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**"RAPID QUENCHING OF MOLTEN LITHIUM-ALUMINUM JETS IN
WATER (U)"**

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Rapid Quenching of Molten Lithium-Aluminum Jets in Water

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Introduction:

The Westinghouse Savannah River Company operates the "K" production reactor at the Savannah River Site in South Carolina. This is a heavy-water moderated and cooled reactor designed for the efficient production of radioisotopes, including tritium. Operation is restricted to temperatures below the boiling point of the coolant at atmospheric pressure; this permits the fuel and many components to be fabricated from aluminum and its alloys.

Control rods for the K reactor are grouped in assemblies of seven rods, called "septifoils". As shown in the schematic diagram in Figure 1, the septifoils have a cooling system that is separated from the fuel cooling system. Cooling water for the tubular fuel assemblies is circulated downward from an upper coolant plenum; cooling water for the septifoils is circulated upward from a coolant header underneath the reactor, and flows through the septifoil housing tubes to cool those control rods that remain inserted during operation. A cross-section of the septifoil housing and tube positions is shown in Figure 2. The number of rods in place during reactor operation may vary from one to five. The control rods are fabricated of aluminum-lithium alloy, typically about 3 weight percent lithium, clad in a thin aluminum jacket.

Safety analysis of potential problems in reactor operation identified a class of incidents in which problems might arise in the control rod cooling system, leading to inadequate cooling and coolant boiling inside a septifoil housing. The result would be voiding of a large part of the space inside the housing, and loss of cooling for part of the cooling rods. Seepage of moderator through the bottom fitting should maintain some depth of water in the bottom of the housing.

A particular concern identified was that overheated cooling rods might partially melt, so that the resulting molten metal could drain into the water at the bottom of the housing. If the resulting melt-water mixture underwent a vapor explosion, it might burst the thin septifoil housing and damage the nearby fuel assemblies. Literature data indicated that molten aluminum-lithium alloy could give energetic explosions under certain conditions.¹ However, the experiments in the literature were artificially

triggered; it was not clear that self-triggering of vapor explosions was likely under the conditions of interest. The experiments described here were undertaken to investigate this question, and to characterize any explosions that might occur.

Fortunately, and atypically for questions of vapor explosions in nuclear reactor accidents, this problem allowed experiments to be done that were fully prototypic in scale and geometry. Calculations of melting rates showed that the maximum amount of molten material present in a septifoil at any given time might range from tens to hundreds of grams, depending on the assumptions made about the holdup of molten material inside unmelted cladding.² In the course of this work experiments were performed on the melting of control rod segments in a furnace, and these suggested that the actual maximum amounts of melt present at any one time would be in the lower part of this range. However, for conservatism, in this study up to one kilogram of molten alloy was contacted with water at a time. The melt was contacted with water inside actual septifoil housing tubes complete with end fitting and spacer. Conditions were varied in an attempt to include those factors that might trigger a vapor explosion. The experimental assemblies were instrumented to measure pressure and temperature, and post-irradiation examination of each experiment was performed to characterize the quenching process.

Experimental

Two different but similar experimental assemblies were constructed, each incorporating similar basic components. A melt delivery system consisting of a resistance furnace and pour spout was positioned over the top of a length of septifoil housing. When the material in the furnace had thoroughly melted and was at the desired temperature, the melt was allowed to drain into the septifoil assembly and be quenched by water there.

The experimental apparatus built at Argonne National Laboratory is shown in Figure 3. This apparatus was located in a remote, blast-resistant cell, with three-foot-thick concrete walls and fourteen-inch-thick steel doors. It was designed to melt and deliver up to 1 kg of molten metal. Its features include a furnace mounted on top of the assembly, a pneumatic mechanism for delivery of the melt, a distribution plate that can distribute melt into one or several channels of the septifoil (Section C-C in Fig. 3), a water-filled interaction vessel, and a cooled accumulator vessel for condensing steam from the assembly.

