SAND80-2043C, TTC-0154

CONF-810217--13

SAFETY ASSESSMENT OF SPENT-FUEL TRANSPORTATION IN EXTREME ENVIRONMENTS[a]

Robert P. Sandoval
and
John P. Weber
Sandia National Laboratories
Transportation Technology Center
Albuquerque, NM 87185

and

George J. Newton
Inhalation Toxicology Research Institute
Lovelace Foundation for Medical Education and Research
Albuquerque, NM 87115

INTRODUCTION: STATEMENT OF THE PROBLEM

This paper deals with a program currently under way at Sandia National Laboratories (SNL) to characterize source term aerosols from radioactive materials exposed to extraordinary environments and to produce improved estimates of environmental and human health consequences. The extreme environments of interest here have been divided into two categories -- those related to sabotage (or intentional) acts and those related to extra severe accidents. Major efforts in this program to date have concentrated on the intentional act activity in which the objective is to develop an experimental data base characterizing the release of radioactive material resulting from the sabotage of a spent fuel transport. This experimental data base would be the primary input to an analysis of the human health and economic consequences for these types of environments.

The origin of the program can be traced back to 1975 when the U. S. Department of Energy (DOE) sponsored a study at SNL to determine the ability of several generic high energy devices to disrupt a large truck spent fuel shipping container. The results of this study indicated that it was indeed possible for certain high energy devices to breach a large spent fuel cask.

---

[a]This work supported by the U. S. Department of Energy under contract DE-AC04-76DP00789.

DISCLAIMER

In 1977, the NRC published a final environmental impact statement on the transportation of radioactive materials by air and other modes, NUREG-0170,[1] which concluded that spent fuel shipments do not constitute a threat to the public health and safety. This same study states that the risk of sabotage of radioactive material transports is sufficiently small to constitute no adverse major impact to the environment or to public health.

However, in 1978, the NRC published a draft environmental assessment of the transport of radionuclides in urban environments. The so-called "Urban Study" evaluated the radiological hazards resulting from the transportation of radioactive material in urban areas for various types of environments including those caused by sabotage. The first rough draft version of this study, SAND77-1927,[2] predicted that several hundreds of latent fatalities could occur from the sabotage of spent fuel shipment systems subjected to certain modes of attack. A second version of the urban study, NUREG/CR-0743,[3] reduced the latent fatalities to less than 100 based upon a re-evaluation of released quantities of radioactive material.

In 1979, the NRC reacted to the initial urban study by requiring additional physical protection measures for U. S. spent fuel shipments pending the availability of credible experimental data supporting or disproving these predictions.

In 1979, the Comptroller General of the United States published a study[4] of federal actions needed to improve safety and security of nuclear material transportation. This study recommended that the NRC and DOE develop experimental data bases concerning the quantity of material that could be released from the sabotage of spent fuel casks.

In response to these data requirements and requirements to support DOE spent fuel programs, the DOE initiated the TSSE program at SNL to evaluate the effects of intentional acts and extreme accident environments on spent fuel shipping systems and to determine experimentally the quantity, size and chemical form of any released material. This source term data base will be used to assess the safety and security of spent fuel transportation.

PROGRAMMATIC APPROACH

Early in the program, it was realized that it would not be feasible from cost or safety standpoints to perform a full scale,

spent fuel cask test in the atmosphere. It was also realized that in order to achieve the programmatic objectives in a cost efficient manner (maximizing the information and minimizing the costs) that a series of scaled tests should be performed. Therefore, the programmatic philosophy was to conduct scaled tests using surrogate and spent fuel before attempting scaled tests on casks filled with actual spent fuel.

The first task is to evaluate the effectiveness of several types of high energy intensive devices (EID's) to aerosolize and disperse spent fuel elements. Also important in this evaluation is the scaling of the aerosol parameters.

A second task of the program is to evaluate the effectiveness of these energy intensive devices to cause a release of material from a 1/4 scale spent fuel cask filled with surrogate fuel (depleted UO_2, zircalloy cladding). This task is being performed at SNL.

The third task involves experiments to determine a correlation between selected radionuclide particulate size distributions produced from disrupted spent fuel and that from surrogate fuel (UO_2 in this case). These experiments are being performed at EG&G/INEL and involve subjecting actual spent fuel pellets as well as surrogate fuel pellets to scaled EID's. These experiments were initiated in 1980, and the experimental results obtained to date will be discussed in this paper.

