CONF-790816--59

MOLTEN CORE DEBRIS-SODIUM INTERACTIONS: M-SERIES EXPERIMENTS

by

E. S. Sowa, J. D. Gabor, J. R. Pavlik, J. C. Cassulo, C. J. Cook, and L. Baker, Jr.

MASTER

Prepared for

ANS/ENS International Meeting

on

Fast Reactor Safety Technology

Seattle, Washington

August 19-23, 1979

UNIVERSITY OF CHICAGO-AUA-USDOE

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

Operated under Contract W-31-109-Eng-38 for the

U. S. DEPARTMENT OF ENERGY

MOLTEN CORE DEBRIS-SODIUM INTERACTIONS: M-SERIES EXPERIMENTS

E. S. Sowa, J. D. Gabor, J. R. Pavlik, J. C. Cassulo, C. J. Cook, and L. Baker, Jr.

Reactor Analysis and Safety Division
Argonne National Laboratory

ABSTRACT

Five new kilogram-scale experiments have been carried out. Four of the experiments simulated the situation where molten core debris flows from a breached reactor vessel into a dry reactor cavity and is followed by a flow of sodium (Ex-vessel case) and one experiment simulated the flow of core debris into an existing pool of sodium (In-vessel case). The core debris was closely simulated by a thermite reaction which produced a molten mixture of UO_2, ZrO_2, and stainless steel. There was efficient fragmentation of the debris in all experiments with no explosive interactions observed.

INTRODUCTION

This paper describes the results of the continuation of the M-Series of molten core debris-sodium interaction experiments, M5, M6, M7, M8, and M9. Experiments M1, M2, and M3 were previously reported [1, 2]. The method of producing the molten core debris was by highly exothermic chemical (thermite) reactions between metals such as uranium and oxides of less active metals. The core debris simulated the situation that would result from core disruption in an oxide-fueled, sodium-cooled, fast breeder reactor.

The purposes of the interaction experiments were to determine fragmentation characteristics, including completeness of fragmentation, particle size distribution, and pressure pulses associated with the interaction. Such studies are applicable to the prediction of the course of events in very unlikely hypothetical core disruptive accidents (HCDAs) in reactor plants. A fragmented bed is readily cooled by the sodium which can permeate the loosely fragmented structure, whereas a bed of dense fuel would not be readily cooled. Two categories of situations are important (1) "In-vessel", and (2) "Ex-vessel". "In-vessel" refers to the situation where molten core debris (fuel and cladding) is ejected from the core region into a sodium-

filled plenum either above or below the core region within the reactor vessel. "Ex-vessel" refers to the situation where the core debris emerges from the reactor vessel and guard vessel and enters a dry steel-lined cavity. In this latter case, the sodium inventory of the reactor vessel would be expected to flow downward through the breach in the reactor/guard vessel and begin to interact with the molten core debris. The early experiments, M1, M2, and M3, simulated in-vessel conditions where molten core debris entered a pre-exisitng sodium pool. Experiment M8 was also an in-vessel simulation. Experiments M5, M6, M7, and M9 were performed under ex-vessel conditions where the core debris began to enter a dry cavity before sodium entry began.

The earlier study [1] had indicated that molten UO_2-sodium interactions are relatively mild and did not result in significant conversions of thermal energy to mechanical work. Only sporadic short-rise time pressure pulses were observed in the gas phase above the sodium pool. Accordingly, no violent interactions were expected in the continuation of the experimental series and the program was designed primarily to investigate the completeness of fragmentation and to characterize the particulate.

EXPERIMENTAL METHOD

In the earlier experiments, M1, M2, and M3, the molten mixture was generated in a thermite reaction vessel and the flow emerged directly downward from the vessel through a flow director into a sodium-filled crucible. One of the concerns expressed about the results of those experiments was that some pre-fragmentation of the core debris might have occurred because of the vigor of the expulsion of material from the vessel. To circumvent this problem in the current series of experiments, a vortex tube injection system was developed that would tend to separate any driving gas from the liquid product mixture and dissipate some of the energy of propulsion through tangential flow in the vortex tube.

Several injection experiments were performed with 3-to-4-kg charges using the vortex tube in the open without any problem. Figure 1 shows individual frames taken from motion pictures. The outline of the spray pattern using the early molybdenum-containing mixture had the general shape indicated in Fig. 1a. The nature of the spray pattern indicated that the mixture was flowing at a significant velocity because of pressures generated within the thermite vessel. However, the thermite mixtures used in tests M1 through M5 are atypical because molybdenum is not normally present in large quantities in fast reactor core loadings. Accordingly, a thermite composition was developed with would simulate core debris more accurately [3]. The composition of the mixture used in experiments M6 through M9 is shown in the second column of Table I. The modified thermite, vortex tube, and pressure venting produced the pattern seen in Fig. 1b.

