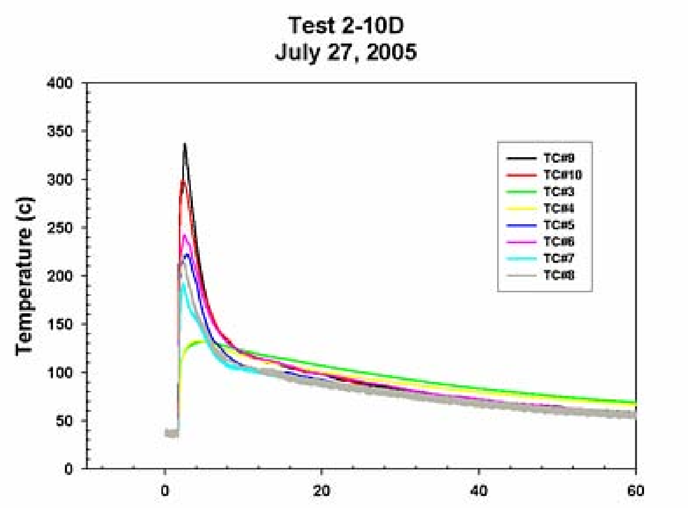


Figure 2.3.5. Test 2/10D Measured Temperatures vs. Seconds

2.4 Test Phase 2/ Phase 3

The Phase 2 / Phase 3 cross-over series of tests was added to the original test design program in 2004, and three of these tests, 2/9/A, 2/9B, and 2/9C were performed [2]. The Phase 2/Phase 3 test matrix is shown in Table 2.4.1. These added tests, basically, an extension of the original Phase 2 program, were intended to bridge the equipment and facility gap between earlier Phase 2 surrogate tests with CeO_2 pellets, performed in the initial vertical explosive-aerosol containment chamber" Figure 2.2.3, and the Phase 3 tests with DUO_2 test rodlets to be performed in the following, new Phase 3 test chamber, shown in Figure 2.5.3. The purpose of these cross-over tests was to exercise and demonstrate the full Phase 3 test system and operational controls, to institute several new techniques, and also to collect surrogate target aerosol data, in a *non-radioactive* test environment.

The final two planned Phase 2 / Phase 3 cross-over tests, 2/9D and 9E, similar to test 2/9C, are planned to be performed at the SNL Gamma Irradiation Facility (GIF) primarily for SNL Technical Area V nuclear operational readiness, procedure check-out, and proof tests for demonstration purposes; also refer to Section 2.5.3. These tests will be handled and performed in a "semi-remote" manner *as if* they contained highly radioactive spent fuel rodlets.

Table 2.4.1. Phase 2/Phase 3 Cross-over Tests Matrix

Test #	Pressure	Dopants	Variables	Date
2/9A	1 bar	no	new test chamber, w/ CeO_2 target rodlet, 4 Marples & 4 LPS, in ECF	8/2004
2/9B	1 bar N_2	no	same, with inert atmosphere, in ECF	8/2004
2/9C	1 bar	no	"blank" w/Zirc tube, for post-test handling operations, 4 Marples & LPS	11/2004
2/9D	1 bar	no	"blank" w/Zirc tube, in GIF, "as if" Phase 4 spent fuel, 4 Marples & LPS	TBD
2/9E	1 bar N_2	no	"blank" w/Zirc tube, in GIF, same, w/ inert atmosphere	TBD

The cross-over tests 2/9A and 2/9B used non-radioactive CeO_2 test rodlets in a Zircaloy-4 cladding tube, very similar to Phase 2 tests. Test 2/9B was a replicate of test 2/9A, but was performed with an internal test chamber atmosphere of inert nitrogen gas [2]. No dopant materials were included in the target rodlets, in order to eliminate potential fission product species residual contamination in subsequent Phase 3 tests. Test 2/9C was a "blank" test, using an empty Zircaloy-4 cladding tube, 203 mm (8 inches) long, and 10.6 mm outer diameter, un-pressurized. This test also served as a test system demonstration for international program participants during the 8th Technical Meeting of the WGSTSC, at SNL, in November 2004. These Phase 2/Phase 3 tests were performed within the new, optimized, Phase 3 explosive-aerosol test chamber, to be used for all the DUO_2 Phase 3 tests, using four replicate Marple impactor and Large Particle Separator units on top, as illustrated in Figure 2.5.3. The first two tests were performed at the SNL Explosive Components Facility, SNL bldg. 905; the third was conducted in the ECG remote Gun Site bldg 6750.

Gravimetric data on particle size distributions and measured temperature and pressure data in the top-mounted particle sampling units were documented in [2]. The post-test chemical and elemental analyses on aerosol particles for tests 2/9A and 2/9B were presented in [3]. Measured CeO_2 respirable fraction (RF) values are included in Section 4.1.

Temperatures and pressures measured within all four aerosol sampling systems on all three performed Phase 2 / Phase 3 cross-over tests was presented in detail, as a function of each thermocouple location, in Section 6.4 of [3]. The measured peak temperatures never increased more than ~ 18 ºC above ambient, to a maximum of about 40 ºC. In addition, the observed post-HEDD peak detonation pressures within the four aerosol sampling systems rapidly increased about 21 psi (1.4 bar), then decreased to ambient pressure within about one minute.

2.5 Test Phase 3

Phase 3 tests use slightly radioactive, unirradiated depleted uranium oxide (DUO_2) pellets in comparable, new Zircaloy-4 cladding tube test rodlets. The complete Phase 3 test matrix and test variables are listed in Table 2.5.1. Preliminary test details and requirements were initially documented in [1] and are still current. Note that the Phase 3 testing "*order*," (A) to (F), in Table 2.5.1 is different than the test/rod numbers. This was modified in order to eliminate potential cross-contamination from the test rodlets that contain non-radioactive fission product dopant disks. Testing with all Phase 3 test rodlets that contain no dopants was performed first. The test chamber and aerosol apparatus used were also decontaminated after (i.e., between) each test performance. Three Phase 3 tests have been completed to date, one in 2005 and two in 2006; the remaining three tests may be performed when funding permits.

Table 2.5.1. Phase 3 Tests: Advanced DUO_2 Surrogate Test Matrix

Test # (order)	Rodlet	Pressure	Dopant	Variables	ECF Date
3/2 (A)	DUR-2	1 bar	no	air (in aerosol chamber)	10/14/2005
3/5 (B)	DUR-5	40 (He)	no	air	1/12/2006
3/1 (C)	DUR-1	1	yes	air	3/09/2006
3/3 (D)	DUR-3	1	yes	N_2 (in aerosol chamber)	TBD
3/4 (E)	DUR-4	40	yes	air	TBD
3/6 (F)	DUR-6	40	yes	N_2	TBD

2.5.1 Phase 3 Test Components

In this section we describe the primary Phase 3 test components, the test rodlets, the explosive-aerosol test chamber, and the test facility used, along with the associated safety and radiological control constraints.

2.5.1.1 Depleted Uranium Oxide Test Rodlets: As part of the WGSTSC program cooperative efforts, the six Phase 3 DUO_2 pellet target rodlets, shown in Figure 2.5.1, were fabricated by CERCA (a Framatome-ANP, AREVA subsidiary) in Romans-Sur-Isère, France for IRSN, for testing at SNL. There is an external center mark on the cladding for each rodlet, locating the center of the internal central pellet, to be used for HEDD jet alignment purposes; these marks are (barely) visible in Figure 2.5.1. Fabrication in, and shipment from France was completed in July 2004; these rodlets were successfully received at SNL in August 2004.

Each test rod contains five 13.8 mm-long pellets of ~ 97% theoretical density DUO_2, with dished ends, pictured in Figure 2.5.2. The DUO_2 pellets used contain 0.2 wt. % of ^{235}U and were obtained from FBFC International (an AREVA subsidiary), in Dessel, Belgium. On average, each of the six DUO_2 test rodlets contains 47.99 g of uranium oxide (ceramic), 42.33 g of uranium, including 0.085 g of ^{235}U. The calculated radiation content/activity is 21 μCi/rodlet or 0.78 MBq/rodlet (specified as 4.699 MBq/all 30 pellets, and 1 MBq = 27 μCi).

The test rodlet design, shown in Figure 2.5.3, particularly the end pieces, was a collaborative effort by IRSN, SNL, and Argonne National Laboratory. The rodlet dimensions, except for total length, are very similar to U.S.-origin pressurized water reactor (PWR) fuel pins. The rodlets are fabricated from Zircaloy 4 cladding tube of 10.6 mm outside diameter, 9.32 mm inside diameter, supplied to IRSN by SNL. Laser end-cap and seal welding were used to fabricate the rodlets. The

Figure 2.5.1 Photograph of Phase 3 DUO_2 Test Rodlets

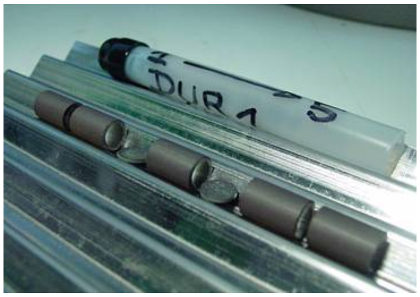


Figure 2.5.2 DUO_2 Test Pellets and Dopants

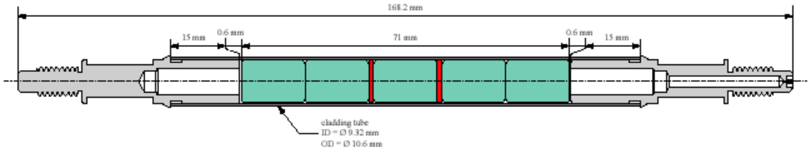


Figure 2.5.3 Schematic of DUO_2 Test Rodlet DUR-4

Figure 2.5.4 Post-test DUO_2 Test Rodlet

Figure 2.5.5 Phase 3 Test Chamber and Aerosol Sampling Systems

completed rodlets were He-leak tested, all welds were X-ray tested, and then the rodlets were shipped to SNL. These tests incorporate the variables of internal rodlet pressurization (1 or 40 bar of He within rodlet end plenum regions, similar to PWR fuel rods at the end of their life, re: NUREG/CR-6831), added non-radioactive fission product dopant disks (as illustrated in Figure 2.5.3, in three of the six rodlets), and an internal aerosol chamber atmosphere (either air or inert nitrogen). A post-test DUO_2 rodlet, with its center-region particulated is shown in Figure 2.5.4.

2.5.1.2 Test Chamber and Aerosol Apparatus: The Phase 3 tests use a (single) vertical, self-contained, explosive-aerosol test chamber, shown in Figure 2.5.5. This test chamber (named "Tweety Bird") is based on the similar but less sophisticated Phase 2 test chamber design shown in Figure 2.2.3. This test chamber was engineered by SNL for total HEDD blast containment (pressure, fragmentation) and for total, leak-tight isolation of all particles produced. The total chamber is approximately 0.6 m-diameter by 1.3 m-high, has a total interior volume of 183 L, and is fabricated out of thick steel to contain the explosive blast (bottom chamber) and contain all aerosol particles produced (top chamber) for sampling. Other details of this test chamber and its pressure and blast qualification test requirements were provided in [3].

The four replicate aerosol particle sampling systems shown on top of Figure 2.5.5 are identical to those used in the Phase 2+ tests and described in Section 2.3.1.3 and in [3]. Each particle sampling system is independently removable from the top of the test chamber, while the aerosol test chamber (below) remains sealed with the use of individual manual-closure valves, preventing the release of radioactive particulates within.

2.5.1.3 Phase 3 Test Facilities and Major Obstacles Overcome: Originally [2], all Phase 3 DUO_2, as well as the Phase 4 spent fuel explosive-aerosol tests, were to be performed at the Sandia Gamma Irradiation Facility (GIF). However, due to appreciable delays experienced in 2004 and 2005 for receiving final approval for the GIF Documented Safety Analysis (DSA) from the DOE-Sandia Site Office (SSO; NNSA), performance of the Phase 3 test program at an alternate facility at Sandia was opened to consideration. During mid-2005, approval was obtained to conduct the Phase 3 DUO_2 tests within the SNL Explosive Components Facility (ECF), Bldg 905, test firing pad Room 1213A. This move resulted in a significant positive impact on test schedule and expense, compared to using the GIF. The level of explosive safety and radiological containment would need to be essentially the same in either the GIF or the ECF facilities. The ECF building had not been used previously for radioactive testing of this type or scope nor was it classified as a nuclear facility.

It was concluded that conduct of the Phase 3 tests was indeed feasible and could be accomplished with the following requirements and test control issues met: (a) need approved documentation of a NEPA amendment for moving the Phase 3 tests from GIF to ECF; (b) need a PHS/HA (Preliminary Hazard Screening/ Hazard Assessment) review and approval; (c) need the development and approval of a formal Operating Procedure (OP) for the test performance, with the use of properly trained personnel; (d) need to arrange for appropriate waste management for the post-test DUO_2 materials, including decontamination between tests; and, (e) need to include appropriate secondary containment of the test chamber within the ECF test room, to address potential release of particulate radioactive materials. All of these issues were successfully addressed, documented (internal Sandia National Laboratories documents) and completed during 2005, and are described in detail in Section 5.3 in SNL Technical Report SAND2006-5674 [3].

The primary barrier to the release of particulate radioactive materials during and after each Phase 3 DUO_2 test is the vertical explosive-aerosol test chamber. To further control the *potential* release of particulate radioactive materials within the ECF, secondary barriers were implemented. The test chamber is located within a Large Walk-in (blast enclosure) Chamber in room 1213A, shown in Figure 2.5.6. The Large Walk-in Chamber floor and instrumentation ports were lined with plastic and Herculite as an additional secondary containment barrier. Furthermore, inside the walk-in chamber is a large portable plastic containment tent, supported by a metal frame about 12 ft-long x 7 ft-wide x 7.5 ft-high (3.6x2.1x2.3 m), illustrated in Figure 2.5.7. The secondary containment tent serves as the barrier for the radiological contamination area and was sealed during each test and all post-test decontamination activities. Access to the primary explosive-aerosol Phase 3 test chamber during the explosive portion of the test was controlled by closing and locking the Large Walk-in Chamber access door via seven hydraulic pins into the Chamber door; this door is interlocked to the monitoring and firing system. Post-test, the aerosol apparatus systems were disassembled and the weights of all aerosol samples were then obtained in an external (directly outside the Large Walk-in Chamber) small plastic glove-box enclosure within room 1213A, shown in Figure 2.5.8. The aerosol samples then individually packaged within the glove box enclosure for off-site transport to the GEL chemistry analysis laboratory. The secondary containment physical controls are accompanied by radiological protection surveys performed by radiological control technicians. All post test work, either in the glove box or containment tent, was supported by radiological control technicians under the direction of the appropriate radiological work permits.

Figure 2.5.6. ECF Walk-In Blast Enclosure

Figure 2.5.8. Glove Box Enclosure

Figure 2.5.7. Secondary Containment Tent

2.5.2 Test Phase 3 Major Results

2.5.2.1 Aerosol Results for Depleted Uranium Oxide: The respirable fraction values for depleted uranium in DUO_2 were calculated from the aerosol measurements taken in the first three Phase 3 tests and are reported in Section 4.1 in Table 4.1.1 and illustrated in Figure 4.1.1. The measured average **RF for DU = 1.32% ± 0.32%** with a 99% confidence interval or ± 0.21% with a 90% confidence interval. Tests 3/2 (A) and 3/1 (C) were performed with a 1 bar internal rodlet pressure, and 3/5 (B) had a 40 bar He internal pressure. There was no observable pressure effect on measured DU RF values – similar to the results for CeO_2 tests, as described in Section 2.2.2.5. The average DU RF value is very consistent with the earlier surrogate cerium oxide sample RF results from all gathered data, **Ce RF = 1.36% ± 0.50%**, or **Ce RF = 0.65% ± 0.23%** based on the Marple impactor data only, both with a 99% confidence interval. In addition, Test 3/1 (C) incorporated fission product dopant disks with Cs, Ru, Sr, and Eu. Results for measured fission product RF and EF values are included in Sections 4.1.2 and 4.2 for comparisons to all other measured fission product results.

From observations of the Marple impactor DU distributions in the respirable size range (illustrated in multiple figures in Appendix A3 in [3], DU particles appear to peak distinctly over the respirable range size of 1.6 to 3.5 μm AED, then increase again from 21 to 35 μm AED. The uranium particle distribution in the aerosol range from about 30 to 100 μm AED, collected with the LPS apparatus, is generally highest in the 30-48 μm cut (for tests 3/2 (A)), then decreases, then increases somewhat in the 82 to 100 μm range (for both 3/2 (A) and 3/5 (B)).

2.5.2.2 Other Phase 3 Test Results and Observations: Detailed observations regarding sampling times, measured temperatures and pressures within the top-mounted aerosol apparatus, and concerns and resolutions of fission product and other background or contamination issues were included in Section 6.3 of [3]. Further observations and results are described below.

External Contamination: The particulate radioactive contamination potentially released during the top-mounted aerosol apparatus assemblies removal operation was monitored and recorded as "*at or below detectable levels*." As such, radiation work permit (RWP) procedures were revised after the first Phase 3 test so that the use of personnel respirators was no longer required for this aerosol apparatus removal operation. During the test chamber flange plate (refer to Figure 2.5.5) removal process for post-test rodlet removal and chamber decontamination prior to the following test, a minimal (detectable) amount of particulate materials were released and collected on the secondary barrier tent floor below with appropriate control materials. As such, test personnel were still required to wear respirators as well as full anti-contamination suits during this procedure.

Internal Apparatus Contamination and Decontamination: Following completion of all sampling of aerosol particle materials, SNL Radiation Protection personnel surveyed and decontaminated the Marple and Large Particle Separator units plus associated sampling system hardware assemblies to defined acceptable radiological levels (< 1000 dpm; actual measured levels were 150-300 dpm, beta-gamma radiation). This decontamination process consisted of using a combination of steam cleaning and manual swabbing/wipe-downs of cloth materials soaked in Coca Cola, then household ammonia. Finally, the secondary containment tent itself was wiped down with household ammonia. The inside of the test aerosol chamber decontamination technique or procedure, as developed during this test phase, involved "sand blasting" of the inside of the test chamber with glass beads, followed by HEPA vacuuming of residual glass, then using steam cleaning. The RWP procedure requires that the residual contamination level must be < 1000 dpm (beta gamma activity); a post-cleaning level of 150-300 dpm was measured and determined to be very adequate.

Target Rodlet Disruptions: The observed effects of an explosive HEDD jet impact on the Phase 3 test rodlets were very similar to the preceding Phase 2 and 2+ tests. For Phase 3 tests, the average Zircaloy tube gap was 27-32 mm in length and the average length of pellet material particulated was 43-47 mm; full details were summarized in Table 6.3 in [3]. There was no observable damage to the welds at the outer ends of the DUO_2 test rodlets, a point of interest.

2.5.3 Remaining Phase 3 Tests and Options

The final three Phase 3 tests 3/3 (D), 3/4 (E), and 3/6 (F) were planned to have been performed at the SNL ECF test facility during FY 2007. However, as a result of unanticipated program funding delays and reductions, these plans were indefinitely postponed. Future budgetary limitations are also expected. Therefore, it is very uncertain that the remainder of test Phase 3 will be performed. We plan on resuming spent fuel sabotage aerosol testing activities at SNL with the re-start of the Phase 4 spent fuel tests (refer to Section 2.6) at the GIF, when or if sufficient test program funding is received. However, when funding is restored, a potential option has been considered to perform some of the remaining Phase 3 DUO_2 tests at the SNL GIF, not the ECF test facility, as follows. We stated earlier in Section 2.4 that the final two planned Phase 2 / Phase 3 cross-over tests, 2/9D and 2/9E with a "blank" Zircaloy tube target rodlet, are planned to be performed at the SNL GIF as preliminary or introductory tests for the Phase 4 program, primarily for SNL Technical Area V nuclear operational readiness, procedure check-out, and proof tests for demonstration purposes. As a viable option, several Phase 3 DUO_2 target rodlets could be used instead of "blank" targets, making the Phase 4 operational test segment more realistic. The only added expenses if this Phase 3 / Phase 4 option is performed are the costs for post-test chemical analysis of the DUO_2 and fission product dopant particles produced, plus staff time for processing an interpretation of resultant data.

The alternate option is to conclude the Phase 3 test program as it currently stands, with no further testing. Existing results for the three tests performed to date wave been very consistent with each other. The Phase 3 data is probably quite sufficient for comparison to to-be-obtained Phase 4 data, in order to measure the spent fuel ratio, SFR. No final decisions have been made on which option can or will be pursued.

As a consequence of the extended time postponement of Phase 3 testing starting in 2006 and extending through 2007, all Phase 3 test apparatus has been decontaminated and removed from within the SNL Explosive Components Facility. The explosive-aerosol test chamber has been placed into remote storage. When Phase 3 is declared complete or no longer needed, the remaining test apparatus and DUO2 test rodlets will be treated as low-level radioactive waste and will be disposed of appropriately as part of the Sandia National Laboratories waste stream [1].

2.6 Test Phase 4

The final phase of this test program, Phase 4, will use segments of actual PWR spent fuel rods, highly radioactive uranium oxide pellets in their original Zircaloy cladding, fabricated into short test rodlets. Four of the Phase 4 rodlets will use high burnup (~ 72 GWd/MTU) spent fuel originating from the H.B. Robinson pressurized water reactor. Another four Phase 4 tests will use a low-medium burnup (~ 38 GWd/MTU) spent fuel originating from the Surry PWR. The overall planned test matrix is presented in Table 2.6.1. Final determination of the spent fuel ratio (SFR) will be based on a comparison of the aerosol particle results from the to-be-obtained Phase 4, actual spent fuel test data, to the Phase 3, surrogate DUO_2 test data. The accurate determination of the SFR was the primary driving force for WGSTSC participants at the initiation of this program.

A significant amount of work and progress has been accomplished on the Phase 4 test program beginning in 2002 and continuing through the present time; this will be summarized herein. The initial feasibility study to evaluate if an experimental program using highly radioactive spent fuel and HEDD explosives could, potentially be conducted safely within available SNL facilities was performed by Sandia National Laboratories [26]; this study concluded that this was indeed possible. A significant effort began during 2003 to get approval to use the facility, the Gamma Irradiation Facility (GIF) Cell 3 in SNL Technical Area V, for the Phase 4 test program. This was a collaborative effort of SNL and the Department of Energy Sandia Site Office (SSO, DOE NNSA) that was successfully accomplished in 2007. Details of the Phase 4 test plan, activities, details, and preliminary requirements were originally documented by SNL in SAND2004-1832 [1].

