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**ORNL EXPERIMENTS TO CHARACTERIZE FUEL RELEASE FROM THE
REACTOR PRIMARY CONTAINMENT IN
SEVERE LMFBR ACCIDENTS***

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*Research sponsored by the Office of Nuclear Regulatory Research, Nuclear Regulatory Commission under Interagency Agreement 40-551-75 with the U.S. Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation.

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ABSTRACT

This paper presents results from aerosol source term experiments performed in the ORNL Aerosol Release and Transport (ART) Program sponsored by the U.S. NRC. The tests described were performed to provide information on fuel release from an LMFBR primary containment as a result of a hypothetical core-disruptive accident (HCDA). The release path investigated in these tests assumes that a fuel/sodium bubble is formed after disassembly that transports fuel and fission products through the sodium coolant and cover gas to be released into the reactor secondary containment. Due to the excellent heat transfer characteristics of the sodium, there is potential for large attenuation of the maximum release.

In LMFBR hypothetical accidents, the fuel is estimated to reach high energy states, up to and greater than 3500 J/g at core disassembly. A technique designated capacitor discharge vaporization (CDV) was developed at ORNL that uses electrical energy to place ~20 grams of uranium dioxide pellets into these high energy states. These fuel samples then "disassemble" in different surrounding environments. Two series of tests are discussed: a) vaporizations in an argon gas environment; and b) vaporizations under a water pool. Vaporizations in a sodium pool will eventually be performed.

In the first series of tests, uranium dioxide samples disassembled and underwent essentially free expansions in an argon-filled vessel. A free expansion was expected to produce maximum vapor and result in an "upper limit" aerosol source. In these tests, up to 6 grams of fuel vapor - which rapidly converted to an aerosol - was formed. Vapor yields produced for high energy inputs were less than adiabatic free expansion maximums determined from preliminary calculations. However, the calculated values depend on knowledge of the fuel energy distribution before expansion. This distribution is not accurately known due to the lack of accurate fuel electrical and thermal properties at high temperatures. Additional work continues to improve the fuel energy state calculations. Measurements of the fuel aerosol primary particle size distributions were also made. The size distributions might be representative of aerosols produced by fuel vapor released directly into the secondary containment, though the aerosol may also partially consist of small droplets produced by fragmentation.

Under-water experiments were performed in the Fuel Aerosol Simulant Test (FAST) vessel which is .61 m in diameter and 1.83 m high. Water levels above the fuel sample were 1.12 m, with a .254 m argon cover gas space above the water. Xenon gas (to simulate non-condensable fission products) was included within the fuel sample containers. Results from tests performed at varied argon and xenon gas pressures and varied water temperatures indicate that rapid bubble condensation prevents the transport of the vapor bubble to the surface, except when the water temperature is initially near the boiling point. These results require cautious interpretation in terms of LMFBR aerosol source attenuation for two reasons: (1) water may not be an appropriate simulant for sodium, and (2) bubbles produced are smaller than those expected to be formed in LMFBR accidents and, so, would be more likely to rapidly condense.

In summary, results from the argon gas and under-water vaporization tests provide evidence that considerable reductions in the aerosol source that could be released to the secondary containment in LMFBR HCDAs are possible. To verify these observations, uncertainties in the estimates of fuel energy states after capacitor discharge must be determined, under-sodium tests must be performed, and the results evaluated and compared to those from under-water tests. Careful scaling of the results from these tests to larger-scale accident conditions must also be performed.

INTRODUCTION

Safety analyses prepared as a part of the licensing process for Liquid Metal Fast Breeder Reactors (LMFBRs) must provide estimates of the radiological consequences - that is, determine the aerosol "source term" - for hypothetical core disruptive accidents (HCDAs). In such accidents, some of the reactor fuel is assumed to reach highly energetic states (fuel temperatures of the order of 6000 K) and to then "disassemble" and produce a fuel/sodium vapor bubble that can transport fuel and fission products through the liquid sodium and cover gas to be released into the reactor secondary containment.

