THE RESPONSE OF SPENT LWR FUEL TO EXTREME ENVIRONMENTS

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INTRODUCTION

The research reported in this paper addresses the radiological source term which could arise when irradiated fuel in transport from a commercial light water reactor is exposed to the extreme environments postulated for some transportation accidents, specifically those involving a fire. The release of spent fuel radionuclides to the environment requires a breach of both the cask and the fuel rod cladding. Past research has given significant emphasis to evaluating the response of the shipping cask to mechanical and/or thermal loads from hypothetical accidents. Less consideration has been given to evaluating the response of the fuel rods to these environments. In this paper, the response of the fuel rods to an extreme thermal event was experimentally evaluated and the quantity of solid fuel material that could be released from the fuel rods to the cask cavity was estimated.

Briefly, the objectives of this study were as follows:

1. Identify those conditions within a transportation cask which might produce fuel-rod cladding failure, emphasizing conditions associated with fires, and
2. Determine by experiment and analysis the nature of the source term so produced.

The release of radionuclides from coolant or deposits on the outer surfaces of the fuel assembly was not addressed in this study.

LITERATURE SURVEY

Two literature surveys were conducted to address the first objective: the first survey was performed to examine the data on all mechanisms by which zircaloy-clad fuel rods might fail; the second to identify which failure mechanisms would most likely occur in a severe fire environment associated with an extraordinary transportation accident. The key findings of these surveys were as follows:

1. For fire temperatures and exposure times defined for licensing purposes by the US NRC\(^1\) and the IAEA\(^2\) regulations, no failures of fuel rods inside the cask are expected to occur.
2. The most likely fuel rod failure mechanism in a severe fire with temperatures greater than 800°C for longer than two hours would be a creep rupture failure induced by high temperatures and the pressure differential across the cladding.
3. There are considerable data in the scientific literature relevant to creep rupture failure, but none which fully characterize the associated radiological source term.
The lack of radiological source term data relevant to creep rupture failure indicates that experiments to better characterize the source term are justified.

SCOPING ANALYSIS OF THERMAL ENVIRONMENTS FROM TRANSPORTATION ACCIDENTS

To determine the likelihood of fuel rod failure as a result of an extreme thermal environment such as might accompany a fire, initial efforts of the program were directed toward characterizing conditions inside the cask. From a safety perspective, only those events in which the cask is breached are significant to this analysis. Thus it is assumed that any medium, such as water, used to cool the fuel rods during transport would be lost as a result of the event. From that point onward, the combination of the decay heat generated by the fuel and the external heat from the fire would act to increase the fuel rod's temperature.

Figure 1 shows calculated transient temperature curves for a single-PWR spent fuel shipping cask exposed to four fire temperatures for 24 hours assuming a 7 kW decay heat load generated by the fuel. This is equivalent to PWR fuel at one year out of the reactor and a burnup of 33,000 MWD/t. Thermal analyses were performed using the TRUMP(3) heat-transfer code. In order to provide a basis for comparison, conservative values as specified by the NRC(1) and IAEA(2) regulations were used for the initial conditions (i.e., such as flame emissivity and cask absorptivity). As can be seen from Figure 1, the effect of the decay heat load on the maximum fuel rod temperature was found to be minor compared to the effect of the heat load from the fire. The curves of Figure 1 indicate that the maximum fuel rod temperatures are less than 100 C above the average fire temperature \( T_{Fire} \). The same is true for decay heat loads of 3 kW and 10 kW. Although accident fire temperatures of this magnitude are extremely unlikely, maximum flame temperatures of about 1050 C(4,5,6) have been observed in some petroleum product fires. However, the rate of heat input to a spent fuel shipping cask from an actual petroleum fire is normally less than that of the hypothetical fire defined in the NRC regulations because realistic flame emissivities and cask absorptivities are less(6) than the conservatively high values specified for the hypothetical fire. The expected times necessary to reach these maximum temperatures are also noteworthy. From Figure 1, it is expected that a fire would have to persist for many hours in order for the spent fuel in the cask to achieve the flame temperature of the fire. In a real situation, that time would be available to take emergency action or the fire's fuel supply could be exhausted long before the spent fuel could achieve the fire temperature. The results shown in Figure 1 were used to plan the experiments described in the next section. These experiments were designed intentionally to fail prototypical irradiated fuel rods to derive
an estimate of failure temperatures and the associated radiological source terms.