The test apparatus (see Fig. 2) contained a reaction vessel which received the molten core debris and the sodium, and whose bottom simulated the base of the reactor cavity. The schematic of the apparatus is shown in Fig. 3.

TABLE I

Thermite Reaction Mixtures

Charge Composition	Mixture Used in M5, g	Mixture Used in M6, M7, M8, M9, g
MoO_3	1200	-----
Fe_2O_3	----	654
CrO_3	165	272
NiO	----	117
MnO_2	----	24
SS shot	----	354
U	3000	1612
Zr	----	409
TOTAL	4365	3442
Theoretical Temperature	3620°C	3035°C

TABLE II

Experimental Conditions and Results for UO_2-Sodium Interaction Experiments

No.	Thermite Mixture	Quantity Entering Sodium, kg	Core Debris Loading kg/m^2 Crucible	Median Particle Diameter μm	Peak Base Temp. °C	Peak Vessel Pressure kPa
Ex-vessel Configuration						
M5	UO_2, Mo	1.8	102	250	970	800
M6	UO_2, ZrO_2, SS	0.76	43	400	225	70
M7	UO_2, ZrO_2, SS	0.72	41	550	400	760
M9	UO_2, ZrO_2, SS	1.00	57	680	625	100
In-vessel Configuration						
M1	UO_2, Mo	0.64	20	510	---	---
M2	UO_2, Mo	1.4	42	300	---	---
M3	UO_2, Mo	3.1	95	200	---	---
M8	UO_2, ZrO_2, SS	1.15	65	300	---	175

EXPERIMENTAL RESULTS

The results from each experiment included the output traces from the electromagnetic flowmeter, the time traces of the reaction vessel pressure, temperature-time traces from thermocouples located on the outside of the base of the reaction vessel (simulated reactor cavity liner), and output traces from the strain gauges also located on the outside of the base of the reaction vessel. A typical experiment is described below in detail and the others are described more briefly. Table II summarizes the conditions and pertinent results for these experiments.

Experiment M5. After ignition of the thermite, the molten charge leaving the thermite vessel contacted TC6 whose output began to rise rapidly. This was taken as time 0. At 0.5 s, power was applied to the solenoid by the parting of the burn wire at the mouth of the vortex tube. At the same time, the strain gauge S-1 at the outer base of the reaction vessel began to read, indicating that the first of the UO_2 contacted the inner face of the reaction vessel.

At 0.7 s, the pressure P-3 in the sodium vessel rose to its maximum level when the ball valve opened, admitting the gas from the driver vessel. Simultaneously, the flowmeter E-1 showed the movement of sodium, which began to flow into the reaction vessel. The sodium flow appeared to be continuous until 2.0 s, when fluctuations appeared as the flow decayed to zero at 3.0 s.

At the time the start of sodium flow was indicated, a pressure pulse was recorded on P-2, the reaction-vessel pressure gauge. This pulse, lasted for only 10 ms and was about 800 kPa in magnitude. The pulse resulted from the initial contact of UO_2 and sodium. At about 2.0 s the strain-gauge reading reached a peak of 9 mV. This coincides with the rise of T-4 toward its peak value of 970°C at 3.0 s.

The initial rise to the peak of the strain gauge resulted from heating of the reaction-vessel inner face from room temperature on contact with the UO_2. As the temperature of the external surface of the vessel base began to increase, the thermal stress and the resulting strain decreased somewhat. However, continued heating of the base by fuel and expansion of the vessel walls in contact with 425°C sodium caused a continuing increase in strain up to 8 s when saturation of the amplifier prevented further recording.

Postexperimental examination of the apparatus showed that, of the 4200-g charge loaded into the thermite container, 1292 g remained in the thermite vessel. The remainder contacted about 8.3 liters of sodium. The pressure spike caused ejection of 185 g of UO_2 upward into the vent baffle, where it adhered to the stainless steel plate. A total of 408 g was retained as a block in the mouth of the vortex tube, and 1802 g was recovered as a particulate from the reaction vessel.

Removal of the sodium showed that fragmentation of UO_2 was substantially complete. However, significant quantities of molybdenum were frozen

to the base as globules. The entering velocity of the sodium was found to be about 10 m/s and fuel and sodium entry were nearly simultaneous.

