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AI-ERDA-13148

QUARTERLY TECHNICAL PROGRESS REPORT
NUCLEAR SAFETY
CHARACTERIZATION OF SODIUM FIRES
AND
FAST REACTOR FISSION PRODUCTS
JANUARY - MARCH 1975

ERDA Research and Development Report

*Prepared for the U.S. Atomic Energy Commission,
Division of Reactor Research and Development,
under Contract Number AT(04-3)-824*

MASTER

Rockwell International

Atomics International Division
8900 DeSoto Avenue
Canoga Park, California 91304

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The preceding Progress Report was AI-AEC-13144.

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Canoga Park, California 91304

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Program: Characterization of Sodium Fires and Fission Products

ERDA Task: 10 - Characterization of Sodium Fires and Fission Products

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Project Manager: H. A. Morewitz

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Principal Investigators: J. P. Heisler, R. P. Johnson, J. M. Otter, C. T. Nelson,
E. R. Specht, C. Guderjahn, R. Koontz, D. Toppen,*
E. U. Vaughan

I. PROJECT OBJECTIVES

- 1) Develop a computer program for calculating two-dimensional, transient natural convection phenomena arising from various postulated pipe breaks and pool fires in LMFBR heat transfer equipment vaults.
- 2) Develop experimental programs and conduct tests that will characterize the behavior of sodium, sodium oxide, fuel, fission product and other aerosols as they might be generated by various postulated LMFBR accidents.
- 3) Determine by analysis and experiment the generation and transport of these aerosols with respect to source (location, type and configuration) release dispersal, agglomeration attenuation, and removal for the entire course of events associated with real and hypothetical accident conditions.

II. MAJOR ACCOMPLISHMENTS DURING FISCAL YEAR 1975

The fluid mechanics portion of SOMIX-1 was developed. Computer runs showed that the code was computationally stable over the entire range of Grashof numbers tested, which covered all flow ranges up to the onset of turbulent flow. The conversion of SOMIX-2 to cylindrical geometry was completed and excellent agreement was achieved with SOMIX-1 for low Grashof number cases.

*Consultant

Calibration of the Laboratory Spray Modeling (LSM) test apparatus was completed for the various liquids used in the test series. Testing of H₂O, H₂O-5 vol % and H₂O-34 vol % ethanol mixtures were completed and sodium testing initiated. Observations of liquid sheet and drop formation, drop size and spatial distribution were made for each test liquid over a range of 3 to 6 m/sec.

III. PROGRESS DURING REPORT PERIOD

A. SUBTASK A – SODIUM SPLASH DISPERSAL

Five liquids: water, water-5% ethanol, water-34% ethanol, sodium, and NaK 78 were chosen as test fluids for the LSM. These liquids have absolute viscosity and densities within $\pm 9\%$ and surface tension values ranging from approximately 30 to 190 dyne/cm. The physical properties of these liquids are summarized in Table 1.

TABLE 1
LSM TEST SERIES – FLUID PHYSICAL PROPERTIES

Liquid	Test Temperature (°C)	Density ρ (gm/cc)	Absolute Viscosity η (cp)	Surface Tension σ (dyne/cm)
H ₂ O	45	1.000	0.596	68.7
H ₂ O + 5 vol % C ₂ H ₅ OH	45	0.986	0.603	54.9
H ₂ O + 34 vol % C ₂ H ₅ OH	45	0.917	0.917	31.1
NaK-78	45	0.867	0.680	103.0
Sodium	150	0.915	0.541	195.0

The liquid jet tests conducted in the LSM were conducted in a manner that would determine the behavior of a liquid jet as it impacts on a perpendicular surface (height of horizontal plate above nozzle opening to pipe diameter ratio was maintained at 4:1 throughout this test series). The behavior of the liquid sheet formed included measuring the radial distribution, the resulting size and distribution of droplets formed when the liquid sheet falls away from the plate,

and the horizontal velocity (if any) of the droplets at the point of separation from the plate. The behavior of identical liquids at the same test velocity on different horizontal plate materials was also determined.

