QUARTERLY TECHNICAL PROGRESS REPORT
REACTOR SAFETY
OCTOBER – DECEMBER 1977

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MASTER

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Program: LMFBR Safety

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SA002: Accident Debris Behavior

Program Manager: R. W. Keaten

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17100, 19000, 21000, 22000

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I. PROJECT OBJECTIVES

- 1) Conduct tests that will characterize the behavior of sodium oxide, fuel, fission product, and other aerosols as they might be generated by various postulated LMFBR accidents.
- 2) Determine by analysis and confirm by experiment the generation and transport of these aerosols with respect to source (location, type, and configuration), for the entire course of events associated with real and hypothetical accident conditions.
- 3) Conduct tests that will determine the effect of molten fuel on reactor structural or sacrificial material.

II. MAJOR ACCOMPLISHMENTS DURING REPORT PERIOD

A. SUBTASK C - AEROSOL LEAKAGE

1. Multiple Bend Leak Path

A multiple bend leak path was constructed to simulate the type of leak path that may exist in a reactor head seal. Tests are being performed to show that aerosols containing sodium combustion products, when passing through leak paths of this type, have large attenuation factors and quickly plug.

The multiple bend leak path, as shown in Figure 1, is composed of four leak paths plus connecting annuli. The leak path LPa represents a rotating

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Figure 1. Multiple Bend Leak Path

plug sodium seal in which the seal is void of sodium. Leak Paths LPb, LPc, and LPd represent the spacing that may occur at the lower margin ring assembly, dynamic (inflatable) seal, and upper margin seal (including the associated assembly), respectively.

A checkout test was conducted using the aerosol generator described in previous reports. An aerosol mixture (Na_2O_x and Na_2CO_3) with a concentration of 15 to 20 $\mu\text{g}/\text{cc}$ (sodium content) was generated for this test. The multiple bend leak plugged in ~ 5 min while passing ~ 217 liters of gas. Calculations showed that 3 to 4 grams of sodium (in the form of $\text{Na}_2\text{O}_x + \text{Na}_2\text{CO}_3$) were required to plug the specimen. A pressure differential of 0.068 atm was maintained during the test.

When the leak path was disassembled, complete plugging at LPa, LPb and the annulus between was observed. The inlet annulus was nearly plugged. Very little aerosol was collected beyond LPb.

2. Simple Model for Plugging of Ducts by Aerosol Deposits

Experiments⁽¹⁾ on the leakage of aerosols through capillaries indicate that complete, partial, or incipient plugging of the flow path by deposits of aerosol material plays a major role in reducing the inhalation hazard, both by decreasing the amount of escaping particulate matter and by increasing the size of the particle agglomerates. However, current theories⁽²⁾ of the deposition of aerosol particles during the flow through ducts ignore the modification of the geometry of the flow by the accumulation of material on the walls. An initial attempt to model this neglected feature is now presented.

The deposits observed are highly irregular in general configuration and dendritic in detailed structure. This complexity is also encountered in the problem of aerosol agglomeration where irregular dendritic agglomerates are replaced by structureless spheres with considerable success.⁽³⁾ This modeling is adapted to the present problem by assigning a mean density, ρ , to the porous deposit while simplifying the geometry to a single growing plug that retains a fixed shape having rotational symmetry about the axis of a cylindrical duct (Figure 2). Use of a single plug reflects the tendency for a larger plug to grow more rapidly than a smaller plug by intercepting more of the flow.

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Figure 2. Schematic of Plug Geometry

With these assumptions, the mass of the plug at any time, $M(t)$, is determined by the value at that time of any one geometrical parameter of the plug, together with the fixed radius of the duct, R . The parameter used is the greatest thickness of the deposit, denoted by $h(t)$. The area of a section of the plug by a plane through the duct axis is then a fixed multiple, α , of $h^2(t)$, and the volume is the product of this area by the length of the circumference described by its center of mass as the plane rotates about the duct axis. The distance of that center of mass from the wall being another fixed multiple, β , of $h(t)$, the mass is