A fourth task in the program is to perform scaling verification experiments on full scale shipping casks filled with surrogate fuel subjected to full scale EID's. The purpose of these experiments is to provide scaling information on aerosol parameters and release fractions, in order to allow scaling to full scale scenarios. These experiments are being performed at SNL in fiscal year 1981.

A fifth task of the program is to perform multi-component aerosol modeling and dispersion studies in order to help determine containment effects, dilution effects and to obtain a better understanding of basic aerosol formation processes in these types of high energy environments.

The sixth task provides for scaled tests using spent fuel. If the previous tasks (1-4) are successful and the experimental data base determined in these tasks is sufficient, then the tests of the sixth task will not be necessary.

EXPERIMENTAL

Six major tests using surrogate fuel have been performed at SNL in support of the second task and five correlation tests using depleted UO_2 and irradiated spent fuel have been performed at EG&G/INEL to date as part of the third task. Table I shows the six major experiments performed at SNL, and Table II shows the five major correlation experiments performed at EG&G/INEL. Of the six major tests performed at SNL, two were performed on full scale simulated cask walls in the atmosphere, and four were performed on 1/4 scale shipping casks in an aerosol containment chamber.

The experimental configuration of the atmospheric tests performed at SNL consisted of six surrogate fuel pins (depleted UO_2) placed between two simulated full scale cask walls (laminated lead and steel) attacked by a full scale EID. The purpose of the two atmospheric experiments was to determine the feasibility and accuracy of making aerosol measurements in the atmosphere. A second objective of these atmospheric experiments was to determine if a uranium mass balance could be made in the atmosphere. Aerosol samplers were

TABLE I

Experiments Performed to Date at SNL

Test No.	Type	Target	Date
1	Atmospheric	Simulated cask Surrogate fuel	7/79
2	Atmospheric	Simulated cask Surrogate fuel	8/79
3	Chamber	Steel target	1/80
4	Chamber	Steel target	2/80
5	Chamber	Surrogate fuel	3/80
6	Chamber	1/4 scale cask Surrogate fuel	6/80

TABLE II

Correlation Experiments Performed at EG&G/INEL During FY 80

Test	Type	Fuel	Results
18 tests	Parameter Evaluation	Depleted UO_2	Experience in aerosol measurement
Correlation #1, 2, 3	In containment	Depleted UO_2	Sieve total; Filter sample; SEM; Cascade impactor
Correlation #4	In containment	Depleted UO_2	Sieve; Total filter sample; Cascade impactor; SEM
Correlation #5	In containment	H.B. Robinson II Spent fuel	Sieve; SEM; Total Filter sample; Cascade Impactor

placed at various strategic locations around the test site to measure size and concentration of the dispersed aerosol. Modified Anderson cascade impactors were used to measure particle size distribution of the aerosol plume. High volume filters were used to measure mass concentration of the aerosol plume. Samplers were analyzed gravimetrically for total mass, and fluorometric analyses was used to quantify the aerosolized uranium mass. It was determined from the results of these atmospheric tests that it was not feasible to accurately account for all of the uranium aerosol mass. However, it was determined that it was possible to measure minute quantities of uranium particulates dispersed in the aerosol plume.

Based on the results of these experiments, it was decided to conduct future 1/4 scale cask/surrogate fuel experiments in an aerosol-containment chamber. A schematic of the aerosol-containment chamber is shown in Fig. 1. Four tests using various types of targets were conducted in this aerosol chamber. The chamber offered the advantages of confining the aerosol cloud and permitting all of the dispersed material to be recovered for mass balance and particle size measurements. Also, a time history of the respirable aerosol cloud could be obtained as well as the size and mass distribution of the total fuel disrupted. Targets in the four experiments were steel blocks (Test #3 & #4), depleted UO_2 fuel pins (zircalloy cladding) placed on steel blocks (Test #5), and a 1/4 scale steel/lead

