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MASTER

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Program: LMFBR Safety

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

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Project Manager: H. A. Morewitz

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I. PROJECT OBJECTIVES

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The objectives of this project are to:

- 1) Develop a computer program for calculating two-dimensional, transient, natural convection phenomena such as those arising from various sodium spill accidents in Liquid Metal Fast Breeder Reactor (LMFBR) heat transfer equipment vaults, head compartments, containment buildings, and secondary heat transfer systems.
- 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.
- 4) Determine the effects of an accident in an LMFBR involving fuel melting by contacting molten UO_2 (a fuel simulant) with stainless steel, sodium, concrete, and various sacrificial materials.

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II. MAJOR ACCOMPLISHMENTS DURING REPORT PERIOD

A. SUBTASK B – SODIUM JET DISPERSAL

Additional tests were conducted to determine some physical properties of burning sodium drops. A multi-image still camera was used to record photographically the terminal speed of sodium drops ranging in size from 0.4 to 0.7 cm initial diameter. The terminal velocity was calculated from these data and was compared with the theoretically calculated values from SOMIX. Close agreement between experiment and the model was obtained.

B. SUBTASK C – AEROSOL LEAKAGE

Tests were performed to document by photography the deposition of particulates during the plugging of capillaries. The photographs show entrance plugging, exit plugging, plugging of capillaries with right angle bends, buildup of deposits along the wall, and dendrite formation due to impaction.

C. SUBTASK D – HIGH TEMPERATURE-CONCENTRATION AEROSOLS

During this report period, five High Temperature-Concentration Aerosol tests were conducted using a dual nozzle injection system which increased the sodium injection rate to 4.4 kg/min. This system modification resulted in minimum aerosol fallout and wall plateout during the spray period and, consequently, higher airborne aerosol concentrations were achieved.

D. SUBTASK M – CHARACTERISTICS OF RELEASED AEROSOLS

Laboratory testing was initiated to determine the characteristics of released sodium oxide aerosols. The laboratory tests involved developing test apparatus and chemical processes which can be used to identify quantitatively the various chemical species of the released aerosols. The chemical species known to be present in released sodium oxide aerosols are Na_2O , Na_2O_2 , NaOH , and Na_2CO_3 . The latter two compounds are produced by exposure to natural atmospheric moisture and carbon dioxide. A differential microgasometer was developed to detect the presence of Na_2O_2 , Na_2CO_3 , and NaOH (by secondary reaction), in the released aerosols, with an accuracy of 2 to 5 μl .

III. ACTIVITIES DURING REPORT PERIOD

A. SUBTASK A – SOMIX CODE DEVELOPMENT

1. Description

SOMIX-1 is a computer program which was developed to study the transient convective motion of low-oxygen gas environments in cylindrical and rectangular enclosures, representing heat transfer vaults of LMFBR power plants. The gas is heated by a spray of hot sodium droplets generated by a break in one of the large sodium pipes in the vault. The resulting high velocity liquid breaks up into a spray of drops. As the sodium drops fall, they react with the low oxygen environment (0 to 2 vol %) in the enclosure, and are simultaneously heated by low-level combustion and cooled by convective heat transfer to the gas and by radiative heat transfer to the walls of the vault.

2. Progress During Report Period

A literature search on the mechanics of falling liquid drops has been completed. The result shows that falling burning sodium drops exhibit many of the characteristics that have been observed for nonburning liquid drops.

Photographic observations ⁽¹⁾ show that the drops approach the shape of continuously oscillating oblate spheroids. Some of these oscillations are started by the elongation which occurs when the drop breaks loose from the hypodermic needle; others result from the motion of the surrounding fluid, particularly in the wake, where intermittent shedding of vortices probably occurs. Photographs of falling drops have indicated the presence of oscillation even after falls of 20 ft. The amplitude of the oscillation increases with increase in size of the drops.