EXPERIMENTAL PROGRAM
A series of experiments using irradiated and unirradiated fuel rods were performed to address the second objective. Single irradiated fuel rods were instrumented and exposed to thermal environments more severe than would be expected from hypothetical accident conditions specified in current regulatory requirements for certification. Only specimens with intact cladding were used in these tests. The heating rate (thermal ramp) was determined from analyses of a typical PWR shipping cask exposed to a constant radiant heat source of 1090 °C. The analyses were performed for a typical state-of-the-art truck cask having a 343 mm inside diameter (ID) and 168 mm thick lead gamma shield. Those analysed indicated that for exposure of the cask to a 1090 °C radiant source, the maximum heating rate for the contents would be about 4 - 5 °C/min. Tests were therefore designed and conducted to reproduce this temperature environment for a segment of a single irradiated fuel rod. Tests of unirradiated zircaloy cladding filled with surrogate fuel pellets were conducted to verify testing procedures and equipment performance. The unirradiated fuel rod tests also provided baseline data for cladding in the as-fabricated condition. The irradiated specimens were taken from fuel rods whose histories were well known and which had been previously examined and characterized. Both the unirradiated and irradiated specimens were approximately 305 mm long. Burst tests conducted on unirradiated specimens of different lengths indicated that this length was sufficient to eliminate end effects on the central 100 to 150 mm portion of the specimen.

Tests were conducted using both an induction furnace and a tube type resistance furnace to heat the fuel pin specimens at the prescribed rate. However, evaluation of the test results indicated that the resistance furnace provided a thermal environment which more closely simulated the uniform axial heating that would be experienced by a fuel pin (and fuel assembly) in a cask exposed to an actual fire. Therefore, results of the tests performed in the resistance type furnace only are reported in this paper. Three tests were conducted in the resistance type furnace as follows: (1) one with an unirradiated specimen at a heating rate of 4.4 °C/min, (2) one using an irradiated specimen at a heating rate of 4.4 °C/min, and (3) one using an irradiated specimen at a heating rate of 1 °C/min. The specimens for all tests were pressurized to 3.45 MPa (500 psig) with helium and then sealed. Figure 2 shows a schematic diagram of the resistance furnace and aerosol/particulate collection apparatus used for the unirradiated and irradiated tests. The particulate collection system consisted of three cascade impactors and sixteen deposition plates in a square array (four per side) surrounding the specimen over its entire length.
RESULTS

Table 1 summarizes the results of the three tests performed using the resistance furnace. The results of the heating ramp tests indicated that, for the magnitude of the initial internal pressure examined, cladding failures of the irradiated fuel rods occurred at a cladding temperature of about 743 °C (1370 °F). The results of Test 3 are considered to be more reliable than those of Test 2 because of a suspected leak in the pressure fitting of Test 2. The test data also indicated that the failure temperature is independent of the heating rate.

### TABLE 1.
SUMMARY OF PERFORMANCE RESULTS FROM TESTS USING RESISTANCE FURNACE

<table>
<thead>
<tr>
<th>PARAMETER</th>
<th>UNIRRADIATED SPECIMEN</th>
<th>IRRADIATED SPECIMEN</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Test 1&lt;sup&gt;a&lt;/sup&gt;</td>
<td>Test 2&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>Initial Pressure MPa (psig)</td>
<td>3.45 (500)</td>
<td>3.45 (500)</td>
</tr>
<tr>
<td>Maximum Pressure MPa (psig)</td>
<td>4.21 (610)</td>
<td>5.00 (725)</td>
</tr>
<tr>
<td>Burst Pressure MPa (psig)</td>
<td>1.38 (200)</td>
<td>1.72 (250)</td>
</tr>
<tr>
<td>Heating Ramp C/min (F/min)</td>
<td>4.4 (8)</td>
<td>4.4 (8)</td>
</tr>
<tr>
<td>Burst Temperature C (F)</td>
<td>860 (1580)</td>
<td>938 (1720)</td>
</tr>
</tbody>
</table>

<sup>a</sup> Heating Rate 4.4 C/min  
<sup>b</sup> Heating Rate 1. C/min

Based upon the results of these tests and the analysis shown in Figure 1, failure of the fuel rod cladding would not be expected to occur until after 4 hours exposure to the NRC regulatory hypothetical fire environment ($T_{fire} = 800$ °C). Thus, fuel pin cladding failure would not be expected to occur for cask exposures under 4 hours. Even if the highly conservative curve in Figure 1 for $T_{fire} = 1090$ °C is considered, cladding failure would not be reached until exposure times greater than 2 hours.