Table 2.6.1. Phase 4 Actual Spent Fuel Tests Matrix

Test #	Pressure ^	Variables	GIF Date
4/1	~ 1 bar * He (rod plenum)	High burnup PWR, ~72 GWd/MTU	TBD (≥ 2009)
4/2	~ 1 *	Air (in aerosol chamber)	TBD
4/3	~ 1 *	N_2 (in aerosol chamber)	TBD
4/4	~ 1 *	N_2	TBD
4/5	~ 1 *	Low-medium burnup PWR, ~38 GWd/MTU	TBD
4/6	~ 1 *	Air	TBD
4/7	~ 1 *	N_2	TBD
4/8	~ 1 *	N_2	TBD

(* as modified in 2006)

2.6.1 Phase 4 Work to Date

2.6.1.1 Spent Fuel Test Rodlets: The Phase 4 high burnup spent fuel pellets/rods originated at the H.B. Robinson PWR, Rod R01, and are currently at Argonne National Laboratory (ANL). 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, and a ^{235}U enrichment of about 2.90 wt. %. Similarly, the low-medium burnup fuel selected 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. Both PWR fuels were supplied to ANL as part of a research program sponsored jointly by NRC, DOE, and EPRI. They are *DOE-owned research material*, not commercial SNF. The selected PWR fuel rods have been fully characterized (non-destructive and destructive characterization of fuel material and cladding) in the ANL Alpha Gamma Hot Cell Facility (AGHC). Characterizations included: visual exams, axial gamma scanning, optical metallography, cladding hydrogen content, and isotopic analyses – for following aerosol and radiological source term material behavior evaluations. The Spent Fuel Characterization Report for the H.B. Robinson fuel rodlets, ANL-05/41 [27] has been completed. The second, Surry isotopic analysis report has been delayed because of a programmatic shutdown delay at the ANL Alpha Gamma Hot Cell Facility (AGHCF); however, Surry rod characterization results from sibling rods are documented in [28].

It is relevant to note that earlier, small-scale explosive-aerosol tests performed by Battelle Columbus Laboratory [10] and Idaho National Engineering Laboratory [11] in the 1980s, for SFR determinations, both used lower-burnup spent fuel originating from the H.B. Robinson reactor. This was described in Section 1.3.

The Phase 4 tests will use the H.B. Robinson and Surry spent fuel short rod segments in their original irradiated Zircaloy-4 cladding, and will be fabricated by ANL into short test rodlets (refer to Figures 5.4 and 5.5 in [2]) with a rodlet end-fitting design essentially similar to the Phase 3 DUO_2 rodlets as shown in Figures 2.5.1 and 2.5.3. Sectioning of 8 three-inch-long (7.6 cm) spent fuel test samples has been completed. This fabrication and characterization work by ANL has been performed under contract to SNL, with DOE (NA and RW) funding. SNL has requested ANL to delay the completion of test rodlet fabrication activities due to funding shortages and because Sandia cannot accept delivery until a DOE-approved agreement is made for interim, off-site storage of the post-test chambers. ANL has agreed to store the spent fuel rodlet sections until requested. Argonne, or an alternate facility will complete fabrication of the spent fuel rodlets at a later date (TBD), including post-welding leak testing and external contamination control, and then transport the rodlets to SNL for testing when Sandia has received DOE authorization to accept them. Following preparation and approval of an Argonne transportation plan, ANL intends to ship all eight test rodlets within a GE-100 transport cask to SNL. This transportation also requires inputs and receipt approval from both DOE SSO and the SNL Transportation Department; this process was initiated in 2005 but has since been put on hold status. Once accepted at SNL, these spent fuel rodlets will be stored at the GIF until each one is used individually in the Phase 4 experiments starting in 2009, or when program funding permits.

Test Rodlet Pressurization: Originally [1-2], all Phase 4 test rodlets were to have been internally pressurized with He gas in their end plenum regions (high burnup rodlets to 44 bar, low-medium burnup rodlets to 33 bar), to be as representative of end-of-life PWR spent fuel as possible, re: NUREG/CR-6831. The test spent fuel rodlets were intended to be similar to large-scale tests done with pressurized surrogate DUO_2 fuel rods in tests performed in the early 1990s [5]. However, the capability and facilities at ANL to internally pressurize the spent fuel rodlets was lost in 2005 and no longer exists. As such, SNL contacted all WGSTSC partners in 2006 regarding the absolute need for short, spent fuel test rodlet fabrication with pressurization. Based on the Phase 2 and Phase 3 experimental respirable fraction results of pressurized and un-pressurized short, surrogate test rodlets, as discussed in Section 4.1.1.1, the existing technical data and interpretation opinion do not support the absolute need for test rodlet pressurization for Phase 4 spent fuel test rodlets. Therefore, WGSTSC participants concurred that the use of un-pressurized short Phase 4 rodlets would be acceptable, that aerosol particle blow-down release from the short test rodlets will not be significant compared to the observed data scatter of all previous experiments.

2.6.1.2 Phase 4 Test Chambers: The explosive-aerosol, vertical test chambers for the Phase 4 spent fuel tests have been designed and are illustrated in Figure 2.6.1; the internal horizontal spent fuel rodlet, aerosol sampling tubes, and the HEDD jet-stop block are visible, as are the four top-mounted, replicate aerosol collection apparatus. These Phase 4 test chambers are very similar to the Phase 3 test chamber, except that they have no flanged access port to the top aerosol chamber. Phase 4 (and Phase 3) test chambers have a top head plate that is 5 inches (12.7 cm) thick, nominally for radiation shielding. The total internal volume of the Phase 4 aerosol top chamber is identical to the Phase 3 aerosol top chamber; compensation was made for the volume loss from the lack of an internal flange support in the Phase 4 top chamber. The Phase 4 test assembly during fabrication consists of two pieces, “top” and “bottom,” that are welded together into a single test chamber assembly prior to delivery. The top piece is designated as “high pressure or vacuum flange, open end (the aerosol chamber),” and the bottom piece is termed the “high pressure or vacuum flange closed end (the explosive containment chamber).”

The Phase 4 test chamber is quality controlled [3] *in accordance with* the American Society of Mechanical Engineers (ASME) code for pressure vessels, Section VIII Division 1, with internal SNL documentation [29-30]. It was explosively over-tested successfully to twice the HEDD-produced pressures expected in planned usage (peak reflected blast pressure of ~800 psi, 55 bar, measured). This chamber was modeled for static stress analyses, welds have been 100% X-rayed in *accordance* with ASME code and dye-penetrant tested, and it has been hydrostatically leak tested [29]. SNL explosive safety personnel have concurred that this "chamber (both the Phase 3 and following Phase 4 chamber iteration) is qualified for production testing and meets the requirements for a hazard classification and storage compatibility Group of 1.4S."

The Phase 4 test chambers have a top-mounted, internal HEDD-jet stop block. The internal components of this stop block consist of alternating plates of mild steel and polypropylene. The purpose of these plates is to stop the very energetic HEDD jet, as well as the less energetic residual HEDD metallic slug or "carrot," within a manageable distance; this distance is appreciably less than 30 cm. The bottom-most plate (hit first by the HEDD jet) is made of steel. The polypropylene plates are critical for keeping this stopping distance to a minimum length.

Each Phase 4 test chamber will be used *one time only.* The chamber is never opened post-test, to prevent the potential release of spent fuel aerosol particles contained inside. Each test chamber will be used once then temporarily stored at Sandia in the GIF floor vault, with the explosively-disrupted, post-test spent fuel rodlet and residual particulates contained within.

Figure 2.6.1 Diagram of Phase 4 Test Chamber and Aerosol Sampling Systems

Each sealed, post-test Phase 4 chamber will be enclosed within a thin corrosion resistant stainless steel overpack (yet to be designed). At a later date (TBD by the SNL Transportation Department and DOE), the overpacked test chambers will be shipped off-site within a GE-2000 transport cask to an approved, limited-term radioactive material temporary storage facility (tentatively at the Idaho National Laboratory) prior to final disposal, when a licensed repository facility is available to accept these chambers. The overpacked SNL Phase 4 spent fuel test chambers will fit within the "Large, 24-inch O.D. DOE Standardized SNF Canisters" for Yucca Mountain, as specified in DOE/SNF/REP-011, Rev. 3, August 1999. Even with the additional overpack and top handling lug, two or possibly three SNL test chambers will fit within one DOE Standardized SNF Canister.

Two Phase 4 test chambers were fabricated and delivered to SNL in 2005; these two are both in storage until needed. They have been purged and backfilled with dry nitrogen gas, then sealed, to minimize potential internal corrosion during storage. The remaining six of eight test chambers will be fabricated as required, and when funding permits.

2.6.2 Phase 4 Test Facility and Summary of GIF DSA

The combination of an explosive, high energy density device and highly radioactive Phase 4 spent fuel test rods gives rise to significant radiological safety testing concerns. These concerns have necessitated extensive facility environmental and safety assessment evaluations, contamination and radiation controls, plus remote handling and post-test, off-site disposition concerns. These same issues significantly increase testing expense and difficulty. The selected test facility [26], the SNL GIF, Test Cell 3, in Sandia Technical Area-V, has been previously operated as a clean facility for gamma irradiation testing only, with no radioactive contamination. Diagrams of the GIF floor plan and cross-sectional view were provided in Figures 8.1 and 8.2, respectively, in SAND2006-5674 [3]. The GIF is classified as a Hazard Category 3 nuclear facility with a DOE SSO approved Documented Safety Analysis (DSA). However, the existing, previous safety basis documentation did not adequately address the use of explosives and fissile materials, specifically spent fuel, due to the lack of description and analysis in the original DSA. An updated GIF DSA and additional Technical Safety Requirements (TSR) were required to be prepared by SNL for DOE SSO approval. Some further details of SNL requirements to support the DSA process, and delays encountered, were discussed in Section 8.1 of [3]. This critical path issue, the GIF DSA resolution and approval process produced appreciable delays to the planned Phase 4 testing schedule that was not successfully resolved until recently.

On June 19, 2007, DOE SSO formally approved the GIF Documented Safety Analysis (DSA) and issued their final Safety Evaluation Report (SER) [31]. Based on a comprehensive technical review and satisfactory comment resolution process, the DOE SSO Safety Basis review Team concluded that the GIF DSA and TSRs adequately identify the controls needed to maintain safe operations of the facility. The Sandia GIF is now authorized for spent fuel sabotage-aerosol, spent fuel ratio (SFR) testing, when funding permits. This represents a significant programmatic success of a former critical path item. The initial SNL support work on this DSA revision was started in 2003, with some support from the NRC. The original SNL submittal of the GIF DSA documentation to DOE SSO took place in 2004.

This recently approved GIF DSA not only authorized SFR testing, but it also involved a significant upgrade to the facility authorization basis. Efforts to address implementation and subsequent verification of the DSA upgrade are currently underway. There are specific implementation requirements for SFR testing at the GIF. The experiment-specific requirements outlined in the DSA essentially require engineering documentation of the explosive-aerosol test chamber that will ultimately be reviewed by an explosive's expert within DOE SSO; this is very similar to documentation previously required by the SNL Explosive Components Facility owners for Phase 3 DUO_2 testing effort, and was satisfied by SNL explosive's experts. Therefore, a majority of the documentation needed already exists. The remainder can be readily generated with minimal cost and associated schedule. The current implementation plan for the GIF does not include addressing the implementation specifics associated with SFR testing. These efforts will progress upon the restart of funded programmatic work.

In addition to the implementing engineering documentation, associated procedures will also need to be developed. The GIF DSA upgrade was sufficiently large that it required re-writing nearly all existing facility procedures. When funding is made available, efforts to write the SFR experiment specific procedures (fuel receipt, operational, post test) will also begin.

2.6.3 Phase 4 Major Obstacles Remaining

Several major obstacles or critical path items remain to be resolved before the Phase 4 test program can be re-started, performed, and brought to a successful conclusion. We have already mentioned the GIF DSA approval (resolved) and the current lack of adequate program funding. The other issues will be described, as well as their impacts, past work and progress, plus current status. These critical items include:

2.6.3.1 Test Rodlets Completion and Shipment to SNL: We have described the postponement in fabrication activities at Argonne National Laboratories in Section 2.6.1.1, due both to funding and Alpha Gamma Hot Cell Facility problems. Rodlet fabrication activities can be restarted in the future, possibly at an alternate facility location with ANL support, and ANL can develop and finalize a transportation plan for the GE-100 cask with test rodlets inside. The most likely hot cell facility to be used for completion of spent fuel test rodlet fabrication is at Oak Ridge National Laboratory. ANL personnel have initiated these potential plans with Oak Ridge and have discussed such plans with both SNL and DOE.

ANL and SNL personnel worked on transport cask internal hardware and related transportation receipt issues in 2005. In addition, transportation pre-planning discussions with SNL Transportation Department and DOE SSO personnel were also conducted during 2005; actual transport priority and post-test spent fuel rodlet transport scheduling by SNL Transportation personnel remains to be done. All of these activities are presently dormant. Finalization of the fabrication and transportation activities is also dependent on the next critical path item.

2.6.3.2 Post-test Transport and Interim Storage of Phase 4 Spent Fuel: SNL presently *cannot* accept ANL spent fuel rodlets without first obtaining an agreed upon *post-test disposal pathway* for the spent fuel rodlets. Sandia Radiation Sciences Center staff and management (responsible for nuclear facilities in SNL Technical Area V) and the DOE SSO have mandated that a spent fuel disposal pathway for these test materials must be specified and authorized before the spent fuel test rodlets can be received at, and tested at Sandia. Discussions between SNL, DOE RW, DOE EM, and Idaho National Laboratory (INL) have been in progress since late 2003 through 2006. These discussions have been to address administrative, site, financial, and technical issues regarding potential agreements on transportation of eight shipments (one test chamber per each GE-2000 cask shipment) of SNL post-test rodlets within sealed, overpacked test chambers to the Idaho National Laboratory site. Transportation would be within DOE-owned and leased GE-2000 transport casks. The tentative plan was for the eight post-test spent fuel containing chambers to be stored in available underground storage silos at the CPP 749 facility at the INL site for an interim period of several decades. DOE and INL would then re-ship these chambers to a licensed federal repository for final disposal, when such repository could accept them. Negotiations on this transportation, temporary storage, and final disposal issue resolution are still in process between DOE RW and other offices. SNL requires a signed DOE agreement to move forward, to resolve this critical path item. In addition, a related critical path issue needs to be addressed on approvals for the use of a GE-2000 transport cask, from SNL to INL.

2.6.3.3 GE-2000 Transport Cask Certificate of Compliance: It is planned to transport each post-test Phase 4 chamber from SNL to the limited-term radioactive material (temporary) storage facility (at INL) within an approved GE-2000 spent fuel transport cask. In order to use the GE-2000 cask, modifications to the cask Certificate of Compliance (CoC) must be made and then approved by the U.S. Nuclear Regulatory Commission (NRC). The typical NRC review times for this activity are on the order of one year. The CoC modification is required because criteria in the current CoC limit the spent fuel to be transported to a maximum burnup of 52 GWd/MTU; the peak burnup of the high burnup Phase 4 test rodlets are about 72 GWd/MTU. During 2005, SNL worked with GE Vallecitos to revise the Certificate of Compliance for the GE-2000 cask and then obtain NRC approval. SNL also discussed preliminary details of this planned work with NRC, for guidance. Under contract to SNL, GE could provide the necessary support to prepare and submit a license amendment to the NRC authorizing the use of the Model GE-2000 cask for transport of high burnup irradiated nuclear fuel sections for SNL needed purposes. GE Vallecitos estimated that this work would take about three months, once initiated. SNL submitted a purchase requisition to GE Vallecitos for performing this work in 2005, but had to postpone the contract due to inadequate program funding. This contract work could be restarted in the future, when funding becomes available, to complete this critical item.

2.6.3.4 SNL GIF Pre-Test Preparation Activities: The option to restart the Phase 4 spent fuel test program has been left open, pending future adequate DOE funding and continued program support. Other areas of work to be performed by SNL TA-V personnel in preparation for the start of Phase 4 testing in the GIF include required test hardware design, procurement, fabrication, and installation activities, plus development of experiment specific procedures. A secondary confinement enclosure needs to be obtained, for assuring that the GIF test cell facility remains uncontaminated. This confinement would surround the explosive-aerosol test chamber and is anticipated to be very similar to the secondary containment tent used in the SNL ECF for the Phase 3 tests. A portable glove box has been located for use and needs to be installed in the GIF test cell to remove aerosol impactor particle samplers post-test, then weigh and package them. This glove box is slightly different than the portable glove box used for the Phase 3 tests in that it is a shielded glove box with a larger internal volume. This glove box also allows for proper negative ventilation during pass-through operations. During fuel receipt activities, it is envisioned that manipulators would be used to handle the test rodlets. The GIF facility has a number of manipulators available for use. Prior to this specific experiment campaign, there has been no previous need for manipulators at the GIF and as such they have previously never been installed. For proof of principle, procedures were developed in August 2006 for use at the GIF which involved removal of solid steel manipulator port plugs and installation of the manipulators themselves. Shortly thereafter the manipulator set to be used for SFR testing was installed in the cell without requiring modifications and subsequently the cell was returned to its normal state. Other activities and support apparatus may be required and will be defined in the future. Efforts for larger support hardware have already been identified. Procurement of these components can take place with associated funding. *None* of the support apparatus and pre-test activities and are considered critical path items.

Further descriptions of the importance for continuing and completing the Phase 4 spent fuel test program and the critical need for obtaining the spent fuel ratio are included in Section 5.1.

3. Recent Testing

Very limited amounts of new testing have occurred since the publication of SAND2006-5674 [3] in late 2006. However, two significant activities have occurred and are presented herein, the Phase 2+ test 2/CSC and the aerosol laboratory calibration studies.

3.1 Phase 2+ Test 2/CSC

On March 8, 2007, SNL successfully performed a single explosive-aerosol surrogate test as an extension of the Phase 2+ test program, test 2/CSC, to quantify respirable, aerosol particulate dispersal and to reduce some topics of uncertainty. This test incorporated multiple surrogate cerium oxide pellets and multiple fission product dopant disks, contained in a Zircaloy-4 cladding tube. Test 2/CSC was performed within a 50 m^3 steel aerosol chamber located within SNL Technical Area 3. Multiple Marple impactors and other aerosol particle collectors were used, plus multiple thermocouples. Primary objectives of this test were to evaluate effects on produced aerosol behavior resulting from lesser amounts of explosive carbon soot (by HEDD detonation in an oxygen-rich environment) and lower test chamber temperatures and pressure in the 50 m^3 steel chamber, in comparison to previous Phase 2, 2+, and 3 test chambers (Figures 2.2.3, 2.3.1, and 2.5.5).

Test 2/CSC was a cooperative effort between personnel in the SNL High Consequence Assessment and Technology Department and the Materials Transportation Testing and Analysis Department, with major support by personnel in Explosive and Firing Systems, Explosive Projects/Diagnostics, and the Aerosol Sciences Departments. Primary support was provided from NRC Office of Nuclear Regulatory Research, combining experimental efforts on (1) evaluating explosive dispersal characteristics of spent fuel materials and (2) explosive dispersal characteristics of NRC-licensed material potentially used in radiological dispersion devices. This second experimental work is directly related to homeland security and is directed at achieving counter-terrorism or homeland security objectives. Secondary test 2/CSC support (test materials, the HEDD, aerosol analyses, labor) was also funded in part through DOE RW activities.

3.1.1 Test 2/CSC Experimental Details

3.1.1.1 Surrogate Cerium Oxide Test Rodlet and Dopants: The surrogate test rodlet consisted of a Zircaloy-4 cladding tube (204 mm long with a 10.6-mm outer diameter), with a total of 9 CeO_2 ceramic pellets (each 7.22 mm long with a 9.33-mm outer diameter, average) and 8 non-radioactive fission product dopant disks (each 1 mm long with a 9.0-mm outer diameter), very similar to the test rodlet in previous test 2/10B. The rodlet contained a center CeO_2 pellet ("C") with four fission product disks ("|") on each side and four CeO_2 pellets on each end. This configuration is illustrated as CCCC||||C||||CCCC. The 8 dopant disks contained cesium iodide (CsI), ruthenium oxide (RuO_2), strontium oxide (SrO), and europium oxide (Eu2O3), at a slightly higher concentration than the six dopant disks contained in the previous test 2/10B; refer to Table 2.3.2 for dopant concentrations. The internal pressure within this test rodlet was 1 bar, ~ atmospheric pressure.

3.1.1.2 Test Chamber, Aerosol Apparatus, and HEDD: Test 2/CSC was performed in a 50 m^3 steel aerosol chamber (actually 48.6 m^3) near Site 9920 at SNL, located at the Coyote Test Field in Technical Area 3. The outside and inside of the chamber are illustrated in Figure 3.1.1. There were also twelve thermocouples and one pressure transducer installed within the test chamber. All of the thermocouples were sampled for the 30-minute test sampling duration at 1-second intervals.

Figure 3.1.1. SNL Site 9920 50 m^3 Steel Test Chamber, External and Internal

Aerosol sampling within the test chamber was performed with 8 Marple cascade impactors (the same type and model as used in all other Phase 2, 2+, and 3 tests), 12 inline total mass samplers, two cyclone separators, and four fans to promote mixing (visible in Figure 3.1.1, internal). The Marple impactors and total mass samplers were placed in protective steel boxes (30.5 cm x 15.2 cm x 30.5 cm), shown in Figure 3.1.2 on the right and left, respectively. These sampling boxes have been labeled #1-12 (Figure 3.1.3, also visible in Figure 3.1.1, internal). The total mass samplers and cascade impactors in boxes #3 and 10 sampled for time 0 to 30 minutes. Boxes #2, 4, 5, 8, and 11 sampled from 0 to 15 minutes and boxes #1, 6, 7, 9, and 12 sampled from 15 to 30 minutes. Boxes #1, 6, 8, and 11 did not contain cascade impactors. The two cyclone separators were located in separate boxes on the floor of the steel chamber. They sampled from 0 to five minutes. Large particles are expected to settle out over the shorter time periods while the smallest, respirable particles will stay suspended over the longer time periods. Particles settling out on the test chamber floor and walls were collected for evaluation post-test with a HEPA vacuum. Post-test, ICP/MS chemical analyses of collected aerosol particles were performed, similar to other Phase 2, 2+, and Phase 3 tests.

