SAND2021-11471C

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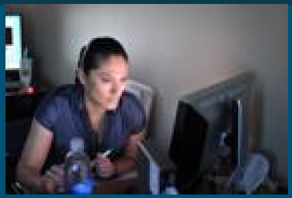
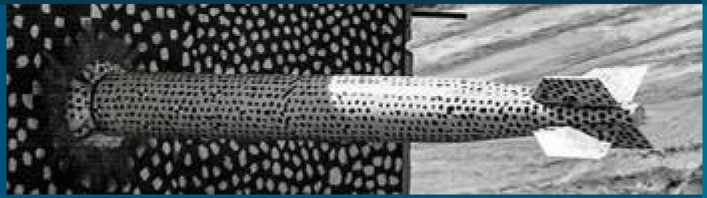
Review of Spent Fuel Reprocessing and Associated Accident Phenomena





National Academy of Sciences

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PRESENTED BY

F. Gelbard

A. Brown, D. Louie, F. Pierce, J. Phillips, and N. Bixler (retired)

Sandia National Laboratories

U.S. DEPARTMENT OF ENERGY NNSA

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Fred Gelbard Biography

B.S. Chemical Engineering, Massachusetts Institute of Technology (MIT)
Ph.D. Chemical Engineering, California Institute of Technology (CalTech)
Ph.D. Electrical Engineering, University of New Mexico (UNM)

- Analysis Research Areas

- Aerosol particle formation, growth, and deposition in nuclear reactor containments.
- Fluid flow and aerosol modeling in spent fuel canisters.
- Flow and heat transfer modeling in burning solid rocket propellant environments.
- Explosively generated aerosol formation and release. (*part of today's presentation*)
- Molten salt nuclear reactor accident and radionuclide release modeling.

- Experimental Research Areas

- Measuring transport of radionuclides through rock for WIPP (Waste Isolation Pilot Plant).
- Measuring temperature and aerosol formed from RTG (Radioisotope Thermal Electric Generator) simulant in a burning solid rocket propellant environment.

Background and Motivation

Two aqueous reprocessing plants for commercial fuel have been licensed in the US.

- Nuclear Fuel Services reprocessing facility at West Valley in 1966 (operated for a few years).
- Barnwell Nuclear Fuel Plant Separation Facility in 1970 (never operated).

Both were licensed under 10 CFR Part 50 by the AEC.

There are no current reprocessing facilities operating or planned in the US.

The major gap is in evaluating the potential source term.

What is the Source Term

- Source terms are releases from a confinement building into the environment.
- Atmospheric releases generally have the greatest and most immediate consequences.
- Source terms from an aqueous reprocessing facility can be created by a range of accident types. Four types considered in this work are:
  - Explosion
  - Fire
  - Nuclear Excursion
  - Leakage from Vessels or Pipes

Typical Source Term

- Initial source term into a room or compartment of the confinement building.
  - Energy insertion.
  - Mass insertion.
  - Physical form, vapors or aerosols.
  - Aerosol particle size distribution needed to determine transport and respirable fraction.
- Vapors and aerosols are transported through confinement to atmosphere.
  - Through doors or penetrations.
  - Through ventilation system.
  - Through filters.