Important issues that need to be addressed to permit estimates of the fuel release via this route from the reactor primary containment in HCDAs are the following:

1. In calculating the "initial" contents of the fuel bubble produced - after the fuel expands to the surrounding system pressure - the expansion is assumed to occur such that the maximum amount of vapor is produced (by an adiabatic expansion). Two issues related to this initial vapor source are: a) whether less than the adiabatic vapor source limit can be assumed; or b) whether during fuel expansion aerosol-sized droplets (by fragmentation) are formed, that could be transported up in the bubble and so add to the initial aerosol source more fuel than assumed by adiabatic expansion.
2. It has been estimated that the fuel/sodium bubble produced would have an ≈ 2 second residence time within the sodium of the primary containment. In this time, there is potential for condensation of the bubble - due to radiation heat loss from the vapor, condensation at the vapor-sodium interface or onto droplets entrained in the bubble, condensation on structure in the bubble path, or fuel condensation due to cooling of fuel vapor by mixing with sodium vapor. The important condensation mechanisms and the likely amount of condensation must be determined. After the fuel has condensed, however, it may convert to an aerosol that could be carried through the coolant by the fission product gases that are present in the bubble or by uncondensed vapors.

This paper presents results from aerosol source term experiments performed in the ORNL Aerosol Release and Transport (ART) Program sponsored by the U.S. NRC. A technique designated capacitor discharge vaporization (CDV) was developed at ORNL¹ to heat portions of uranium dioxide fuel simulant samples to the high energy states (up to and greater than 3500 J/g) that could be produced at core disassembly in HCDAs. These fuel samples are then allowed to "disassemble" in different surrounding environments. Two series of the tests are discussed in this paper: a) vaporizations in an argon gas environment; and b) vaporizations under a water pool. Vaporizations in a sodium pool

will eventually be performed; however, in this paper the relation of the water test results to sodium tests will be discussed.

DESCRIPTION OF CDV TECHNIQUE AND TEST FACILITY

A schematic of the fuel samples used in the capacitor discharge experiments is shown in Figure 1. Each sample consists of a stack of UO_2 pellets, weighing ~ 20 grams and about .1 m in length, that is surrounded by about 30 grams of UO_2 microspheres (microsphere diameters are $\sim 350 \mu\text{m}$). The pellets and microspheres are contained in a quartz tube.

Sample heating by the CDV technique is performed in two stages, preheat and capacitor discharge. Heating of the fuel to high temperatures requires rapid heating rates of the order of 10^6 Watts/gram. This is not possible when the fuel samples are at room temperature, where their electrical resistance is $\sim 10^6$ ohms. However, UO_2 has a highly negative dependence of resistivity with temperature-proportional to T^{-5} . Samples are preheated at various levels of constant power until a final level of ~ 2000 Watts is reached where a substantial fraction of the pellet mass is expected to be molten. The presence of the microspheres permits pellet melting without melting of the quartz tube. At this point, the sample resistance is typically ~ 0.5 ohms. A two-second "down time" between preheat and capacitor discharge is used; this results in a more uniform temperature distribution through the pellets at the start of capacitor discharge. The capacitor discharge phase typically lasts for 2-5 ms, during which energy input rates to the fuel are at ~ 10 MW levels. Energy input to the fuel occurs until the internal fuel pressure bursts the quartz tube. Capacitor discharge voltage and current data are recorded to permit the amount of energy input to the fuel sample to be determined.

The fuel sample schematic shown in Figure 1 indicates that a steel tube (.13 mm wall thickness) surrounds the quartz tube; for tests discussed here that were performed in an argon gas environment, this steel tube was not present. For the under-water, and later under-sodium, tests, the presence of the steel tube permits energy input to the fuel without shunting to the surrounding liquid. In the gap between the steel and quartz tubes (this gap has a volume of $\sim 10^{-5} \text{ m}^3$) xenon gas is inserted. Xenon is used to simulate non-condensable fission products that could be in HCDA fuel bubbles. The amount of xenon present is one of the variables in the under-water and under-sodium tests.