The melt furnace/injector was 3.75 in inside diameter and eight inches high. It was wrapped with 1/16 in coaxial heater that could deliver 2 kW of heat. Injection of the melt was achieved by having a cutter actuated by a pneumatic piston cut through a vitreous aluminosilicate fiber crucible inside the furnace, which contained the melt.

The interaction vessel was nine inches in diameter, and was filled with water up to three inches from its top flange. Nearly all of the septifoil housing used in the test was submerged in the water. An aluminum mounting rod one inch in diameter was used to locate the top of the housing two inches above the water level, and one inch below the bottom of the injector tube. Two stainless steel rings with threaded positioning rods were used to locate the septifoil laterally under the injector tube opening. During the tests, the plenum space above the water was purged with argon. For all tests except #4 two gas samples were withdrawn from the plenum above the water at ten and fifteen seconds after the melt injection.

The steam accumulator was a plastic container partially filled with water and vented. It was connected to the interaction vessel by a heated, insulated one inch copper line. The end of the line was capped, and steam was emitted into the accumulator below the water level through sixty-four 1/4 in holes. The water for condensing steam was initially at room temperature. In the first three experiments a one-gallon plastic container was used; in the final two a 2.5 gallon plastic container replaced it.

Figure 3 shows the thermocouple and pressure transducer mountings in the interaction vessel. The thermocouples were chromel-alumel, mounted in a 1/16 in stainless steel sheath. A pressure transducer reading 0 to 1000 psia was mounted near the bottom of the interaction vessel, and a 0-2000 psia transducer was located near the center of the vessel. A transducer reading 0 to 100 psia measured the plenum pressure above the water. The thermocouple readings were recorded by Digitrend Model 210 during the heatup phase. A Honeywell 1858 Visicorder and a Honeywell 101 magnetic tape unit were used to record temperature and pressure data. Heat flow to the furnace and to the interaction vessel were regulated by 0-1000°C Weather Measure Model PTC-1 temperature controllers.

The apparatus built at Brookhaven National Laboratory is shown schematically in Figure 4. Blast shielding is provided by enclosing the septifoil housing in a six-inch, schedule 120 stainless steel pipe. (The maximum amount of alloy melted was approximately 120 g. An analysis showed that the pipe enclosing the septifoil would resist damage from an explosion involving a much larger amount of melt and water.) This structure is 36 inches long and has a 150-lb slip-on flange welded at each end. (H and J in Fig. 4) The bottom is penetrated by a liquid-filled dip leg for a pressure transducer, and by a gas injection line. The top has five penetrations: a half inch centered hole for the injection of the melt from the crucible A, two steam vent lines that direct steam into a water reservoir for condensation, a thermocouple port, and a gas bubbler to mix the water in the blast vessel to ensure uniform temperature. The water pool temperature, the melt temperature, and the water pool pressure are recorded at a rate of 100 Hz by a HP1000 data acquisition computer. The calibrated, filtered and amplified data are transferred to magnetic tape for storage. Debris from the experiments was collected, characterized, and photographed.

The melt crucible used in these experiments is shown in Figure 5. It is constructed of stainless steel, oxidized at 800°C for 24 hours in air so that it would resist attack by the molten alloy. The crucible is constructed from a four inch long piece of two inch diameter stainless steel tubing, welded at one end to a two inch diameter slug of solid stainless steel. This slug is milled to provide a conical seat for the rod that opens and closes the drain hole, and the half inch drain into the center of the septifoil housing. The crucible can be inerted with argon, which is necessary to avoid air oxidation of lithium. The argon was maintained at a slight overpressure, which also served to help force the melt through the drain hole and out of the crucible. The rod that seals the drain is connected to a pneumatic, spring-loaded cylindrical piston, which is operated electrically to open the drain line. A microswitch on the piston is connected to the data logging system and serves as an event marker for the experiment.

The crucible is placed in a cylindrical clamshell resistance heater. A ceramic fiberboard insulation block, "H" in Figure 5, insulates the crucible from the blast vessel and avoids heat loss. Additionally, stainless steel shims were positioned between the insulation block and the flange during the heat-up phase to minimize heat loss from the crucible.