$$M(t) = 2\pi \alpha \rho [R - \beta h(t)] h^2(t) . \quad \dots (1)$$

A mass balance can be derived by equating the rate of change of the mass obtained by differentiating Equation 1 to the deposition rate provided by suitable assumptions about the flow and deposition mechanisms. The suspended mass concentration of particulates is supposed to be an arbitrary function of time, $C(t)$. The volumetric flow rate through the duct may vary in time because of the changes in plug size and also for other reasons, and is accordingly denoted by $Q[t, h(t)]$. The ratio of the deposition rate to the interception rate that would result if the particles were moving on straight lines parallel to the axis is called the collection efficiency of the plug. It is assumed to depend on the ratio of plug height to duct radius, $h(t)/R$, but not on flow rate nor particle

size, and is therefore denoted by $E[h(t)/R]$. These assumptions, with their implied neglect of the inertia of the particles, are not necessarily correct, but can be tested by comparing the properties of the model with experiments.

In terms of these functions, the deposition rate is expressed as

$$\dot{M} = C(t)Q[t, h(t)]E[h(t)/R]\pi \{R^2 - [R - h(t)]^2\} / \{\pi R^2\}. \quad \dots (2)$$

Here the product, $C(t)Q[t, h(t)]$, represents the rate at which particulate mass is carried past the plug by the flow and can be eliminated by using the total mass thus transported up to the time, t , as the independent variable instead of t itself. Then Equations 1 and 2 give

$$dM/dm = 2\pi\alpha\rho[2Rh(m) - 3\beta h^2(m)]dh/dm, \quad \dots (3)$$

$$dM/dm = E[h(m)/R] \{1 - [1 - h(m)/R]^2\}, \quad \dots (4)$$

respectively. Finally, taking $x = h/R$ as a new variable, the mass balance resulting from Equations 3 and 4 can be solved for dm/dx in the form

$$dm/dx = 2\pi\alpha\rho R^3(2x - 3\beta x^2)/[(2x - x^2)E(x)]. \quad \dots (5)$$

The total particulate mass, $\bar{m}$, transported past the plug before its growth blocks further passage of particulates, is obtained by integration of Equation 5 from $x = 0$ to $x = 1$, corresponding to growth of the plug from $h = 0$ to $h = R$. The result is

$$\bar{m} = 2\pi\alpha\rho R^3 \int_0^1 dx (2x - 3\beta x^2)/[(2x - x^2)E(x)]. \quad \dots (6)$$

Since the values of α , β , and ρ , and the form of the function $E(x)$ remain unknown, the only useful content of Equation 6 is that

$$\bar{m} = KR^3, \quad \dots (7)$$

where the constant K is unknown, but is independent of the concentration $C(t)$, the flow rate, $Q[t, h(t)]$, and the size of the particulates.

A preliminary test of Equation 7 is presented in Figure 3. The lower five points were obtained from experiments⁽¹⁾ on sodium carbonate aerosols passing through smooth capillaries. The other two points represent "inadvertent experiments" in which investigations of cover-gas systems were interrupted by blockages in the ducts.^(4,5) The empirical support for Equation 7 is seen to be modest but encouraging.

Figure 3. Data on Plugging Mass vs Duct Size

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B. SUBTASK D — HIGH TEMPERATURE-CONCENTRATION AEROSOLS

The high temperature-concentration aerosol (HTCA) tests conducted to date have been designed to simulate the conditions within a gas bubble caused by an HCDA in an LMFBR system. Estimates have been made of the initial HCDA bubble temperatures and aerosol concentrations, i.e., 3000 to 4000°C gas temperature and $2-3 \times 10^3 \text{ g/m}^3$ aerosol concentration. The tests conducted in

the HTCA test series have used sodium oxide aerosols to simulate the UO_2 - PuO_2 -Na aerosols produced in an HCDA. Preheated sodium is sprayed into the HTCA test apparatus under high atomization gas pressure and allowed to rapidly burn in an oxygen-rich atmosphere to form a sodium peroxide aerosol. Due to the rapid sodium burning, gas temperatures exceeding 2000°C and aerosol concentrations approaching $1 \times 10^3 \text{ g/m}^3$ have been obtained. These test conditions provide an adequate base on which to characterize the behavior of the aerosols.