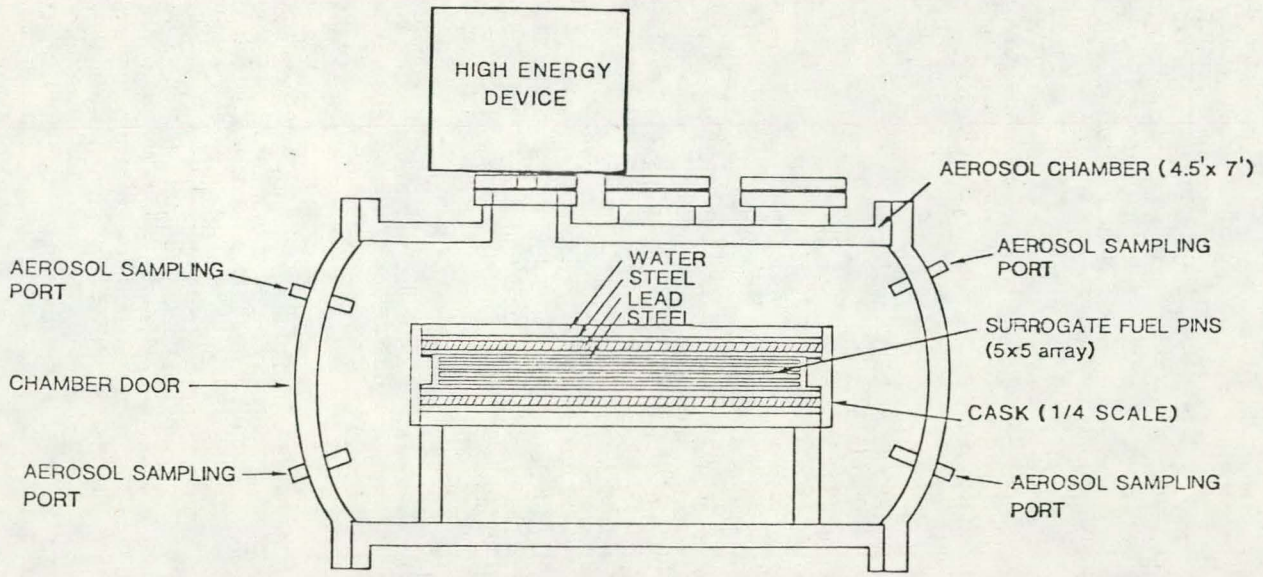


Figure 1. Schematic of Aerosol Containment Chamber

cask containing 3-ft long sections of full scale, UO_2 surrogate fuel pins (Test #6). Again, the purpose of the sixth experiment was to determine the quantity and chemical and physical characteristics of the released UO_2 aerosol.

As shown in Fig. 1, the EID was mounted external to the chamber and detonated through a port in the chamber. Between the EID and the chamber was an explosive isolation valve. This valve was designed to close milliseconds after the detonation of the EID in order to reduce the quantity of combustion products entering the chamber. Five sampling ports penetrated the chamber in various locations. These ports were closed before and during detonation by pneumatically operated valves. The valves were then opened after detonation in order to allow sampling of the aerosol cloud.

Since no single aerosol instrument can size particles from 0.01μm to 2 mm diameter, a battery of instruments was used to size the aerosol in these experiments (Table III). In addition to this aerosol instrumentation, energy dispersive x-ray and fluorometric analysis were used to determine the quantity of various elements