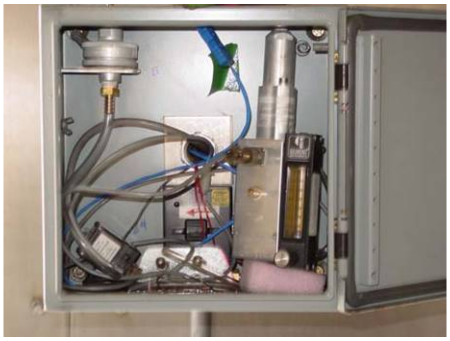


Figure 3.1.2. Total Mass Sampler and Marple Cascade Impactor

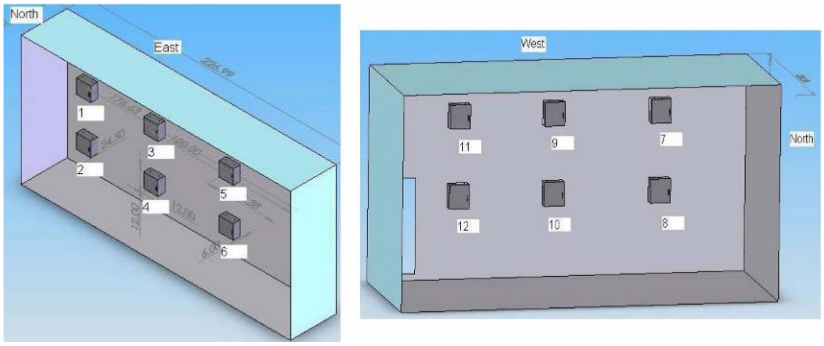


Figure 3.1.3. Numbering for Aerosol Sampler Boxes

The aerosol particle masses collected were adjusted for sampler inlet efficiency and inter-stage losses, and settling losses over stirred sampling times; calibrations were calculated from multiple earlier, non-related tests by the SNL facility owners. The calibration efficiency multipliers for the 50 m^3 test chamber are illustrated in Figure 3.1.4. There is more uncertainty for particles > 20 µm AED if the stirred settling assumption (well stirred and uniformly mixed) is not met.

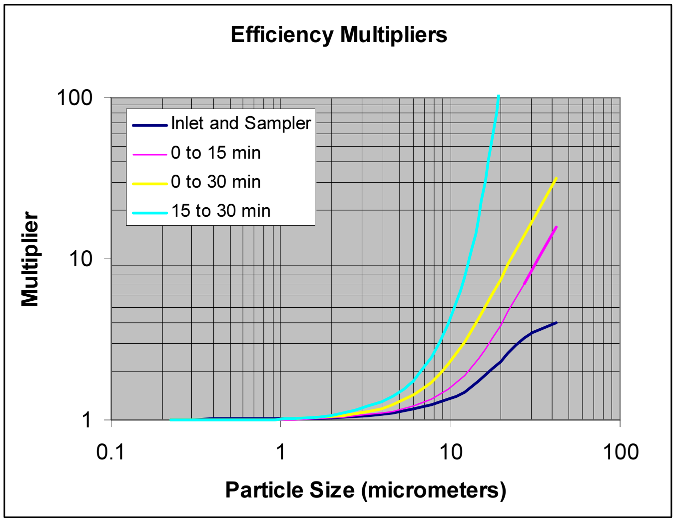


Figure 3.1.4. 50 m^3 Chamber Efficiency Multiplier Calibrations

Figure 3.1.5. Test 2/CSC Rodlet and Support Apparatus

A "blank" test was performed on March 6, 2007 to determine background levels for the fission product dopant chemical species of interest within the 50 m^3 chamber and apparatus, specifically cerium, cesium, ruthenium strontium, and europium. One cascade impactor and four total mass samplers were used in this test and impactor media were chemically analyzed by ICP/MS by two independent chemical laboratories. The results of this "blank" test showed negligible background levels of Ce, Eu, and Ru; however, elevated levels of Cs and Sr were present and quantified in the background test. These background concentration levels were taken into account (subtracted) when analyzing the Test 2/CSC data for measured Cs and Sr RF and EF values.

The precision HEDD explosive used in this test ("CSC1"), and the HEDD-to-rodlet stand-off distance was identical to that used in all other tests in this program at SNL. Details of the HEDD were provided in [18]. The HEDD is located within the fragmentation shield on the right in Figure 3.1.5, the surrogate target rodlet at the center, and the HEDD jet stop block at the left in the same figure, all inside the 50 m^3 test chamber.

3.1.2 Test 2/CSC Major Results

3.1.2.1 Target Rodlet Disruption: The observed effects of an explosive HEDD jet impact on the test 2/CSC rodlet appeared very similar to the preceding Phase 2 and 2+ tests. There was a gap in the Zircaloy tubing of ~27-(32 mm average)-36 mm; photographs of the post-test rodlet appear to show a larger disrupted gap, but the rod ends had moved outward. The center CeO_2 pellet, all 8 dopant disks, and two CeO_2 pellets on either side of the center were particulated; the remaining two CeO_2 pellets on each end were intact.

3.1.2.2 Temperature Measurements: The temperatures within the 50 m^3 test chamber were of interest because they were anticipated to be significantly lower than temperatures measured within the Phase 2, 2+, and Phase 3 sealed aerosol test chambers; refer to Section 2.2.2.7 and Table 2.3.3. Measured results from the thermocouples are shown in Figure 3.1.6. Thermocouples 1 and 6 (on the fixture near the rodlet) were averaged, shown as Temp 1&6. Thermocouple 7 was on the wall, and thermocouple 12 was ~1 cm from the wall. The remaining thermocouples were in their corresponding boxes. Temperature profiles for thermocouples directly across from one another were similar and were averaged for plotting purposes. These data indicate temperatures inside the large chamber did not stay elevated for long periods of time, as the peak value is just over 40°C (105°F). The pressure transducer did not capture any notable data.

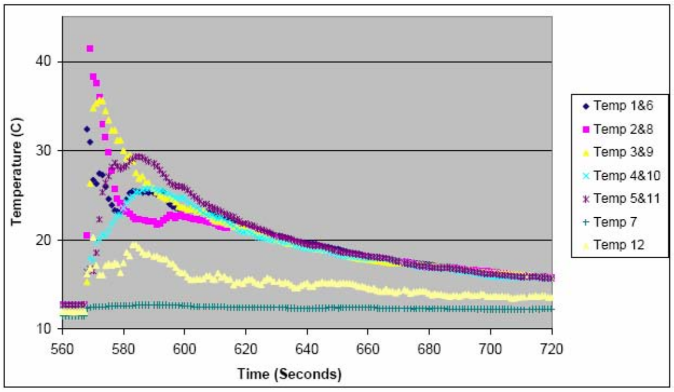


Figure 3.1.6 Test 2/CSC Measured Temperatures

3.1.2.3 Test 2/CSC Aerosol Results: The respirable fraction (RF) and integrated and differential enrichment factor (IEF) values for Ce in the CeO_2 and the fission product dopants cesium, ruthenium, strontium, and europium calculated from the aerosol measurements taken in test 2/CSC are summarized in Table 3.1. These values *are based on all samples collected.* Listed uncertainty values are the calculated 99% confidence intervals. Test 2/CSC RF, IEF, and differential EF values are also incorporated into multiple tables and figures within Sections 4.1 and 4.2, following, for direct comparisons to all other measured RF and IEF results.

Table 3.1. Test 2/CSC Measured RF and IEF Values

Element	Respirable Fraction RF ± 99% CI	Integrated Enrichment Factor IEF ± 99% CI
Cerium (Ce)	0.95% ± 0.45%	
Cesium (Cs)	13.5% ± 12.1%	15.0 ± 15.4
Ruthenium	9.8% ± 16.5%	22.2 ± 6.1
Strontium	14.0% ± 5.6%	15.6 ± 6.0
Europium	5.05% ± 2.11%	5.7 ± 0.7
Zirconium	1.60% ± 1.05%	

The Ce, Eu, and Zr measured RF values for test 2/CSC compare well with other Phase 2+ tests performed. The comparison of test 2/CSC measured RF results with tests 2/10B, 2/10C and 2/10D RF values is illustrated in Figure 3.1.7. Little data is available from these tests for comparison of Sr and Ru RF values. Test 2/CSC cesium RF values are observed to be a little lower, but within a factor of < X2 in magnitude. Despite the overall agreement of the Phase 2+ tests measured RF values, a slight shift was noted within the respirable particle size range distribution for both Ru and Cs. The primary, peak particle size range for Test 2/CSC was around 0.10 µm AED for Ru and 0.51 µm for Cs; both had secondary, lower peaks around 3.5 µm. The primary peak for both Ru and Cs in the other Phase 2 and 2+ tests performed (refer to Section 4.1.2) was around 3.5 µm with very little if any peak at the smaller size range. A possible explanation for these peak respirable particle size differences is that the reduction in the amount of soot and soot agglomeration in the large 50 m^3 test chamber (compared to the Phase2 and 2+ small, sealed, and somewhat oxygen-deficient test chambers) decreased the agglomeration of Ru and Cs onto the less plentiful, slightly larger carbon soot particles (soot peaked at about 3.5 µm AED). While this difference in soot content between test chambers may appear to result in an experimental bias, the overall effect on both respirable fraction (RF) and integrated enrichment factor (IEF) measured values is minimal, essentially a random uncertainty, since both RF and IEF values extend over the entire respirable particle size range of 0 to 10 µm AED. However, in regards to the measured differential enrichment factors (EF) as a function of size range, presented in Section 4.2, a systematic bias is appropriate since the peaks in EF values may be displaced towards a slightly smaller respirable size.

The test 2/CSC measured integrated EF values for Cs, Eu, and Ru also fit within the measured range of values from the similar data from Phase 2+ test 2/10C and Phase 3 test 3/1C, as shown in Figure 3.1.8; the test 2/10B Cs and Eu values are, however, appreciably higher. Again, very little Sr and Ru data were available for comparison.

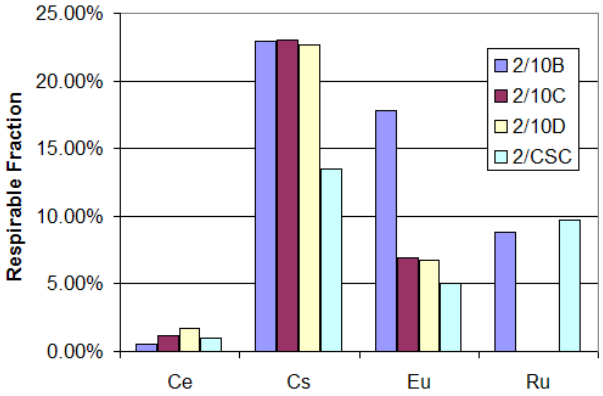


Figure 3.1.7. Comparison of Phase 2+ Test RF Data for Ce, Cs, Eu and Ru

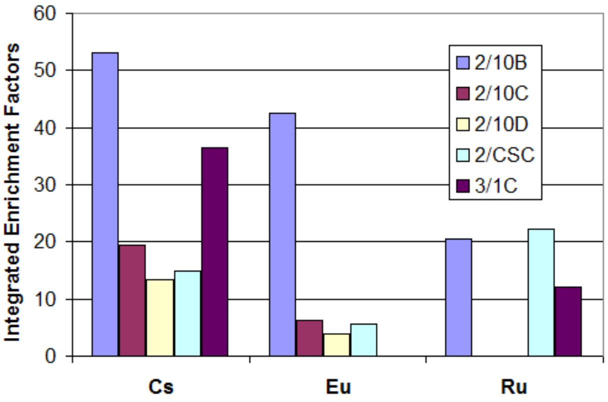


Figure 3.1.8 Comparison of Phase 2+ and 3 Test IEF Values for Cs, Eu, and Ru

In general, the measured RF and IEF data from Test 2/CSC compare quite well with similar data from Phase 2, 2+, and Phase 3 tests (refer to Sections 4.1 and 4.2) conducted in the smaller, hotter, higher-pressure-gradient, and oxygen-deficient explosive-aerosol chambers used therein. These dissimilar conditions between tests did not have a major impact on the source term data. For instance, particle deposition was observed in the internal sampling tubes in the vertical explosive-aerosol test chamber as previously reported [3], be described further in the following Section 3.2. This observed deposition is due presumably to thermophoretic, diffusiophoretic, and turbulence effects, and was suspected [3] to add some uncertainty or underestimation to measured RF values by possibly as much as a factor of two. However, consistent RF results from test 2/CSC in the well-calibrated 50 m^3 chamber suggest that explosively generated soot, plus higher internal temperatures and pressures in the vertical test chamber do not have a significant impact on the source term data, probably by less than a factor of two, or well within the calculated confidence intervals to be presented in Sections 4.1 and 4.2 --- except for cesium IEF values which are discussed in more detail in Section 4.2. *This decrease in uncertainties may be the most important finding from test 2/CSC.* The consistency of the measured RF and EF values between the vertical chamber tests and the 50 m^3 chamber test strongly suggest that the sampling line losses observed in the vertical chamber tests are likely to be from larger aerosol particles that most probably have some sort of inertial deposition mechanism such as turbulence or unsteady flow; this is described further in the following section.

3.2 Aerosol Inlet Losses and Laboratory Calibration Testing

The sampling efficiency of the particle collectors used in our tests, the Marple impactors and Large Particle Separator apparatus is very high and is a function of particle size, rapidly changing temperatures and pressure within the sampling system. For respirable size particles, the efficiency is close to 100%. In our previous FY 2005-2006 technical report SAND2006-5674 [3], we noted that appreciable particle deposition losses have been observed in the aerosol sampling hardware lines within and adjacent to our sealed, high-internal temperature, explosive-aerosol test chambers, as used for most Phase 2, 2+, and Phase 3 tests. We have considered what could cause this observed deposition. The possibilities are thermophoresis, diffusiophoresis, and turbulent deposition effects. For thermophoresis, caused by large thermal gradients within the aerosol collection chamber and sampling lines, to be the cause, gas cooling would need to occur predominantly in the lines where most of the deposition has occurred. This would also be the case for diffusiophoresis. Diffusiophoretic effects may result in deposition losses on test surfaces on which water vapor condense. The water vapor is generated as a byproduct in the explosive detonation/ oxidation, and may reach super-saturation as the sampled gas cools in the sampling lines. Since flow is laminar in the sample lines above the test chamber and upstream of the isolation ball valve, and that ball valve produces a turbulent jet as does the sample ball valve during opening and closing, if turbulent deposition is causing the observed deposits, they are occurring in a consistent location. As such, we considered all respirable fraction aerosol results previously reported in our technical documents [2-3] as being *preliminary* in nature, until the completion of test 2/CSC and some SNL laboratory calibration testing addressing these losses.

In our explosive-aerosol, sealed test chambers used for aerosol particle sampling, there are a number of mechanisms that can alter the aerosol distribution during its transport form the chamber to the sampling/collection instruments. The temperature and pressure excursion during and after the detonation of the HEDD declines rapidly and drops from about an observed (Phase 2 and 2+ tests) maximum of 330 °C (603 K) (and in several limited instances, 833-1113 K) and 50 psia (~ 3.4 bar) at about 1 second after ignition to 920K and 35 psia (~2.4 bar) at the initiation of sampling to 480K and 19 psia (~1.3 bar) at the completion of sampling; refer to Section 2.3.2.3. This rapid variation in temperature and pressure during the sampling process can complicate interpretation of the collected sample. The sampling train from the Large Particle Separator (LPS) and forward to, and past the Marple (refer to Figure 4.9 in [3]), was seen to have little variation in temperature but will experience the pressure variation seen in the chamber. Sampling flow is controlled by a critical orifice at nominally 2 liters per minute volumetric flow at the upstream conditions, i.e., temperature not very different from ambient and pressure declining from about 35 psia to 19 psia (~3.4 to ~2.4 bar) during the sample period. The inlet and sample lines and sampling train will have been pressurized to 35 psia (~2.4 bar) at the initiation of sampling. The sample line up to the ball valve will see the full pressure excursion. The gas will cool as it flows into the sampling lines to the LPS. The sampling flow in the lines sees a temperature of the ambient room and a pressure on the order of the chamber pressure. The dead volume of the sampling train is 111 cm^3 and this volume is pressurized at the chamber pressure which is falling during the sampling period introducing an expansion that to a small extent counteracts the sample flow. Since the pressure in the chamber arises from the thermal input of the HEDD, the ratio of temperature to pressure is constant, and one can estimate a volumetric flow into the sampling tube of about 1.75 actual liters per minute at the initiation of sampling, increasing to just under 2 actual liters per minute at the end of the 10 second sampling period. There is an isolation ball valve with a 0.25 inch (6.4 mm) internal diameter at the end of the internal sampling tube. The Reynolds number through this valve is about 400, which is sufficient to produce a turbulent jet into the 0.375 inch ID (9.5 mm) sampling line going from the isolation ball valve to the control ball valve. During operation, the control ball valve opens and closes resulting in the temporary generation of a turbulent jet into the ball valve itself, and into the sampling line going from the control ball valve to the LPS. At that point, just before the LPS, temperature measurements show very little variation, going from the ambient room temperature to about 10 K above ambient. This indicates that gas cooling has taken place upstream of the LPS and the rest of the sampling train. Minor deposition has been observed in the initial sampling line upstream of the isolation ball valve, and significant deposition (on the order of the mass collected in the LPS and impactor) has been observed in the sample lines from the isolation ball valve to the LPS.

Calibration work to assess the magnitude of the deposition loss effects potentially resulting from thermophoresis, diffusiophoresis, and turbulent loss effects was planned and initiated, but equipment problems coupled with personnel availability prohibited planned completion. Some data were taken from the laboratory calibration effort, but the data was inconclusive because of aerosol charging uncertainty caused by a defective neutralizer. We have, however, been able to assess composition data from tests that indicate most of the material deposited in the sample lines between the isolation ball valve and the LPS reflects the composition found in the larger particles rather than that of the respirable < 10 µm AED particles. This suggests that the observed material deposited in the sampling lines is predominantly larger particles and most probably deposited by turbulence. Furthermore, comparison with the large 50 m^3 chamber test 2/CSC, in which aerosol samples were taken without the inlet tube, valves, or LPS in the sampling train, and under conditions without the possibility of thermophoresis, diffusiophoresis, or turbulence acting on the sample collection, we see values for respirable fraction of the pellet matrix material (CeO_2 or UO_2) similar to those determined from the previous smaller chamber tests (without adjustment for small particle loss). Although the laboratory calibration work planned has not been brought to a fully satisfactory conclusion due to the problems encountered, further calibration work cannot be anticipated until a later time, when full, or further program testing is restored. Aerosol data collected on the Large Particle Separators, with data relevant to 30 to 100 µm AED particles, still may have a larger degree of residual uncertainty and remain to be fully interpreted.

Therefore, based on the available results and interpretations from both the initiated laboratory calibration work to date, plus the measured aerosol results from tests 2/CSC in the 50 m^3 test chamber, the measured RF values reported in this document, ± their calculated confidence intervals, are reported as final data, no longer referred to as preliminary.

4. Aerosol Data and Interpretations

In this section, we summarize the relevant respirable and aerosol particle results obtained from all Phase 2, 2+, Phase 2/Phase 3, and Phase 3 test performed from 2002 through 2007. This allows us to provide a thorough interpretation and to inter-compare results from all tests. The aerosol data for surrogate cerium oxide, depleted uranium oxide, and dopant fission product materials are considered prototypical of similar data to be obtained from actual spent fuel materials in later Phase 4 tests in this program. In addition to some new data, other data and analyses presented in this section are updated and in some cases revised from those documented earlier in [2-3] and presented at numerous WGSTSC technical meetings over the same time period.

Following the HEDD jet impact with the test target rodlets, aerosol particulates are generated rapidly. As measured, more than 10 g/m^3 of respirable particles were generated in the test chamber; a large fraction of this material, approximately half, is carbon soot from the explosive device. The high soot concentration in sealed test chambers limited the total sampling time to 10 seconds, in order to prevent Marple impactor overloading. The particle masses on each stage of the particle impactor or Large Particle Separator were directly measured after each test. The sample volume is taken as the flow through the orifice at the orifice temperature, which is taken as ambient temperature prior to the test.

We quantified the aerosol particle size distributions of all major elemental species involved in the explosive-aerosol/sabotage process, including Ce or depleted U from the pellets, Cs, Ru, Sr, and Eu fission product species, Cu from the HEDD, and Zr from the Zircaloy-4 cladding tube. From observations (in Appendix A in [3]) of the CeO_2 distributions in the ~0 through ~20 μm AED size range, CeO_2 particles appear to peak broadly over the 3.5-9.8 μm respirable size range. No distinct Ce or CeO_2 particle pattern was observed in the aerosol range from about 30 to 100 μm AED, collected with the LPS apparatus. The observed DUO_2 particle size distribution peaks quite strongly at about 3.5 μm AED, then decreases.

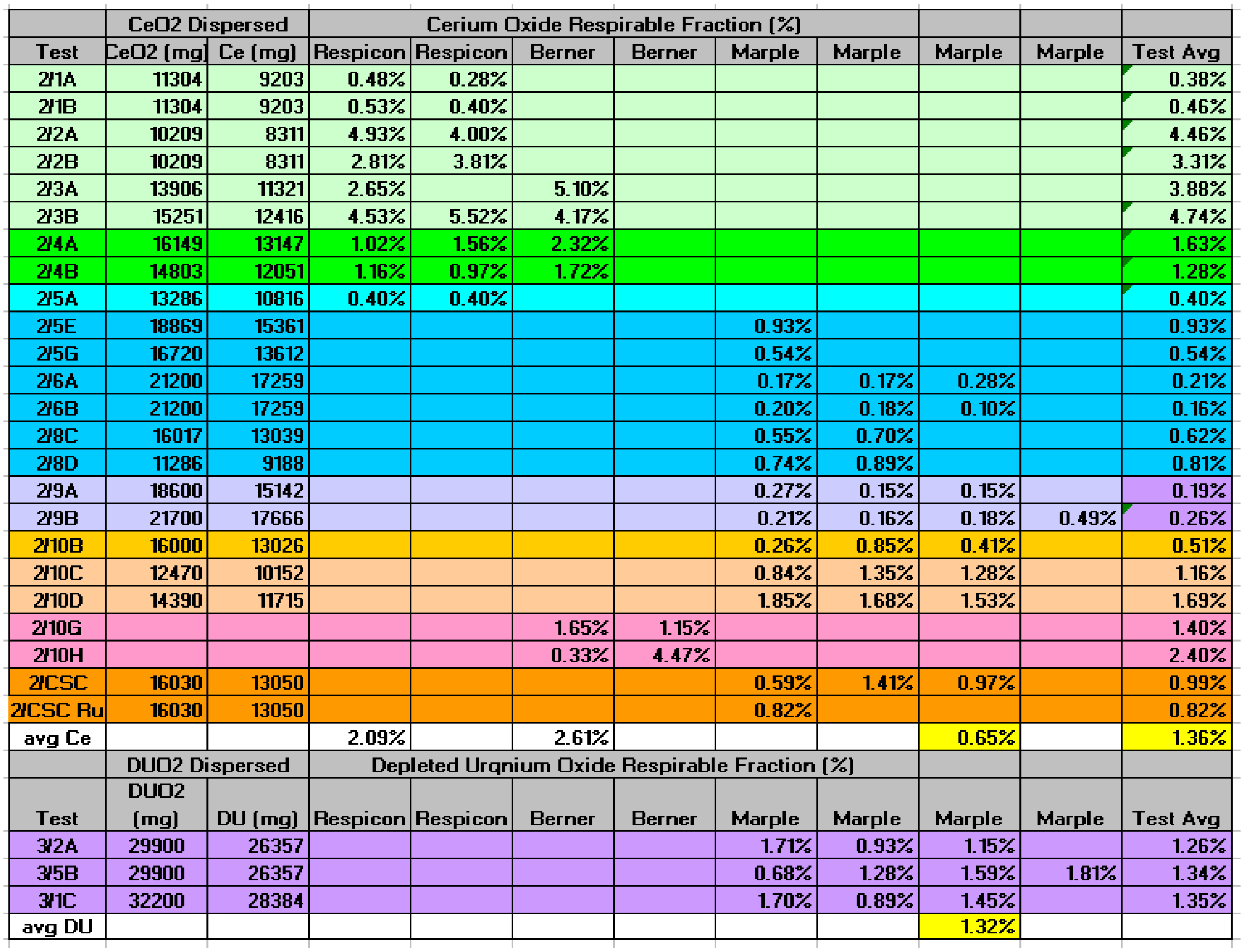
4.1 Respirable Fraction Results

The respirable fraction (RF) produced when the HEDD jet impacts a target rodlet is the fraction of a specific material (cerium oxide, uranium oxide, zirconium, fission product, etc.) *produced* (in the rod swept_volume, over the entire range of particles of size 10 μm AED and smaller) divided by the total amount of those material particulated.