Figure 2 is a simplified illustration of the facility in which the tests are performed. The capacitor system consists of six "banks," each containing ten capacitors. Each capacitor can store 3 kJ of energy at 2500 Volts, so the total capacitor energy storage capability is 180 kJ.

The tests in an argon gas environment were performed in the Containment Research Installation-III (CRI-III) vessel. This vessel has a $.56 \text{ m}^3$ volume,

and is equipped with aerosol sampling equipment to permit measurement of the airborne aerosol concentration vs time (by filter sampling) and determination of the size distribution of the primary aerosol particles (by collecting samples on photomicrograph grids and viewing these under an electron microscope). High-speed movies (at 10,000 frames/sec) are taken of fuel heating, sample rupture, and fuel expansion.

Under-water (and later under-sodium) tests are performed in the Fuel Aerosol Simulant Test (FAST) vessel. This is a .46 m³ vessel that is 1.83 m high and .61 m in diameter, roughly 1/10 the linear dimensions of the primary containment for the proposed Clinch River Breeder Reactor. The vessel is instrumented to permit measurement of the pressure transients produced due to bubble expansion and collapse, and also to make measurements of the properties of aerosols transported to the argon cover gas above the liquid. High-speed movies are taken during the water tests, and an acoustic detection system will be used to track the bubbles formed in under-sodium tests.

ARGON GAS FUEL VAPORIZATION EXPERIMENTS

Fuel vaporization experiments were performed in an argon gas environment (in the CRI-III vessel) because it was expected that after sample rupture, the fuel would undergo an essentially free expansion in the argon gas environment.

After the fuel vapor is produced, it rapidly cools, due to mixing with argon gas, and forms an aerosol. Measurements are made of the mass of airborne material in the argon gas as a function of time; extrapolation to zero time permits the initial airborne mass to be estimated. This measured airborne mass can be compared to the amount of fuel calculated to have vaporized, in an assumed free expansion after capacitor discharge occurs.

More than 50 capacitor discharge experiments have been performed at various levels of preheat and for different capacitor bank charging configurations. In these tests, the argon gas pressure in the test vessel was ~.101 MPa. Results from three of these tests, all having the same preheat level before capacitor discharge and spanning the range of energy inputs, are summarized in Table I.

Table I

CDV Energy Inputs and Measured Aerosol Yields for Selected CDV Tests Having 2200 Watt Final Preheat (In Each Test, the Pellet Mass Was ~22 Grams and the Microsphere Mass Was ~36 Grams)

Test	CDV Energy Input (kJ)	Measured Aerosol Yield (grams)
CDV 22	8.0	.54
CDV 17	29.7	1.67
CDV 51	43.9	6.82

In a previously published report,² aerosol yields from a number of tests were compared with yield calculations from models for the electrical preheat, capacitor discharge, and the subsequent fuel expansion. These calculations used a temperature distribution calculated for preheat as input to the capacitor discharge calculation, and assumed an adiabatic fuel expansion, after capacitor discharge, to the surrounding argon gas pressure. For low CDV energy inputs (like that in CDV 22) the calculated yields were less than those measured. However, for higher energy inputs (like those in CDV 17 and 51) calculated vapor yields were a few times the measured aerosol yields.

However, two effects were not included in these preliminary calculations that could considerably change the results. These are:

1. An apparent increase in the electrical resistivity of UO_2 as it is heated through melt - this information comes from recent CRI-III experiments;³
2. The influence of the cooling of the fuel sample during the two-second time period between preheat and capacitor discharge.