The LSM, shown pictorially in Figure 1, was designed to expediently conduct the tests described above and determine the efficiency of a spray restricting device which deflects a large fraction of the resulting liquid sheet into a separate reservoir. This sheet-restrictor is required when used on larger pipe diameter jet tests to be conducted in the LTV and on the full scale water tests to be conducted at the large Hydraulic Laboratory at Rocketdyne. The restrictor used in the LSM tests deflected a 300° sector of the liquid sheet, formed on the underside of the horizontal plate by the impaction of the vertical jet into a reservoir. Only 60° of liquid sheet is spread on the underside LSM lid. This device proved satisfactory for all tests conducted. Figure 2 shows a schematic of the LSM test configuration.

Jet velocities chosen for the test series were respectively; 3.04, 4.27, 5.18 and 6.08 m/sec. Two tests were run with each liquid at these velocities to obtain experimental data and photographically document the test. The technique used to determine drop size and mass distributions was to freeze the drops in liquid nitrogen and screen these materials through U.S. standard testing sieves. The experimental results obtained for all of the test fluids are tabulated in Table 2. Figures 3, 4, and 5 show the mass fraction of accumulated droplets measured as a function of radial distance from the nozzle. At 3, 4, and 5 m/sec jet velocities it was observed that 100% of the sheet separated from the lid to form a bell-shaped radial mass distribution of droplets. With each test fluid and at test velocities of 6.09 to 6.34 m/sec a horizontal component developed which consisted of smaller diameter droplets originating directly at the point of jet impaction. The horizontal velocity of these droplets was calculated to be between 3.81 and 5.38 m/sec at their impaction with the LSM wall 0.95 meters from nozzle. As the surface tension of the test fluid was decreased, the mass fraction associated with this horizontal component increased from 0.03 to 0.34 wt %.

The mean drop size, D_{50} , ranged from 0.5 to 0.6 cm over the range of 3 to 9 m/sec jet velocity. Figure 6 shows the D_{50} drop sizes for water, 5 and

Figure 1. Laboratory Spray Modeling (LSM) Test Apparatus

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Figure 2. LSM Test Configuration

TABLE 2
LSM EXPERIMENTAL DATA SUMMARY

Test Fluid	Test Velocity (m/sec)	Drop Size D ₅₀ (cm)	Sheet Dispersion Radius (cm)	Deviation of D ₅₀ (%)
H ₂ O	3.04	0.578	38.0	11.57
	3.04	0.570	38.0	
	4.27	0.526	42.0	
	5.18	0.603	45.0	
	6.09	0.636	55.0	
	6.09	0.612	55.0	
	6.09	0.650	55.0	
H ₂ O + 5 vol % ethanol	3.04	0.546	37.0	8.34
	3.04	0.707	37.0	
	4.27	0.653	42.0	
	5.24	0.629	47.0	
	6.19	0.634	55.0	
	6.19	0.683	55.0	
H ₂ O + 34 vol % ethanol	3.07	0.434	30.0	11.24
	4.32	0.615	34.0	
	5.33	0.624	38.0	
	6.34	0.596	43.0	
	6.34	0.631	43.0

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Figure 3. LSM Water Jet Tests

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Figure 4. LSM 5 vol % Ethanol - Water Mixture Jet Tests

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Figure 5. LSM 34 vol % Ethanol - Water Mixture Jet Tests

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Figure 6. LSM Water Jet Tests [Droplet size as a function of velocity (all fluids)]

34% ethanol-water mixtures. To determine the effect of drag coefficient resulting from interaction of the liquid sheet with various LSM lid materials, tests were run with each test fluid (excluding sodium and NaK) at a velocity of 5.2 m/sec on wetted Lucite, stainless and mild steel. The D_{50} drop size deviation resulting from the water tests increased from 0.60 to 0.74 on mild steel, to 0.80 on stainless steel and to 1.00 on Lucite coated with a wetting agent. Figure 7 shows these drag coefficient effects. Figure 8 shows similar results for 34% ethanol-water mixtures except that the alcohol dissolved the wetting agent coated on the Lucite, and the mean drop size decreased.