TABLE III

Sampling Instruments Used in 1/4 Scale Chamber Tests

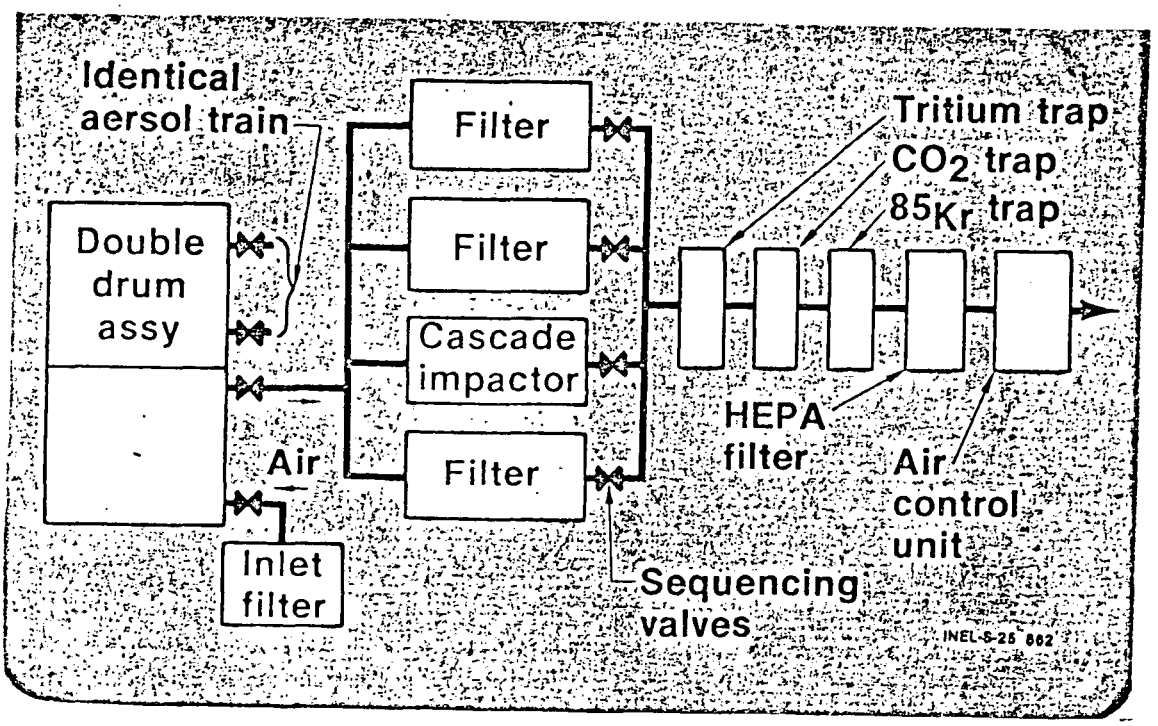
Instrument & Applicable Size Range	Purpose of Samples	Analytical Method
LMJ cascade impactors 0.5-12 μm	Aerodynamic size, geometric standard deviation of total aerosol mass and UO_2 mass.	Gravimetric (Cahn micro-balance) and fluorometry for determination of uranium.
Point-to-plane electrostatic precipitator TEM and SEM 0.01-12 μm	Particle morphology, count distribution and elemental distribution.	Transmission and scanning electron microscopy and energy dispersive x-ray analysis.
Filter 37 mm, sequential, 0.01-12 μm	Provide a time history of total and uranium mass after EID detonation. Samples for surface area measurements.	Gravimetric for total mass and uranium by fluorometry, BET nitrogen absorption for surface area measurements.
Filter 37 mm, front surface reentrant filter 0.01-12 μm	Sequential filter samples from filters inside chamber to compare with filters obtained by extractive techniques to address aerosol line losses.	Gravimetric for total mass of aerosol and uranium by fluorometric techniques.
[a]Condensation nuclei counter 0.001-12 μm	Total count of aerosol particles vs time after detonation.	Optical light scattering instrument.
[a]Electrical aerosol analyzer Diameter < 1.0 μm	Size distribution parameters for particles less than 1.0 μm.	Electrical mobility.
Sieves 38-2000 μm	Provide size distribution data on larger particles of surrogate spent fuel.	Mechanical sieving followed by weighing for total mass and fluorometry for uranium determination.

[a]Data from these instruments are not included in this report.

in particles collected by the electrostatic precipitators. The sampling procedure was designed to provide a time history of the respirable aerosol in the chamber. From the time history, calculations could be performed to determine the initial release parameters. By developing such techniques on casks containing surrogate fuel, results can be extrapolated and used in tests on casks containing spent fuel where sampling might not be permitted until considerable time after detonation.

Aerosol size parameters as a function of time were determined from cascade impactor samples obtained at various time intervals after detonation. Similarly, filter samples provide a time history of the change in mass concentration. Changes in particle morphology were shown by sequential electrostatic precipitator samples and changes in number concentrations were shown by continuously recording condensation nuclei counters.