4.1.1 CeO_2, DUO_2, and Zr Measured Respirable Fraction Results

Respirable fractions for CeO_2, DUO_2, Zr, and fission product Cs, Ru, Sr, and Eu have been calculated from the aerosol measurements taken in all the tests to date, based upon the measured aerosol size and chemical (elemental) concentrations, and the amounts of those materials dispersed into the test chamber. The measured respirable fraction results are summarized in Table 4.1.1 and Figure 4.1.1 for surrogate CeO_2 and DUO_2 pellet materials. New data for tests 2/10G and 2/10H (performed by Fraunhofer ITEM and EMI) and 2/CSC are included. The measured, average **RF for CeO_2**, based on the most recent Marple impactor data only (for tests 2/5E and later, considered our best quality and largest amount of interpretable data), was **RF = 0.65% ± 0.23%** with a 99% confidence interval (CI) or ± 0.15% with a 90% CI. Similarly, using <u>all</u> measured data, including Marple plus older Respicon and Berner impactor data [2-3], the average CeO_2 **RF was 1.36% ± 0.50%** with a 99% CI or ± 0.32% with a 90% CI. Results from older tests (2/2A through 2/3B, in particular) may have higher RF values due to use of different, partially vented square-box aerosol chambers [1], as previously illustrated in Figures 2.2.1 and 2.2.2.

Table 4.1.1. Respirable Fraction Test Results for CeO_2 and DUO_2



	CeO2 Dispersed		Cerium Oxide Respirable Fraction (%)								
Test	CeO2 (mg)	Ce (mg)	Respicon	Respicon	Berner	Berner	Marple	Marple	Marple	Marple	Test Avg
2/1A	11304	9203	0.48%	0.28%							0.38%
2/1B	11304	9203	0.53%	0.40%							0.46%
2/2A	10209	8311	4.93%	4.00%							4.46%
2/2B	10209	8311	2.81%	3.81%							3.31%
2/3A	13906	11321	2.65%		5.10%						3.88%
2/3B	15251	12416	4.53%	5.52%	4.17%						4.74%
2/4A	16149	13147	1.02%	1.56%	2.32%						1.63%
2/4B	14803	12051	1.16%	0.97%	1.72%						1.28%
2/5A	13286	10816	0.40%	0.40%							0.40%
2/5E	18869	15361					0.93%				0.93%
2/5G	16720	13612					0.54%				0.54%
2/6A	21200	17259					0.17%	0.17%	0.28%		0.21%
2/6B	21200	17259					0.20%	0.18%	0.10%		0.16%
2/8C	16017	13039					0.55%	0.70%			0.62%
2/8D	11286	9188					0.74%	0.89%			0.81%
2/9A	18600	15142					0.27%	0.15%	0.15%		0.19%
2/9B	21700	17666					0.21%	0.16%	0.18%	0.49%	0.26%
2/10B	16000	13026					0.26%	0.85%	0.41%		0.51%
2/10C	12470	10152					0.84%	1.35%	1.28%		1.16%
2/10D	14390	11715					1.85%	1.68%	1.53%		1.69%
2/10G					1.65%	1.15%					1.40%
2/10H					0.33%	4.47%					2.40%
2/CSC	16030	13050					0.59%	1.41%	0.97%		0.99%
2/CSC Ru	16030	13050					0.82%				0.82%
avg Ce			2.09%		2.61%				0.65%		1.36%
	DUO2 Dispersed		Depleted Urqnium Oxide Respirable Fraction (%)								
Test	DUO2 (mg)	DU (mg)	Respicon	Respicon	Berner	Berner	Marple	Marple	Marple	Marple	Test Avg
3/2A	29900	26357					1.71%	0.93%	1.15%		1.26%
3/5B	29900	26357					0.68%	1.28%	1.59%	1.81%	1.34%
3/1C	32200	28384					1.70%	0.89%	1.45%		1.35%
avg DU									1.32%		

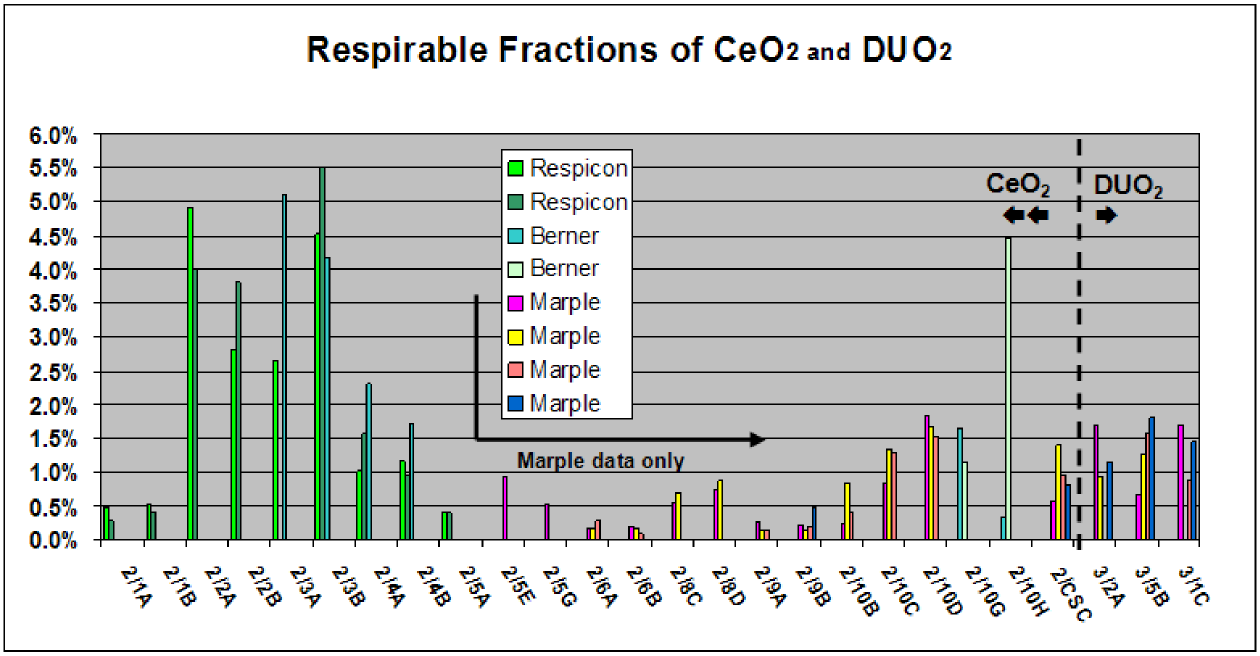


Figure 4.1.1. Respirable Fraction (RF) Results for CeO_2 and DUO_2

The measured, average respirable fraction value for **DUO_2 is RF = 1.32% ± 0.32% at 99% CI** or ± 0.21% at 90% CI, based on all data from the first three Phase 3 tests. This DUO_2 measured RF value is very close to the average measured CeO_2 RF value of 1.36% ± 0.50% (all data) or a factor of two higher than the CeO_2 RF value of 0.65% ± 0.23% for Marple data only.

Both the DUO_2 and CeO_2 RF values are significantly below the "conservatively estimated" UO_2 spent fuel RF value of 5% used in earlier analyses [4]. For additional comparison with historical data and calculations described in Section 1.3, Luna et al. [13] derived DUO_2 RF values from earlier large-scale SNL [6] and GRS [5] DUO_2 test results; these calculated surrogate DUO_2 RF values ranged from 1.7 to 5.3 for one-wall cask penetration, or 2.8 to 12 for two-wall cask penetration. The experimentally measured DUO_2 RF = 1.32% ± 0.32% from our current SNL-WGSTSC program is appreciably lower than all of the previous calculations.

An effect that has been considered is that the CeO_2 RF represents the small tail of a distribution produced by fragmentation. Sieve analysis of the CeO_2 in the debris indicates a mass mean debris size on the order of several hundred micrometers over the various tests, with observed variation [3]. Even with highly repeatable fragmentation distributions characterized by small variance in the mean and spread of the distribution, the integral mass in the small tail is subject to larger variation.

There are noticeable variations in measured RF values within individual tests using replicate particle samplers, as shown in Figure 4.1.1. These variations are difficult to specify, but are most likely due to both statistical variations and possible differences in experimental conditions and sampling between particle collectors, plus following particle chemical analyses. In addition, by comparing the assorted CeO_2 RF results grouped by test series, as shown in Figure 4.1.1, we can also make several observations and/or implied results.

4.1.1.1 Rodlet Pressurization Effects: Phase 2 CeO_2 explosive-aerosol tests 2/6A and 2/6B were specifically performed with internally pressurized (helium gas) rodlets at 27.6 bar and 40 bar, respectively, to evaluate the effects of pressurization. Compared to later Phase 2/Phase 3 tests 2/9A and 2/9B performed with un-pressurized (1 bar, atmospheric pressure) rodlets in a similar vertical explosive-aerosol test chamber, no discernable effect of rodlet pressurization on measured CeO_2 RF can be noticed, e.g. a presumed increase in measured RF values, as evident in respective data bars in Figure 4.1.1. Adding other test results to this comparison from other un-pressurized rodlets in tests 2/5A, 2/5E, 2/5G, 2/8C and 2/8D, all performed in the identical test chamber as 2/6A and 2/6B, no specific significant effect of pressurization on RF can be obtained among the various tests. Similar results were obtained with the first three Phase 3 DUO_2 tests, test 3/2 (A) and 3/1(C) at atmospheric pressure and test 3/5 (B) pressurized with 40 bar of He gas within the rodlet plenum. The measured DUO_2 respirable fractions for these Phase 3 tests, shown in Figure 4.1.1, are essentially the same, regardless of pressure within. The conclusion here is that the *internal pressurization effect in the plenum region of short test rodlets* (from 1 to 40 bars) is not a significant variable when compared to the total amount of particulates released from a relatively large (~27 mm) length of the rod impacted/particulated by the HEDD jet. The measured differences or scatter between results from multiple, replicate Marples impactors used on individual tests appear greater than the differences between non-pressurized and pressurized rodlet versions of several comparable tests. That is, the potential effects due from short rodlet pressurization in these tests seem to be smaller than test-to-test and sample-to-sample variations. Based on these experimental results and observations, short spent fuel test rodlets for Phase 4 testing will be fabricated without internal pressurization, i.e., with 1 bar He gas; WGSTSC partners agreed on this conclusion [3].

4.1.1.2 Aerosol Chamber Temperature and Soot Effects: Based on the measured RF values shown in Table 4.1.1 and Figure 4.1.1, it is also difficult to determine any conclusive effects on measured CeO_2 RF values produced when comparing results from a sealed, high internal temperature, vertical test chamber (tests 2/5E through 2/9B and 3/2A through 3/1C) with results from semi-open chambers (tests 2/10B through 2/10D) plus similar semi-open square box chamber (tests 2/4A and 2/4B) compared to the 50 m^3 chamber (lower internal temperature, lower soot cont) in test 2/CSC or the low internal temperature vertical elutriator test chamber used in tests 2/10G and 2/10H, performed in Germany. The areas of concern between these different test configurations are potential effects of explosive by-product carbon soot and chamber temperature rise (plus potential thermophoretic effects), from the HEDD detonation. No major trends are obvious and there is a large amount of data scatter.

4.1.1.3 Zirconium RF Results: Table 4.1.2 presents all available measured respirable fraction results (for particles of size ≤ 10 μm AED) for zirconium from the Zircaloy-4 cladding tube; Figure 4.1.2 illustrates this zirconium RF data. The measured respirable fraction average for the **Zr RF = 1.13% ± 0. 24%** based on the Marple data only from both CeO_2 and DUO_2 tests, with a 99% CI (or ± 0. 15%, with a 90% CI). The Zr RF average based on all collected data was, quite similarly, 1.32% ± 0.29% with a 99% CI (or ± 0. 18%, with a 90% CI). These values are comparable to the cerium oxide respirable fractions measured for all data, even though zirconium (Zircaloy-4 cladding) is a ductile metal, not a brittle ceramic like the CeO_2 ceramic pellets.

The total amount of zirconium material disrupted by the HEDD jet and dispersed is based on the observed gap in the target rodlet plus the measured zirconium mass loss. Zirconium metal fragmentation may be dominated by explosive-mechanical disruption; small fragments or chunks of Zircaloy metal were obvious in photographs of Phase 2 chamber debris collected [1, Figure 19a]. However, the zirconium can also partially melt and oxidize into particles as a result of the high-temperature HEDD jet impact. High speed video photography was performed during tests 2/0, 2/1A, and 2/1B [1]. Rapid oxidation (burning) of the zirconium was clearly evident, occurring within the first ~0.3 seconds after detonation. Zirconium oxidation is also suggested by the appreciable amount of zirconium found in the smaller, respirable impactor size ranges. From observations of the Marple impactor zirconium distributions in the ~0 through 10 μm AED respirable size range, and even up to ~35 μm (Appendix A in [3]), zirconium is not one of the most prevalent respirable materials produced by the HEDD jet explosive impact process. However, respirable zirconium particles appear to peak somewhat over the range size of 1.6 to 10 μm AED (maximized, frequently, at about 1.6 to 6 μm AED).

4.1.2 Fission Product Respirable Fraction Measured Results

4.1.2.1 Cesium: Fifteen tests with CeO_2 test rodlets, and one so far with DUO_2 test rodlets, have incorporated fission product dopants. Table 4.1.3 presents the respirable fraction results for fission product dopant cesium for all tests to date. Figure 4.1.3 illustrates all these cesium RF data.

The cesium respirable fraction average was **Cs RF was 24.9% ± 5.6%** of dispersed mass based on all CeO_2 target Phase 2 and 2+ collected data with a 99% CI (± 3.6% with a 90% CI); there was no reason to distinguish between Marple and earlier Respicon and Berner particle sampler data in this case. Based on the DUO_2 Phase 3 data only, the Cs RF average was somewhat higher, **45.0% ± 16.1** with a 99% CI (± 10.3% with a 90% CI).

Table 4.1.2. Respirable Fraction Results for Zirconium

	Zr Dispersed	Zirconium Respirable Fraction (%)							
Test	Zr (mg)	Respicon	Respicon	Berner	Marple	Marple	Marple	Marple	Test Avg
2/1A	2990	0.41%	0.21%						0.31%
2/1B	2990	0.28%	0.26%						0.27%
2/2A	3035	1.85%	1.61%						1.73%
2/2B	3002	1.48%	1.94%						1.71%
2/3A	3249	1.89%		4.87%					3.38%
2/3B	3249	2.23%	2.61%	3.04%					2.63%
2/4A	3769	0.84%	1.17%	3.45%					1.82%
2/4B	3249	1.10%	0.90%	2.95%					1.65%
2/5A	3509	1.18%	1.34%						1.26%
2/5E	2859				2.92%				2.92%
2/5G	3249				1.29%				1.29%
2/6A	3510				1.12%	1.30%	1.27%		1.23%
2/6B	3510				1.24%	1.39%	1.44%		1.36%
2/8C	2989				1.32%	2.12%			1.72%
2/8D	2729				2.08%	2.35%			2.22%
2/9A	3930				0.58%	0.79%	0.51%		0.63%
2/9B	3280				1.78%	1.52%	1.63%	1.31%	1.56%
2/10B	3930				0.62%	0.76%	0.64%		0.67%
2/10C	2900				0.72%	0.80%	0.85%		0.79%
2/10D	3670				0.81%	0.69%	0.67%		0.72%
2/CSC	4160				0.84%	2.25%	1.70%		1.60%
avg all		1.25%		3.58%			1.27%		1.44%
Test	Zr (mg)	Respicon	Respicon	Berner	Marple	Marple	Marple	Marple	Test Avg
3/2A	3540				1.09%	0.81%	0.82%		0.909%
3/5B	3900				0.32%	0.61%	0.71%	0.93%	0.640%
3/1C	1490				0.75%	0.39%	0.67%		0.606%
avg all							1.13%	1.10%	1.32%

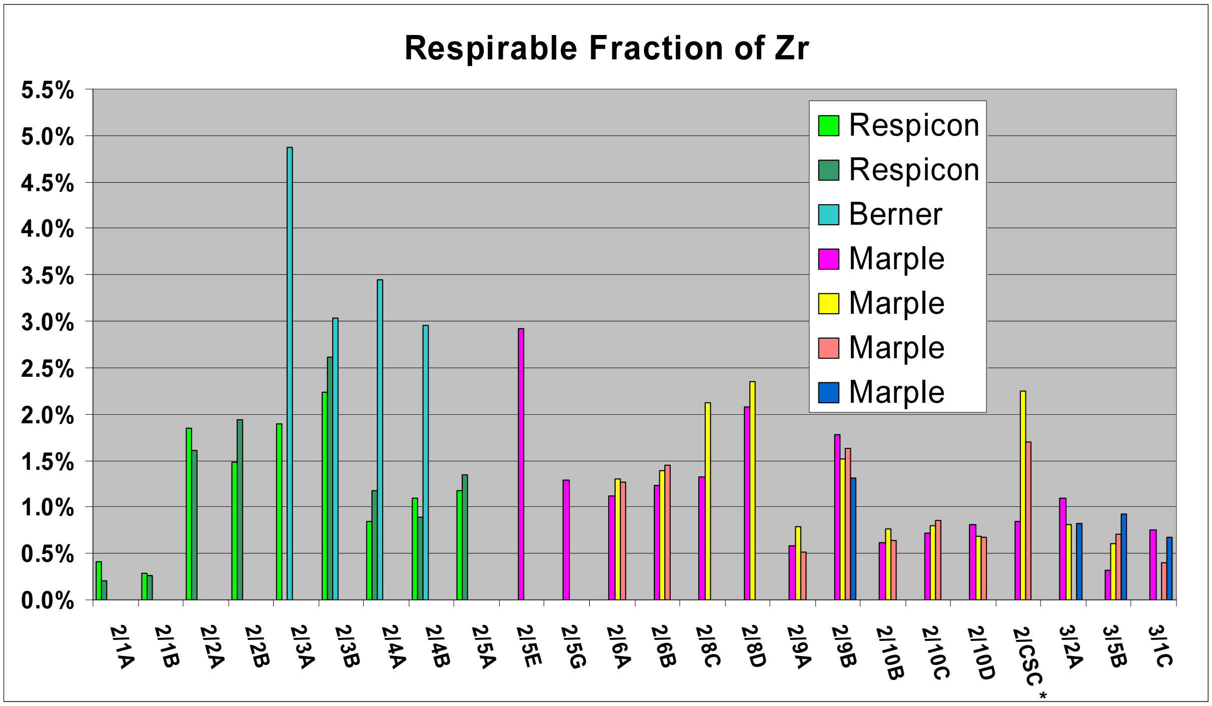


Figure 4.1.2. Respirable Fraction Results for Zirconium

Table 4.1.3. Cesium Dopant Measured Respirable Fraction Results

	CsI Dispersed		Cesium Respirable Fraction (%)								
Test	CsI (mg)	Cs(mg)	Respicon	Respicon	Berner	Berner	Marple	Marple	Marple	Marple	Test Avg
2/4A	32.6	16.7	24.40%	32.39%	10.34%						22.38%
2/4B	30.4	15.5	23.17%	16.91%	11.63%						17.23%
2/5A	31.0	15.9	28.50%	28.22%							28.36%
2/5E	22.6	11.6					42.44%				42.44%
2/5G	18.9	9.7					27.40%				27.40%
2/6A	30.2	15.4					17.16%	20.96%	22.26%		20.13%
2/6B	35.8	18.3					26.72%	28.12%	36.84%		30.56%
2/8C	33.9	17.3					41.88%	72.87%			57.37%
2/8D	33.9	17.3					44.86%	46.42%			45.64%
2/10B	98.8	50.5					16.30%	25.77%	26.84%		22.97%
2/10C	217.0	111.0					14.54%	30.29%	24.10%		22.98%
2/10D	217.0	111.0					24.42%	23.19%	20.39%		22.67%
2/10G					22.85%	23.15%					23.00%
2/10H					4.97%	5.10%					5.04%
2/CSC	118.3	60.5					3.96%	11.21%	12.33%	26.49%	13.50%
avg all			25.60%		13.01%					27.51%	24.85%
Test	CsI (mg)	Cs(mg)	Respicon	Respicon	Berner	Berner	Marple	Marple	Marple	Marple	Test Avg
3/1C	33.9	17.3					53.00%	32.67%	49.45%		45.04%
avg all										29.39%	26.36%

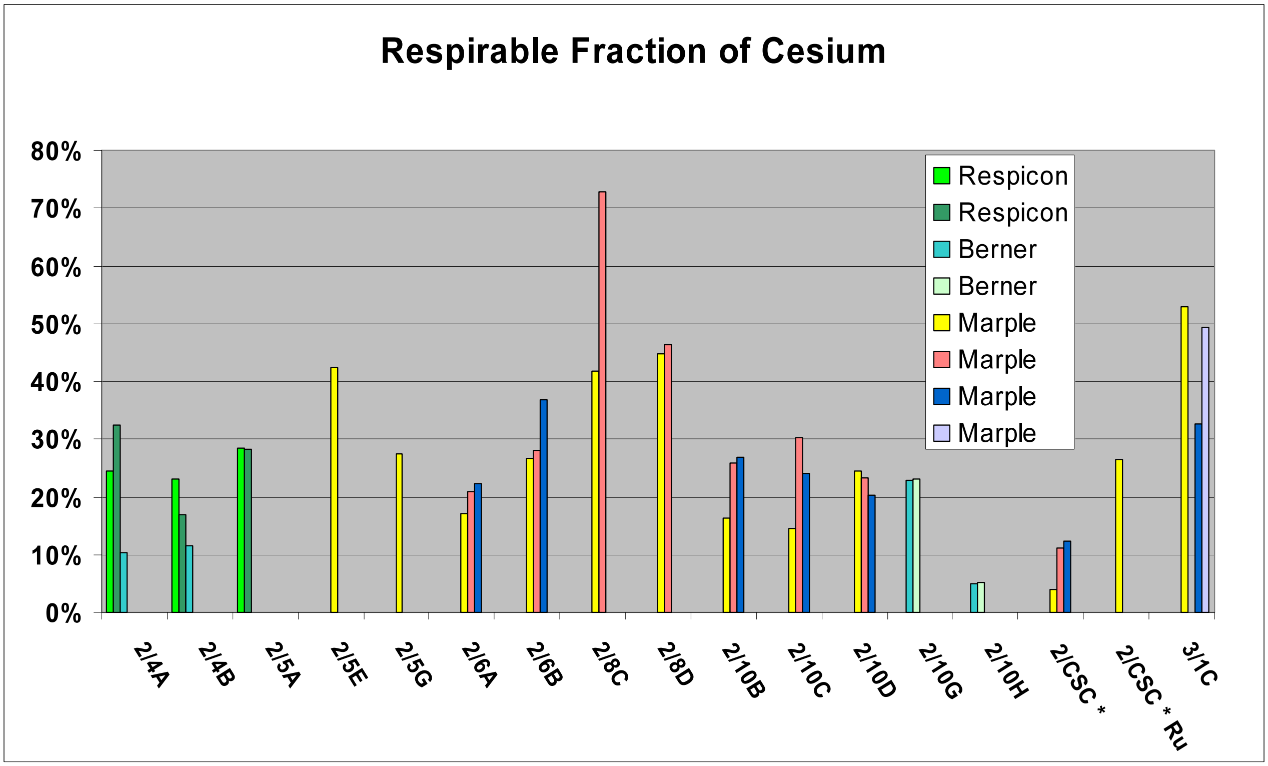


Figure 4.1.3. Respirable Fraction Results for Cesium Dopant

NOTE: Data bars for test “2/10CSC” and “2/10CSC Ru” in Figure 4.1.3 originate from the same, singular test, but from separate Marple samplers, and from different chemical analyses performed by two separate analytical laboratories.