In the Brookhaven experiments a variety of parameters were varied to investigate possible triggering mechanisms. These variations included variations in water temperature; the presence of an obstacle a few inches above the melt surface; dropping a rod section in a side channel at the same time as the melt was entering the water; creating a stratified temperature distribution in the septifoil, with hot water on top and cold water beneath; and injecting gas near the bottom to provide turbulence. The rod drop was triggered by one of two methods; either the rod was suspended by a monofilament line that was in the path of the falling melt, or a cable attached to the piston that controls the furnace drain was used to release the rod. Both methods appear to have functioned as designed, in that the rod struck the water surface while melt was falling through the water.

All the Brookhaven experiments were run at a melt temperature in the range of 645°C to 660°C. Under these conditions the melt flowrate was in the range of 100 to 200 g/sec. The experiments were designed to deliver 100 g of melt from the crucible; this was not achieved in all experiments because of various experimental factors.

Experimental Results

Argonne Experiments

The Argonne experiments are summarized in Table 1. The thermal conditions were identical in all tests: the melt temperature was in the range of 680° to 700°C, ensuring that the alloy was fully molten with minimal superheat. The water temperature in all experiments was 85°C.

In the first experiment the melt was poured as a single stream into one of the peripheral channels of the septifoil; in the other it was poured through the perforated plate that divided the stream among five adjacent peripheral channels around the septifoil. In each case an eleven inch section of control rod was located in the bottom of each channel into which melt was poured. The effect of water depth was investigated: in tests 1, 2 and 5 the depth was twenty-four inches inside the housing; in tests 3 and 4 it was sixteen inches. The melt mass was 0.5 kg in the first test and 1.0 kg in the remainder.

No explosive events occurred in any test, nor was there any damage to the septifoil housing and structure. No significant pressure pulses were detected by the two transducers in the interaction vessel outside the septifoil. Some pressurization of the plenum space was observed in all tests, ranging up to about 0.15 MPa.

Boiling of the water in the septifoil forced substantial amounts of water out of the septifoil housing. Table 2 shows the water losses from the septifoil, along with estimates of the amount of vaporization produced by the thermal energy of the melt. The amount of water ejected was much greater than could be accounted for by vaporization; it was evidently blown out by steaming. In the two tests at low water level the septifoil was found to be nearly dry.

Metallic debris was recovered as loose rubble in the bottom of the septifoil. Figure 6 shows an x-radiograph of the bottom of the septifoil following the first test. The debris accumulated in all channels. Similar results were found in the other tests. Additionally, a fine white powder was found adhering to the debris; x-ray diffraction analysis at the Savannah River Technology Center identified lithium aluminum carbonate hydroxide, $\text{Li}_2\text{Al}_4(\text{CO}_3)_3(\text{OH})_{12}\cdot 3\text{H}_2\text{O}$ and aluminum hydroxide, $\text{Al}(\text{OH})_3$, among the components of the powder. (Presumably the carbonate was formed by the reaction of a hydroxide with air during handling.) The presence of these hydroxides shows that some chemical reaction with the water occurred; additional evidence was found from pH measurements of the septifoil water, which was in the range of 10 to 12, and from the measurement of significant amounts of hydrogen in the gas plenum samples. Chemical reaction of melts with water was observed and quantitatively measured by Nelson and his co-workers in smaller scale experiments.³ These results appear to be consistent with his findings.

Unexpected and interesting temperature transients were observed during these experiments. An example is the temperature trace of a thermocouple in one of the injector channels during the third test (Figure 7). Upon melt injection, the trace shows the melt temperature. Within two seconds this decreases to about 100°C, suggesting that saturated steam is flowing through the channel. Then, unexpectedly, the temperature increases well above the initial melt temperature. The only source of heat in the system is chemical reaction; a plausible explanation for the temperature increase is

that a chemical reaction between steam and residual melt adhering to the channel walls produced the heat. Temperature increases were also noted in the melt crucible after it was opened to the septifoil. These might also be attributed to the reaction of steam with residual material.