The correlation tests at EG&G/INEL performed during 1980 consisted of four depleted UO_2 tests and one actual spent fuel test. The aerosol measurement train for the EG&G/INEL correlation test is shown in Fig. 2. The experimental aerosol measurement train consists of a double 55-gal drum assembly and filter and cascade impacter samplers. Also included in the aerosol measurement train are gas traps in order to quantify and characterize the gases released from the spent fuel pellets. The results of the spent fuel correlation experiments will be reported in greater detail in the next section.



Experimental Results and Analyses

The SNL atmospheric tests conducted in July and August of 1979 demonstrated the feasibility of obtaining aerosol measurements following full scale explosive attacks on simulated cask walls and surrogate fuel. Although an estimate of the respirable UO_2 aerosol released in the plume was made based on average UO_2 mass concentration and extrapolating to the estimated cloud size, no mass balance was possible because of the large loss of surrogate fuel to the surrounding area. Based on the average uranium mass concentration (8.8 $\mu g/m^3$), and an estimated aerosol plume volume (83,000 m^3), the aerosol plume

was estimated to contain approximately 0.7 g of uranium aerosol smaller than 10 μm aerodynamic diameter.

The fourth chamber test performed in June of 1980 (Test #6 in Table I) subjected a dry 1/4 scale steel/lead cask (containing a short section of a 5 x 5 array of full scale surrogate fuel pins) to a 1/4 scale EID. No explosive isolation valve was used in this chamber test because previous tests had indicated that less than 2 percent of the measured chamber aerosol was combustion products from the explosive device. A total respirable UO_2 aerosol mass of 0.78 g was measured at time 12 seconds after detonation. Using standard error propagation techniques, the uncertainty in the measured respirable UO_2 mass was calculated using known experimental uncertainties of the measured parameters (uranium mass fraction, chamber volume and total mass concentration). The estimated error of this measurement (0.78 g) is about $\pm$ 0.05 g.

This measured respirable aerosol mass is the basis of the estimated respirable release fraction. Based on a mass loss of 216 g of cladding and depleted UO_2 (which was determined by weighing fuel pins before and after detonation) the aerosolized release fraction for Test #6 was approximately 0.36 percent. This number is based on the quantity of mass loss and not upon the total inventory of the cask. Approximately 96 percent of the UO_2 mass (172.32 g) lost from the event was accounted for in this experiment. Because of the experimental confidence in the precision of the measured respirable UO_2 (0.78 $\pm$ 0.05 g), the other 4 percent of the total UO_2 mass (7.18 g) was believed to be of particle sizes greater than 30μm which were deposited, but not collected, on surfaces inside the test chamber.

Figure 3 shows a time history of the aerosol mass within the chamber and is based upon sequential filter sampling. The exponential decay of aerosol mass in Fig. 3 indicates that the first filter sample (used for respirable release calculations) from 12 seconds to 3 minutes after detonation is consistent with all subsequent filter samples obtained. Sequential cascade impactor samples were also taken to give a time history of aerosol size distribution parameters. Following a peak size of approximately 3.5μm aerodynamic diameter seen at approximately 12 minutes after detonation, the aerosol size stabilized to a mass median aerodynamic diameter of 2μm after 30 minutes. Similar trends were observed for the standard deviation (which stabilized to approximately 2.0 after 30 minutes).

The fraction of total aerosol mass that was uranium dioxide ranged from 7 percent at 0.2 minute to about 2 percent for times 10 minutes after detonation. Fluorometric determination of uranium mass was verified by energy dispersive x-ray analysis using a lithium-drifted silicon detector and a multi-channel analyzer. Total SEM field scans, small particle SEM analysis and large particle SEM analysis indicated that the percentage of uranium ranged from 1.6 to 1.9 percent for times greater than 10 minutes after detonation.