In a recent series of CRI-III tests (done in argon gas) the preheat level before capacitor discharge was set well below that expected to produce fuel melt. A typical set of capacitor discharge voltage and current data from these tests is shown in Figure 3. The abrupt decrease in current (corresponding to an ~25% resistance increase) at 2.9 ms is not observed in tests in which the pellets are melted prior to capacitor discharge during preheat. An increased UO_2 resistivity at temperatures greater than melt would lead to more energy being deposited to the microspheres (and less to the pellets) than previously expected; this would reduce the calculated vapor yield.

Results of parametric calculations for the influence of a resistivity increase by a factor of 2 through melt (MR = 2 corresponds to this, MR = 1 corresponds to no change) and also for the two-second cooling period between preheat and CDV are shown in Figure 4. These results indicate a) that the cooling of the sample between preheat and CDV must be included in vapor yield calculations, and b) yields calculated assuming a resistance increase through melt are, for a given energy input, considerably less than calculated without the melt resistance change. These results strongly suggest that the UO_2 electrical properties at high temperatures need to be accurately determined to allow confidence in the comparisons of measured and calculated vapor yields from CRI-III argon gas experiments.

Figure 5 shows the cumulative primary aerosol size distribution from three CRI-III tests. As shown, the aerosol was log-normally distributed; this distribution was found to be relatively insensitive to the CDV input energy for each test. This may be similar to the primary aerosol distribution that would be produced when fuel vapor is released from an HCDA bubble to the reactor cover gas and secondary containment. It is not certain, however, that these particles do not come from a combination of aerosol nucleation and liquid fragmentation when UO_2 liquid is dispersed after capacitor discharge.

A series of tests performed in cooperation with Sandia Laboratories personnel⁴ may help to answer this question; in these tests, the debris produced immediately (less than 3 ms) after capacitor discharge is sampled in a vacuum environment. The size distribution of dispersed droplets can be determined and will be compared to primary aerosol size distributions.

UNDER-WATER FUEL VAPORIZATION TESTS

Nearly thirty under-water tests have been performed in the FAST facility within the past year. As outlined in Table II, these tests have been performed over a wide range of system pressures and temperatures, and also with

Table II

Test Parameters Varied In FAST Water Tests Performed To Date (All Tests Performed With Water Height Above Test Sample - 1.12 m)

Test Parameter	Values Used
System (Water Pressure = P_w)	$P_w = .025 \text{ MPa},$ $.101 \text{ MPa},$ 2.02 MPa
System (Water) Temperature = T_w	$T_w = 298 \text{ K},$ 363 K
Xenon Gas Pressure In Test Sample = P_x	$P_x = .101, 505 \text{ MPa}$ for $P_w = .101 \text{ MPa}$
	$P_x = .505 \text{ MPa}$ for $P_w = .025 \text{ MPa}$
	$P_x = 2.02 \text{ MPa}$ for $P_w = 2.02 \text{ MPa}$

large variations in the pressure of the xenon gas in the 10^{-5} m^3 volume in the test sample. These test conditions were varied for the following reasons:

1. Variation of system pressure should cause the amount of fuel vapor produced in the bubble to vary, and so should influence bubble size and bubble dynamics;

2. Variation of system temperature should cause the amount of water vaporized to vary. If a large amount of water vapor is formed, it may carry condensed fuel aerosol through the water to the cover gas;
3. One analysis for UO_2 condensation, neglecting radiation heat loss, in HCDA-sized (~ 3 m diam) bubbles indicates⁵ that fission product gases in the bubbles can greatly reduce the fuel condensation rate. Variation of the xenon gas pressure may provide insight as to whether this is the important condensation mechanism, or if radiation heat transfer dominates. The xenon also provides a medium for transport of condensed fuel through the coolant.

All of the tests discussed in this paper were performed under the same pre-heat conditions and had CDV energy inputs in the 30-40 kJ range. Therefore, we can assume all fuel samples had roughly the same energy content when bubble expansion began. In addition, all tests were performed with the liquid height above the test sample set at 1.12 m.