The Laboratory Test Chamber (LTC) has been used as an expansion vessel during the HTCA test series. The LTC is coupled to the HTCA test apparatus through a 1.2-cm diameter pipe fitted with a series-mounted ball and sliding gate solenoid valve. At the completion of the HTCA sodium injection period, indicated by a rapid drop in test apparatus gas pressure, the connecting valves are opened and ~ 6 to 10 liters of HTCA gas is transferred to the LTC, thereby diluting the aerosols by a factor of 1:400. The aerosols which are also transferred to the LTC are subsequently analyzed for concentration. Fallout slides placed on the floor of the LTC provide a means of determining the initial aerosol size. For all HTCA tests where the maximum aerosol concentration exceeded 150 g/m^3 , large agglomerates ranging in size from 100 to $250 \mu\text{m}$ diameter were observed on the LTC fallout slides. From these data it is postulated that the aerosols in the HTCA test apparatus rapidly agglomerate in a short time period, then are subject to removal by plateout and fallout. Aerosol size measurements, obtained ~ 25 to 30 s after the spray period, show that at that time only aerosols in the 1-4 μm range exist.

An improved internal wall plateout and floor fallout aerosol sampling device has been developed and installed in the HTCA test apparatus. This sampling device is comprised of two wall plateout stations and a single floor fallout station. The surfaces to be exposed to the vessel atmosphere during a test consist of 16 each removable tabs which are positioned under a shield canopy. Upon actuation of the sampling device drive motor, these tabs are withdrawn from under the respective wall or floor shield canopies at the rate of 2.6 s/tab. The tabs are removed from the test vessel at the completion of a test and analyzed for aerosol plateout or fallout deposition. Figure 4 is a photograph showing a typical fallout or plateout sampling device. In the figure, alternate tabs have been removed from the main slide for demonstration.

Figure 4. HTCA Plateout and Fallout Sampling Device

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The HTCA aerosol plateout and fallout histories previously reported⁽¹⁾ were incorrectly labeled. This figure is correctly shown here as Figure 5. Operational checkout of the improved internal plateout and fallout sampling device was made on HTCA Test No. 21. Although little quantitative data were obtained during the test due to failure of a slide-gate valve located on one of the aerosol sizing sampling ports, some qualitative fallout and plateout data were obtained. During the test, the fallout and plateout sampling device was actuated at the beginning of the sodium spray period. This allowed measurements to be taken both during and after the spray. Post-test analysis showed that a fraction of the available aerosols were removed due to impaction on the test apparatus walls and floors. It is expected that from the additional experimental data obtained with the improved fallout and plateout sampling device, complete time-dependent aerosol histories can be obtained on future HTCA tests.

C. SUBTASK F — LARGE SCALE MOLTEN FUEL TESTS

1. Introduction

A facility has been constructed for arc melting and pouring large quantities (up to 100 kg) of UO_2 or steel for use as simulants for the debris which might result from a reactor accident.

Melting takes place within a large water jacketed tank (Figure 6). The melting crucible is water cooled copper about 25 cm in diameter and 20 cm deep. The crucible has a layer of solid stainless steel on the bottom. The melt is heated by a high current arc which passes from a graphite electrode to the UO_2 surface and then through the UO_2 , the stainless steel, to the copper crucible. Some UO_2 remains frozen in a skull next to the copper or stainless steel while the remainder in the center is melted by the arc.

The several uses of the molten UO_2 and steel are to: (1) study the production, agglomeration, and dispersal of fuel and sodium aerosols, (2) test the interaction of molten material with sodium, particularly in regard to any mechanism which might produce overpressure, (3) determine the rate of melt-through of the reactor vessel, underlying concrete, and/or sacrificial material used to retard melt-through, (2) study methods of post-accident heat removal,

a. HTCA AEROSOL PLATEOUT HISTORY

b. HTCA AEROSOL FALLOUT HISTORY

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Figure 5. Typical HTCA Aerosol Histories

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Figure 6. Arc Melting Furnace

and containment rupture problems in general, and (5) test the effect of the presence of fission products (using stable isotopes) in any of the above.

During the past quarter, several melts and pours of UO_2 have been made onto graphite and ordinary steel plate. In one instance 15 kg of UO_2 was melted and poured onto a steel plate. The molten UO_2 exhibited a low viscosity and spread out in an even layer on the plate without melting the steel except at one edge. This test demonstrated the fluidity of molten UO_2 and its tendency to spread out into a configuration favoring rapid heat transfer.