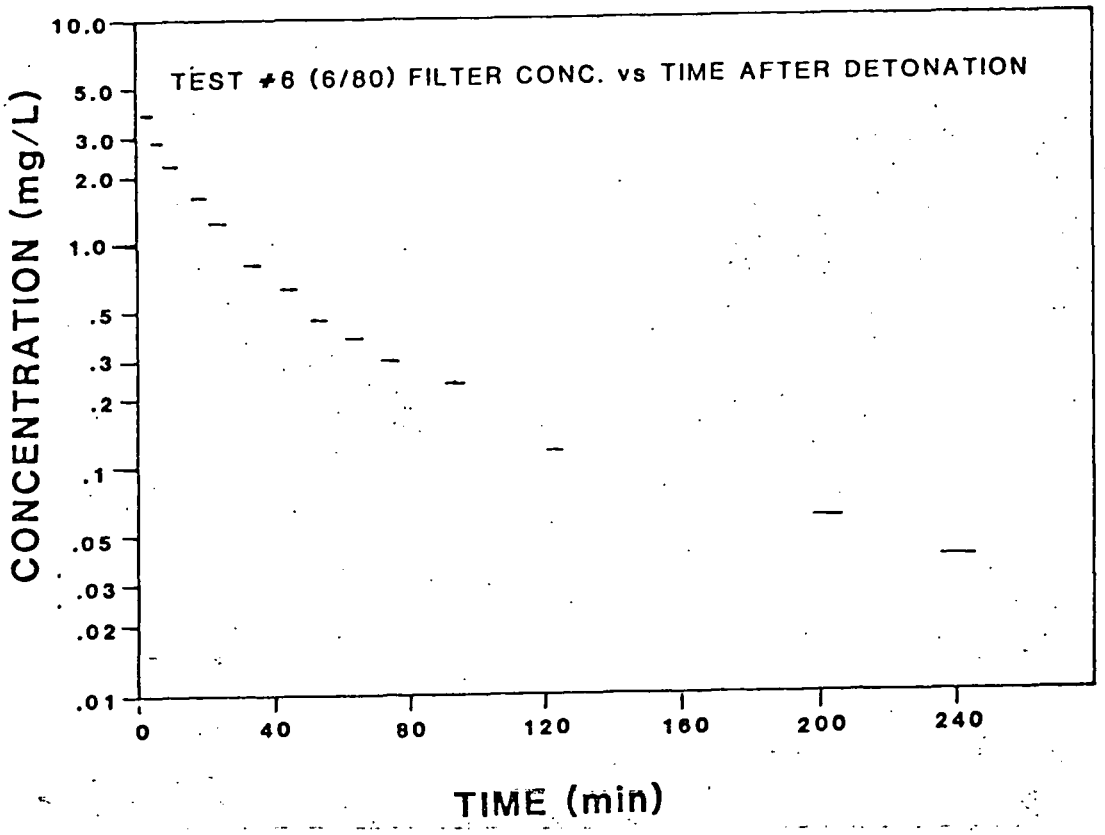
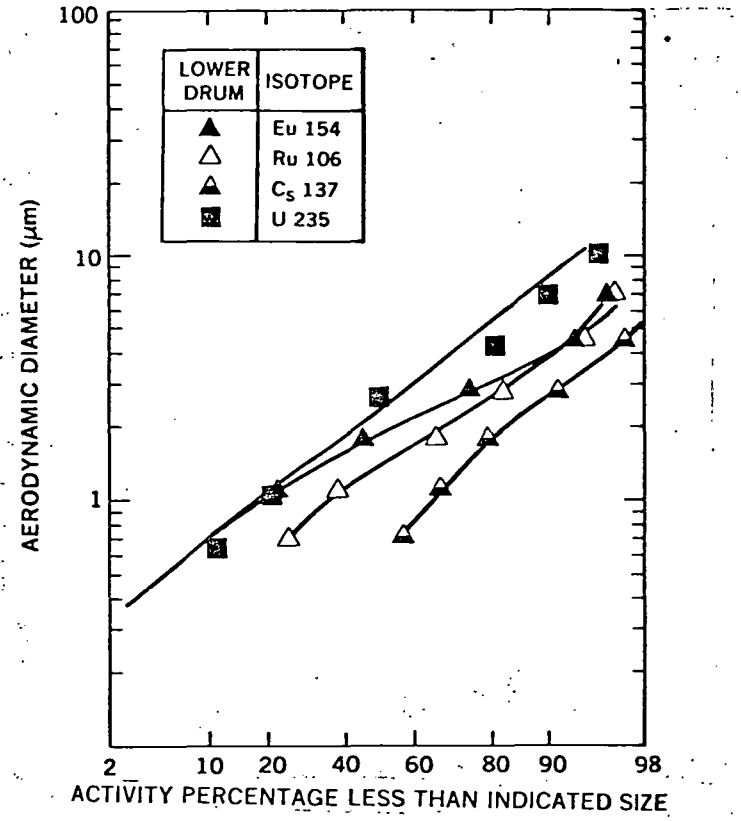