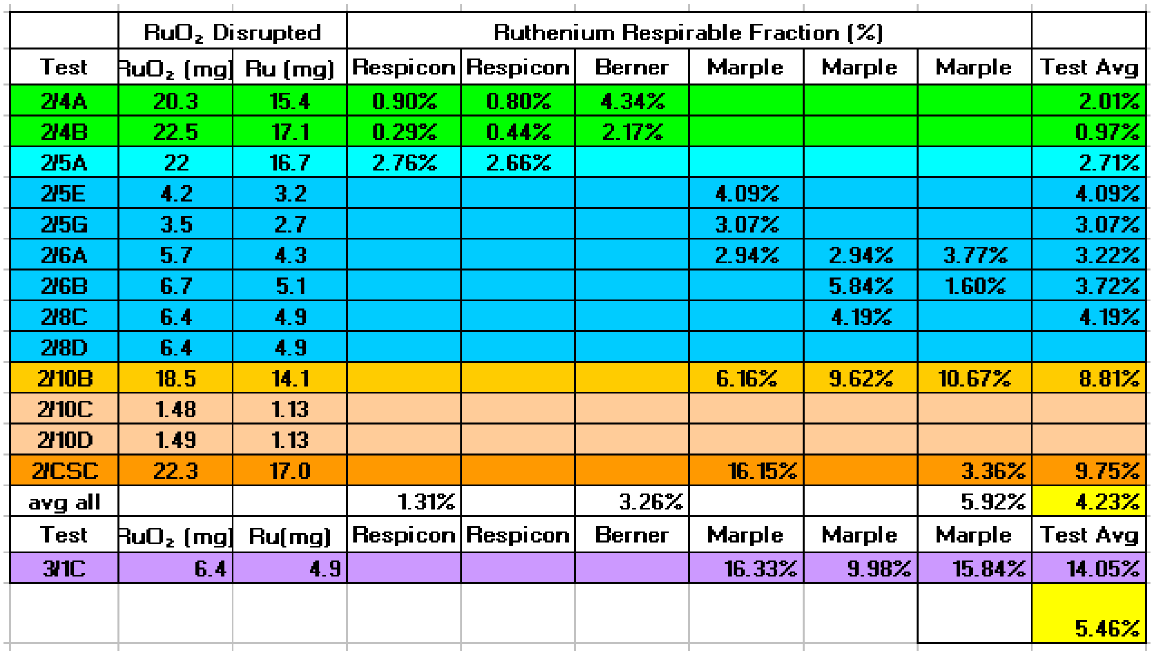
4.1.2.2 Fission Dopant Physical Form: Based on measured RF results shown in Figure 4.1.3, there was *no observed significant difference* in the measured cesium RF values as a function of the physical forms/distributions of the dopant material used, whether: (a) in external, non-homogeneous solid chemical wells (tests 2/4A, 2/4B, 2/5A); (b) or in adjacent resin-based disks (tests 2/5G through 2/8D, 2/10B, 2/CSC, and 3/1C); (c) or with a more nearly homogeneous distribution of these internal dopants within the cerium oxide pellet (2/10C, 10D, 10G, and 10H) - - similar to the situation expected in actual spent fuel, irradiated UO_2 pellets. All of the fission product material in all test cases was within the HEDD jet particulated zone. There are, however, more noticeable differences for measured cesium integrated enrichment factors, IEF, as a function of dopant distribution to be discussed in Section 4.2.

4.1.2.3 Cesium Respirable Size Distributions: From observations of the Marple impactor data in tests 2/10B-10D for cesium respirable particle size distributions (as illustrated in multiple figures in Appendix A [3]), cesium particle or concentration distributions appear to peak over the respirable range size between 0.5 to 3.5 µm AED. As described in Section 3.1.2.3 for test 2/CSC, the observed cesium concentration for this test also peaked at around 3.5 µm AED. This observed minor shift in particle size peak has no impact on the measured RF value; the particle peaks were all within the respirable size range of 0-10 µm AED. The cesium iodide used as a fission dopant in these surrogate material tests melts at 899 ºK (626 ºC) and boils at 1553 ºK. It may undergo phase changes and volatilization when impacted by the hot HEDD jet. It is obvious that thermally volatilized cesium is preferentially found sorbed onto respirable particles, particularly in the 0.5 to 3.5 µm AED size range, correlating well with the distribution of both copper and carbon soot. The close correlation of cesium and copper particle size distributions suggests a parallel vaporization of both materials in the localized hot interaction zone of the HEDD jet and subsequent formation of a copper fume incorporating the volatile elements of target material such as cesium. There is no similar correlation with the particle size distribution of brittle matrix material which presumably has a larger particle size fraction than the hot jet interaction zone. The Cs particle distribution in the aerosol range from about 30 -100 µm AED, collected with the LPS apparatus, is generally highest in the 30 - 48 µm segment, then decreases.

4.1.2.4 Background Interferences: After all the Phase 2+ tests and the first Phase 3 test 3/5 (A) had been performed and the ICP-MS chemistry analyses were received, it became obvious that there were very high anomalous concentrations of barium, boron, and aluminum present (orders of magnitude too high), and a fairly high, almost constant concentration of strontium. The glass fiber filter media used as particle collection substrates in the Marple impactors and the Large Particle Separators presented a problem in that they contained background levels of some of the fission product dopants comparable to those released in the tests. This impurity concentration problem was most notable for Sr but may be in effect for Ru and Eu as well. Measured concentrations of cesium in the particle analyses were appreciably higher and were not affected by interferences. The known, analyzed background concentrations of Sr, Ru, and Eu have been subtracted out in the tables and figures herein, and in evaluations. However, residual higher levels of uncertainty may remain. These problems and solutions (replacing impactor substrate media and background or impurity level subtractions) were discussed in detail in Section 7.4 of SAND2006-5674 [3].

4.1.2.5 Ruthenium: Table 4.1.4 presents all measured respirable fraction results for fission product dopant ruthenium. Figure 4.1.4 illustrates all of these ruthenium RF data. Zero or non-detectable levels of Ru have been ignored; this may result from the low levels of Ru present in the tests and the subsequent low levels in the samples, making detection difficult, giving us a fairly high uncertainty in Ru respirable fraction. The resultant ruthenium respirable fraction average was **Ru RF = 4.2% ± 2.2%** of dispersed mass based on CeO$_2$ target Phase 2 and 2+ collected data (with a 99% CI). Based on the DUO$_2$ Phase 3 data only, the Ru RF average was somewhat higher, Ru RF = 14.1% ± 7.7%. Based on <u>all</u> collected data the Ru RF = 5.5% ± 2.7% of dispersed mass (with a 99% CI). Further uncertainties for these Ru RF values are discussed below.

Table 4.1.4. Ruthenium Dopant Measured Respirable Fraction Results



	RuO$_2$ Disrupted		Ruthenium Respirable Fraction (%)						
Test	RuO$_2$ (mg)	Ru (mg)	Respicon	Respicon	Berner	Marple	Marple	Marple	Test Avg
2/4A	20.3	15.4	0.90%	0.80%	4.34%				2.01%
2/4B	22.5	17.1	0.29%	0.44%	2.17%				0.97%
2/5A	22	16.7	2.76%	2.66%					2.71%
2/5E	4.2	3.2				4.09%			4.09%
2/5G	3.5	2.7				3.07%			3.07%
2/6A	5.7	4.3				2.94%	2.94%	3.77%	3.22%
2/6B	6.7	5.1					5.84%	1.60%	3.72%
2/8C	6.4	4.9					4.19%		4.19%
2/8D	6.4	4.9							
2/10B	18.5	14.1				6.16%	9.62%	10.67%	8.81%
2/10C	1.48	1.13							
2/10D	1.49	1.13							
2/CSC	22.3	17.0				16.15%		3.36%	9.75%
avg all			1.31%		3.26%			5.92%	4.23%
Test	RuO$_2$ (mg)	Ru(mg)	Respicon	Respicon	Berner	Marple	Marple	Marple	Test Avg
3/1C	6.4	4.9				16.33%	9.98%	15.84%	14.05%
									5.46%

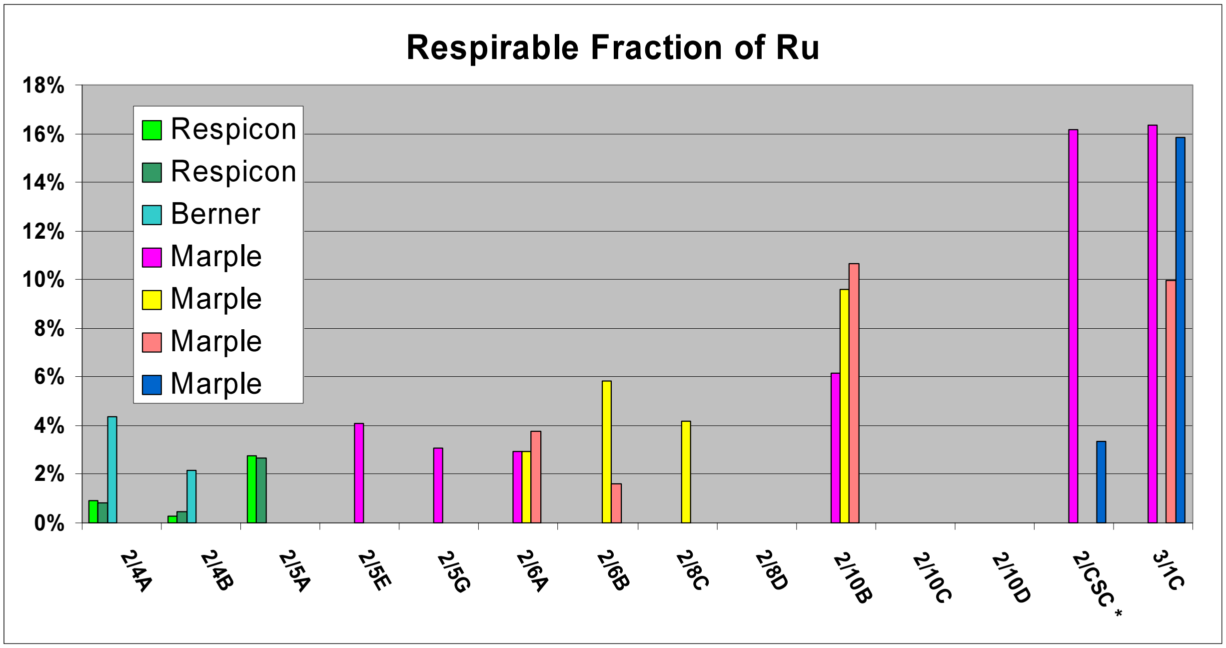


Figure 4.1.4. Respirable Fraction Results for Ruthenium Dopant

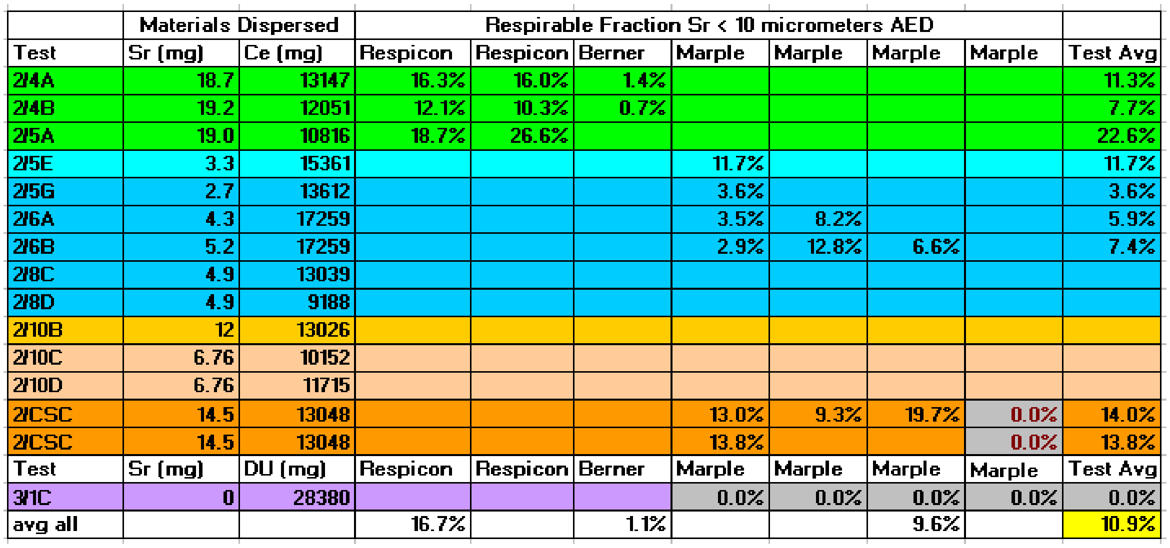
It was originally expected that the volatile ruthenium fission product dopant would behave similarly to the volatile cesium fission product dopant and have similar measured RF values. This was, however, not the case as the Ru RF values were significantly lower than the Cs RF values. Ruthenium and its multiple available oxidation states can form more complex chemical forms than cesium can. Ru metal (used as a dopant in tests 2/10 C, D, G, and H) is not thermally volatile, but it can oxidize to a thermally volatile form. Therefore, Ru RF values may not be directly related or comparable to measured Cs RF values.

Further Ru Uncertainties: There remains a high level of uncertainty in the chemically analyzed and reported Ru amounts present in the collected particles, even with subtraction of known impurity levels; this may be due to differences in Ru dissolution and analysis procedures used by different laboratories, particularly in the case of test 2/CSC. It is also possible that higher levels of Ru were actually incorporated into the internally doped pellets used in tests 2/10C, D, G, and H than has been reported, based on the chemical analysis of a few similar internally doped pellets. The concentrations of Ru detected in particle analyses were in many cases much larger (+50% to +100%) than the amount placed in the test as dopant [3]. As such, further interpretations of the ruthenium data RF values will not be made at present.

Ru Particle Sizes: Ruthenium fission product dopant respirable particle size distributions were quite similar to those for cesium, but at a much lower concentration. Ruthenium particles in test 2/10B, for example, appear to peak over the respirable range size of 1.6 to 3.5 µm AED, although they are found over the entire range of 0 to 10 µm. The ruthenium particle distribution in the aerosol range from about 30 to 100 µm AED, collected with the LPS apparatus, is generally highest in the 30-48 µm size range, then decreases. It seems obvious that the thermally volatilized ruthenium species that did form, similar to the cesium, are preferentially found sorbed onto respirable particles, particularly in the 1.6 to 3.5 µm AED size range, correlating well with the distribution of copper and carbon soot respirable particles.

4.1.2.6 Strontium: Table 4.1.5 presents the integrated respirable fraction results for fission product dopant strontium for all tests to date. Figure 4.1.5 illustrates all these strontium RF data. Zero values or non-detectable levels of Sr have been ignored; this results from the low levels of Sr present in the tests and the subsequent low levels in the samples, making detection difficult, and giving us a fairly high uncertainty in Sr respirable fraction values. Measured background or impurity levels of strontium have also been subtracted. The resultant strontium respirable fraction average was **Sr RF = 10.9% ± 4.1%** with a 99% CI based on all collected data from CeO_2 tests. No useable, reliable Sr data was gathered from the Phase 3 DUO_2 tests. Because of the high levels of strontium in filter substrate media and resultant background subtractions, no further interpretation of fission product strontium aerosol results are provided. The strontium dopant, strontium oxide (SrO), melts at 2693 K and boils at ~3270K; as such, it is unlikely to undergo phase change.

Table 4.1.5. Strontium Dopant Measured Respirable Fraction Results



	Materials Dispersed		Respirable Fraction Sr < 10 micrometers AED							
Test	Sr (mg)	Ce (mg)	Respicon	Respicon	Berner	Marple	Marple	Marple	Marple	Test Avg
2/4A	18.7	13147	16.3%	16.0%	1.4%					11.3%
2/4B	19.2	12051	12.1%	10.3%	0.7%					7.7%
2/5A	19.0	10816	18.7%	26.6%						22.6%
2/5E	3.3	15361				11.7%				11.7%
2/5G	2.7	13612				3.6%				3.6%
2/6A	4.3	17259				3.5%	8.2%			5.9%
2/6B	5.2	17259				2.9%	12.8%	6.6%		7.4%
2/8C	4.9	13039								
2/8D	4.9	9188								
2/10B	12	13026								
2/10C	6.76	10152								
2/10D	6.76	11715								
2/CSC	14.5	13048				13.0%	9.3%	19.7%	0.0%	14.0%
2/CSC	14.5	13048				13.8%			0.0%	13.8%
Test	Sr (mg)	DU (mg)	Respicon	Respicon	Berner	Marple	Marple	Marple	Marple	Test Avg
3/1C	0	28380				0.0%	0.0%	0.0%	0.0%	0.0%
avg all			16.7%		1.1%			9.6%		10.9%

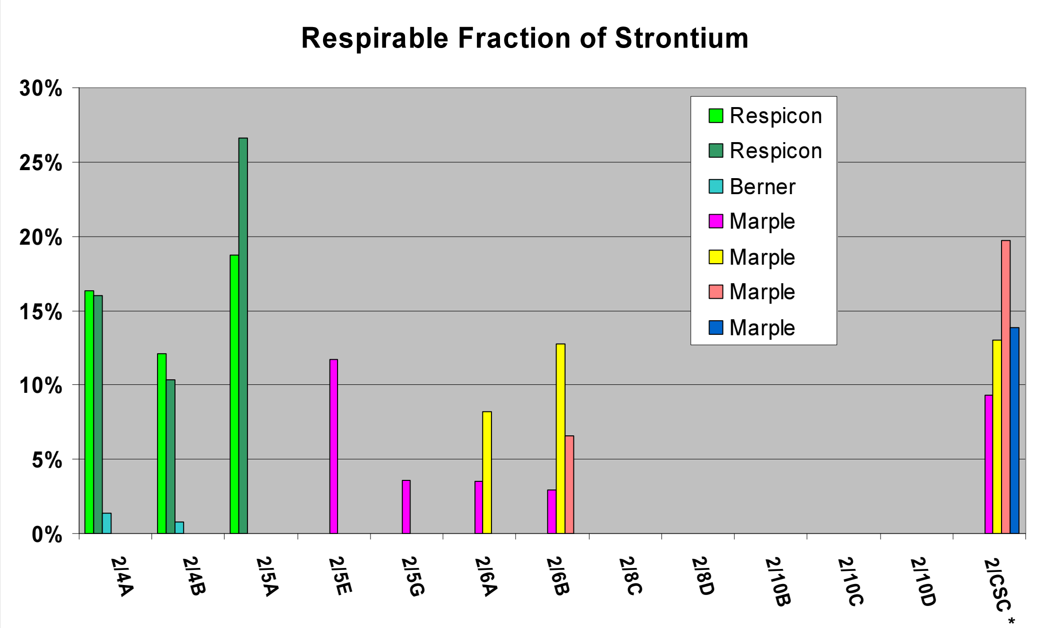
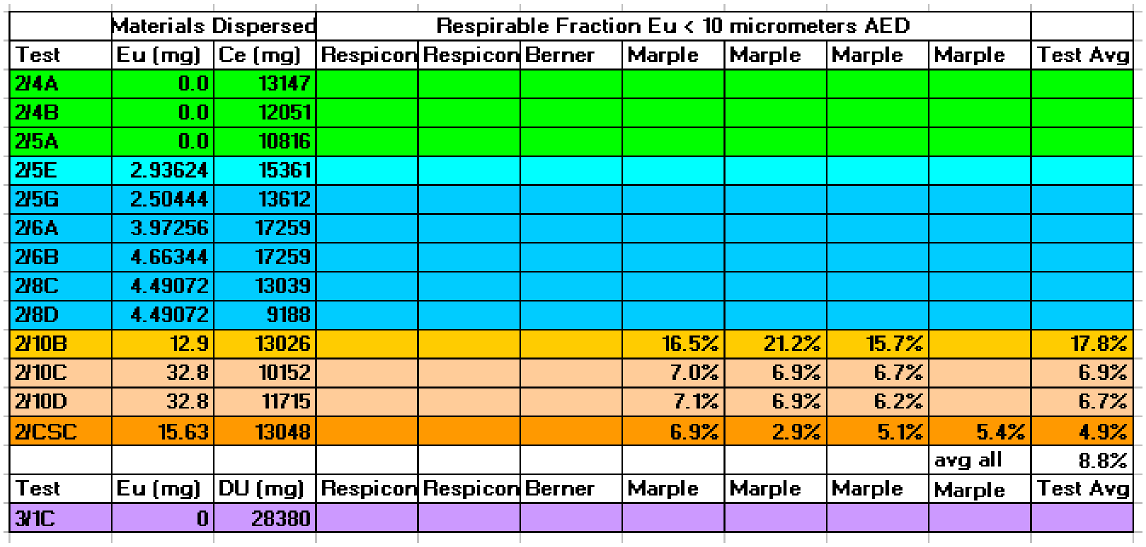


Figure 4.1.5. Respirable Fraction Results for Strontium Dopant

4.1.2.7 Europium: Table 4.1.6 presents the integrated respirable fraction results for fission product dopant europium for all tests to date. Figure 4.1.6 illustrates all these Eu RF data. Zero values or non-detectable levels of Eu have been ignored; this results from the low levels of Eu present in the tests and the subsequent low levels in the samples, making detection difficult, giving us a very high uncertainty in Eu respirable fraction values. The resultant respirable fraction average for Eu was RF = 8.8% ± 3.9% of dispersed mass based on all collected data (with a 99% CI). Since Eu, present as a non-thermally volatile Eu_2O_3 dopant, was detected at only about twice the background level, no further interpretations are made. However, for these tests, there appears to be a slight, broad Eu concentration peak over the 3.5-9.8 µm AED range.