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Figure 7. LSM Water Jet Tests [Droplet size as a function of velocity (water only)]

Design and fabrication of the stationary (and falling) sodium drop test apparatus have been completed. These tests will study the burning rate of a sodium droplet, the burning phenomena, and subsequent heat transfer to its environment. Figure 9 shows a schematic of the stationary sodium drop test apparatus.

B. SUBTASK C – SOMIX CODE DEVELOPMENT

Fromm's second order subroutines were converted to cylindrical geometry and programmed into SOMIX-2. Computer runs were made with a Grashof number of 10^{-5} as a trial problem. The runs were numerically stable. However,

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Figure 8. LSM Water Jet Tests [Droplet size as a function of velocity (34% alcohol in water only)]

the calculated results did not correspond to those obtained with SOMIX-1. The discrepancy was discussed with Fromm at IBM-San Jose on January 10, 1975, following which he sent us a slightly revised version of the program along with a sample problem using his first order approximation. An approximate conversion of this to cylindrical geometry gave results which corresponded reasonably well with SOMIX-1. Work was started on a rigorous conversion of the revised version of the code to cylindrical geometry.

Sodium drop drag and combustion models were completed for SOMIX-1. The drop drag routine, using an offset chopped cosine drop distribution, was

9005-4548

Figure 9. Layout of Stationary Sodium Drop Test Apparatus

programmed on the time-share computer and is working satisfactorily. The drop combustion and gas heat transfer programs are currently being checked out on the time-share computer.

The oxygen diffusion and transport equation was developed in a form suitable for programming into SOMIX-1.

Work has been started on developing the turbulent gas flow equations.

C. SUBTASK D – HCDA AEROSOL SOURCE TERM

A Test Plan, TP-707-130-013, was prepared to conduct the extended HCDA aerosol source term experiments. These tests have application in determining

the attenuation of HCDA aerosols which are trapped in a bubble rising through a liquid medium. The aerosols will be introduced with various ratios of condensable gases.

Photographs of further tests, conducted using the test procedure DTP-707-130-002, were made of a bubble having an approximate composition of 90 to 95% condensable steam at 120 psig and 5 to 10% noncondensable air. No aerosols were added to the bubbles during these scoping tests. Figure 10 shows the time sequence history of this bubble.

D. SUBTASK E - AEROSOL LEAKAGE

Fabrication and installation of the aerosol optical density sensors, gas flow accumulator, test fixture plugging sensor, and aerosol sampling system have been completed. Preliminary calibration of the sensors has been completed using sodium oxide aerosol.

A test procedure document,* DTP-707-130, has been completed.

A laboratory for the analysis of sodium samples is being furnished with a flame photometer, pH meter, and supporting equipment. Installation is 30% completed.

E. SUBTASK F - FUEL AND FISSION PRODUCT RELEASE FROM BURNING SODIUM

1. Experimental

A reaction vessel was built and a stoichiometric mixture of Na_2O and UO_2 for the formation of Na_3UO_4 was added with a small quantity of metallic sodium used as a binder.⁽²⁾ The mixture was added into the reaction vessel in an inert argon glove box. The reaction vessel was closed and heated at 800°C for 48 hr. The gas was then evacuated and the sodium removed by vacuum distillation. The sample is presently being analyzed by x-ray diffraction to see if the pattern agrees with that reported in Reference 2.

*R. P. Johnson and C. T. Nelson "Test Procedure for the Aerosol Leakage Tests," DTP-707-130

a. Time = $T - 0.0078$ sec

b. Time = 0.00 sec

c. Time = $T + 0.032$ sec

d. Time = $T + 0.079$ sec

Figure 10. Time History of 90 to 95% Condensible Bubble

2. Modeling Development

A literature review of a series of thermodynamic studies of the Na-U-O system has verified the stability of sodium uranates under potential LMFBR operating conditions.⁽²⁻⁵⁾ Mass spectroscopic studies of the vaporization products⁽⁴⁾ showed that no uranate was released.