The major measurements made were of the pressure transients due to bubble expansion and collapse and of the amount of aerosol transported to the cover gas; visual results, from high-speed movies, were also produced. Pressure measurements were made using a Kaman Sciences transducer mounted 23 cm from the test sample. A typical pressure record is shown in Figure 6. The early-time pressure pulse is due to the initial sample rupture, while the later pulses occur due to the collapse and re-expansion of the bubble. We will call the time between the first and second pulses, T_p , the period between bubble formation and the first bubble collapse.

Values of the bubble period, T_p , as a function of test conditions are presented in Table III. As expected, increasing the system pressure causes the bubble period, T_p , to decrease. Two other results that are interesting to note are:

1. Increasing the xenon gas level for a fixed system pressure had very little effect on the initial bubble period. This could mean that the presence of the inert gas has little influence on bubble condensation during the time, T_p ;
2. Increasing the system (water) temperature to its boiling point resulted in a substantial increase in T_p (from 55 to 80 ms). This is a good indication that a significant amount of water was vaporized during the time, T_p .

For all tests except those performed at high water temperatures, no fuel vapor or aerosol was transported to the argon cover gas region (based on mass

Table III

Measured Initial Bubble Periods T_p For Various Test Conditions (For 1.12 m Water Height)

Test Conditions	Initial Bubble Period T_p (ms)
$P_w = P_x = 2.02 \text{ MPa}$ $T_w = 298 \text{ K}$	~ 4
$P_w = P_x = .101 \text{ MPa}$ $T_w = 298 \text{ K}$	~ 60
$P_w = .101 \text{ MPa}$ $P_x = .505 \text{ MPa}$ $T_w = 298 \text{ K}$	~ 55
$P_w = .101 \text{ MPa}$ $P_x = .505 \text{ MPa}$ $T_w = 363 \text{ K}$	~ 80
$P_w = .025 \text{ MPa}$ $P_x = .505 \text{ MPa}$ $T_w = 298 \text{ K}$	~ 180

measurements made in the gas space). Visual evaluation of the high-speed films for all tests indicates that there was an aerosol in the bubble within 100 to 200 ms of the start of bubble formation. This is an indication that in the water tests done at low water temperatures, there is no bubble rise phase - all condensation occurs in the early expansion and contraction phase.

In the water tests with water temperatures of ~ 360 K, however, measurements in the cover gas indicate that ~ 1 gram of fuel is transported through the water. Assuming that the bubble contains roughly 2 grams of fuel vapor initially (based on argon gas test results), this means that $\sim 5\%$ of the initial source seemed to be transported to the cover gas in these tests. The likely mechanism for this transport is a water vapor bubble produced by cooling of the hot fuel vapor and liquid in the bubble.

The water experiments were done as a prelude to performing sodium tests, mainly to permit visual observation of the formation and dynamics of fuel vapor bubbles. Interpretation of the condensation results (i.e. total condensation in 100-200 ms) in terms of LMFBR source term prediction should be done cautiously at this time for two reasons: water is not necessarily a good simulant for sodium, and the bubbles formed in these tests are considerably smaller than those possible in HCDA's.

The main reason that water is not a good simulant for sodium is that the thermal conductivity of water is much less than that of sodium. Based on results obtained in the under-water tests, one might expect that bubbles will collapse even more rapidly in sodium. This is certainly possible, but, since the potential for water vaporization is large, it is also possible that water vapor is being produced in the bubble. This water vapor could mix with much hotter UO_2 vapor and cause very rapid UO_2 condensation. This mixing-condensation mechanism may not be present in sodium tests.

Because bubbles produced in FAST tests are much smaller than those possible in HCDAs, rapid condensation in water or sodium tests may not mean rapid condensation in HCDAs. HCDA bubbles may be as large as 1.5 m in radius and are expected to have a roughly 2-second rise time. In contrast, bubbles produced in the FAST water tests seem to have radii as large as .1 m. This means that the surface-to-volume ratio for FAST bubbles may be 15 times that for HCDA bubbles. The condensation that occurs in the FAST tests is likely to be more rapid as a result of the large bubble surface-to-volume ratios.