The effect of these oscillations is to increase the drag resistance of the drops, particularly for the larger drop sizes. The effect of such oscillations on drag resistance has been investigated by Hughes and Gilliland ⁽²⁾, who have developed a semi-empirical procedure for estimating the drag coefficient of falling liquid drops. The distortion of the drop, which is assumed to be an oblate spheroid with its axis in the direction of motion, is given by the following equation for the height/width aspect ratio of the oblate spheroid:

$$h = 1 - .002185 R_e^{2.35} / Su \quad \dots 1$$

Equation 1 was developed from tabular data presented in Reference 1. Su is the inverse (Ohnesorge number)⁽²⁾ and is given by:

$$Su = g \sigma D \rho_g / \mu_g^2 \quad \dots 2$$

g = gravitational acceleration

σ = interfacial surface tension of the drop

D = diameter of the equivalent spherical drop

ρ_g = gas density

μ_g = absolute gas viscosity

The true Reynolds number, Re' , for the distorted drop is found from:

$$Re' = Re / h^{1/3} \quad \dots 3$$

Using this Reynolds number, drag coefficients CD_d and CD_s are obtained for circular discs and solid spheres, respectively. The drag coefficient, CD , for the deformed drop is then given by the following equation, which is based on the experimental data of Riabouchinsky⁽³⁾:

$$CD = CD_s \left(1 + (1-h)(CD_d / CD_s - 1) \right) / h^{2/3} \quad \dots 4$$

The above procedure will be incorporated into the SOMIX code, with appropriate physical property values being determined from the falling sodium drop burning test data.

3. Next Report Period Activities

The theoretical correlations described above and the (h) results of the falling sodium drop tests (Subtask B) will be incorporated into the SOMIX code.

B. SUBTASK B - SODIUM JET DISPERSAL

1. Description

It became of interest to provide experimental data on the terminal velocity of falling-burning sodium drops to test the drop velocity subroutine in the SOMIX Code. This was accomplished by using a multi-image still camera to photograph the burning sodium drops produced from a sodium drop generator. An 8 x 10 cm still camera was converted to a multi-image still camera by mounting a 25-cm diameter, continuously rotating shutter wheel in front of the camera lens. The rotating shutter was fabricated with 12 equally-spaced 1.2 x 2.5 cm openings along a circumference. When this shutter rotated at 1750 rpm, the time between openings (open-closed) was determined to be 1.29 ms. A schematic of the multi-image still camera is given in Figure 1. A shutter chopped streak photograph was taken of each falling drop after falling ~ 13 meters. By measuring the distance between images, the drop velocity could be determined.

2. Test Results

During these tests, the size of sodium drops produced by the drop generator ranged from 0.51 to 0.76 cm diameter (initial size). As a drop fell into a 14.7-meter high test cell, ignition occurred and the drop accelerated to its terminal velocity under burning conditions. The chopped streak still photographs shown in Figures 2 and 3 show the flight of burning sodium drops 2.5 meters before striking an inert gas filled receiver on the floor of the test cell. The two horizontal light traces on each figure result from stationary "pin" lights positioned 30.48 cm apart (vertically), and in the same view plane. These traces provided an absolute scale to measure drop fall distance. Each trace was scanned with an optical projection comparitor to determine the number of light images in a fixed distance. Table 1 presents the measured terminal velocities of the successful drop tests.

Comparison of the experimental and theoretical terminal velocities were sufficient to validate the drop velocity subroutine in SOMIX for the range of drop size studied.

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Figure 1. Schematic of Multi-Image Camera to Measure Sodium Drop Velocity

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Figure 2. Shutter Chopped Streak
Photo of 0.75 cm Diameter
Burning Sodium Drop
 $v = 10.9 \text{ m/s}$

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Figure 3. Shutter Chopped Streak
Photo of 0.51 cm Diameter
Burning Sodium Drop
 $v = 9.5 \text{ m/s}$

TABLE 1
TERMINAL VELOCITY OF FALLING SODIUM DROPS

Test No.	Initial Drop Diameter (cm)	Theoretical Drop Diameter After Falling 14.7 Meters* (cm)	Terminal Velocity After Falling 14.7 Meters (m/s)
1	0.760	0.754	11.0
2	0.755	0.747	10.9
3	0.508	0.439	9.5
4	0.517	0.449	9.8

*Calculated by measuring the mass/drop collected in the inerted receiver on the test cell floor and using the sodium density at boiling temperature, 0.739 g/cm^3 .

3. Future Tests

Velocity measurements will be made on burning drops of the size range 0.3 to 0.5 cm initial diameter.