Table 4.1.6. Europium Dopant Measured Respirable Fraction Results



	Materials Dispersed		Respirable Fraction Eu < 10 micrometers AED							
Test	Eu (mg)	Ce (mg)	Respicon	Respicon	Berner	Marple	Marple	Marple	Marple	Test Avg
2/4A	0.0	13147								
2/4B	0.0	12051								
2/5A	0.0	10816								
2/5E	2.93624	15361								
2/5G	2.50444	13612								
2/6A	3.97256	17259								
2/6B	4.66344	17259								
2/8C	4.49072	13039								
2/8D	4.49072	9188								
2/10B	12.9	13026				16.5%	21.2%	15.7%		17.8%
2/10C	32.8	10152				7.0%	6.9%	6.7%		6.9%
2/10D	32.8	11715				7.1%	6.9%	6.2%		6.7%
2/CSC	15.63	13048				6.9%	2.9%	5.1%	5.4%	4.9%
									avg all	8.8%
Test	Eu (mg)	DU (mg)	Respicon	Respicon	Berner	Marple	Marple	Marple	Marple	Test Avg
3/1C	0	28380								

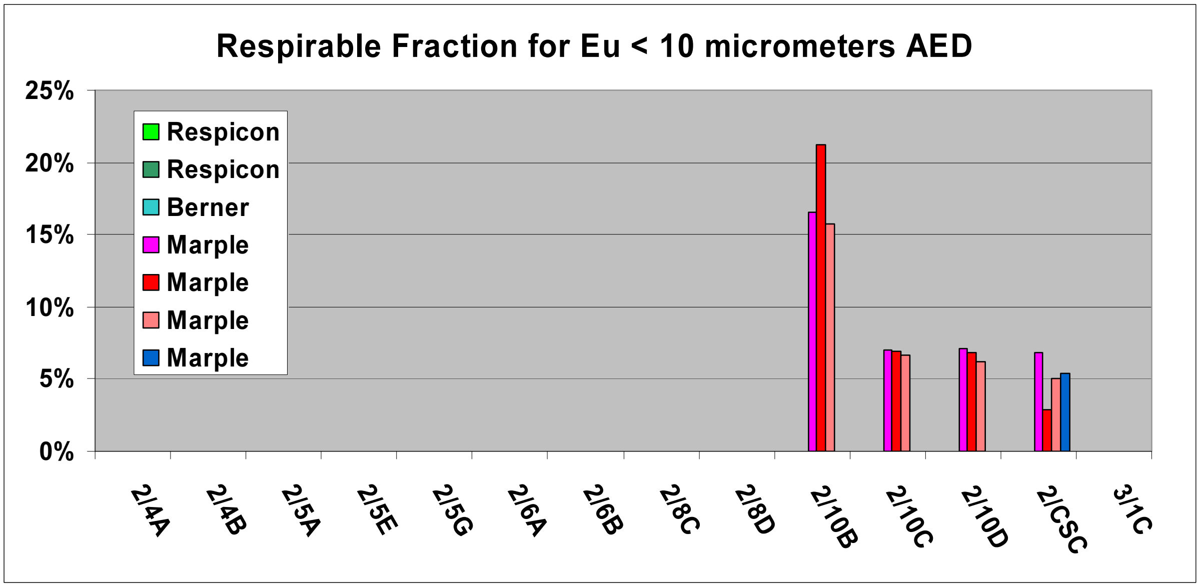


Figure 4.1.6. Respirable Fraction Results for Europium Dopant

4.2 Measured Enrichment Factor Results

4.2.1 Cesium Integrated Enrichment Factor

The measured enrichment factor (IEF) values for cesium fission product, integrated over the respirable size range of 0-10 µm AED, are listed in Table 4.2.1 and shown in Figure 4.2.1. The measured, average IEF for Cs, using all measured data for the CeO_2 Phase 2 and 2+ tests only, the **Cs IEF was 44 ± 17** with a 99% confidence interval (CI) or ± 11 with a 90% CI. (Note: A previously reported Cs IEF value of 369 for test 2/6B has been removed as an obvious outlier point and is not included in current calculations.) The measured, average enrichment factor value for **DUO_2 Phase 3 tests only is Cs IEF = 36 ± 7** at 99% CI or ± 4 at 90% CI. Similarly, using all measured data for the CeO_2 and DUO_2 tests, the total average Cs IEF was 43 ± 16 with a 99% CI or ± 10 with a 90% CI.

The measured cesium IEF values measured for tests 2/7A and 7B and 2/10E, 10F, and 10I, all with cesium dopant within surrogate German glass HLW targets, are also presented in Table 4.2.1. Comparison of data between tests using glass targets and those using CeO_2 pellets must consider the different fragmentation behavior of each as it affects the RF and EF. It is interesting to note that the IEF for the cesium in the glass tests 2/10E, 2/10F, and 2/10I displays similar behavior to the CeO_2 tests, but has much lower EF values for the particles on the order of 2 µm and less (in Section 4.2.2). This may result from differences in fragmentation behavior between the glass and CeO_2 pellets used in the tests.

Note: The aerosol backup filter was damaged in test 2/10I, as described in Section 2.3.1.2. There were no reliable absolute data (Cs RF) available from this test, but the relative results (Cs IEF in Table 4.2.1) are still expected to be reasonable, but to have somewhat limited reliability.

Table 4.2.1. Cesium Dopant Integrated Enrichment Factor Results

	Cesium Integrated Enrichment Factor (IEF)											
Test	Cs (mg)	Ce (mg)	Respicon	Respicon	Berner	Berner	Marple	Marple	Marple	Marple	Test Avg	Test Std
2/4A	33	13147	23.9	20.7	4.5						16.4	10.4
2/4B	30	12051	19.9	17.5	6.8						14.7	7.0
2/5A	31	10816	70.8	69.9							70.4	0.6
2/5E	23	15361					45.6				45.6	
2/5G	19	13612					50.8				50.8	
2/6A	30	17259					101.8	123.0	79.0		101.3	22.0
2/6B	36	17259					132.7	155.8			144.2	0.0
2/8C	34	13039					76.7	104.2			90.4	19.5
2/8D	34	9188					60.8	52.3			56.6	6.0
2/10B	50.5	13026					63.2	30.3	65.9		53.1	19.8
2/10C	104	10152					17.2	22.5	18.8		19.5	2.7
2/10D	133	11715					13.2	13.8	13.3		13.4	0.3
2/10G					13.8	20.1					17.0	4.4
2/10H					14.9	1.1					8.0	9.7
2/CSC	60.5	13048					7.952	6.718	12.711		15.0	12.0
2/CSC Ru	60.5	13048					32.505					
3/1C	17.34	28380					31.162	43.684	36.587	34.130	36.4	5.3
avg all			37.1		10.2		44.0	avg Ce all	36.4	avg DU	43.3	avg all
2/7A		(glass)			167							
2/7B		(glass)			84							
2/10E		(glass)			11.5							
2/10F		(glass)			7.1							
2/10I		(glass)			11.7							

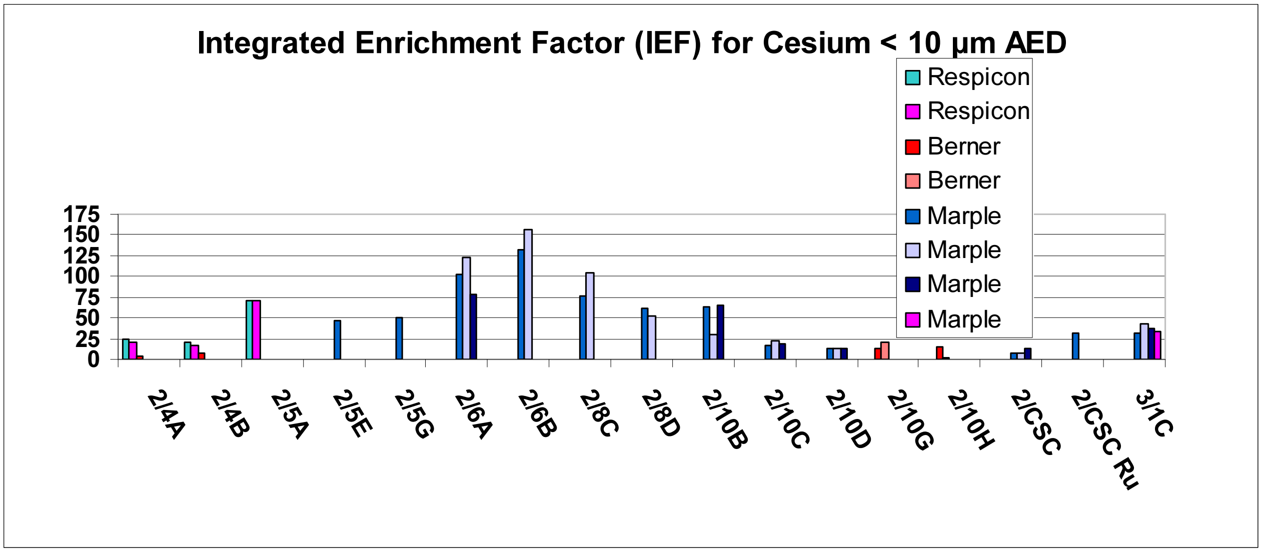


Figure 4.2.1. Enrichment Factor Results for Cesium Dopant

> [NOTE: In Figure 4.2.1, data bars for test 2/10CSC and "2/10CSC Ru" originate from the same, singular test, from multiple Marple samplers, but from different chemical analyses performed by two separate laboratories.]

The integrated Enrichment Factor (IEF) has been defined as equal to the respirable fraction of Cs divided by the respirable fraction of Ce or DU in the pellet material. Since the Enrichment Factor is based on ratios, it is less subject to uncertainties and sampling losses, such as potential thermophoretic and diffusiophoretic deposition effects, than are respirable fraction determinations.

We found a rather large standard deviation for Cs IEF values in the CeO_2 tests, as visible in Figure 4.2.1. These differences can be preliminarily interpreted to be due to higher internal temperatures in fully enclosed test chambers vs. semi-open test systems and/or to differences in distribution of the cesium dopant within the test rodlet/pellets, as discussed below. It is conceivable that actual Cs IEF values may be appreciably lower than reported herein, dependent on differences in test setup. A complementary representation of the cesium IEF data is given in Figure 4.2.2 in an attempt to separate potential influences of temperature and/or cesium dopant distribution.

1. **Test Chamber Temperature Effects:** A few tests (2/10G and 2/10H) in a Fraunhofer semi-open, vertical elutriator chamber (Figure 2.3.3) and in the SNL 50 m^3 test chamber, both with low internal temperatures, all had Cs IEF average values less than 20. Tests 2/6A through 2/10B, and 3/1C, with significantly higher internal temperatures, all have noticeably higher observed Cs IEF values; refer to Table 4.2.1 and Figures 4.2.1 and 4.2.2. There is no similar temperature-related difference observable from the cesium RF measured data between "hotter" and "cooler" test chambers. Tests 2/4A and 2/4B ("square box," Figure 2.2.2, no thermocouples installed) and tests 2/10C and 2/10D also had measured Cs IEF values less than 20, but were performed within a higher temperature test system (refer to Section 2.3.2.3) than in the "cooler" test chamber used for the German test system for glass tests 2/10G and 2/10H (Figure 2.3.3) with low cesium IEF values. Figure 4.2.2 suggests that this temperature difference or effect may be a significant variable affecting Cs IEF values, but it may not be a sufficiently adequate, defensible explanation yet; further study would be required. The limited number of low cesium IEF results (less than 20) measured within test Phase 2+ are supported further by earlier tests conducted in Germany [35]. However, temperatures within an actual spent fuel transport rail cask potentially breached by a HEDD jet sabotage event can actually be at high temperatures, up to about 400 °C, which were reached in the SNL self-contained aerosol chamber tests.

2. **Dopant Distribution Effects:** It appears, for example, that the Cs IEF values for tests 2/10C, 2/10D, 2/10G, and 2/10H, with the CsI dopant thermally diffused through/nearly homogeneously within the base CeO_2 pellets, and tests 2/7B plus 2/10E and 2/10F, with cesium dopant melted within German glass HLW targets, are lower in magnitude compared to Cs IEF values for tests 2/5E through 2/10B, with non-homogeneous, adjacent fission product dopant disks (shown in Figures 4.2.1 and 4.2.2). Cesium IEF values in the recent test 2/CSC with similar adjacent dopant disks in a 50 m^3 test chamber are also similarly lower in value (average Cs IEF = 15). However, this low cesium IEF could also be attributed to the low test temperature within test 2/CSC. The data correlation of the linear fit in Fig 4.2.2 for dopant disk tests increases with addition of test 2/CSC to the data sample. In addition, tests 2/4A and 2/4B, with dopant chemical inserted into discrete non-homogeneous "wells" in pellets also have low measured Cs IEF values, but probably with relative high test temperatures. Such disparate observations confound a clean interpretation of dopant distribution effects.

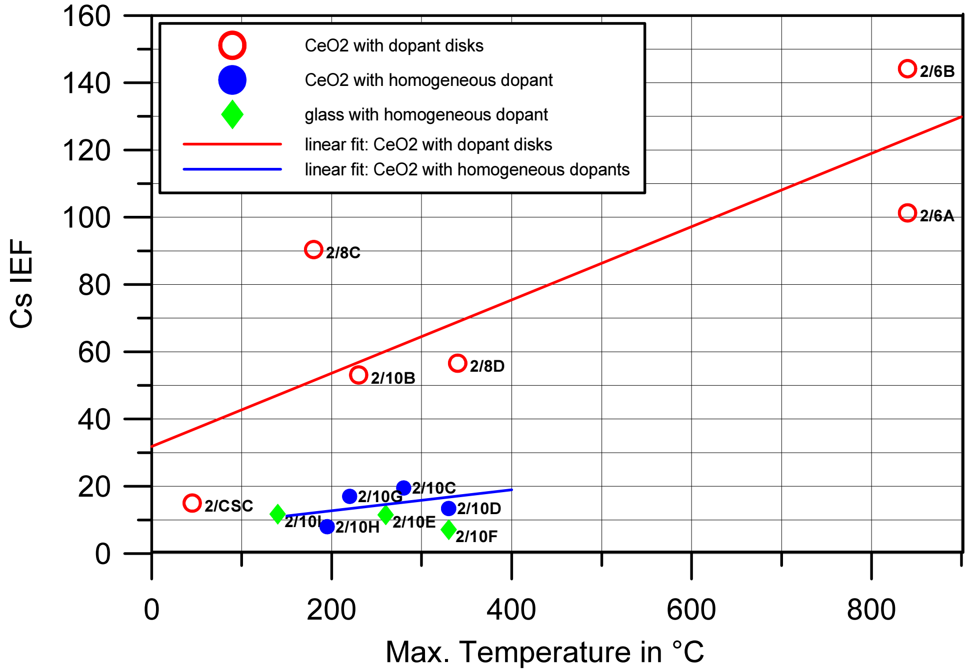


Figure 4.2.2. Temperature Dependence of Cesium IEF Values for different Dopant Distributions and Target Materials

4.2.2 Cesium Differential Enrichment Factor

In the Phase 2+ tests performed both at SNL and Fraunhofer Institut (ITEM and EMI), the differential Enrichment Factor for cesium was determined as a function of particle size at each size range from material collected in the Marple impactor and LPS, and with Berner impactors in Germany. Representative Cs differential EF results from tests 2/8C and 2/8D, 2/10B, 2/10D, 2/10C and 2/10/G, 2/10E and 2/10F (HLW glass pellets), 2/CSC and 2/CSC (linear), and 3/1(C) are illustrated in Figures 4.2.3 -4.2.11. For tests 2/10C and 2/10D for example, the cesium EF was observed to have a multi-modal distribution, falling from an EF of ≥ 100 at ≤ 1 µm AED to ≤ 10 at ~10 µm, then increases slightly to ~10 or above at about 30 to 50 µm before decreasing again toward ~1 or less at larger particle sizes approaching 100 µm. The secondary peak was observed with Marple impactor data up to ~35 µm AED, plus LPS and Berner impactor data, and sieved impact debris at larger sizes. The second maximum needs further investigation as it has been observed in most, but not all tests. Results for test 2/10G were somewhat similar, but slightly lower in magnitude and with the secondary peak at a larger size. However, when the Cs EF data for test 2/CSC are plotted on a linear-log (not log-log) scale in Figure 4.2.10, the dual maxima structure, and its relative significance, is much harder to discern. Similar dual maxima peak trends were observed in almost all other tests performed at SNL, but not as much in tests performed by Fraunhofer. The cesium EF data for the DUO_2 tests, Figure 4.2.11 are not as pronounced, having a flatter decrease with increasing size.

Besides the credible explanations on the effects of higher internal test chamber temperatures impacting measured EF values, as mentioned previously, there may be two other effects going on in regards to the significant variations observed in cesium EF values. There is reason to believe that the mechanism producing the cesium particle distribution (Cs vaporization and subsequent condensation on the smaller sized particles, most probably soot) is not necessarily related to that producing the cerium, surrogate material particle distribution (mechanical shock and fragmentation of a brittle material into small particles). While the EF is a measurement that can be made with good precision in a given size range (the sampling efficiencies cancel nicely), the value in a given test can vary considerably. This is true especially since the mechanical fragmentation particles in the small size range are the tail of the larger distribution.

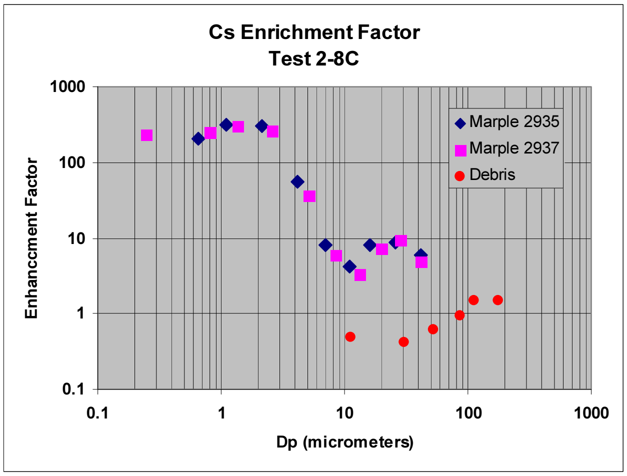


Figure 4.2.3. Differential Cesium EF, Test 2/8C

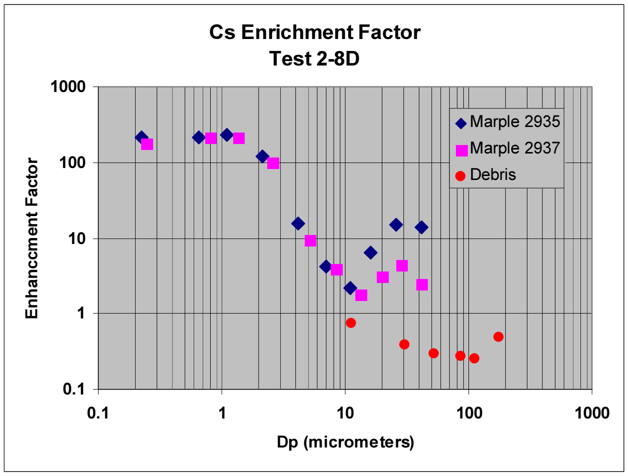


Figure 4.2.4. Differential Cesium EF, Test 2/8D

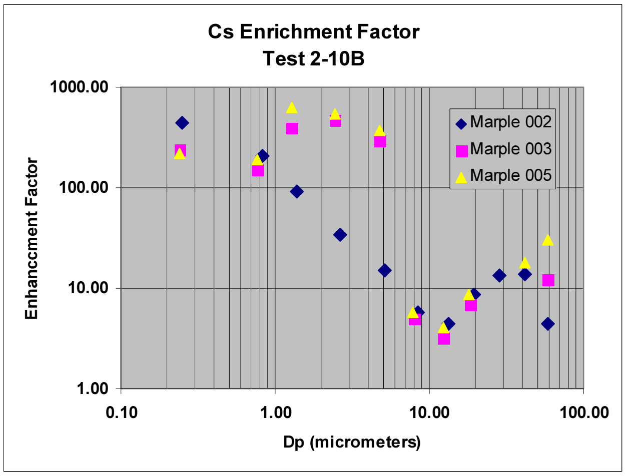


Figure 4.2.5. Differential Cesium EF, Test 2/10B

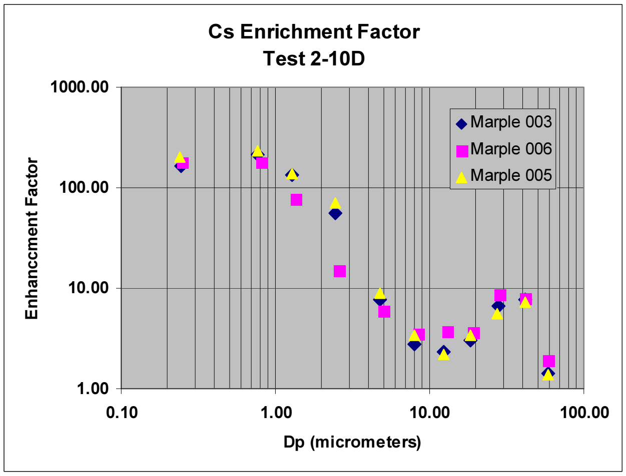


Figure 4.2.6. Differential Cesium EF, Test 2/10D

* Dp = Diameter of particle, mid-point.

While the larger particle distribution can be very repeatable, the tails can vary considerably. Additional analysis of fragmentation behavior and debris distribution may be worthwhile to ascertain the variation in the particle mass contained in the tails of the fragmented pellets. Consider that the mass fraction of cerium oxide less than 10 µm AED is only a few percent of the fragmented cerium oxide and that it will vary from test to test as we have seen and discussed above. Slight changes in the CeO_2 particle distribution can produce considerable changes in the distribution in the small tail of the distribution. This variation, coupled with a separate source of variation in the soot distribution and amount of Cs available gives two separate particle distributions that we then ratio to get the Cs EF. Given all these sources of variability in EF, it is somewhat remarkable that we see such similarity in the size dependent, differential EF plots.