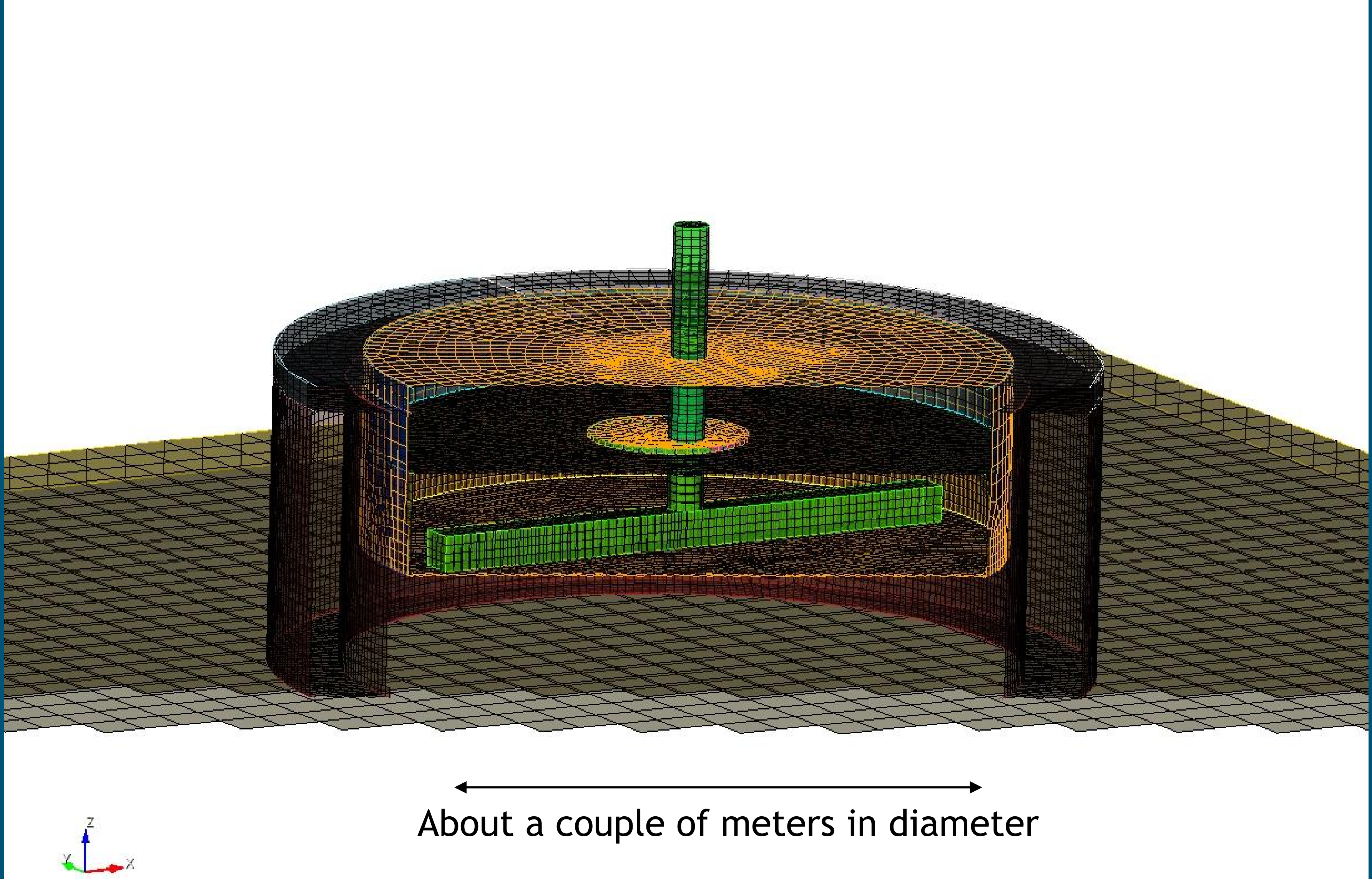
Most Significant Accident Types

- **Chemical explosion (Structural Damage & External Release)**
  - Process controls fail to maintain temperature or concentration limits of aqueous or organic solutions.
  - Released energy can exceed 1 GJ (over milliseconds).
- **Fires (Structural Damage & External Release)**
- **Nuclear excursion (Mostly local but can initiate external releases)**
  - Process controls fail to maintain concentration limits on fissile isotopes (U-235 and Pu-239).
  - Released energy can exceed 20 MJ per excursion (over seconds), with a total of 1 GJ (over tens of minutes).
- **Pipe or vessel leaks (Mostly local but can initiate external releases)**