C. SUBTASK C - AEROSOL LEAKAGE

In these experiments, tests were performed to document photographically the particulate deposition in capillaries when the capillaries are exposed to a sodium oxide aerosol [concentration of 0.5 to $3.5 \mu\text{g}/\text{cm}^3$; particle sizes of 1.8 to $2.5 \mu\text{m}$ (AED_{50})]. The humidity ranged from 2800 to 5200 ppm. For all experiments, the vessel pressure was maintained at 1.068 atm. The capillaries were 1.8 to 2.0 mm diameter x 5 cm.

Figure 4 is a photograph of entrance plugging where the particulates do not follow the air stream and impact at the entrance. When such a buildup of oxide occurs and gas leakage through the capillary exists, the oxide plug

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Figure 4. Capillary Entrance Plugging

performs as a filter. Plugging at the capillary exits are shown in Figures 5 and 6.

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Figure 5. Capillary Exit

During these tests, plug creep occurs and considerable mass may accumulate before plugging finally ends the oxide flow. When plugging does not occur, large agglomerated masses are expelled from the capillary exit. This is shown in Figure 7 where a strike plate, placed approximately 19 cm from the capillary exit, captured the ejected mass. These particulates are shown

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to be considerably larger than respirable size particulates (i.e., $> 10 \mu\text{m}$).

Figures 8 and 9 show a typical deposit of material along the capillary wall at magnifications of X 40 and X 100, respectively. The small particulates vary from 2 to 4 μm . Figure 10 shows the deposit near the capillary exit.

(Figures 5, 6 and 7 are orientated so that the gas flow is from right to left.)

Plugging inside the capillary is shown in Figure 11.

Capillaries with right-angle bends are shown in Figures 12 and 13. Leak paths of this type have a preference for particle deposition in the capillary bend region where impaction occurs.

An experiment was performed to show what would happen when particle sizes greater than 0.95 μm (AED_{50}) were removed and the remaining particles allows to leak through a capillary (0.3 mm diameter x 1.1 mm) and strike a plate

← Figure 6. Capillary Exit

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Figure 7. Expelled Particulates

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← Figure 8. Wall Deposition
(X 40)

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Figure 9. Wall Deposition
(X 100)

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← Figure 10. Wall Deposition
Near Capillary Exit
(X 40)

1.7 mm from the capillary exit. The flow through the capillary was maintained at $100 \text{ cm}^3/\text{min}$. The results can be seen in the oblique photograph (Figure 14). At the impact region, a dendrite column of oxide (approximately 0.06 mm diameter $\times$ 1.3 mm) was produced normal to the surface. This column would eventually have entered the capillary and produced a plug if the flow had been allowed to continue.

These photographs show that particulates do not flow through leak paths in the same manner as gases, that leak paths do plug, and that many particulates which exit the leak paths are larger than the respirable size.

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Figure 11. Internal Plugging

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← Figure 12. Plugging in Right Angle Bend

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Figure 13. Plugging in Right Angle Bend

Figure 14. Dendrite Formation by Impaction (Oblique View)

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D. SUBTASK D - HIGH TEMPERATURE-CONCENTRATION AEROSOLS

1. Description

Modifications to the High Temperature-Concentration Aerosol (HTCA) test apparatus have been checked out with the five tests conducted during the report period. Each test was initiated by injecting 100 g of sodium, which was pre-heated to 540°C, into the 430-liter test vessel. The sodium injection rate was increased to 4.4 kg/min by the addition of two nozzles; the transfer time for the 100 g of sodium used during these tests was reduced to 1.35 s. During the current tests, measurements were made of vessel pressure, lower and midplane vessel gas temperature, sodium injection rate, and oxygen enrichment injection rate. Mechanically operated incremental wall plateout and floor fallout sample strips, and an automatic LVDT fallout detector, both mounted in-vessel, were also used.

Test numbers 14 and 15, the first two tests conducted during this report period, were initiated at atmospheric pressure and temperature conditions. For Tests 16, 17 and 18, the vessel atmosphere was first enriched with oxygen, then evacuated to 0.66 bars. This procedure established an initial atmosphere which had approximately the same amount of oxygen at partial vacuum as that of standard air at atmospheric pressure.