Experiment M6. This experiment was performed in the same way as M5 except that the stainless steel thermite mixture was used. Very few steel particles were frozen to the base and as a result the thermal transient on the base was much more mild than in M5.

Experiment M7. It was planned to have a 5-s delay before injection of the sodium. However, premature entry of ∿23% of the sodium produced results very similar to those of M6. The experiment was repeated as M9 where a rupture disc was used to prevent inadvertent injection.

Experiment M8. The stainless steel thermite mixture was dropped into a preexisting pool of sodium. There was no attack on the base and fragmentation was complete.

Experiment M9. In this experiment, there was a 5-s delay in the injection of any sodium. The delay provided time to freeze a layer of the oxide-steel mixture. A map of measured layer depths is shown in Fig. 4. The average depth was 3.3 mm. The remainder of the material was fragmented.

Characterization of Particulate. After the experiment, the sodium was separated from the thermite debris by reaction with 100% ethanol and the sodium salts dissolved in a water wash. The particulate debris was wet-seived to separate particles in the range 53 to 5560 mm. The fine particle (-53 to +0.1 μm) distribution was measured by sedimentation with a Sedi-Graph 5000 particle size analyzer.

The particle size distributions are shown in Fig. 5. The results show that molybdenum formed particles much larger than UO_2, Fig. 6, whereas stainless steel particles were only 2 to 4 times larger than the UO_2, Fig. 7. The UO_2 particles were irregular and had sharp edges indicating fragmentation from a solid material. The metallic particles tended to be rounded and smooth indicating fragmentation in the molten state.

DISCUSSION OF RESULTS

The results of the experiments presented herein confirmed those of the earlier experiments in that no vapor explosions were observed. The interaction events could be described qualitatively as rapid heat transfer and transient boiling processes without the generation of damaging pressure pulses.

The first ex-vessel experiment, M5, used the same thermite mixture employed in the early series of in-vessel experiments, which generated a mixture of molten UO_2 and molybdenum metal at a temperature approaching 3400°C. The core debris mixture was vigorously ejected from the thermite reaction vessel as a spray in the experiment.

The remaining experiments were performed with the new thermite mixture, which is believed to be a more accurate simulation of the oxide-fuel debris that would be generated by an HCDA in a sodium-cooled fast reactor. The mixture produced an oxide phase consisting of 77 wt % UO_2 and 23 wt % ZrO_2 which has properties similar to mixed oxide fuel. The metal phase is stainless steel. The relative proportions of the steel and oxide phases (31 wt % steel) were similar to that of FFTF driver fuel (35 wt % steel). The theoretical temperature of the thermite-generated material was 3035°C, which is close to that expected to result under HCDA conditions.

In the ex-vessel experiments, fragmentation was substantially complete for the steel-containing core debris when there was little or no delay in the entry of sodium to the simulated reactor cavity. When the entry of sodium was delayed for slightly more than 5 s, a frozen layer of core debris having an average thickness of 3.3 mm adhered to the cavity base. In the experiment with the molybdenum-containing core debris, the UO_2 was fragmented; however, the molybdenum was present as large globules, some of which were fused to the steel base. This behavior was believed due to the high density and high melting temperature of molybdenum which are not characteristic of the metal phase of actual core debris. The settling and fusion of molybdenum to the steel base resulted in a severe thermal transient at the base. Only a mild thermal transient was observed in the experiments that involved the steel-containing debris. The transient was equivalent to that expected from the sudden contact of freezing UO_2 with cold steel in which there is no general melting of the interior surface of the steel and the exterior surface does not exceed the sodium temperature so long as there is prompt sodium entry.

The results of the in-vessel experiment were very similar to those of the previous series of in-vessel experiments. The steel-containing debris was completely fragmented into particulate having a median particle size of 300 μm. There was no material found adhering to the base. It is concluded that there was no effect of the change in core debris properties on the fragmentation characteristics under in-vessel conditions.