Chemical Explosion Characteristics

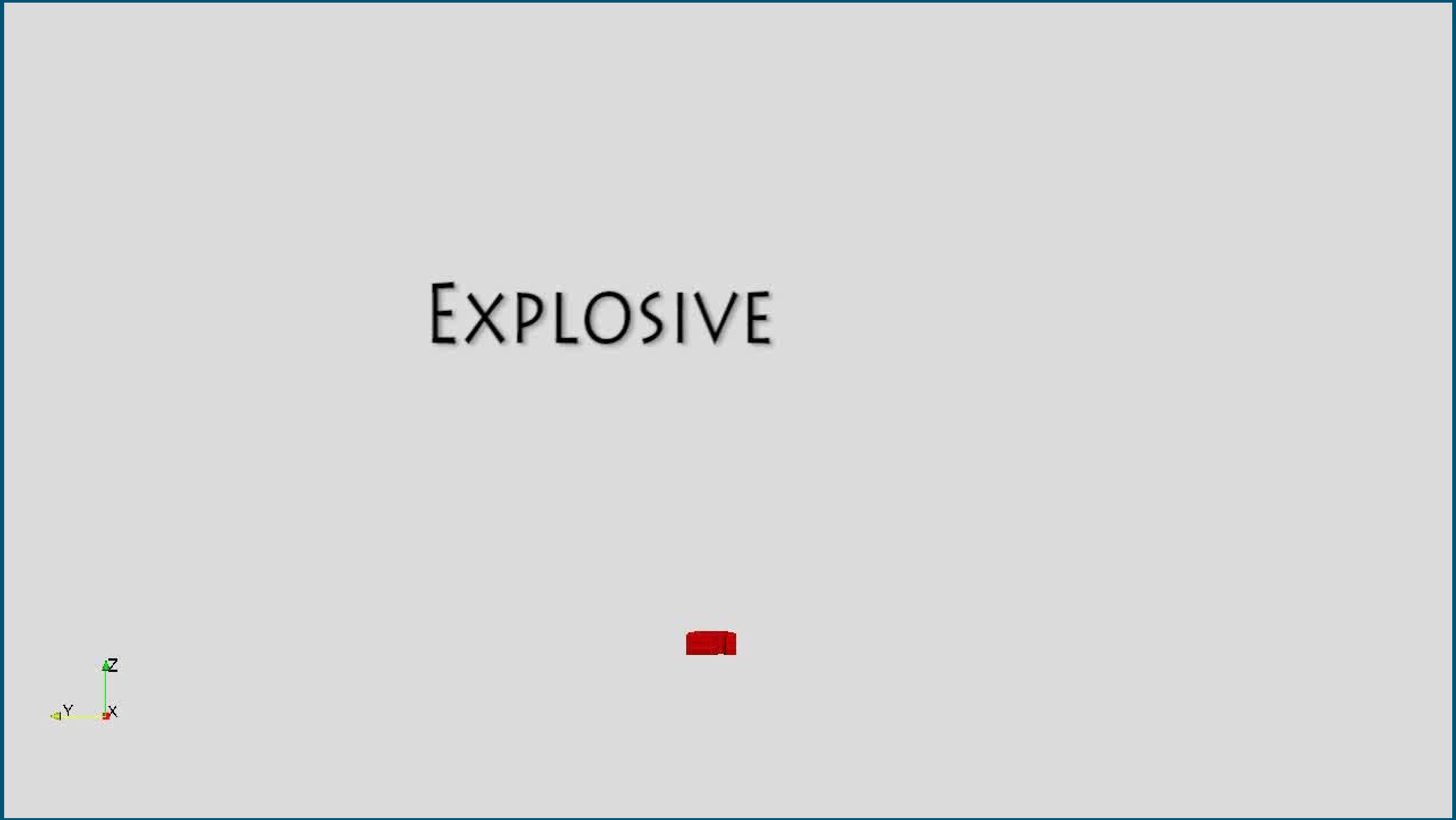
- Initiating event (e.g. red oil from TBP-Tributyl Phosphate)
- Rapid release of energy within a closed vessel
- Generation of pressures that can burst vessel
- Ejection of liquids and vapors from vessel
- Creation of aerosols by shear-induced breakup of ejected globules
- Change of phase
- Aerosol agglomeration, evaporation/condensation, and deposition
- Damage to walls, doors, and ventilation system

Discretized Model of Reprocessing Vessel (preliminary)



About a couple of meters in diameter

Simulated Explosion and Aerosol Release in Vessel (700 MJ)



Comparison To Conservative Estimates from DOE Handbook 3010-94 (2000)

DOE Handbook: "Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities: Volume I"

- Release is a product of five factors: (1) Material-at-Risk, (2) Damage Ratio, (3) Airborne Release Fraction, (4) Respirable Fraction, and (5) Leak Path Factor.
- MAR x DR = 1
- ARF x RF = 0.2 : 0.1 : 0.07 (upper bound : median : lower bound)
- LPF =1 for structure destruction

50 MJ explosion: 10^{-9} respirable fraction (preliminary)

700 MJ explosion: 10^{-7} respirable fraction (preliminary)

We have the capability to predict liquid particle aerosol formation and release from explosive events using basic principles.