SUMMARY

In summary, the following points can be made:

1. Fuel vaporization tests have been performed in an argon gas environment. The purpose of these was to compare the assumption of an adiabatic fuel expansion after capacitor discharge with the quantities of fuel aerosol

airborne in the experiments. Such a comparison would tend to indicate the margin available in using this assumption to establish maximum aerosol source due to an HCDA and will serve as a base line to establish the attenuation potential due to the bubble contact with sodium in later experiments. The comparison with the adiabatic assumption is very promising but not yet definitive primarily due to lack of knowledge of UO_2 properties (particularly electrical resistivity) at high temperatures. Measurements have been made of the primary aerosol size distribution produced in the argon tests. It is not yet clear that the airborne particles are produced by aerosol nucleation due to mixing of fuel vapor with argon gas, by fragmentation of liquid drops to small sizes, or by some combination of the two.

2. Under-water fuel vaporizations have been performed for a variety of test conditions. Rapid bubble condensation has been observed to occur in these tests, in roughly 100 to 200 ms after capacitor discharge. When the water was heated to near its boiling point aerosol was transported through the water to the argon cover gas above it. This is the only condition so far in which transport of fuel to the cover gas has occurred; it is likely that the fuel was transported in a water vapor bubble produced in these tests. This result implies that if significant sodium vaporization occurs in an HCDA (for instance, when sodium near the core is close to its boiling point) the possibility exists for fuel transport through the coolant in a sodium vapor bubble. However, water is not a good simulant for sodium, so the water test results should be evaluated with caution. In addition, the fact that the FAST tests are small scale implies that rapid bubble condensation could occur, and the results for under-water and under-sodium tests should be carefully interpreted and scaled.

ACKNOWLEDGEMENTS

This research is sponsored by the Office of Nuclear Regulatory Research, Nuclear Regulatory Commission, under Interagency Agreement DOE 40-551-75 with the U. S. Department of Energy under Contract No. W-7405-eng-26 with the Union Carbide Corporation Nuclear Division.

The authors wish to acknowledge the efforts of J. M. Rochelle (of the Instrumentation and Controls Division at ORNL) and H. E. Robertson in performing the experiments described in this paper.

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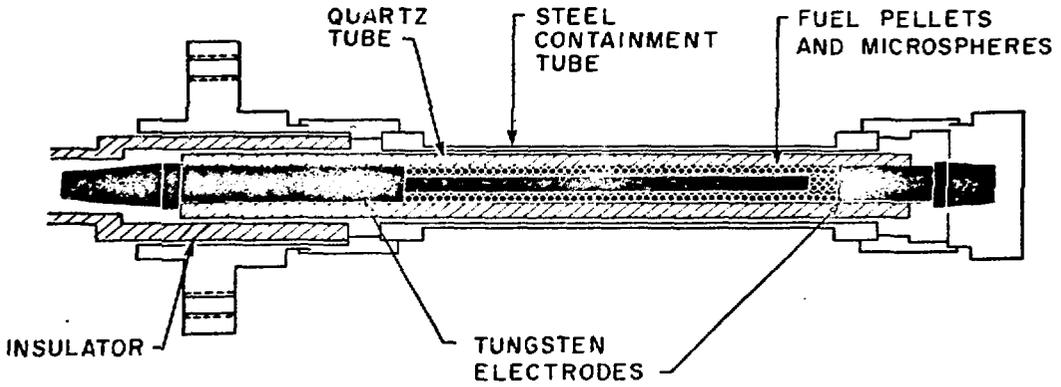


Fig. 1. Schematic of Test Sample Used In Capacitor Discharge Experiments; Sample Mounted Horizontally In Test Vessel As Shown. Steel Containment Tube Present In Under-Water Tests, Not In Argon Tests Discussed In This Report