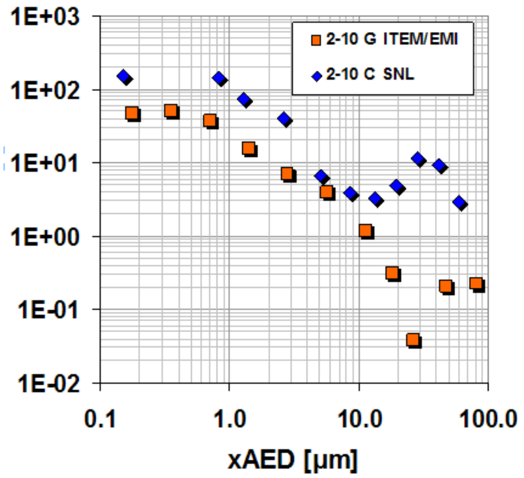


Figure 4.2.7. Differential Cesium EF, Test 2/10C & 2/10G

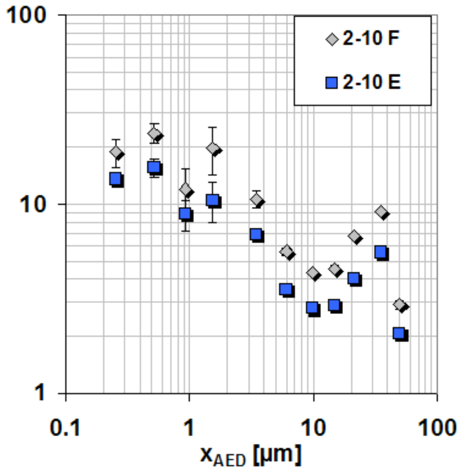


Figure 4.2.8. Differential Cesium EF, Test 2/10E & 2/10F (HLW glass)

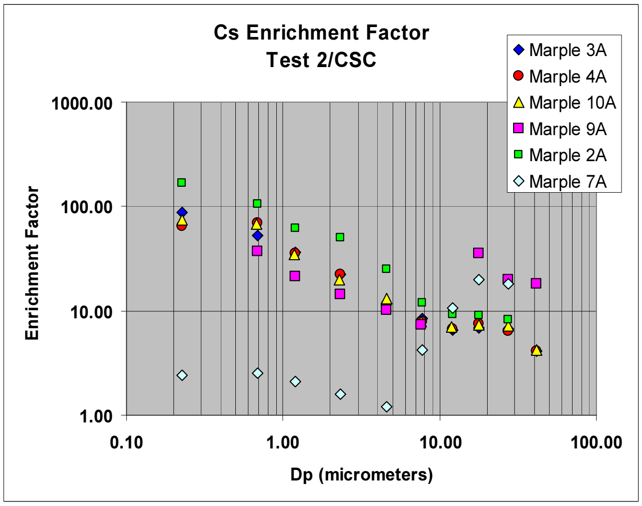


Figure 4.2.9. Differential Cesium EF, Test 2/CSC

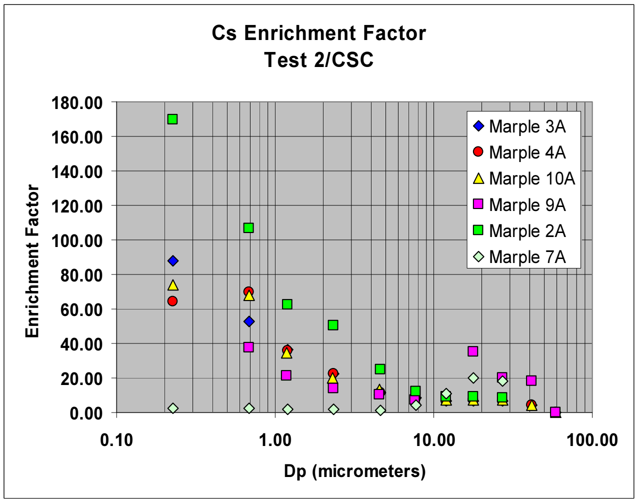


Figure 4.2.10. Differential Cesium EF, Test 2/CSC (linear-log scale)

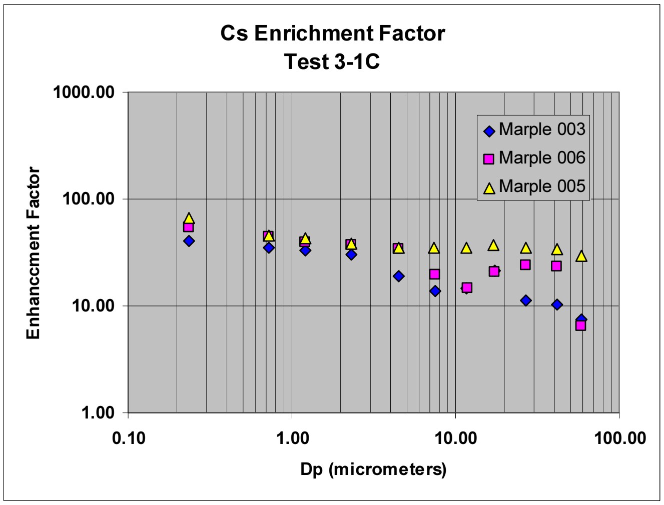


Figure 4.2.11. Differential Cesium EF, Test 3/1C

There are a few previous, historical cesium EF values measured from actual spent fuel pellet tests available, as described in Section 1.3, for comparison to the Cs EF values presented above. Researchers at Battelle Columbus Laboratory [10] reported a Cs EF value of ~ 16, for "sub-micron" particles, while INEL [11] researchers reports Cs EF values of ~56 for < 0.2 μm AED particles and ~35 for < 1 μm AED particles. These values are significantly lower than the comparable Cs EF values in the range of ~ 120 to 500 measured for ≤1 μm particles in tests 2/8C, 2/8D, 2/10B, 2/10C, and 2/10D (all performed in high internal chamber temperature tests) and Cs EF values of ~ 9 to 90 for ≤1 μm particles measured in tests 2/10F, 2/10G, and 2/CSC (all performed in "low" temperature test chambers). These data provide some supporting results that there is a temperature related effect on Cesium EF values; however, the internal temperatures within earlier INL and BCL test systems is not known. Cesium EF measured values of ~ 60 to 80 for ≤1 μm particles in the high- internal test chamber for test 3/1C with doped DUO$_2$ pellets again makes the effects of temperature on enrichment factors more difficult to establish.

4.2.3 Other Fission Products Integrated and Differential EF Values

The measured enrichment factor (IEF) values for ruthenium, strontium, and europium fission products, integrated over the respirable size range of 0-10 μm AED, are shown in Figures 4.2.12 through 4.2.14, respectively. The measured, average IEF for Ru using all measured data (excluding zero or non-detected values) was Ru IEF = 10.2 ± 4.9 with a 99% confidence interval (CI). Similarly, measured, average IEF for strontium was Sr IEF = 108 ± 86 with a 99% CI, and for europium, Eu IEF = 12 ± 11 with a 99% CI. As described in Section 4.1.2 and Section 7.4 in [3], there were significant impurity concentration problems and background interferences inherent in the impactor media used, for Ru, Sr, and Eu; concentration corrections were applied. In addition, the fission product dopant concentrations used for Ru, Sr, and Eu were appreciably lower than for cesium dopant. As such, there *are significant residual uncertainties* for the fission product Ru, Sr, and Eu IEF values presented. No further interpretations of these data can be presented at this time.

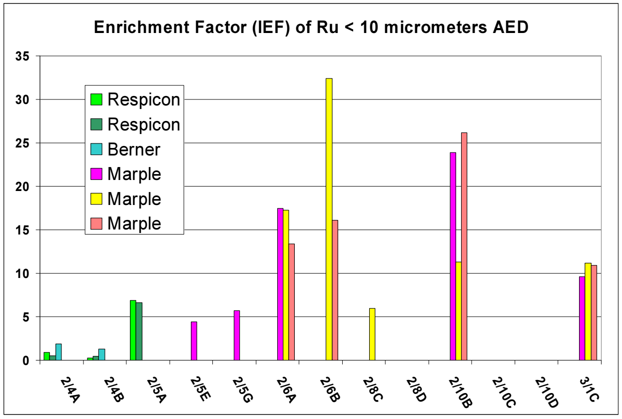


Figure 4.2.12. Ruthenium IEF Results

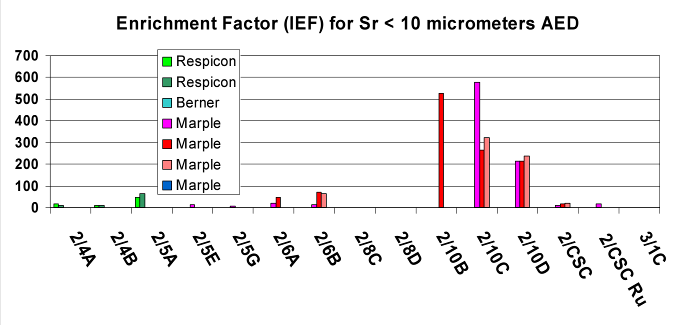


Figure 4.2.13. Strontium IEF Results

Figure 4.2.15 illustrates the test 2/CSC measured europium differential EF values. Figures 4.2.16 and 4.2.17 provide similar test 2/CSC measured strontium and europium differential EF values, respectively. Finally, Figure 4.2.18 shows Eu differential EF results for test 2/10C (by SNL) and 2/10G (by Fraunhofer ITEM and EMI).

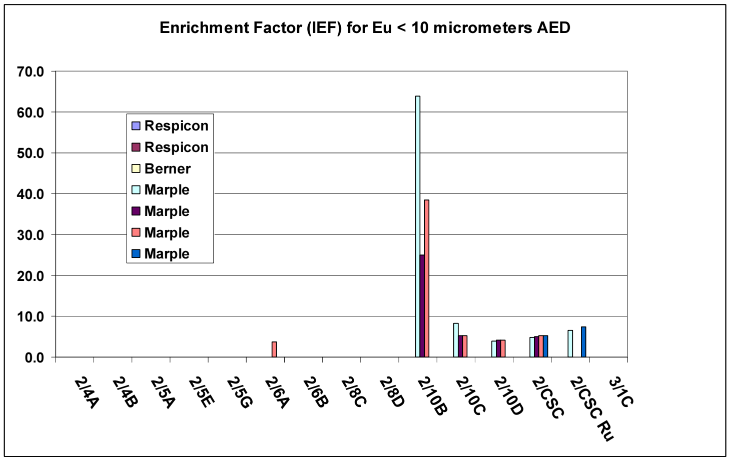


Figure 4.2.14. Europium IEF Results

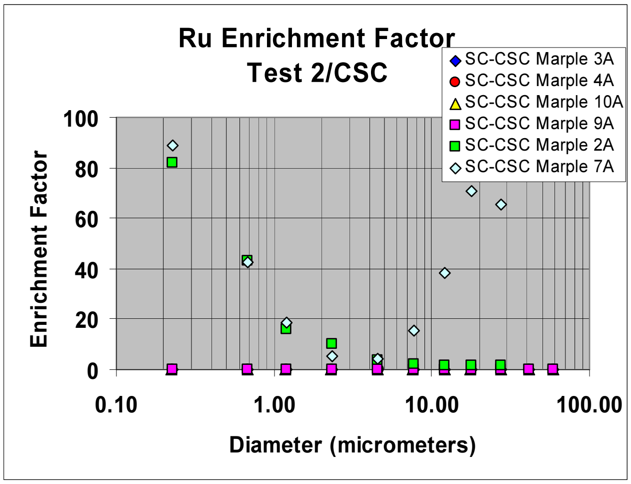


Figure 4.2.15. Ru Differential EF Results (linear), Test 2/CSC

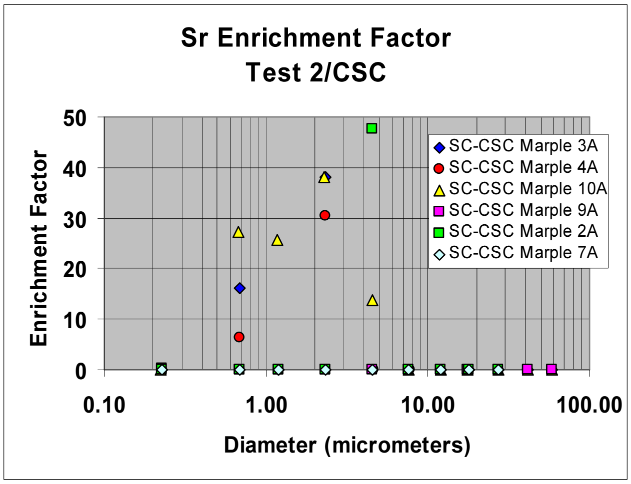


Figure 4.2.16. Sr Differential EF Results (linear), Test 2/CSC

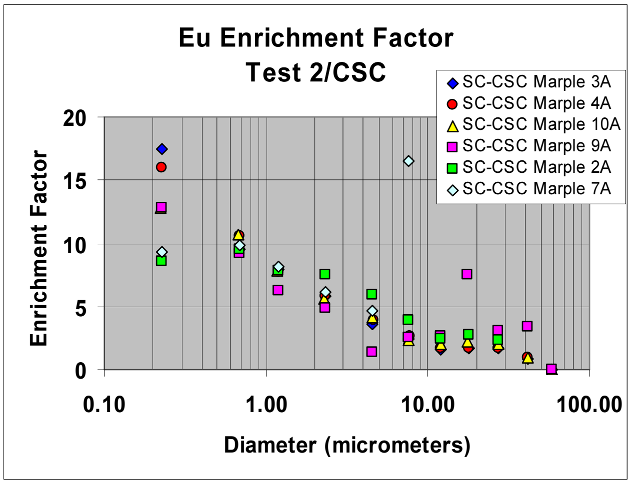


Figure 4.2.17. Eu Differential EF Results (linear), Test 2/CSC

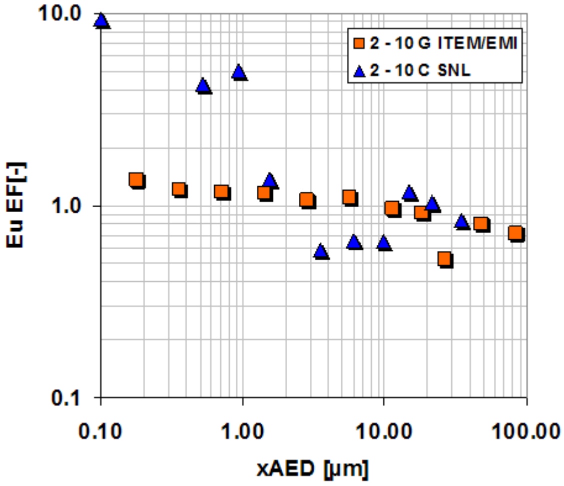


Figure 4.2.18. Eu Differential EF Results, Test 2/10C and 2/10G

5. Test Program Summary, Findings, and Related Items

The casks used for spent nuclear fuel transport and/or storage are extremely resistant to releasing any significant fraction of their contents, even in very severe accident conditions. However, concerns about the possibility of radiological sabotage attacks against spent nuclear fuel shipping and storage casks have been rekindled in recent years, both within the United States and elsewhere. This increased interest is particularly due to: the proposed increase in transport of spent fuel, e.g., to the planned Yucca Mountain Repository or other national or international facilities; increased number of proposed independent spent fuel storage installations; plus, the increased availability of explosives technology and materials. In some intentional, plausible sabotage scenarios, such as an attack employing explosive armor-piercing weapons, i.e., a high energy density device (HEDD), it is possible that a cask could be penetrated. Then, a small percentage of aerosolized particles produced within from disrupted fuel rod and pellet materials could be released as a radiological inhalation dispersal hazard. If released to the environment in a significant quantity, the spent fuel respirable particles have the potential to cause radiological consequences.

Measurements of the actual amounts, nuclide content, and size distributions of the particles produced from spent fuel and related surrogate materials are essential for predicting the significance of aerosol releases from the cask and their potential radiological inhalation impacts. These measurements have been the motivation or driving force behind the overall spent fuel sabotage aerosol measurement program described in this technical report. These aerosol source-term data are the necessary input information for follow-on modeling studies needed to quantify respirable hazards, associated radiological risk assessments, vulnerability assessments, and potential cask physical protection design or safeguards modifications. The need for accurately quantifying this aerosol information has been strongly supported by program participants in the U.S., Germany, France, and others, as described in Section 1.2, as part of the International Working Group for Sabotage Concerns of Transport and Storage Casks. WGSTSC partners need, and helped coordinate this research and subsequent modeling plus assessments, all relevant to radiological sabotage dispersal events. WGSTSC partners have provided cooperative design inputs, participation, and supplemental testing to the overall program effort.

This multinational, multiphase spent fuel sabotage test program has been performed primarily at Sandia National Laboratories since 2001, under closely controlled and simplified experimental conditions -- as required for ease of interpretation. Test results to date have quantified and characterized aerosol particles produced when the products of a HEDD interact with and explosively particulate test rodlets that contain pellets of either surrogate materials (cerium oxide, depleted uranium oxide, German HLW glass) or actual spent fuel. We also compare our results to existing earlier, very limited aerosol results [5-11], as summarized in Section 1.2.

The primary purpose of this comprehensive, interim final technical report is to present a summary of test program details and to document all major results and interpretations obtained since test program initiation through 2007, for use in on-going and future assessments. Measured aerosol results include: respirable fractions produced; produced particle size distributions and morphology; measurements of volatile fission product species enhanced sorption – enrichment factors onto respirable particles; and, status on determination of the spent fuel ratio, SFR, needed for scaling studies. This report also serves as a “place holder” until testing at Sandia National Laboratories can be reinitiated in the future, following the current program suspension, as program funding permits. Further, planned testing activities of the overall WGSTSC program are discussed in Section 5.2.

This current "simplified single short-rodlet" testing portion of the overall WGSTSC program, as described in Section 2, has been designed to provide reliable, quantified source-term input data to parallel follow-on modeling efforts of near-field aerosol release and dispersion of respirables and aerosols to the environment, including computational fluid dynamics, and radiological consequence assessments. Several modeling studies have been initiated to tie these WGSTSC test programs and evaluations, both previous [2-5] and expected to-be-performed in the future, together; this is discussed further in Section 5.4.

5.1 Current Test Program Status, Results, and Findings

Over the last two years, we have performed three Phase 3 tests using slightly radioactive, non-irradiated depleted uranium oxide pellet rodlets, as summarized in section 2.5, plus one surrogate CeO_2 test in a separate 50 m^3 test chamber, as described in Section 3.1. Results from the Phase 3 DUO_2 tests are required to determine the spent fuel ratio, along with results from the still to-be-performed Phase 4 tests with actual spent fuel rodlets, as presented in Section 2.6. Some progress has been made to initiate the Phase 4 tests at SNL, and formal DOE SSO approval *has been received* to use the Sandia GIF facility for this explosive-aerosol testing. Several technical and administrative actions remain before this testing can continue, and were described in Section 2.6.3.

We have also analyzed and completed interpretation of a large body of aerosol data presented within this technical report for the purpose of inter-comparisons of respirable fraction and integrated enrichment factor measured data obtained under slightly different, planned test conditions (test chambers, internal temperatures, homogeneity of fission product dopant distribution, etc.), and with the intent of reducing uncertainties. Table 5.1 presents a summary of the respirable fraction and integrated enrichment values measured in this test program, as described primarily in Sections 3 and 4, along with calculated confidence interval (CI) uncertainties, plus relevant comments. These RF and IEF values are recommended for use in subsequent respirable-aerosol particle modeling studies and for use in future radiological dispersal and consequence assessments.

Table 5.1. Spent Fuel Sabotage Program Aerosol Results

Parameter	Measured Value	99% CI	90% CI	Comments
CeO_2 RF $_{(best)}$	**0.65%**	± 0.23%	± 0.15%	based on Marple data only, 32 measurements
CeO_2 RF $_{(all\ data)}$	**1.36%**	± 0.50%	± 0.32%	based on all data, 57 measurements
DUO_2 RF	**1.32%**	± 0.32%	± 0.21%	based on 10 measurements in Phase 3
Spent Fuel RF	**- TBD -**			(requires conduct of Phase 4 tests)
SFR	**- TBD -**			(requires conduct of Phase 4 tests)
Zr RF	**1.32%**	± 0.29%		based on all data, 59 measurements
Cs RF $_{(CeO_2)}$	**24.9%**	± 5.6%	± 3.6%	based on CeO_2 tests, 37 measurements
Cs RF $_{(DUO_2)}$	**45.0%**	± 16.1%	± 10.3%	based on one DUO_2 test, 3 measurements
Ru RF $_{(CeO_2)}$	**4.2%**	± 2.2%		based on all CeO_2 tests, 21 measurements
Ru RF $_{(DUO_2)}$	**14.1%**	± 7.7%		based on one DUO_2 test, 3 measurements
Sr RF	**10.9%**	± 4.1%		based on CeO_2 tests, 19 measurements
Eu RF	**8.8%**	± 3.9%		based on CeO_2 tests, 13 measurements
Cs IEF $_{(CeO_2)}$	**44**	± 17	± 11	based on CeO_2 tests, 36 measurements; BUT, may vary as function of temp. or dopant form
Cs IEF $_{(DUO_2)}$	**36**	± 7	± 4	based on one DUO_2 test, 4 measurements
Ru IEF	**10.2**	± 4.9		* based on all data, 22 measurements
Sr IEF	**108**	± 86		* based on CeO_2 tests, 24 measurements
Eu IEF	**12**	± 11		* based on CeO_2 tests, 16 measurements
				* high resid.uncertainties, background subtractions

The respirable fraction (RF) values measured in this program for surrogate CeO_2 and DUO_2 tests are quite similar, CeO_2 RF = 0.65% ± 0.23% (or 1.36% ± 0.23%) or DUO_2 RF = 1.32% ± 0.32%, respectively, all at the 99% CI. Our measured RF values are for respirables *produced* within a test chamber, not the realistically much smaller amounts *released* [5] to the environment through a hole in a sabotage-damaged cask. There are very few earlier measured or estimated RF values available for comparison to our results. In an evaluation documented in 2001, Luna et al. [13] derived RF values from earlier (1980s and 1990s) large-scale, transport cask-relevant SNL [6] and GRS [5] DUO_2 tests, with measured releases of respirable particle from HEDD-damaged casks. These calculated surrogate DUO_2 RF values [13] ranged from 1.7 - 5.3% for one-wall cask penetration, or 2.8 - 12% for two-wall cask penetration. The measured surrogate CeO_2 and DUO_2 RF values from the SNL-WGSTSC test program are appreciably below, but not inconsistent with, the 5% RF value for spent fuel estimated in an earlier repository related transportation-sabotage analysis [5]. This suggests that the estimated respirable particle release predicted in that earlier analysis is likely to be appreciably conservative, i.e., the radiological consequences of a sabotage event on a spent fuel transport cask may not be as significant. Further results from the Phase 4 spent fuel tests, plus future consequence assessment calculations are still needed to confirm this inference and to successfully complete this program.