Figure 4 summarizes the cascade impactor results for the spent fuel correlation test (#5), performed at EG&G/INEL. It shows the particle aerodynamic diameter as a function of activity percentage less than stated size for four isotopes measured in these experiments: Europium-154, Ruthenium-106, Cesium-137 and Uranium-238. Not shown in this figure is Cerium-144 which closely follows the particle size distribution for Europium-154. Figure 4 shows that the more



volatile fission products (Ruthenium-106 and Cesium-137) fractionate to smaller particle sizes. More than 98 percent of Ruthenium-106 is smaller than 9μm aerodynamic diameter, and approximately 98 percent of Cesium-137 is smaller than 5 μm aerodynamic diameter. Ruthenium readily oxidizes to ruthenium tetroxide, and ruthenium tetroxide has a high vapor pressure at 45°C. Similarly, cesium oxide boils at 1280°C and has a high vapor pressure at 621°C. The material temperature produced by the high velocity impact pressures is believed to be greater than 1200°C; therefore, Ruthenium and Cesium are expected to be vaporized in these experiments.

The primary objective of the EG&G/INEL correlation tests was to provide a correlation between the particle size distribution for selected radionuclides of spent fuel and that of our surrogate fuel (Uranium-238). It appears at this point in the program that a correlation between spent and surrogate fuel aerosol parameters is achievable and that it is feasible to relate the release fractions of surrogate fuel to selected radionuclides of spent fuel.

SCALING CONSIDERATIONS

Experiments to obtain a release fraction for the full scale scenario are now underway at SNL, but the release fraction results of the 1/4 scale cask test can be compared to that of a full scale event by using a scaling calculation to extrapolate the aerosol parameters from 1/4 scale to full scale. Certain assumptions must be made a priori to allow extrapolating the measured 1/4 scale release fraction to that for full scale. The following assumptions are made in the scaling of our release fraction: (1) a total UO_2 respirable aerosol mass of 0.78 $\pm$ 0.05 g measured at 0.2 minute after detonation in the 1/4 scale test is used to compute a full scale respirable aerosol release fraction; (2) calculations of fractions of released airborne respirable aerosol for full scale events are made based on the number of damaged fuel rods; (3) assuming a three-PWR assembly cask of the type used in the urban study (1.4 t of heavy metal), approximately 400 rods would have a net mass loss for the reference EID used in this program. Again, this assumes the longest path of interaction. Extrapolation of 0.78 g for a 1/4 scale, single-assembly casks to a three-assembly, full scale cask results in approximately 31.2 g of respirable radioactive aerosol released from a full scale event. Thirty-one grams of respirable radioactive material is approximately 0.0023 percent of the total heavy metal inventory of the three-PWR assembly reference truck cask. These scaling calculations are based on a linear extrapolation of 1/4 scale test results. Verification of these scaling calculations await the results of our scaling tests planned for June 1981.

Consequence Analysis

The expected health consequences were calculated using the extrapolated release fraction of .0023 percent as the primary input to the consequence reactor safety model called CRAC[5]. The CRAC consequence model was used to examine the health consequences for several reasons. First, there is considerable experience available in the use of this model. Second, it allows consequences to be estimated out to considerable distances from the release point. Third, parameters may be varied in the model to explore the effects of radiation exposure pathways. And, fourth, CRAC was one of the consequence models used in the Urban Study, which is used for comparison in this study. The detailed population distribution employed in these models is equivalent to Manhattan, New York City (approximately 16,000 people per square kilometer). The total population distribution used in this model closely approximates the

actual population within 500 miles of the assumed release point. One hundred sequences of New York City weather conditions representative of weather near the release point were used in these calculations. The spent fuel radionuclide inventory used for CRAC has been generated using the fuel burnup code ORIGIN assuming light water reactor fuel with 33,000 MWd/t of heavy metal burnup at 40 kw/kg power density and 150 days cooling. A truck-mounted cask is assumed to contain radionuclides equivalent to 1.4 t of heavy metal charged to the reactor. The estimated time of release was mid-afternoon. A thermal source in CRAC was used to account for the effects of high explosives lofting the material (thus reducing the close ground level concentrations). All the consequence estimates have been made with the population in place. No attempt was made to model or account for evacuation to avoid early exposure. Deposition velocities for a 2 μm particle were assumed to be approximately 0.01 m/s. These deposition velocities are consistent with our experimental data.