Airborne release data for small scale experiments on the combustion of sodium containing PuO_2 have been reported by Chatfield.⁽⁶⁾ In one experiment when 0.47 grams of sodium containing 0.09 gm PuO_2 finely dioxide powder (unknown size distribution and manufacture) was ignited, $0.18\mu\text{Ci}$ of plutonium was released. Chatfield states that a reaction between PuO_2 and sodium must have occurred. Chatfield did not identify the plutonium isotope. If one assumes that it was Pu^{239} , the release fraction is about 3.3×10^{-5} . If any other isotope of plutonium is assumed except Pu^{242} , the release fraction is even lower.

If the Chatfield results can be extrapolated to large scale sodium fires in which the PuO_2 contents are similar to those which are allowable in the CRBRP primary sodium (0.1 ppm), then plutonium will not contribute appreciably to the hazard in ordinary spills. However, there are several cases where a larger plutonium contamination of primary sodium could potentially occur and produce important radiological consequences. These cases are:

- 1) A fuel transport cask, transporting a defective fuel subassembly, could leak sodium onto the floor of the reactor containment building (RCB) or the reactor service building
- 2) The melt-through of the reactor vessel by the internal debris following a HCDA could mix the full core inventory of molten fuel with the 1200°F sodium, resulting in finely divided plutonium oxide, Na-Pu-O, and fission products dispersed in the hot sodium.
- 3) A used cold trap loaded with fission products, sodium oxide, Na-Pu-O, UO_2 , PuO_2 , and sodium could ignite in air while being removed from the reactor system.

The Chatfield results also indicates that if uranium oxide is released in the same fraction as the PuO_2 then ordinary chemical means cannot be used to measure the release fraction in the present experiments.

F. SUBTASK G – AEROSOL MODEL IMPROVEMENTS

A brief study was made of the dependence on containment height of the gravitational agglomeration efficiency ($\alpha\epsilon$) and settling velocity correction (α) for HCDA aerosols. α was related to an empirical correlation of settling velocity with the number (N) of source particles in the agglomerated particle and its internal structure. ϵ was determined from fits of aerosol transport calculations (HAA-3B code) to measurements in LTV, a 30 ft (9.14 in.) high chamber. An expression for α as a function of N, particle structure, the particle size frequency distribution, and the particular accident history was derived. Using HAA-3B results for containment vessels with different heights, a first-approximation solution was obtained for $\alpha\epsilon$ as a function of height for two accidents. The results, shown in Figure 11, indicate a weak dependence on height for both a given volumetric source rate and a given volume-integrated source rate. Based on these results, the variation of $\alpha\epsilon$ over a range of different hypothetical accidents (source rates, source particle sizes, etc.) is expected to be greater than the variation with height for a given accident.

TI-707-130-036 by J. M. Otter, "Concerning the Dependence on Containment Height of the Efficiency of Gravitational Agglomeration and Settling of HCDA Aerosols," was written in January 1975. The need to reexamine previous experiments in terms of the values of N and to perform additional experiments was identified. It was concluded that experiments to characterize α and $\alpha\epsilon$ for design height containment vessel (≈ 170 ft) can probably be performed in the LTV (30 ft high).

G. SUBTASK J – HIGH TEMPERATURE PROPERTIES OF FUEL MIXTURES

A laboratory for measuring the high temperature physical properties of fuel mixtures was established. Two furnaces were set up – a small arc melting furnace (Figure 12) and an induction melting furnace (Figure 13).