The tested specimens visibly swelled over the entire length in a manner considered representative of the swelling which a pin would experience in an actual thermal event. The test of the irradiated specimen heated at a rate of 4.4 C/min indicated that about 60% of the measured particulate release was deposited in the lines of the
collection system. Only about 10% of the particulates were collected by the impactor. The remaining 30% of the released material was deposited on the deposition plates. A smaller total release of aerosol and particulate mass was measured for the irradiated specimen heated at a rate of 1 C/min than that for the test with a 4.4 C/min heating rate. Gamma scans (using Eu-154 as a fuel matrix tracer) of the collection system, deposition plates and sampling lines were made to determine the amount of spent fuel collected. Alpha spectra for some of the deposition plates were also obtained to determine the amounts of Pu-238, Pu-239, Pu-240, Am-240, Am-241, and Cm-244 present. No attempt was made to track and measure the volatile fission products such as Cs-137 and Ru-106. The total mass of spent fuel particulates released from the 4.4 C/min and the 1 C/min heating rate tests was estimated to be 6051 µg and 460 µg, respectively. The smaller total release is attributed to the smaller hole size measured in the cladding of the specimen heated at 1 C/min compared to that measured for the 4.4 C/min heating rate test.

ESTIMATED RELEASE FRACTIONS
The particulate release data obtained from the two resistance furnace tests on irradiated specimens were used to estimate the release of spent fuel mass from a full PWR fuel assembly when its cask is subjected to a similar thermal environment. To ensure that the estimated full-scale release is an upper bound, two conservative assumptions were made. They are:

1. The release from each fuel rod is 6051 µg, the larger of the two measured releases, and
2. The release from a 12-foot fuel rod is linearly proportional to the release from the 1-foot long test specimen even if there is just one failure point in the full-length rod. This is an extremely conservative assumption because it is expected that the pressure-induced flow of gases and particulates through the rupture orifice is a local and transient process, and those portions of the fuel in the cladding removed from the failure point could only contribute a relatively small fraction to the total release by diffusion processes.

Assuming that all fuel pins in a single PWR fuel assembly were to fail, then an upper bound estimate of a full scale release from a single PWR fuel assembly to the cask cavity is 19.2 g of the solid fuel per assembly. Since there are approximately 465,000 g of radioactive fuel in a typical PWR assembly, an upper bound estimate of the fraction of the total solid fuel mass in an assembly available for release from the cask cavity to the environment is $4 \times 10^{-5}$ (0.004%). This is a highly conservative
FIGURE 1. CALCULATED TRANSIENT TEMPERATURE RESPONSE OF THE HOTTEST FUEL PIN IN A PWR SHIPPING CASK EXPOSED TO VARIOUS FIRE TEMPERATURES ($T_{\text{FIRE}}$) FOR 24 HOURS (DECAY HEAT = 7 kW).

FIGURE 2. SCHEMATIC OF TEMPERATURE FAILURE THRESHOLD BURST TEST APPARATUS AND PARTICULATE COLLECTION SYSTEM USING RESISTANCE FURNACE.
estimate of the fuel mass available for release because this number does not include deposition of released fuel on surrounding fuel rods, fuel rod gap, and the interior cask wall. Results from the thermal tests performed in this study indicated that approximately 90 percent of the released fuel was deposited on nearby surfaces. In a full scale event, it is very probable that a similar fraction of the released material would be deposited on cask cavity walls and fuel rod surfaces. It is emphasized that these estimates apply only to the solid fuel matrix inventory of the reactor fuel and no attempt was made to measure the release of the more volatile fission product radionuclides.

REFERENCES