In addition, our measured fission product cesium respirable fraction average was Cs RF = 24.9% ± 5.6% of dispersed mass based on CeO_2 target data, and RF = 45.0% ± 16.1% based on the DUO_2 Phase 3 data only, both with a 99% confidence interval. Both of the cesium RF values are significantly greater than the measured RF values for the surrogate CeO_2 and DUO_2 pellet matrix materials; this indicates significant enrichment or enhanced sorption of the volatile fission product dispersed onto adjacent respirable particles. The measured integrated enrichment factor for cesium was Cs IEF = 44 ± 17 based on CeO_2 target data and Cs IEF = 36 ± 7 based on the DUO_2 Phase 3 data only. Several tests with surrogate CeO_2 and German HLW glass targets measured at both Sandia and Fraunhofer yielded Cesium IEF values below about 20. The effects of lower test chamber internal temperatures and fission product dopant distribution within the test system have been suggested, and described in Section 4.2.1 to explain the lower measured Cs IEF values; however, these interpretations are not conclusive and are still under evaluation among test partners.

Differential cesium enrichment factors as a function of particle size have also been measured and presented in section 4.4.2. In many CeO_2 tests performed within a sealed, high-temperature test chamber, at respirable particle sizes of ≤ 1 μm AED, Cs EF values ranged from ~120 to 500; within the lower-temperature test chambers, similar measured Cs EF values ranged from ~10 to 80. The Cs EF value at ≤ 1 μm AED for the Phase 3 DUO_2 test ranged from ~60 to 80. Previous, small-scale explosive-aerosol tests performed in the 1980s at Idaho National Engineering Laboratory [11] and at Battelle Columbus Laboratory [10] on actual spent fuel pellets, as described in Section 1.3, yielded cesium EF values of ~16 and ruthenium EF values of ~ 5, both in the "sub-μm" range [11]. For the Battelle tests [10], EF values were also measured for "sub-μm" respirables < 0.2 μm, cesium EF = ~ 56, antimony EF = ~ 11, and ruthenium EF = ~16; similarly, for respirables < 1 μm AED, cesium EF = ~35 and ruthenium EF = ~10. We also measured RF and IEF values for fission product dopants ruthenium, strontium, and europium, as summarized in Sections 4.1 and 4.2, and in Table 5.1, above.

Summary of Significant Findings: Some of the more important observations and conclusions described in Section 4, and to a more limited extent, Sections 2 and 3, are summarized as follows.

1. Sintered pellets of cerium oxide, CeO_2, behave as a representative brittle material and serve as a satisfactory chemical and ceramic surrogate for UO_2 fuel and spent fuel materials.
2. The HEDD (CSC1) used throughout this test program behaved in a very consistent and precise manner. The observed amounts of target rodlets' fragmentation/particulation (e.g., the swept volume) were very consistent.
3. The explosive-aerosolization of surrogate and slightly radioactive targets can be safely and totally confined (self-contained) within the vertical test chamber (Phase 3 and 4 versions) and attached aerosol apparatus developed in this program. The Phase 4 test system is nearly identical.
4. No significant stratification of respirable and aerosol particles below about 20 µm AED occurred in the test apparatus and during the sampling periods used, in this program.
5. The presence of significant amounts of soot particles from the confined HEDD detonation was observed to somewhat shift particle size distributions within the respirable range, but has a minimal effect on the overall respirable fractions measured.
6. Testing results are repeatable, but given that we are trying to capture the tail of a distribution (e.g., primarily the respirable particles in the 0-10 µm AED range), uncertainties are high.
7. Based on our measured RF values for the pellet matrix material (CeO_2 and DUO_2), available earlier RF results and estimated values for UO_2 obtained from limited prior tests and subsequent release evaluations were overly conservative, i.e., they were too high in value.
8. Internal rod pressurization in the plenum area of short test rodlets used in this program does not significantly affect the measured respirable fraction values.
9. Measured RF values were not significantly affected by variations in internal test chamber temperatures or the physical form (homogeneity) of the fission product dopants. However, it is possible that the measured integrated enrichment value (IEF) for cesium may vary as a function of internal temperature and dopant homogeneity; Cs IEF results are still being evaluated.
10. The measured high RF values for dopant fission product cesium (and other fission products, in a less definitive manner), and high observed IEF values are a clear indication that a significant amount of the cesium is both mechanically particulated and thermally volatilized. That vapor then preferentially sorbs and is enriched onto adjacent respirable particles of particulated fuel or surrogate pellet materials, copper particles from the HEDD jet, soot particles, and other jet-impacted hardware or cask materials.

Particle Deposition Uncertainties: Observed particle deposition in the sampling lines in the vertical explosive-aerosol test chamber, due presumably to thermophoretic, diffusiophoretic, and turbulence effects, can add some uncertainty or underestimation to measured Respirable Fraction (RF) values. We initiated an aerosol apparatus laboratory calibration effort, described in Section 3.2, to quantify this particle deposition; however, this effort was not satisfactorily completed. We were, however, able to assess particle composition data from these tests that that were completed. These data indicate that most of the material deposited in the sample lines between the isolation ball valve and the LPS reflects the composition found in the larger size particles rather than that of the respirable < 10 µm AED particles. This suggests that the observed material deposited in the sampling lines is predominantly larger particles and most probably deposited by turbulence. The effect of this larger particle deposition on measured respirable fraction values, and their uncertainties is, therefore, presumed to be limited. Potential techniques for minimization of turbulence within the self-containment, explosive-aerosol test chamber system do not appear to be readily achievable, particularly at this late stage of the total test program.

In addition, consistent results from test 2/CSC in the well-calibrated 50 m^3 test chamber suggest that explosively generated soot, plus higher internal temperatures and pressures within the vertical test chamber do not have a significant impact on the source term data, except possibly for the cesium IEF values, with uncertainties probably noticeably less than a factor of two. The estimated uncertainty in measure RF values is probably on the order of ±20%, or less than calculated and reported confidence intervals. The consistency of the measured RF and IEF values between the vertical chamber tests and the 50 m^3 chamber test strongly suggest that the particle line losses observed in the vertical chamber tests [3] are most likely to be from larger aerosol particles that most probably have some sort of inertial deposition mechanism such as turbulence or unsteady flow.

11. This consistency observation, above, from test 2/CSC is also a significant finding.

5.2 Potential Future Testing

5.2.1 Importance of Phase 4 Tests, SFR

As stated earlier in this report, the major goal of this program is the measurement of a more accurate and precise value for the Spent Fuel Ratio for respirable particles. This SFR determination requires performance of explosive-aerosol testing of actual spent fuel in Phase 4 tests, in comparison to parallel testing with the primary “surrogate” material, unirradiated depleted uranium oxide in the Phase 3 tests. Phase 4 testing is the culmination of this entire spent fuel sabotage program effort in support of cooperative U.S. DOE, NRC, and International WGSTSC participant’s goals and data needs. Reliably measured SFR values are needed to permit scaling to other geometries, from a simplified single short fuel rodlets as tested in Phase 4, to rod bundles in casks, by means of supporting modeling studies. The measured SFR values, available after the completion of Phase 4, will provide a better data bridge to previous large-scale surrogate (DUO_2) explosive-aerosol cask tests [5-6] performed earlier in both the U.S. and Germany, and to future evaluations and consequence assessments. Measurements of respirable fraction values for actual spent fuel and RF and EF values for multiple fission products therein, eliminate most of the experimental *simulations* necessitated by using surrogate and dopant materials in the earlier test phases. Actual, *not surrogate* (supporting and “next best available”) values will be used in subsequent modeling and aerosol release or dispersal consequence assessment studies. The source-term data quality obtained from multiple and replicate-measurement actual spent fuel Phase 4 testing is paramount, particularly since there is such a limited amount of earlier test results available [4,10,11]. In brief, Phase 4 spent fuel testing is the most important part of the entire program as envisioned and needs to be completed. It is, however, also the most difficult, expensive, and time-consuming segment of the test program to plan, set-up, and perform successfully.

There is an extremely limited amount of available measured SFR data, as described in Section 1.3, and the SFR values that do exist have a very high uncertainty. The 1980s, small-scale DUO_2/spent fuel tests performed at Battelle Columbus Laboratory [10] with NRC sponsorship determined a SFR value of ~3. The similar tests performed at Idaho National Engineering laboratory [11], performed with SNL and DOE cooperation, determined a SFR value of ~5.6, based on wet sieve measurements of aerosols produced; particle impactor data was not available due to partial sample losses during these analyses. A SFR value of ~0.53 was calculated based on available gravimetric data. These described experiments and subsequent analyses [4, 6, 14-15] predicted an aerosol spent fuel ratio 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 radiological consequences of a successful sabotage attack. These SFR value uncertainties were a prime reason for the initiation of the current SNL-WGSTSC program, particularly the Phase 4 spent fuel tests.

A large amount of effort and funding has already been expended for planning and initiating the Phase 4 program, to make the future conduct of Phase 4 tests a success. These efforts are clearly visible by the amount of time and effort that has been extended towards development of an experiment explosive-aerosol apparatus than can effectively measure in replicate the aerosol data of interest, while preventing radioactive particulate releases to the GIF test cell environment. Two of these test chambers have been fabricated and qualified to date. Other major hardware items to support Phase 4 testing have already been procured and are awaiting use. The existing "surrogate materials" data have shown consistency and can be effectively modeled. Characterization data exists for the Phase 4 spent fuel test targets [27] for proper data modeling post-test. DOE SSO has also authorized the use of the GIF as a test facility to perform the Phase 4 tests. The option to restart and then complete the Phase 4 spent fuel test program at SNL remains open, pending future adequate funding and support from program sponsors.

5.2.2 Future Real-Scale Surrogate Testing

Plans were initiated by GRS and Fraunhofer Institut (ITEM and EMI) in 2006 to perform joint-WGSTSC partner ("common project") large, or "real-scale" tests in Germany with multiple bundles of surrogate material rods in an actual cask-volume enclosure, with representative cask wall materials and a large HEDD device. These planned large-scale tests are to measure the relative amount of aerosols ***released*** (internal distribution and amounts blown out) through the sabotage-produced hole in the "cask." They are intended to be a logical extension of prior large, cask-scale tests performed by both SNL in the 1980s [6] and by GRS in the 1990s [5] using surrogate DUO_2 rods in bundles, within casks, and also an extension of the single rodlet SNL-WGSTSC tests.

An informal ranking of process uncertainties and of test parameter importance was performed by WGSTSC members starting in 2006. Independent results from GRS, IRSN, and SNL participants were merged into tables for further evaluation. Some of the conclusions of this ranking exercise were that the uncertainty estimates from different participants strongly differ from each other, as based on differing starting assumptions. New, real-scale experiments should at least concentrate on: pressure build up and cask blow down, and HEDD jet release. The parameter variations within the test matrix should at least include single-wall and full-penetration, plus free gas volume. Test objectives should include: learning more about the release process (jet, blow-down, full penetration); measurements of pressure/temperature, inside/outside source term for fuel assemblies (initially presumed to be bundles of Zircaloy or stainless steel tubes with cerium oxide pellets and dopant fission products within a limited length of the rods; target materials may be modified in the future); consideration of differences between air/helium atmosphere (oxidation concerns); combine surrogate fuel assembly test results with single rodlet SNL test results; and, understand/simulate/transfer German Gramat [5] large-scale cask DUO_2 test results from the 1990s. A project proposal was developed, starting from this ranking exercise, which consists of a small-scale test matrix to investigate most of the above mentioned processes and questions, and few real-scale experiments to verify scaling assumptions and to validate numerical modeling.

This two-year test program originally was intended to start in late 2006, but did not occur to date, due to German funding difficulties and ongoing discussions of the project in Germany. The basic test program is planned to be performed with German funding by Fraunhofer ITEM / Fraunhofer EMI and GRS personnel mainly using available equipment at Fraunhofer facilities. Optional test extensions will include contributions (staff, materials, test apparatus, evaluations …) from other WGSTS members, but with no exchange of funding. As soon as the project starts, final decisions and details of this test program will be defined by GRS and Fraunhofer personnel, with WGSTSC inputs.

5.3 Other Insights and Modeling Studies

Several parallel modeling studies have been initiated by WGSTSC partners to help tie together results and interpretations from the current "simplified single test rodlet" program, previous large- or cask-scale surrogate tests performed by SNL in the 1980s [6] and GRS in the 1990s [5], and the planned large-, real-scale GRS test programs. Most of the modeling studies have focused on interpreting and replicating results from the previous SNL and GRS tests [5-6], where the amount of respirable materials released from the penetrated casks to the surrounding volumes were measured and ranged from 7.7×10^{-4} to 4.6×10^{-3} for the SNL tests and 6.0×10^{-5} to 3.2×10^{-4} for the German tests [12]. Luna and Yoshimura et al. [4,12-13,33-34] have conducted several modeling evaluations of these tests. A newer modeling program was initiated at SNL in FY 2005 but has not been funded since then.

In 2004, the WGSTSC working group began cooperative modeling considerations on the phenomenon of the release of aerosol materials outside of a cask [32] including modeling to simulate the past tests [5-6]. The objectives of these simulations were to develop analytical tools in order to interpret or to describe mechanical behavior of the fuel rod during the HEDD jet impact. This modeling has permitted the WGSTSC to identify the most influential parameters which govern the aerosol particle release. One overview of the recent WGSTSC modeling efforts has been summarized by Autrusson et al. [32]. These modeling efforts are intended to demonstrate: the capability to interpret experimental observations from the past tests; to predict the differences to be observed between sabotage-damaged actual spent nuclear fuel or tested surrogate cerium oxide pellet rodlets; to model a more sophisticated test or real case with multiple fuel bundles in a cask; and, to extrapolate to another type of fuel rod (fresh fuel, different burn-up of irradiated fuel, MOX, etc.). These WGSTSC modeling studies [32] are evaluating the driving phenomenon concerning the release of material, e.g., internal pressures, temperatures, hole sizes, aerosol transfers and deposition within the cask, SNF rod bundles deformation response, etc.

To give a preliminary answer to these questions [32], WGSTSC participants agreed to define a "common problem" benchmark study to simulate the interaction between a HEDD and a simplified mock-up of shipping cask with simplified models or numerical simulations using a hydrodynamic computer code. The parameters which were studied were the wall penetration (depth and diameter), number of breached pins, pressure and temperature build up, blow down release from the cask, total source term with particle size distribution, source term (mass aerosolized, mass respirable and mass deposited near field). Each parameter was quantified within the range of agreed upon uncertainties in order to support the decision process on future experimental and numerical work of the WGSTSC. Some conclusions from this benchmark study were already considered in the latest German project proposal (described in the previous Section) which will include both, experimental and numerical analyses. Further results from this "common problem" benchmark evaluation will be documented in the future.

5.4 International Cooperation and Information Sharing Agreements

Since the initiation of the International WGSTSC in 1999, a major, agreed upon goal of all participants has been to continue cooperation to improve analytic capabilities through information sharing and collaborative research and development plus modeling, to understand the potential adverse public health effects and environmental impacts of radiological sabotage directed at or associated with the transport and storage of civilian nuclear material or other civilian radioactive materials [32]. This goal requires an open sharing of relevant information, data, and analyses between all U.S. and foreign participants, within the limits of the security requirements of all national entities. Previously stated objectives of this sharing include: 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; to provide technology transfers and support to U.S. NRC, IRSN (France), GRS (Germany), and other vulnerability studies, by providing data and analyses for computer modeling of HEDD attacks against nuclear materials; and, to support DOE and non-US participants national authorities' assessments of the physical protection requirements for nuclear materials in use, storage, and transport.

This sharing intent was reached, initially, by gentleman's agreement among all participants and documented in a formal test program plan document [1]. It should be further clarified that all the information documented to date in WGSTSC technical meetings and formal technical reports is non-classified information in nature, specified as unclassified, unlimited release (UUR). Sandia has been specifically directed by our major program sponsor, the U.S. Department of Energy, to document our results in a UUR manner. The discussion of non-classified, but sensitive information on relevant vulnerability studies, or financial information, has been presented or discussed in closed WGSTSC technical meetings (not open to the public) and was labeled as Official Use Only.

However, it has been specifically recognized by all parties that information to be generated in the future, particularly from the spent fuel Phase 4 tests, explosively-particulated by a HEDD, may be sensitive information requiring a higher degree of security classification. In order to prevent any perceived security violations or breach of export control regulations, the sharing of such security-controlled information requires a formal Multi-Lateral Agreement (MLA) between national entities for WGSTSC participants. This MLA has been in the planning stage since about 2003 and is discussed further in the following Sections.

5.4.1 WGSTSC Memorandum of Understanding

The informal, "gentleman's agreement" between WGSTSC participants to collaborate and share information was recognized by all to be somewhat inadequate in the view of many national supporting, i.e, funding or regulating agencies. As such, a short-term solution, a less formal agreement for further cooperation, e.g., a "memorandum of understanding (MOU)" signed by Technical Support Organizations (TSO) as SNL, IRSN and GRS was brought up for further consideration in 2006. This cooperative MOU agreement is intended to provide a working arrangement between all Parties and TSO at a level below a formal Multi-Lateral Agreement (MLA), not to substitute for a MLA. The MOU *does not* include provisions for sharing of security classified information. Subsequently, IRSN offered a draft "Agreement for collaboration concerning the radiological release resulting from the rupture of a fuel rod segment by a conical shaped charge (Draft March 2003/ Revised June 2006)." This draft MOU for consideration was a revision of a draft MLA that IRSN had suggested in 2003. Some content from "Action Sheet No. 3, a signed, earlier agreement on the same topic between DOE NN (now NA) and CEA management, with IPSN (now IRSN) and SNL key personnel, from March 2000, was also incorporated. It was agreed by all participants that even if a separate, formal MLA is approved by the U.S. DOE and Department of State (DOS), the French and Germans governments probably wouldn't be able to formally ratify it for several years -- hence the desire for a less formal agreement.

A significantly revised Draft MOU, based on the IRSN "Agreement for Collaboration," was submitted to the SNL Legal and Intellectual Properties Departments and to DOE, for comments and suggestions in August 2006. SNL Legal replied that "It is clear from the previous agreement and the content of this agreement that this is a document that requires DOE signature, not Sandia signature. Sandia can negotiate the proper terms for the document, if DOE desires, but we will need DOE to assist with this document. Since DOE is not a party, and Sandia cannot accept work under a MOU, all work and exchanges will need to be under a separate agreement." No further progress at SNL has been made since that time in regards to a Memorandum of Understanding. However, this agreement process has shifted back to work on a formal MLA, with significant effort by both DOE and the U.S. Department of State (DOS). Progress in this regard has been made, as follows.

5.4.2 WGSTSC Draft Multi-Lateral Agreement

At the 5th Technical Meeting of the International WGSTSC, in Cologne, Germany, in 2003, IRSN offered a draft document, "Project Annex No. 1 For Collaboration Concerning The Radiological Release Resulting From The Aerosolization Of A Fuel Rod Segment By A Conical Shaped Charge, Under The Agreement For Cooperation Concerning The Radiological Impacts From Sabotage Of Nuclear Material In Storage And Transport" as a preliminary draft for a Multi-Lateral Agreement, for all participants to discuss and review. Informal review comments were submitted back to the IRSN by all WGSTSC participants at the Technical Meeting, including SNL, NRC, DOE RW and DOE General Counsel (GC).

Both NRC and DOE worked on finalizing this Draft MLA and getting their formal agencies approval to submit it to the U.S. Department of State for formalization and finalization. The MLA, as did the pre-draft IRSN "Project Annex," includes sections or provisions covering the topics of: Agreement Objectives; Areas of Cooperation; Forms of Cooperation; and, Management. There are further Project Annexes covering: Exchange Of Personnel; Equipment; Samples And Materials; Transfer of Information and Equipment; Intellectual Property Rights; Business-Confidential Information; Additional Parties (WGSTSC Members); Funding (Restrictions); General Provisions; and, Duration, Amendment, Termination And Withdrawal. There is a Signatures page for controlling organization participants.

Finalizing and approving (on the U.S. side only) was a lengthy, difficult, multi-year process, and required an extensive U.S. multi-agency review. The process was so time-consuming that the less-formal Draft Memorandum of Understanding was brought back for WGSTSC re-consideration in 2006 (as discussed in the previous section). MLA approvals from the NRC and DOE were both finally received in 2007. In late July 2007, the Multi-Lateral Agreement (MLA) was approved by the U.S. Department of State, and defines specifically how our data (both non-classified and classified) would be shared with the international WGSTSC partners. The MLA has been sent to the German, French and British WGSTSC participants for their review/concurrence. The MLA is considered "draft" only in the sense that the non-U.S. parties haven't signed it yet. DOE RW anticipates that the MLA (if it doesn't require too lengthy a foreign government approval process) will take precedence over the less-formal MOU, making the MOU unnecessary.

Both the MLA and the MOU will be interactively discussed with all WGSTSC participants in the near future, to obtain a better understanding of the potential required time-frame required for these agreements (MLA and/or MLA/MOU) to be implemented.

In conclusion, there continue to be significant benefits for all U.S. and participating foreign governmental, regulatory, competent authority, and test WGSTSC organizations involved for the successful conduct and completion of this spent fuel sabotage explosive-aerosol test program and associated modeling work, including:

1. Reliable, measured source-term data are provided for supporting separate and future modeling analyses of release of respirable aerosol particles and atmospheric dispersal resulting from an intentional, plausible sabotage events on SNF or other radioactive materials.
2. The measurement of the spent fuel ratio plus aerosol source term data and information from these tests and analyses allows extension or scaling to other types of transportation terrorist events via modeling.
3. A basis is provided for evaluating appropriate levels of physical protection, safeguard requirements, and preventative strategies for nuclear materials in use, transport, or storage (including the Yucca Mountain repository transportation program). This provides a direct benefit in lowering the amount of required physical protection for shipments while providing the appropriate amount of security to shipments, i.e., shipments are not “over-protected.”
4. Measured data help guide and validate technical bases for transport and storage regulations of spent nuclear fuels (10 CFR Parts 71, 72, and 73) based on older, limited information [1-2, 5] and provide further validations of sabotage vulnerability studies. And,
5. The continuing, successful conduct of this International WGSTSC program leverages total shared testing, modeling, capabilities, and benefits over all international U.S., German, French, and other participants, with significant, shared technical and policy benefits.

5.5 Further Supporting Documentation

The initial SNL-WGSTSC explosive-aerosol, spent fuel sabotage test program plan, overall design, requirements, experimental details, and results as of the end of FY 2003 were included and documented in Sandia Technical Report SAND2004-1832 [1]. The FY 2004 test and data summary document SAND2005-4446 [2] provided an update and modest revision to that test plan. A summary of the Phase 1 test program and results was documented in SAND2005-5873 [18]. The FY 2005 to first-half of FY 2006 technical report, SAND2006-5674 [3], provided another update and large collection of available data and interpretations. *This* Interim Final Report comprehensively documents all available results and interpretations as of the end of 2007, and summarizes the current status of the overall test program.

Multiple other conference/symposium presentations, WGSTSC technical meeting presentations, and supporting or review documents have been prepared, formally approved, and are available from the Sandia National Laboratories Technical Library as unclassified, unlimited release documents. These are listed chronologically in the References/Further Supporting Documentation section at the end of this report.

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