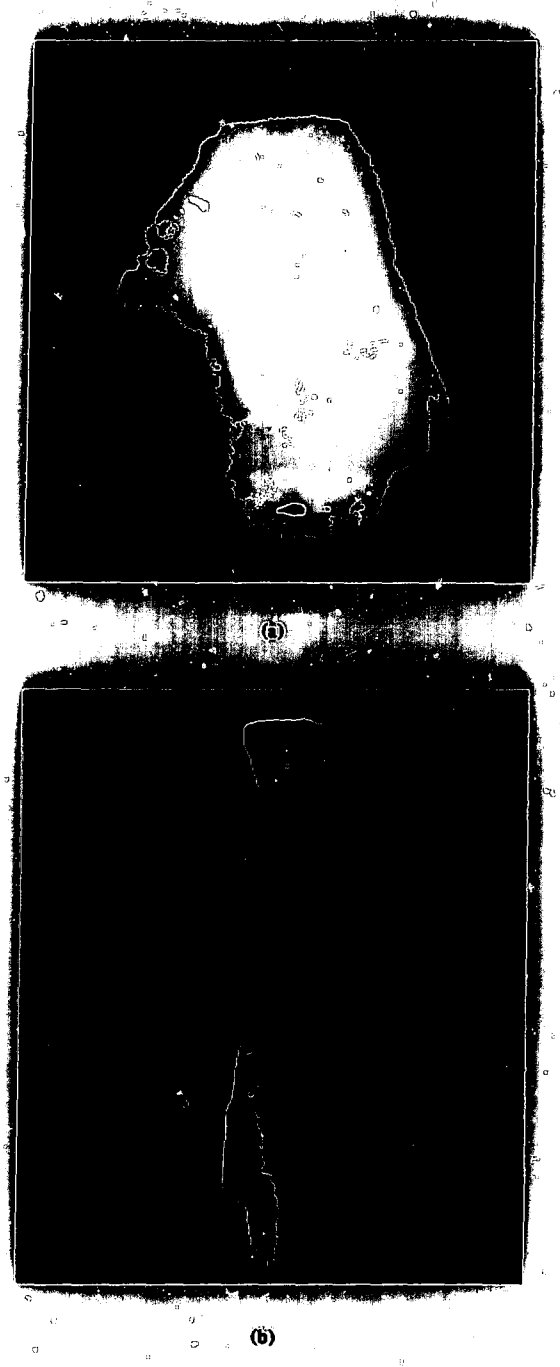
An important result of the experiments is the demonstration of the tendency to form particulate debris. The median particle diameter ranged from 200 to 680 μm for all of the experiments with the stainless steel particles formed 2 to 4 times larger than the core debris particles. The particulate bed formed a highly porous configuration upon the base of the reaction vessel. Every effort was made during these experiments to maintain the compositions and ratios similar to those expected in an LMFBR. Consequently, it is strongly indicative that similar behavior can occur from interactions involving larger quantities of core debris constituting a substantial fraction of an LMFBR core. On the basis of fuel quantity and cavity surface, the experiments are about 1/5000 scale.

REFERENCES

1. T. R. Johnson, L. Baker, Jr., and J. R. Pavlik, "Large-scale Molten Fuel-Sodium Interaction Experiments,", Proc. Fast Reactor Safety Mtg., April 2-4, 1974.

2. T. R. Johnson, J. R. Pavlik, and L. Baker, Jr., "Postaccident Heat Removal: Large-scale Molten-Fuel-Sodium Interaction Experiments," ANL-75-12, February 1975.

3. L. Baker, Jr., et al., "Investigation of Thermite Type Chemical Reactions to Generate Molten Core Materials for Reactor Safety Experiments," Trans. Am. Nucl. Soc., 22, 443 (November 1975).



(b)

Fig. 1. Frames from high-speed motion pictures of ignition tests: (a) molybdenum-containing mixture; (b) steel-containing mixture.

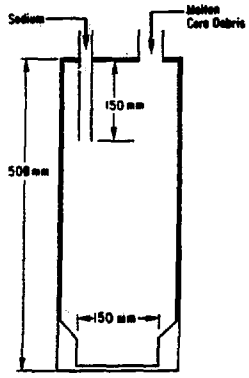


Fig. 2. Reaction vessel configuration used in Ex-vessel Experiments.

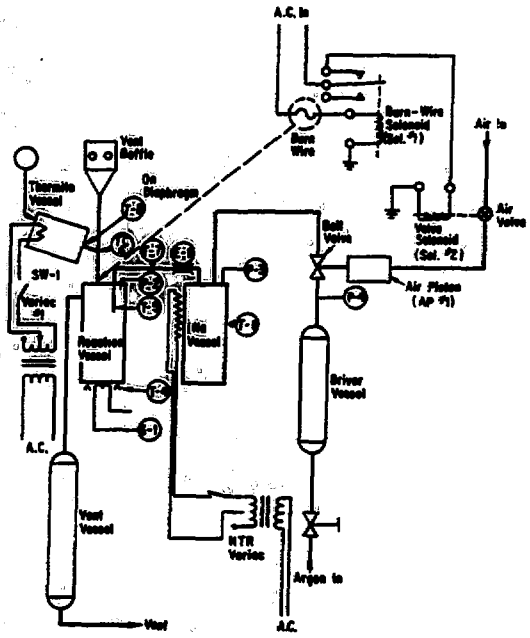


Fig. 3. Schematic of apparatus used in Ex-vessel Experiments.

