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DURING SEVERE ACCIDENTS (U)**

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Phillip G. Ellison, P.R. Monson, and M.L. Hyder

Westinghouse Savannah River Laboratory
Savannah River Site
Aiken, SC 29808

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Phillip G. Ellison, P.R. Monson, and M.L. Hyder

Westinghouse Savannah River Laboratory
Savannah River Site
Aiken, SC 29808

ABSTRACT

An assessment of fuel melt behavior during hypothetical overheating and melting incidents was made for nuclear reactors that use aluminum alloy fuels, which are typically composed of an aluminum/uranium alloy with an aluminum cladding. The assessment was based on analyses of several overheating incidents and on the results of a number of experimental test programs.

The analyses indicated that the fuel failed in three distinct stages. At temperatures below melting, blistering of the cladding was observed, followed by cracking of the aluminum cladding at higher temperatures. When low-burnup fuel melted, it flowed in rivulets through the gaps in the cracked cladding over the oxidized cladding surface, and higher-burnup fuels exuded a metallic molten foam through the cracks.

INTRODUCTION

Fuels composed of an aluminum/uranium core and aluminum cladding are widely used in research reactors and also in the Savannah River Site (SRS) production reactors. This composition has the advantage of excellent heat transfer properties along with being relatively easy to fabricate and reprocess. It does, however, have a relatively low melting temperature, typically 650 degrees Celsius. This is only a problem when some abnormal condition prevents normal cooling. Depending on the extent of such conditions, the result can be localized fuel damage or even extensive reactor core melting.

Several studies of fuel overheating were made as part of the safety evaluations of these reactors. Additionally, several incidents involving varying amounts of fuel damage have actually occurred. Table I shows the sources of these incidents and a summary of the information accumulated is given in the following sections.

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Table 1. Nuclear Metallic Fuel Melt Behavior Data Base

<u>Data Source</u>	<u>Blisters</u>	<u>Cracks</u>	<u>Flow From Cracks</u>	<u>Flow Regime</u>		
				<u>Film</u>	<u>Rivulet</u>	<u>Foam</u>
SRL Out-of-Pile Tests		X	X			X
SRL Annealing Tests	X					
ATR Annealing Tests	X					
SRS In-Pile Incidents	X	X	X		X	X
Westinghouse Test Reactor		X	X		Unknown	
SPERT Tests		X	X		X	
SRL SPERT Tests		X	X		X	
TREAT Experiments	SMALL SAMPLES FOR CHEMICAL OXIDATION EFFECTS ONLY					

DATA SUMMARY

Blistering

Annealing of Al/U fuel plates and tubes has shown that a threshold exists for fuel failure from extensive blistering, which is caused by cracking in the aluminum matrix. This observation was established by post-test metallographic examination of annealed fuel specimens. Internal gas pressure causes the cracks to grow and to exert pressure on the fuel-clad interface. Blistering occurs when the gas pressure and clad temperature have reached a critical condition that causes the clad to locally debond from the fuel. There are several sources for the gas that exist in the fuel matrix. It can be absorbed during fabrication, from gas-forming impurities in the matrix aluminum, and from fission products. The onset of blistering is not a strong function of burnup, leaving gas-forming impurities and gas absorption during manufacturing as the major contributors to blistering. The following categories of blisters were observed.

- Small blisters, typically two to three millimeters in diameter, where separation occurs at the fuel/clad interface
- Intermediate blisters, several millimeters in diameter and similar to the small blisters formed at the fuel-clad interface

- Large blisters, typically several centimeters in diameter, that occur in the fuel core itself

Studies have been conducted at SRS and at the Idaho National Engineering Laboratory (INEL)¹ of the swelling of uranium and aluminum/uranium alloys upon annealing. Studies with aluminum/25 weight percent uranium alloy indicated that annealing at 400 degrees Celsius caused very little swelling, but temperatures from 475 to 550 degrees Celsius resulted in extensive cracking or blistering of the fuel cladding. Swelling is low until about 10 degrees Celsius below the melting point of the fuel. However, above this temperature, extensive deformation is noted and release of fission gas begins to occur as a result of fission-gas bubble agglomeration on UAl4 grains.

All the in-pile overheating incidents that occurred at SRS caused blistering near the regions of localized burnup.

Clad Cracking and Fuel Melt Flow Regime

Al/U fuels fail in three distinct stages during a severe core damage accident. In the first stage, blistering occurs; in the second, the clad cracks; and in the third stage, molten fuel material drains from the cracked, but still solid, cladding. Information is available from a wide range of in-pile melting incidents in which the latter two stages were observed, including the SRS overheating incidents and the Westinghouse test reactor accident. Both plate and tubular elements failed in this manner during in-pile experiments conducted in the Special Power Excursion Reactor Test (SPERT). Tubular elements heated by induction also failed in the same way during out-of-pile experimental programs.

The following paragraphs provide additional information to support these observations.