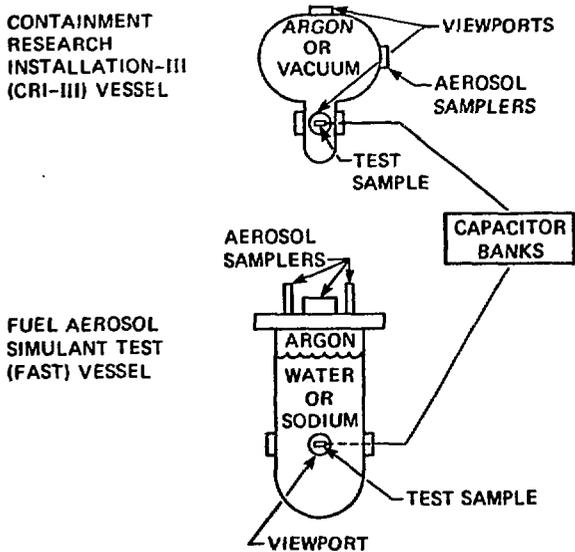


Fig. 2. Test Facility For Capacitor Discharge Fuel Vaporization Experiments

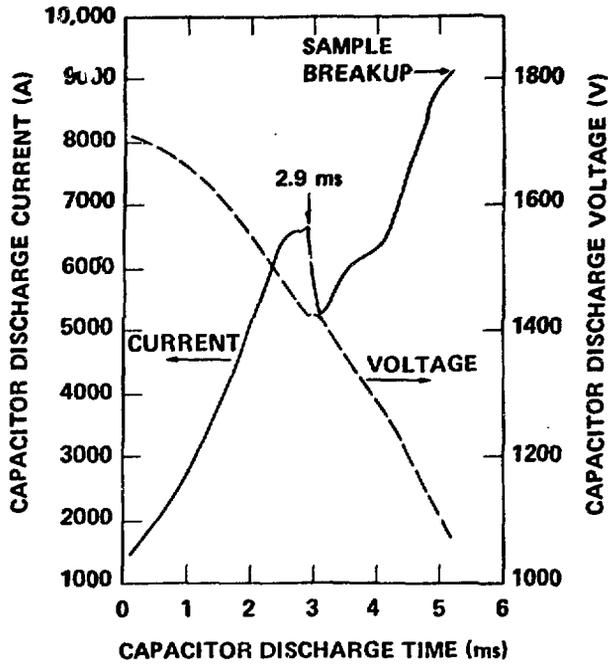


Fig. 3. CDV Voltage And Current Data From CDV 83, Test Where Preheat Level Used Did Not Produce Fuel Melt. Drop In Current At ~ 2.9 ms Attributed To Increase In UO_2 Electrical Resistivity When Fuel Is Heated Through Melt.