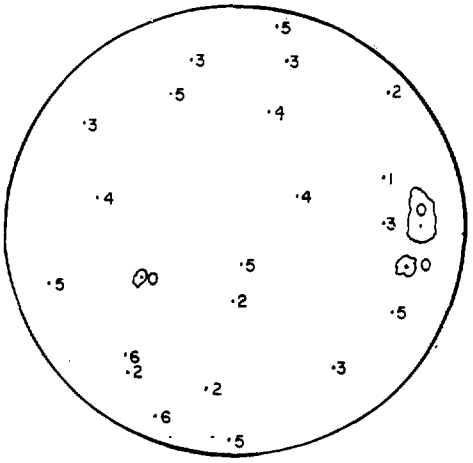


Fig. 4. Map of frozen layer depths in mm from base of reaction vessel, Experiment M9.

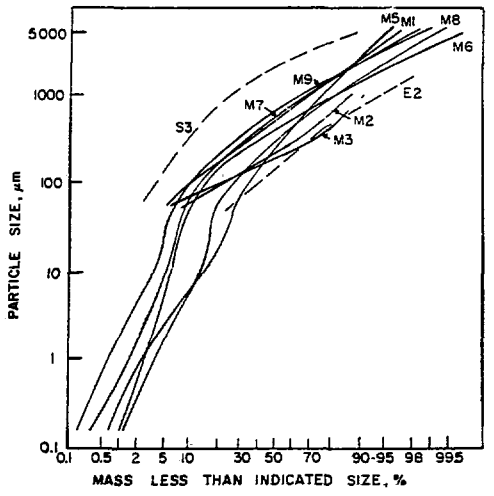


Fig. 5. Comparison of particle size distributions of material from M-series Experiments with range of sizes from In-pile Experiments.

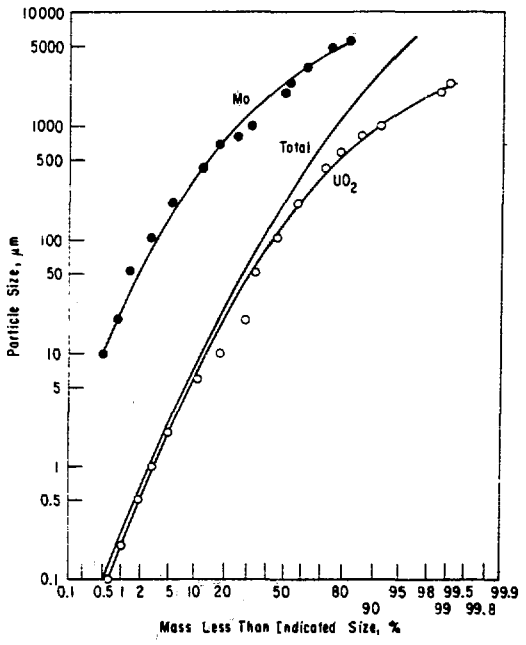


Fig. 6. Particle size distributions for the metal phase and the oxide phase, Experiment M5.

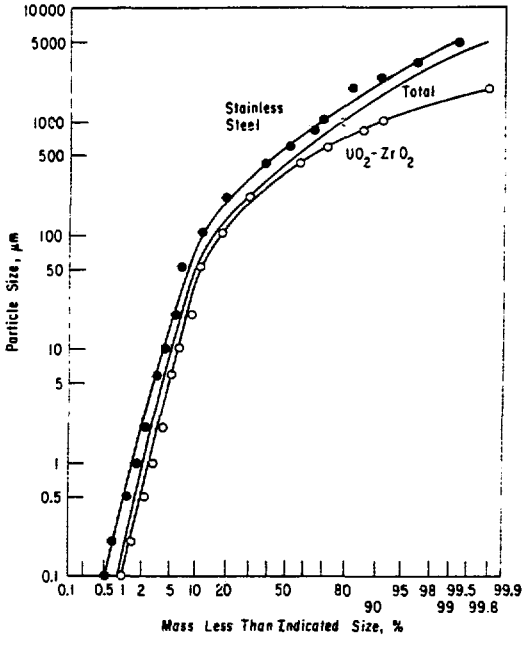


Fig. 7. Particle size distributions for the metal phase and the oxide phase, Experiment M6.