Simulated Fuel Tube Melting Studies

Several experiments were performed at Savannah River Laboratory (SRL) with flowing steam, in which, a one-inch diameter Al/U tube clad with 8001 aluminum alloy was inductively heated to the melting point. The experiments indicated that the fuel cladding cracked; and the molten fuel core flowed out the cracks and down the surface of the tube, freezing in place as an agglomerated sheet on the unheated portion of the tube. Post-test examination of the tube indicated cracking had occurred along the grain boundaries of the cladding. It is believed that clad cracking occurred because of grain boundary melting and because of the pressure exerted by the five-to-six-percent volume increase of the melted fuel core. Frozen fuel droplets were found on the surface of the cladding indicating that the fuel was not wetting the cladding surface. Post-test photographs showed that the trailing edge of the droplet contact angle with the cladding surface was close to zero degrees.

Experiments were also conducted with 1100 aluminum alloy tubes³, which were heated in an induction furnace to failure. Failure occurred before bulk melting and resulted in the formation of axial through-wall cracks along the surface of the tubes. Surface examination of the tubes indicated that the cause of the failure was grain boundary melting of the alloy. Large pieces of the tubes dropped off leaving openings to the inner parts.

In-Pile, Post-Incident Fuel Melting Analysis

Californium-252 was produced at SRS using special metallic Al/U fuel element assemblies of three concentric fuel tubes. Each tube consisted of six feet of enriched Al/U alloy clad with 1100 aluminum on the inside and 8001 aluminum alloy on the outside. The cladding and fuel were

metallurgically coextruded to form an adequate heat transfer bond to the fuel. Six of the fuel elements experienced localized melting during early 1970.

Post-failure analysis revealed that the fuel failures were caused by the formation of cladding holes and cracked cladding with localized Al/U melting. Several other tubes showed signs of blistering and pitting. Rib marks were noted on the surface of the tubes, and melting occurred in patches parallel to the rib lines. Failure resulted from burnout in patches on the cladding surface, which was caused by the rib effects. Heat transfer to adjacent tubes and coolant occurred through the ribs, limiting melting to the center of some of the rib circles. Some of the failures resulted in fuel draining and freezing on the outer clad surface. In other failures, the molten fuel remained in place and did not move from the failure location.

A metallic fuel assembly used at SRS to produce plutonium failed as a result of partial melting after 33 days of irradiation in early 1970. The failure involved more than 50 percent of the assembly. Fuel failure was accompanied by clad blistering, clad cracking, Al/U core melting, and drainage through cracks in the cladding. Post-failure analysis indicated that the fuel drained as rivulets on the surface of the cladding and froze at cooler locations that were not experiencing dryout. Fission-gas bubble agglomeration caused the cladding material to expand, doubling its transverse thickness in some locations of the assembly. Fission-gas bubbles were present throughout the molten fuel core material, indicating the onset of significant fuel foaming.

In April 1960, a fuel element failed in the Westinghouse test reactor and released fission products into the coolant system². Coolant boiling experiments were being conducted on this metal-fueled, water-cooled reactor before the failure occurred. These boiling experiments resulted in the melting of one high-powered fuel assembly in the core of the reactor. Examination of the accident indicated that the fuel core melted inside the solid cladding and the molten core drained through cracks that formed in the cladding. The molten fuel refroze in the coolant channel at a low-power region of the assembly, forming a porous blockage in the bottom of the fuel assembly.

Special Power Excursion Reactor Test

Six fuel tubes similar to those used in the SRS reactors were melted in SPERT I in 1958³. These two-foot-long fuel tubes were an aluminum alloy with 31-weight-percent uranium. The fuel was clad with aluminum. The tube was formed by coextrusion of the fuel and cladding. The coolant flow for the fuel tube was downward as in the SRS reactors.

The experiments were conducted by placing the test reactor on a short reactor period, which caused the metallic fuel tubes to overheat and partially melt. Visual and radiographic examination of the melted fuel tubes showed that the fuel material had melted and flowed out of cracks in the cladding. This occurred because the melting point of the fuel was 20 degrees Celsius lower than the melting point of the cladding. In no case did the fuel assembly spacer ribs melt. The following three factors prevented the ribs and the local region near them from melting.

- The cladding of the tubular fuel design was thicker at the ribs than in the rest of the cladding.
- The ribs acted as an effective heat transfer medium, conducting and convecting heat to the surrounding tube and to the flowing coolant.
- The molten fuel flowed from the core beneath the ribs and reduced the heat source as soon as a crack in the cladding occurred.

Molten fuel drained as rivulets to the bottom of the test assembly in these experiments. Near the bottom of the assembly, the rivulets collected on a cold surface as an agglomerated sheet that

flowed as long drops, or jets, to the bottom of the test assembly. Some of this melt was carried by the flowing coolant out of the test assembly. For the most severe melting case, in which about 25 percent of the fuel melted, about 7 percent of the molten fuel was recovered as a particulate. The particulate was in the form of jagged flakes and agglomerates of once molten particles.