Brookhaven Experiments

The Brookhaven experiments are summarized in Table 3. As in the Argonne experiments, no explosions occurred. In some experiments, however, pressure pulses as high as 18 psi were observed by the transducer.

First Test Series. The first series of five tests (101 to 105) was run to evaluate the apparatus and to study the effect of water temperature. In these experiments the water pool depth was at least 86 cm, the melt temperature was 660°C, and the overpressure in the crucible that forced the melt through the drain orifice was 2 psi. Water temperature was varied systematically from 100°C to 33°C.

The most interesting observation from these experiments was pressure spikes ranging up to 18 psi. These were observed primarily at water temperatures of 60°C or 80°C. (Figure 8) In these experiments, and in the experiment at 100°C, some of the quenched debris was carried out of the septifoil housing through the steam vents or the top of the vessel by the venting steam. Copious steam emission was observed at the higher temperatures, but not at the two lowest temperatures. At lower temperatures melt quenching was a more orderly process, and pressure pulses were very small. Much of the melt was found quenched and retained along the separator webbing. In general little oxidation of the melt was observed, and the melt was recovered as small rounded or irregular particles.

The second test series (201 through 207) was run to examine the effect of shallow water, and of several potential triggering mechanisms. In this series the water depth was about 30 cm, and in all tests the water temperature was 60°C. This was the temperature in the prior series at which the most violent boiling and pressure spikes were observed. However, in none of these experiments were any large pressure spikes observed.

The first variation in conditions was inadvertent; in test 201 the pressure transducer dip leg was found to have entered the central channel several inches above the water surface. This would be expected to divert and break up the falling stream. However, little difference was found between this test and test 202, which was a similar test without such a diversion. In both cases most of the melt was recovered as loose rubble on the septifoil floor.

In the remaining tests in this series a section of control rod 2.22 cm in diameter and 26.7 cm in length was dropped into the water at the same

time as the melt. The melting of a nylon monofilament line by the falling metal was used to trigger the rod drop. The purpose of these experiments was to determine whether the shock of the rod hitting the water could serve as an explosion trigger. (In experiments reported in the literature, involving larger quantities of aluminum melt, the shock from a blow on the exterior of the vessel has triggered explosions.) In this case the shock from the falling rod was observed to be relatively small; the pressure transducer recorded a maximum pulse of about 0.25 psi from the rod impact. The mode of rod release and timing was varied, but there was little difference in the results of the experiments, which resembled those from tests 201 and 202. In each case some of the frozen metal was found adhering to the septifoil webbing, and the remainder in the bottom of the septifoil housing.

The third test series (301 to 304) involved a variety of experiments on potential explosion triggers. There was considerable variation in the conditions among these experiments.

Test 301 was an experiment in thermal stratification, and was designed to test whether this condition could serve as an explosion trigger. Stratification was achieved by heating the septifoil near the top. The septifoil was filled to its maximum level. A temperature of 73°C was reached near the top of the septifoil, while near the bottom the temperature was 25°C. Much of this gradient is believed to have been just below the heated zone, perhaps amounting to as much as 10°C/cm. No unusual pressure events were observed when the melt was dropped into the septifoil. The debris was found to have frozen after passing into the colder water, and most of it was hung up on the septifoil webbing in the form of rather large fragments at about the 50 cm mark.

Tests 302 and 303 were designed to simulate vigorous boiling of the water in the septifoil, by the injection of nitrogen into the base of the septifoil after the water had been heated to near 100°C. In run 302 the water depth was 60 cm, and the vigorous bubbling had the unintended effect of cooling the delivery system so that only a portion (23 g) of the intended melt actually reached the septifoil. In test 303 the water depth was reduced to 30 cm and a larger amount, about 58 g, of melt was delivered to the septifoil. In neither case was any unusual pressure pulse or other unexpected event observed.

Test 304 was a rod drop experiment similar in many respects to the 200 series of tests: the water depth was about 30 cm, and the same rod was used. The water temperature was higher, about 82°C. The principle difference in this experiment was that the rod was released by a cable attached to the piston opening the melt valve, so that there was no delay associated with melting of the monofilament line. Again there was no unusual occurrence in this run, and the solidified melt was recovered as a loose pile of debris from the bottom of the septifoil.