Fire Characteristics

- Initiating event or induced by explosion or leak.
- Significant release of energy, but over longer time than explosion.
- Insertion of mass (soot and radioactive solutes) and energy.
- Aerosol agglomeration, evaporation/condensation, and deposition.
- Induction of chemical explosions.

Beaker-scale fire of burning Kerosine/TBP/Radionuclides for model validation (Mishima and Schwendiman, BNWL-B-274, 1973)

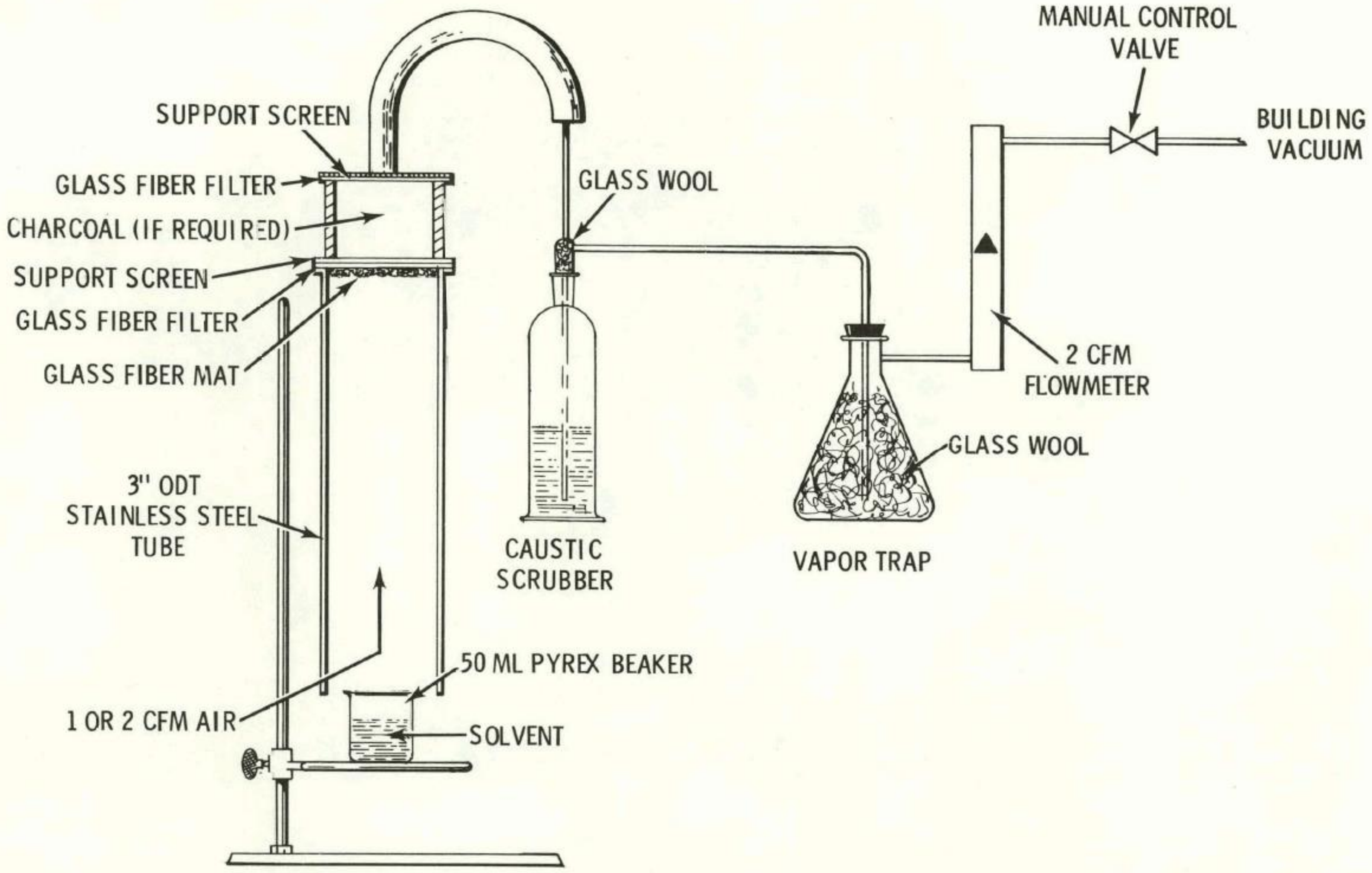


FIGURE 1b

SCHEMATIC DIAGRAM OF EXPERIMENTAL APPARATUS

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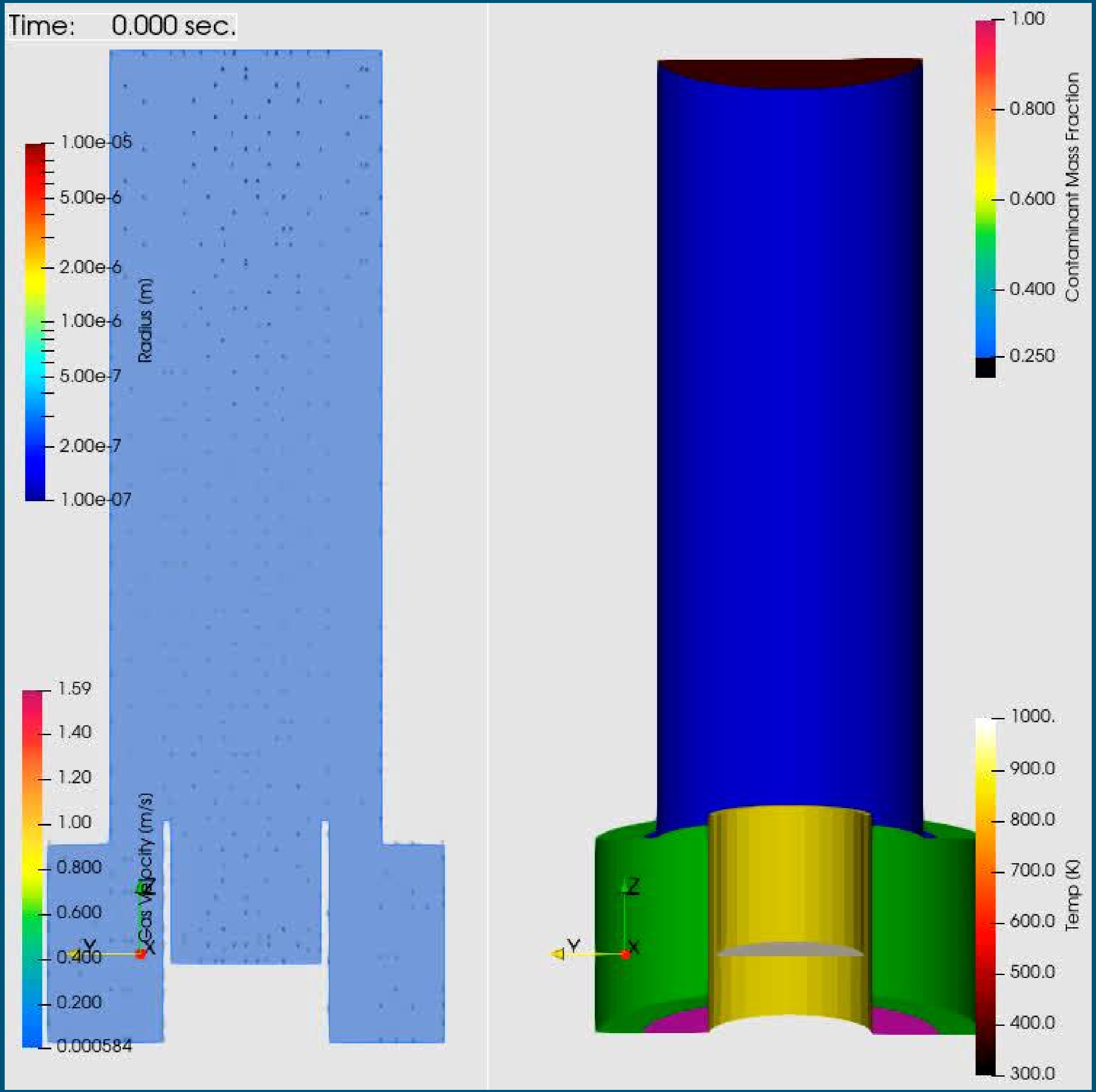
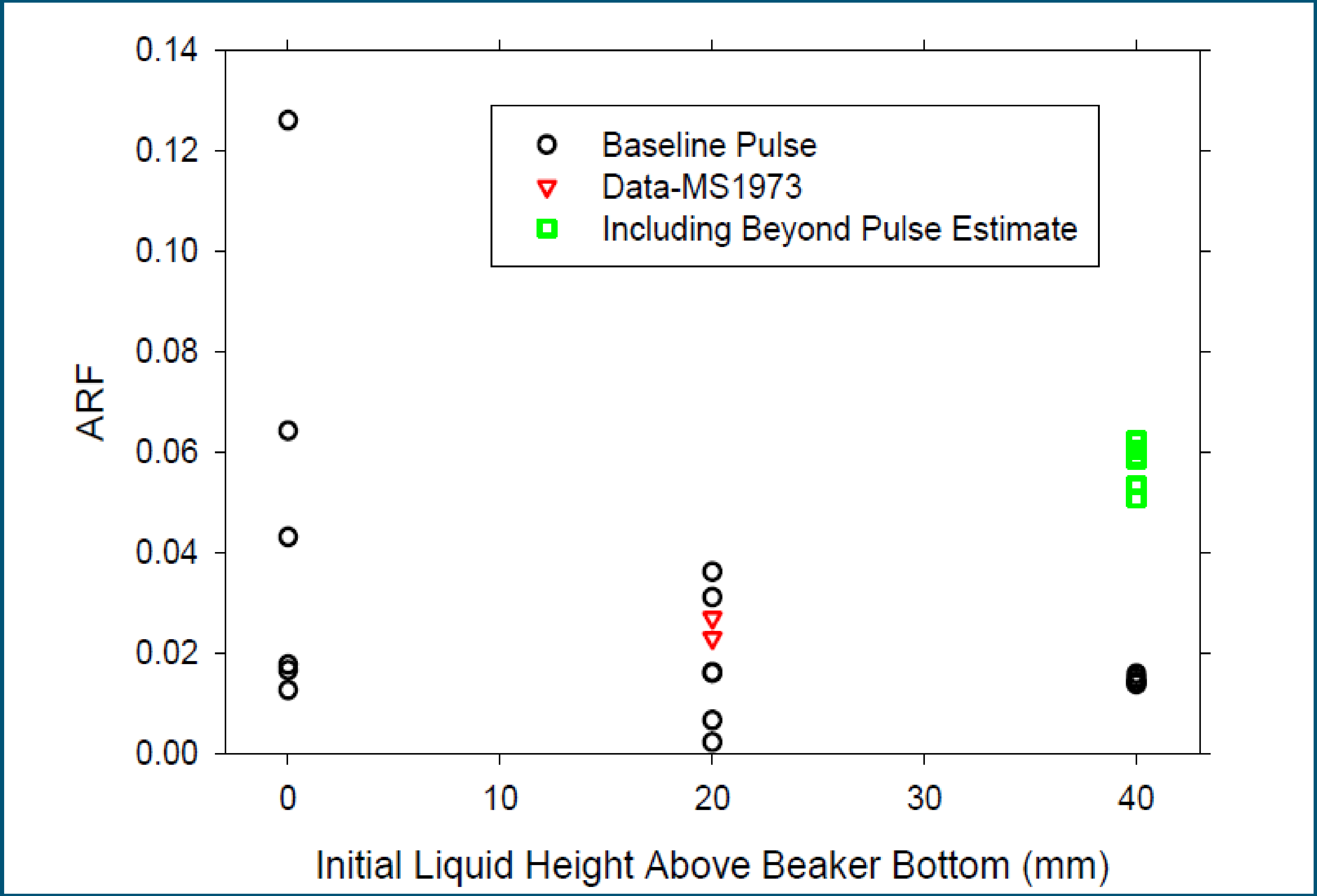
BNWL-B-274