2. Test Results

The specific objectives of tests 14 through 18 of the HTCA test series were to characterize the aerosols produced from a rapid injection and burning of 100 g of sodium. Fulfilling these objectives required simultaneous operation of the test instrumentation and adequate aerosol sampling.

In the HTCA test series, samples which were taken at the point of maximum concentration (end of spray) were a mixture of 3 to 10 μm diameter particles and very large, 100-200 μm diameter, particles (see Figures 15 and 16). The presence of these large diameter aerosols indicates rapid agglomeration, probably due to turbulence in the sodium burning zone during the spray period. The LVDT fallout sensor showed over 95% of the fallout occurred within 15 to 20 seconds. Analysis of the incremental wall plateout samples showed that over 95% of the plateout occurred within a 10-second period. A mass balance history of the airborne aerosol, wall plateout and fallout has been made. Figure 17 shows

100 μm
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Figure 15. HTCA Aerosol Sample
Collected at End of Spray

200 μm
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Figure 16. HTCA Aerosol Sample
Collected at End of Spray

Figure 17. HTCA Aerosol History

this mass balance as fractions of the respective components. Measurement of aerosol size after 1 min shows that only particles 3 to 5 μm remain airborne. These remaining aerosols agglomerate, fallout, and plateout with the same time dependence as if they had been formed in the lower concentration range, i.e., 10 to 100 g/m^3 .

Plateout and fallout materials were chemically analyzed to determine the effect of the high gas turbulence, during the early test period, on the radial fallout and axial plateout distribution. Several 2.5 cm^2 fallout planchets, placed radially on the vessel floor, were analyzed for total sodium content. These analyses show that there is an approximate factor of 2 difference between the fallout mass at the outer vessel boundary and at the center of the vessel. This is attributed to the downward gas flow (created by the sodium spray burning) on the vessel centerline. Also, the wall plateout deposition is higher, by the same factor, in the lower vessel section than in the vessel wall area near the injection nozzle.

Six additional sampling ports have been installed in the HTCA lower vessel head. These sampling ports will be used to take additional aerosol samples for sizing and concentration measurements during the rapid aerosol agglomeration and fallout period. During the next report period, the aerosol concentration will be increased from 370 g/m^3 (100 g sodium injection) to approximately 700 g/m^3 . Comparison of the rapid agglomeration characteristics observed with the 370 g/m^3 concentration tests and the higher concentrations will be made.

E. SUBTASK M - CHARACTERISTICS OF RELEASED AEROSOL

A Warburg syringe manometer with a compensating bulb has been modified to determine the chemical species of sodium combustion product aerosol, assuming that the species are a combination of Na_2O , Na_2O_2 , NaOH , and Na_2CO_3 . The modification consists of adding a three-way microvalve with fluid reservoirs containing H_2O and $\text{HCL-H}_2\text{O}$. During operation, each fluid is allowed to react with the sample located in the sample cavity. The gases evolved are measured in the conventional manner, by manipulating the micrometer to restore the fluid in the capillary to the reference line and recording the volumetric change. The differential microgasometer is maintained at constant temperature by immersion in a water bath.

F. SUBTASK F - PROPERTIES OF HIGH-TEMPERATURE FUEL MIXTURES

1. Introduction

A facility for arc melting and pouring large quantities (10 to 100 kg) of UO_2 or stainless steel has been constructed. The UO_2 will be used as a simulant for fuel which might melt during a reactor accident.

The several uses of the molten UO_2 are to (1) study the production, agglomeration, and dispersal of fuel aerosol, (2) test the interaction of molten UO_2 and sodium, particularly in regard to any reaction-producing overpressure, (3) determine the rate of melt-through of the reactor vessel, underlying concrete, and/or sacrificial material used to retard melt-through, (4) study post-accident heat removal and containment rupture problems in general, and (5) test the effect of the presence of fission products in any of the above.

2. Facility Description

At present, the arc furnace, power supply vacuum system, and cooling system have been installed.