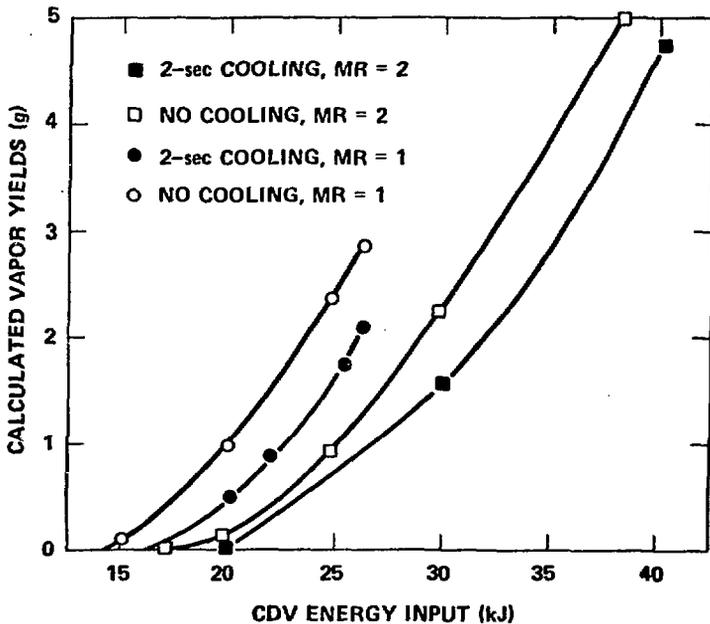


Fig. 4. Parametric Calculations For Vapor Yield vs CDV Energy Input. Initial Condition For Each Case Was Calculated Temperature Distribution After Preheat With Pellet Center Temperature = 3100 K, Slightly Below Fuel Melt. MR = Assumed Change In UO_2 Electrical Resistivity That Occurs As Fuel Is Heated Through Melt. "2 Second Cooling" Refers To Time Between Preheat and CDV, When Sample Is Not Being Heated.

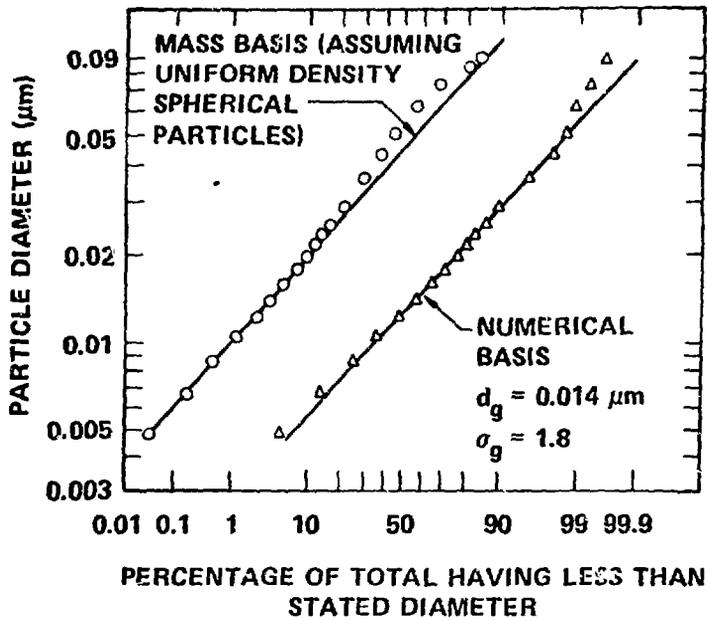


Fig. 5. Cumulative Primary Particle Size Distribution Data From Three CRI-III Argon Tests; 5330 Particles

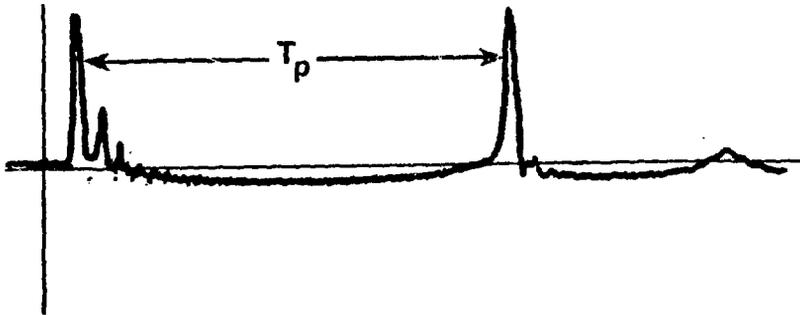


Fig. 6. Typical FAST Under-Water Test Pressure vs Time Data (from FAST 22). Pressure Pulses Shown Occurred At 0.9, 58.3, and 86.8 ms After Sample Breakup And Had Magnitudes Of 1.45, 1.40, and 0.23 MPa, Respectively. Pressure Was Measured \approx 23 cm From Test Sample. Time Between First Two Pulses Is Time Noted As T_p In Text.