Discussion and Conclusions

These experiments indicated that under the conditions of interest a steam explosion that would damage the septifoil is unlikely. This is consistent with what is known of spontaneous triggering of steam explosions:

- Spontaneous triggering of Al or Li-Al melts is never observed at the 10 g level, according to Nelson³;
- Spontaneous triggering has been observed at the 10-20 kg level in a variety of experiments involving molten aluminum and its alloys⁴;
- However, even at this scale spontaneous triggering is unusual when the melt temperature is near the melting point⁴.

There was some concern that the chemical reaction of lithium with water might trigger a damaging steam explosion. Clearly, reaction occurred, and there is evidence for locally high temperatures as a result, but it did not lead to a metal fire or an explosion.

None of the triggering methods involving plausible phenomena associated with an accident, which were investigated in these experiments, triggered a steam explosion. No explosion occurred in the Argonne experiments even though boiling was so vigorous as to force nearly all the water from the septifoil.

The experimental results from these experiments were combined with other information, including probabilistic data, that had been developed on incidents that might cause inadequate control rod cooling. The resulting analysis showed that the likelihood of an accident that would propagate to cause significant core damage was acceptably small.² This conclusion was accepted by review bodies prior to the restart of K Reactor in 1992.

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Table 1. ANL Test Conditions

	A	B	C	D	E	F
1	Test Number	1	2	3	4	5
2						
3	Water height above rods, in	13	13	5	5	13
4	Number of rods and orifices	1	5	5	5	5
5	Melt mass, kg	0.5	1	1	1	1
6	Melt delivered, kg	0.4	0.5	0.8	0.8	0.8
7	Delivery rate, kg/sec	0.3	0.9	0.9	0.9	0.9
8	Melt temperature, °C	680-685	680-685	685-690	690-700	685-690
9	Melt jet diameter, cm	1.4	1.2	1.2	1.2	1.2

Table 2. Water Ejection

	A	B	C	D	E	F
1	Test No.	1	2	3	4	5
2						
3	Volume in septifoil before test, cc	2940	2400	1525	1500	2400
4	Volume remaining after test, cc	2110	1100	0	0	370
5	Net water ejection, cc	830	1300	1525	1500	2030
6	Maximum vaporized water, cc	154	193	309	309	309
7	(Thermal energy only)					

Table 3. Parameters for BNL Tests

Test	Injected Melt Mass (g)	Melt Temperature (°C)	Water Depth (cm)	Water Temperature (°C)	Trigger Method
101	36.2	647.8	91	100	--
102	76.5	671.8	86.4	80	--
103	74.9	664.0	86.4	61	--
104	57.7	660.6	86.4	42	--
105	80.7	656.5	86.4	33	--
201	98.8	662.0	30.5	62	Obstacle
202	71.8	654.9	30.5	60.5	--
204	102.1	658.0	30.5	60.5	Rod Drop
205	110.3	658.4	30.5	60.3	Rod Drop
206	109.0	660.4	30.5	60.2	Rod Drop
207	76.5	656.5	30.5	62.0	Rod Drop
301	98.4	662.1	91.4	25 to 73	Stratified Water
302	23.0	658.6	61.0	99.6	Gas Injection
303	57.6	670.4	30.5	100	Gas Injection
304	91.4	664.6	30.5	81.5	Rod Drop

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Figure 1. Schematic diagram of the reactor coolant system

Figure 2: Cross-Section of a Septifoil with Rods Inserted

Figure 3. ANL Test Apparatus

Figure 4. BNL Test Apparatus (Schematic)

Figure 5. BNL Crucible and Furnace (Schematic)

*Photo available
for final
submital*

Before Test

After Test

Figure 6. X-Radiographs of Septifoil Before and After ANL Test 1

Figure 7. Temperature in Injector Channel, ANL Test 3

Figure 8. Pressure pulses observed in Expts. 102 and 103.
The maximum pressure is about 18 psig (103)

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