The melting crucible is double-walled copper with cooling water, flowing between the walls, directed toward the bottom and tangentially on the sides. The electric current passes as an arc from a graphite or tungsten stinger electrode to the UO_2 surface and then through the UO_2 to the copper hearth. During operation, some UO_2 remains frozen in a skull next to the copper, while the remainder is melted in the center by the high current. About 100 kg of UO_2 can be contained, of which 70 kg can be melted. Pouring the melt is accomplished by quickly withdrawing the stinger and tilting the crucible by means of motor drive. The melt can either be poured within the tank or through a large valve in the bottom of the tank.

The power supply for the melter consists of eight heavy-duty welder supplies with outputs connected in parallel to make a total of 10,000 A. All are now connected. During operation, the supplies are switched off or on as needed.

3. Safety

Considerable emphasis has been placed on the safe operation of the facility because of the large amounts of power involved. In addition, handling the relatively large amounts of powdered UO_2 involves the possibility of personnel

contamination. A safety report (N707NSA190001) has been reviewed by the Health and Safety Organization, the Isotope Committee, and an ad hoc Management Committee; the recommendations of these organizations have been put into effect.

The principal recommendation was to provide a circuit that automatically shuts off the power supplies in case of a ground fault. The power supplies are operated "floating," i.e., without either of the high current leads connected to ground. A single short to ground would not produce dangerous effects; however, two shorts might cause the vaporization of metal. The ground fault detector operates by supplying a 1 kHz signal to the high current leads and to ground. This signal is detected by a detector with an input filter turned to 1 kHz. Should a fault occur on either lead, the signal will be grounded out, and therefore not detected. With the signal missing, the detector will open relays to shut off the power supply.

Another safety consideration is the possibility of a water leak from the water-cooled crucible into the melt. Rapid vaporization of water could possibly overpressurize the furnace tank. To alleviate this problem, a rapid-acting valve has been installed to open an exhaust line to a large vacuum pump. Exhaust from the pump is routed to absolute filters and a stack. In addition, one of the large hinged doors of the tank is held closed by negative tank pressure augmented by spring pressure. The door would partially open to the furnace room, if the pressure rises ~ 0.25 psi above atmospheric pressure. The furnace room is connected to the atmosphere via a 10-ft by 10-ft roll-up door which will be open during all tests.

4. Furnace Startup Tests

In carrying out the startup tests, it has been necessary to solve some operational problems. These are discussed below.

While melting stainless steel, the arc would occasionally drift to the wall of the crucible rather than to the melt. As current was increased, the tendency became more severe. To alleviate this problem, three argon jets have been installed in order to direct a flow of cold argon around the periphery of the crucible. An automatic pressure control on the furnace tank opens the vacuum line to the pump, and thus maintains constant pressure in the tank as argon is injected through the jets.

The argon injection proved effective, in that the onset of arcing to the crucible wall was delayed and the furnace could be operated at a higher power level. It is planned to install more jets, as three were insufficient to eliminate the problem completely.

The new jets will be installed on the rod holding the stinger rather than on the crucible. A circular stainless steel disk will support the jets. The disk will also aid in reflecting radiant heat to the crucible, and will provide a site for condensation of much of the UO_2 vapor rather than the UO_2 condensing on the furnace tank walls.

Determining a method of removing UO_2 pellets from the aluminum tubes in which they were supplied turned out to be more difficult than anticipated. The UO_2 was previously used in critical experiments and was obtained as a surplus item. It has been found necessary to partially dissolve tubes rather than just open the ends to remove the UO_2 . Sodium hydroxide solution dissolves the aluminum quite rapidly without affecting the UO_2 or permeating into it.

About 25 kg of the UO_2 has been placed in the crucible with a central stainless steel starting electrode. Preliminary melting tests have been successful in keeping the arc directed into the melt rather than towards the crucible wall. Higher power operation should result in melting most of the charge.

5. Conclusions and Planned Activities

Some operational problems have been solved. It is planned to begin immediately with large-scale melting of UO_2 using a graphite stinger. A more expensive tungsten stinger will be used if the graphite electrode results in excessive carbide formation. Aerosol samples, taken from the tank, will be used to determine concentration and size distribution of UO_2 particles condensing from vapor in an argon atmosphere.

Stainless steel will be inbedded within the UO_2 to determine its effect upon UO_2 as it vaporizes. It is expected that some foaming and frothing will occur. The melting process will be monitored by closed circuit television. In addition, post test examination of the residue will be made.

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