SPERT Experiments

A series of experiments were performed in the SPERT I reactor to investigate the effects of rapid transients on metallic fuel plate behavior⁴. The experiments progressed in energy yield to the point where a reactor core was substantially melted and then destroyed by a steam explosion. Several conclusions were drawn from the SPERT data by the investigators. The most important conclusion relative to metallic fuel behavior was that, subsequent to thermal distortion, molten fuel escaped through cracks in the unmelted clad. The fuel material in this series of experiments flowed as rivulets down the surface of the fuel plates. The rivulets were thick enough to bridge the gap between adjacent fuel plates in many cases.

Transient Reactor Test Facility Experiments

The Transient Reactor Test (TREAT) facility is an experimental test reactor located at INEL. It was used in assessing the reaction of small metallic fuel plates to rapid heating transients. The fuel plates were made of Al/U alloy and clad with an alloy of aluminum similar in composition to those used in SPERT. The reactor periods that drove the fuel to destruction ranged from 0.108 to 0.285 seconds.

The plates that were subjected to moderate energy inputs fused into a sphere. Peak temperatures in these experiments were less than 1200 degrees centigrade. The plates became incandescent and chemically reacted with the water surrounding the molten fuel.

At the highest energy inputs, the fuel samples ignited and sustained under-water burning of the samples. Molten fuel temperatures in the tests exceeded 2000 degrees centigrade. Extensive fragmentation of the molten fuel also occurred during these high-energy transients.

In almost all of these experiments, the small fuel samples and cladding material fused into a uniform molten mass. Strong evidence of the effects of surface tension was seen upon melting. The fuel plates collapsed into a thin ribbon as the fuel melted and began to fall. In some of these experiments, the interaction of the fuel with the coolant produced an extensive void in the fuel to the point that the fuel floated in the water.

Molten Metal Flow Regime

The previous review of the in- and out-of-pile melting experiments and the SRS incidents showed that the molten alloy tends to bead up on the surface of the cladding. In those incidents in which a small localized burnout occurred or a small region overheated because of clad debonding, the molten fuel material did not move from the site of the melting.

In other cases, the molten material flowed in rivulets near the ribs of the assemblies or bridged large gaps between adjacent fuel plates, indicating that the fuel alloy did not wet the cladding surface. Molten fuel may not spread on the surface of aluminum clad fuel elements because of the nonwetting of molten aluminum on aluminum oxide⁵. In these conditions, the fuel melt will drain and the leading and trailing edges of the fuel deposit will form an angle with the solid surface, which is the contact angle between the melt and the solid. The contact angle is a lumped parameter description of the physical and chemical factors responsible for the nonwetting of the solid by the molten material. The solid is considered incompletely wet if the contact angle is

finite, and this results in a fuel drainage regime described as a rivulet-type flow. Molten rivulet droplets will not drain until their mass exceeds a certain critical value, which is a strong function of the receding and advancing contact angles. Molten rivulets may remain in place without moving until they are captured by an advancing melt front. The rivulets will move downwards until their mass falls below the static value and then will stop and freeze in place.

In film flow, the draining process is limited solely by frictional resistance to flow, whereas, rivulet flow is limited by both frictional resistance and interfacial forces. The film quickly covers the surface and becomes quite thin in comparison with channel thickness.

Foam Flow

Metallic foams resulting from the agglomeration of fission gas and other gas bubbles can occur in fuel melts⁶. Experimental data from out-of-pile tests on uranium and aluminum/uranium alloys indicate that foams do occur. Metallic uranium foams were seen to form in the following manner.

- Cracks form in the fuel specimen and rapidly increase in length and width.
- The cracks appear to grow in width more than in length.
- When the metal begins to melt, the jagged edge of the cracks become smooth.
- Bubbles form and then become round from the surface tension forces.
- Bubbles grow by agglomeration and form a low-density foam.

Test evidence indicates that metallic fuel foams occur with low burnup. One metallic fuel element that failed at SRS because of localized overheating exhibited the onset of foaming. Fission gas bubbles were seen in the post-incident metallographic examinations. The foam began to form in the fuel and then expanded to force its way through the cladding cracks.

Measurement of the Fuel Alloy Fluid Point

Experiments were conducted at SRL to determine the fluid point of molten Al/U alloy. Small droplets of the alloy were cast and suspended from a balance, heated to melting, and the fluid-point temperature was measured. These experiments demonstrated that the Al/U alloy begins to flow at its eutectic point with no superheat. The fuel alloy behaves in the same manner as a homogeneous solid does upon melting. At the eutectic point, the temperature of the alloy remains constant until the free unbound aluminum is molten. The temperature then increases until the phase transitions of the Al/U compounds begin. At the phase transition temperature, the liquid temperature remains constant until sufficient energy is absorbed to complete the transformation of the Al/U compounds to the next metallic phase.

Summary and Conclusions

The following conclusions were derived from the out-of-pile and in-pile experimental programs and post-test examinations of failures of metallic fuel assemblies that led to melting.

- Whether at power or during decay heating conditions, undercooled fuel overheats and fails in the following distinct stages.
- Clad blisters.

- Clad cracks.
- Fuel melts and flows through cracks in the cladding.
- Metallic fuels melt and flow at the eutectic point. For low-burnup fuel, rivulets form. For fuel with a sufficient inventory of noncondensable gases, a fuel foam develops and the fuel drains as a low-density metallic froth.

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