The arc melting furnace has demonstrated a capability of melting small samples of ceramic material, including UO_2 , even though the ceramics are room temperature insulators. Figure 14 shows some of the melted samples of ceramics. The dark color is the result of contamination by tungsten from the electrode. In particular, UO_2 and a mixture of UO_2 and SS were melted. In melting the mixture of material, the SS was volatilized quite extensively. Metallurgical

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Figure 11. $(\alpha\epsilon)$ vs Building Height

Figure 12. Arc Melting Furnace

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Figure 13. Induction Melting Furnace

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Figure 14. Samples of Melted Ceramics

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examination of the samples is underway to determine the extent of foaming and expansion of the UO_2 by SS vapor.

Induction melting of UO_2 will take place in a tungsten crucible situated in a large vacuum tank. Initially, a Lepel 10 kw high frequency generator will be used for melting. A 100-kw Inductotherm machine is also available. The tungsten crucibles will be surrounded by several layers of heat shielding. Material properties experiments are planned for measuring the vapor pressure, viscosity, surface tension and density of UO_2 alone and, UO_2 with additions of stainless steel or simulated fission products. It will also be possible to pour molten UO_2 onto steel.

Planning continued for large scale (50 to 100 kg) melting experiments at the Santa Susana laboratory. Several suitable sites for experimentation have been located. A large arc furnace for melting UO_2 has been returned to AI from ORNL. The furnace will be installed at Santa Susana along with a 10,000 ampere power supply. The molten UO_2 from the furnace will be used to study material properties of UO_2 -SS mixtures and the aerosol production from molten fuel-coolant interactions.

A test plan was written which covers experimentation over a 5-yr period. At the end of this time, the improved understanding of the properties of UO_2 -SS-FP molten mixtures should make it possible to reliably estimate the production and size distribution of aerosols resulting from various hypothetical core disruption accidents in LMFBR's.

IV. NEXT REPORT PERIOD ACTIVITIES

A. SUBTASK A – SODIUM SPLASH DISPERSAL

Efforts will be directed toward completion of the following tests; (1) LSM NaK jet tests conducted at 3-9 m/sec, and additional water jet tests at high velocities, (2) single sodium drop burning tests, (3) falling sodium drop tests and (4) free convection tests conducted in the LTV.

B. SUBTASK C – SOMIX CODE DEVELOPMENT

The sodium drag force, combustion, and heat transfer models will be incorporated into SOMIX-1. With inclusion of the oxygen transport equation, this

will bring the SOMIX-1 development to a point where pipe rupture studies can begin. Checkout runs of the complete code will then be made simulating various pipe rupture accidents.

Modification of SOMIX-2 to cylindrical geometry will continue with effort being concentrated on Fromm's first and second order approximations. The development of turbulent flow models will be carried to the point where the basic turbulence transport equations will be developed and the auxiliary equations required to define production and dissipation of turbulence will be identified.

C. SUBTASK D – HCDA AEROSOL SOURCE TERM

Tests will be initiated to determine the attenuation factors for aerosols in a simulated HCDA bubble rising in a water medium. Procurement of a test vessel [an extension of the Spray Test Vessel (STV)] to conduct small scale simulated HCDA aerosol bubble tests in sodium will be initiated.

D. SUBTASK F – FUEL AND FISSION PRODUCT RELEASE FROM BURNING SODIUM

The synthesis of Na_3UO_4 will be verified. Various mixtures of UO_2 , Na_3UO_4 and sodium will be made. Measurement of the release fraction (as a function of temperature) in various gaseous environments will be attempted, using a modified transpiration apparatus previously developed at AI

E. SUBTASK I – AEROSOL SOURCE FROM FUEL VAPOR

A 75 Kjoule condenser bank discharge system will be set up for exploding wires of UO_2 and sodium. This equipment will enable the measurement of the amount and size distribution of aerosols following vaporization of UO_2 into a sodium saturated gas environment to simulate HCDA conditions.

F. SUBTASK J – HIGH TEMPERATURE PROPERTIES OF FUEL MIXTURES

UO_2 and $\text{UO}_2 + \text{SS}$ will be melted in the induction furnace. The density and surface tension of the material will be measured at 3000°C . Equipment for measuring vapor pressure and viscosity will be installed.

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