2. Melting Procedure

A 10-cm layer of stainless steel was first melted in the bottom of the crucible. (A large temperature gradient develops across the stainless steel and allows the solid UO_2 in the crucible bottom to be hot enough to conduct electricity. Then, sintered UO_2 pellets were placed on top of the stainless steel and melted several times so as to eliminate the interpellet voids.

As a final step, the electrode was withdrawn, and the molten content of the crucible was quickly poured onto a plate below. The height of the pour varied between 56 cm and 25 cm as the crucible was tipped. Figure 7 is a photograph of the graphite electrode and the tipped crucible after pouring. Note the tilted crust on surface of the central region. Some "icicles" of UO_2 are seen extending over the edge. The stainless steel fragments shown on the crucible edge and on the catcher below were parts of rods originally placed across the crucible to serve as temporary electrodes to start the arc. This procedure was necessary because UO_2 is effectively an insulator below $\sim 600^\circ\text{C}$. Some erosion of the graphite electrode can be seen at the top of Figure 7 since the electrode had a rounded end when new.

The melting process required ~ 75 min. During this time, the current was slowly increased so as to evenly heat the crucible contents. At times, heavy smoke appeared and reduced the visibility of the surface. The current was then decreased until the smoke cleared. Fresh argon was continuously bled into the furnace tank and the excess gas pumped away through an automatic pressure regulator valve which maintained the argon pressure at 0.5 atm. The argon temperature increased to about 400 to 500°C during the melting. After

pouring, the furnace and melt were allowed to cool and the furnace was evacuated and backfilled with air.

Figure 7. Crucible After Pouring UO_2

3. Results of Pours Onto Graphite

Tests A and B consisted of two pours of molten UO_2 (4.8 and 14.2 kg) onto a flat graphite surface. In both cases, the UO_2 poured without splattering through argon gas and froze into a pancake shape upon contact with the graphite. Little or no chemical reaction took place because the graphite quickly conducted the heat away from the spread out UO_2 .

Figures 8 and 9 show the top and bottom of the solidified UO_2 after Test B. Some stainless steel rods can be seen protruding from the melt. These rods were used to start the arc and probably fell from the crucible edge. During cooling, the UO_2 cracked into several pieces, and these are shown on edge in Figure 10. The pour shape was roughly rectangular, 22 by 23 cm by 2.5 cm thick. The density of two small pieces was measured by weighing in air and under water. The measured density was 9.84 and 9.92 g/cc. (The theoretical density of UO_2 is 10.96 g/cc.)

Figure 8. Solidified UO_2
Top View (Test B)

Figure 9. Solidified UO_2
Bottom View (Test B)

Figure 10. Solidified UO_2
Edge View (Test B)

4. Results of Pours Onto Carbon Steel

In Tests C and D a 50-cm by 46-cm by 0.64-cm thick carbon steel plate was placed so that the UO_2 poured onto the center of the plate. The plate was separated from a graphite-lined pan underneath by 2.5-cm thick graphite blocks placed at each corner. Figure 11 shows the results of Test C. After the pour, 12.2 kg of UO_2 remained frozen as a 0.6 cm layer on the plate, and 3.0 kg ran over the edge. Figure 12 shows the plate after the frozen UO_2 layer was removed. No melting of the plate was apparent except for the two small areas near one edge where the UO_2 ran over and under the plate. Here the molten UO_2 formed a sandwich around the plate. (The hole shown in the plate was originally present and some UO_2 flowed through it.) The plate was slightly warped by uneven heating. Similar results were observed in Test D.

An interesting result of Tests C and D was the small amount of damage to the steel plate. (A similar result has been reported in Reference 6.) Only two small regions near the plate edge were melted even though the UO_2 remaining on the plate originally contained sufficient enthalpy* to extensively melt the steel, not considering any heat lost by radiation. A transient thermal analysis is made in the following section.

5. Thermal Analysis of Pour Onto Carbon Steel

After UO_2 contacts and runs over a metal sheet, it begins to cool by radiation from the upper surface and by direct conduction to the sheet. The molten UO_2 forms a crust on the upper side and lower sides as shown in Figure 13. Heat is supplied as the UO_2 yields up the latent heat of fusion. Finally, the crust grows in size from both sides until it meets in the center.