Burning Tributyl Phosphate (TBP)/Kerosine (Preliminary)



Experimental validation of ARF (Airborne Release Fraction) for burning kerosine/Tributyl Phosphate (TPB)/radionuclide in 50 ml beaker



Reactivity Excursion

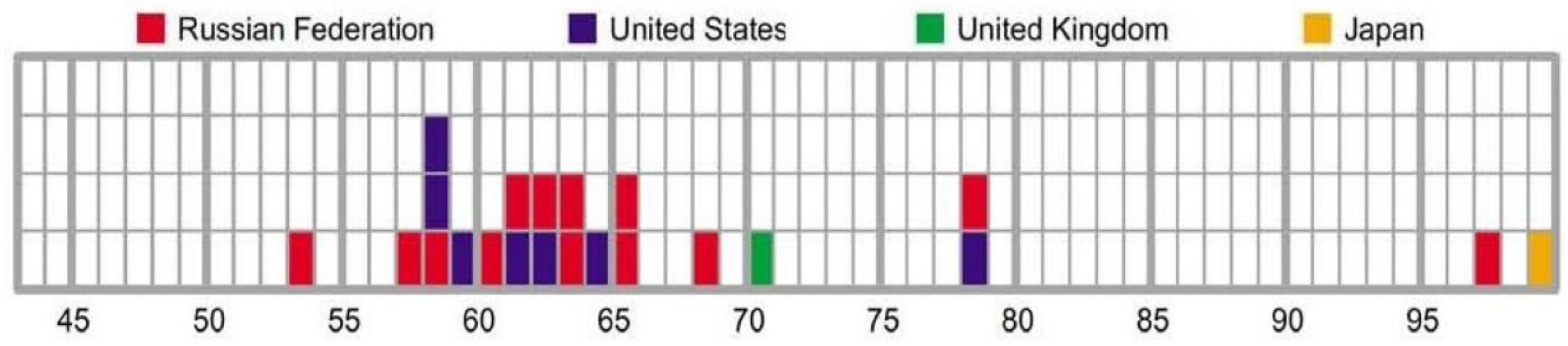


Figure 4-1 **Chronology of process criticality accidents. Plot shows number and location of accidents by year from 1945-2000. [McLaughlin 2000]**

- Fissile materials and neutron moderators are present with water or concrete surrounding the vessel.
- Initiating event.
- Pulse energy insertion which generates pressure that can burst vessel.
- Ejection of radioactive solids, liquids and vapors.
- Creation of aerosols by shear-induced breakup of ejected globules.
- Aerosol agglomeration, evaporation/condensation, and deposition.

Pipe or Vessel Leaks (Spills)

- Initiating event or result of explosion.
- Creation of aerosols by shear-induced breakup and fracturing of ejected globules.
- Change of phase.
- Aerosol agglomeration, evaporation/condensation, and deposition.
- Fires can be induced.

Summary of Estimating a Release

- Correlations in DOE-Handbook-3010-94 have been used to estimate released mass.
- Many assumptions are highly conservative.
- Some correlations are based on bench-scale experiments that often do not capture important large-scale phenomena.
- Alternative approach is to use a basic-principles model to estimate the creation of aerosols.
- Initial focus is on chemical explosions and fires because these are most likely to damage confinement and produce the largest source term.

Presentation Key Points

- Presentation is on proven reprocessing technology, with demonstrations of aerosol production from a red oil explosion in a reprocessing tank and a fire in a small beaker.
- Sandia has developed and is developing mechanistic source terms for risk-informed decision making.
- Plan is to supplement DOE-HDBK-3010 data and correlations for important source terms with basic-principles modeling.
- We expect to obtain more realistic source terms by using our SIERRA codes to substantiate the legacy, limited experimental data, and engineered judgement used in the development of DOE-HDBK-3010.