Table IV shows the results of the CRAC calculation in total latent cancer fatalities, early morbidities and early fatalities. Because the source terms used never produced the threshold dosage for early fatalities and early morbidities, the number of early fatalities and morbidities predicted are zero. The latent cancer fatalities are divided into initial latent cancer fatalities which are a result of initial exposure and total latent cancer fatalities which are a result of long-term exposure to ground contamination and inhalation. The second version of the Urban Study predicts approximately

TABLE IV

Preliminary CRAC Code Results[a] for This Study

Population to 500 Mile Radius	
Early Fatalities	0
Early Morbidities	0
Total Latent Cancer Fatalities (mean/peak)	5/25

[a]Based on measured release of 0.0023 percent.
Early Fatalities: within 1 year after exposure.
Early Morbidities: illnesses within weeks after exposure.
Latent Cancer Fatalities: any time after exposure.

490 peak total latent cancer fatalities and approximately 130 mean total latent cancer fatalities. The preliminary results of this experimental study indicate that 25 peak latent cancer fatalities and 5 mean latent cancer fatalities are possible. This is a factor of 20 to 25 less than the latent cancer fatalities predicted in the Urban Study. It is also interesting to note that the early fatalities indicated by the Urban Study range from 0.2 to 19 and the early morbidities range from 1 to 76. Again, these predictions of early fatalities and early morbidities are zero.

Peak thyroid and bone marrow dose in rems was also calculated as a function of distance from the release point. At a distance of 30 meters from the release point the bone marrow dose was calculated to be 190 mrems, and the peak thyroid dose was calculated to be 100 mrems. At one mile from the release point the peak bone marrow dose was calculated to be 20 mrems and the peak thyroid dose was calculated to be 10 mrems. The Protective Action Guide (PAG) threshold for these distances is 1 rem. The peak bone marrow and thyroid dose for distances of 30 meters or more from the release point are significantly less than the PAG threshold.

SUMMARY

In summary, preliminary estimates of the health effects and/or consequences resulting from a malevolent attack on a spent fuel truck shipment in downtown New York City has been made. This estimate is based upon a measured quantity (0.78 ± 0.05 g) of respirable radioactive material released from a 1/4 scale event. A linear extrapolation from the 1/4 scale event to the generic full scale event has been made and an aerosolized release fraction (0.0023 percent) of the total heavy metal inventory of a three-PWR assembly truck cask has been calculated. Although scaling of the source term parameters is tentative at this point in the program, a full scale experiment is planned in 1981 to verify the scaling methodology used in these calculations. A preliminary correlation between spent fuel and surrogate fuel source terms has been shown to be feasible and that radionuclide size partitioning can be determined experimentally. Finally, it has been shown, based on our preliminary experimental source term data, that a maximum of 25 total latent cancer fatalities could occur, assuming a release in downtown New York City. This is 20 times smaller than the latent cancer fatalities predicted in the Urban Study.[3]

REFERENCES

1. Final Environmental Statement on the Transportation of Radioactive Material by Air and Other Modes, Vols. I and II, NUREG-0170, Office of Standards Development, U.S. Nuclear Regulatory Commission, Washington, DC, December 1977.

2. A. R. DuCharme et al., Transport of Radionuclides in Urban Environs: A Working Draft Assessment, SAND77-1927, Sandia National Laboratories, Albuquerque, NM, May 1978.

3. N. C. Finley et al., Transport of Radionuclides in Urban Environs: Draft Environmental Assessment, NUREG CR-0743 (SAND77-1927), Sandia National Laboratories, Albuquerque, NM July 1980.

4. Federal Actions Are Needed to Improve Safety and Security of Nuclear Materials Transportation, Comptroller General of the United States, Report to Congress, EMD-79-18, May 7, 1979.

5. I. B. Wall et al., Overview of the Reacot Safety Consequence Model, NUREG-0340, U. S. Nuclear Regulatory Commission, Wash., DC, October 1977.