*The enthalpy difference of molten UO_2 at 3100°K is about 1.4×10^3 j/g with respect to the melting point of steel (1700°K). Thus, the 12.1 kg of UO_2 remaining on the plate after the pour could provide 1.2×10^7 j to heat the plate and melt it. To melt steel at 1700°K requires about 1000j/g. As the steel plate weighs 12 kg, 1.3×10^7 j would completely melt it, roughly the amount of heat available. Thus, if there had been good heat transfer, the steel plate would have extensively melted.

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Figure 11. Frozen UO₂ Layer on Carbon Steel Plate

Figure 12. Plate After UO_2 Removal (Test C)

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Figure 13. UO_2 Freezing on a Steel Plate
(Model for thermal analysis using
TAP-4 Code)

TABLE 1
INPUT PARAMETER FOR TAP-4F CODE

	Steel	Solid UO ₂	Liquid UO ₂
Melting Point (°C)	1430	2860	
Initial Temperature (°C)	100		3000
Thermal Conductivity (cal/cm-s-K)	$0.0298 + 3.85 \times 10^{-3}T$	$2.5 \times 10^{-9}T^2 - 9.21 \times 10^{-6}T + 0.0152$	0.0245
Specific Heat (cal/g)	0.12	$2.49 \times 10^{-8}T^2 - 3.43 \times 10^{-5}T + 0.0746$	0.12
Slab Thickness (cm)	0.63	0.63	
Density (g/cm ³)	8.0	9.8	8.5

The governing equation for the one-dimensional slab geometry is as follows:

$$\frac{\partial T_i}{\partial t} = \alpha_i \frac{\partial^2 T_i}{\partial x^2} ,$$

where

$i = 1$ for molten phase

$i = 2$ for solid phase

T_i = temperature ($^{\circ}\text{K}$) of phase i

t = time (s)

α_i = thermal diffusivity of phase i

The phase change boundary condition which represents the movement of the zone of solidification is:

$$K_1 \frac{dt_1}{dx} = K_2 \frac{dt_2}{dx} = \Delta H_f \rho \frac{dx(t)}{dt} , \quad \dots (9)$$

where

ΔH_f = heat of fusion of UO_2

K_1, K_2 = thermal conductivity of molten, solid phase

Initial conditions are: $T_i = T_i(0)$ at $t = 0$.

These equations may be solved numerically using a computer code called TAP-4.⁽⁷⁾ The UO_2 region is divided into Regions 2 to 21 and the steel into Regions 22 to 42. Time steps of 0.005 s were taken. The input parameters for two phases of molten UO_2 and for steel are given in Table 1.

Figure 14 shows the result of one computer run. The calculation shows that the steel temperature will reach the melting point if the UO_2 has zero porosity and thus, the maximum thermal conductivity. If the thermal conductivity of the UO_2 is reduced by 16% porosity, the calculation indicates that the steel will not melt. The steel temperature almost reached the melting point. In the experiment, the steel did not melt except at the edge where there was a thick

Figure 14. Temperature vs Time After Contact
(TAP-4 Simulation)

layer of UO_2 surrounding it. Actually, the maximum temperature of the steel must have been 200° or more below its melting point as the steel plate did not sag even though it was supported at the four corners by small blocks of graphite.

6. Conclusions

Tests C and D seem to indicate that even relatively thin layers of steel could potentially provide considerable protection against immediate melt-through from molten UO_2 fuel on a steel-lined reactor cavity. The fuel would quickly spread out evenly into a more coolable configuration (with higher radiation losses) without necessarily penetrating the liner. These questions of insulation at the steel- UO_2 interface, radiation losses from the UO_2 , and other heat transfer related problems will be the subject of further detailed investigations using the TAP 4 Code.

III. NEXT REPORT PERIOD ACTIVITIES

A multiple bend leak path representing a leaking reactor head seal will be built. Attenuation tests will be tested using particulate concentrations of ~ 40 and 500 g/m^3 , with pressure differentials of 0.7 and 2 atm, respectively.

Studies of particulate attenuation through straight capillaries will continue.

The high temperature-concentration aerosol tests to be conducted during the next period will emphasize collection of experimental data to determine the time-dependent fallout and plateout of aerosols during a test. Preparations are being made to modify the incremental fallout and plateout sampling device to obtain these data.

The tests to be performed at the arc melt test facility will include melting larger and larger quantities of UO_2 and pouring this melt onto stainless steel, ceramic and concrete samples.

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