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SPENT FUEL RESISTANCE TO INTERNALLY PRODUCED CLADDING DEGRADATION

R.E. EINZIGER
D.A. CANTLEY
J.C. KROGNESS
D.E. STELLRECHT
V. PASUPATHI

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DEGRADATION

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R.E. Einziger, D.A. Cantley, and J.C. Krogness

Westinghouse Hanford Company
Richland, Washington

and

D.E. Stellrecht and V. Pasupathi

Battelle Columbus Laboratories
Columbus, Ohio

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INTRODUCTION

Current U.S. nuclear policy precludes reprocessing of spent fuel to recover unused fissile material. Consequently, unmodified spent fuel is being studied by the Office of Nuclear Waste Isolation (ONWI) as a final waste form for geologic disposal. The intact cladding and pellets of this waste form would prevent migration of potentially harmful radionuclei in a geologic repository. The cladding, however, may degrade during disposal due to exterior corrosion or in response to fuel rod internal conditions at disposal temperatures. Blackburn[1] identified and evaluated a number of potential degradation mechanisms. These include mechanical overload, stress rupture, stress corrosion cracking, rapid fracture of flawed cladding, internal hydriding and cladding oxidation. After evaluation, he concluded that stress rupture and stress corrosion cracking were the most likely mechanisms to be operative. Since both of these phenomena are thermally activated, it should be possible to preclude degradation by limiting the maximum temperature during disposal. Blackburn estimated a maximum permissible isothermal temperature for 100 year storage of 321°C. This temperature, however, is dependent upon the validity of his underlying assumptions which involve the failure mechanism, the cladding material properties and the cladding physical condition.

Obviously, direct verification of this temperature limit at expected repository operating conditions cannot be made in a laboratory time frame, but requires that either Blackburn's assumptions be validated or that they be shown to be conservative. To accomplish this purpose, a testing program was established to identify and evaluate internally induced fuel rod degradation mechanisms. Since all the apparent degradation mechanisms are temperature dependent, initial verification tests were conducted at elevated temperatures to obtain results in a short period of time. The primary objectives of these tests were to determine potential mechanisms leading to cladding breach and to improve and validate the basis for the maximum allowable cladding temperature for safe geologic disposal. This paper discusses the results of the first series of tests conducted for this purpose.

TEST PROCEDURES

Both tests contain six unmodified Westinghouse designed prepressurized PWR rods from Turkey Point Unit 3. Whole, intact fuel rods were used to ensure that the performance and breach mode obtained were real consequences of rod properties and internal conditions and not the consequence of artifacts resulting from sample preparation. Pertinent characteristics of the rods are summarized in Table 1. Each rod was nondestructively examined before the test to determine length, diameter, surface condition, axial fission product distribution, and any internal or external cracks that were detectable by the eddy current technique. In addition, companion rods from the same fuel assembly were destructively examined to determine rod internal pressure, fission gas content, exterior surface oxidation and fuel cladding corrosion.

The details of the test apparatus[2] are schematically shown in Figure 1. Each furnace is housed within a steel-lined lead-filled shield to provide the proper personnel biological protection. It has seven independently controlled heat zones. The axial temperature distribution is measured with chromel-alumel thermocouples. Each furnace contains a test rig capable of holding six individually encapsulated fuel rods. The present tests each contain three rods encapsulated in helium and three rods encapsulated in air. The capsule pressure, initially near 0.14 MPa at operating temperature was continuously monitored with a pressure transducer to detect rod breach.

The rods were initially brought to temperature over a 240-hour period during the summer of 1979 and then ran continuously except for minor shutdowns due to power outages prior to the interim examination recently conducted. The intent of these tests was to run until breach occurred, and then to conduct examinations to verify the breach mechanism. Analyses using unirradiated stress rupture properties,[1] the MATPRO creep equation,[3] and estimates of rod internal pressures based on examination of companion rods were made to estimate breach times. Results of these analyses are shown in Table 2. For the test at 510°C and 571°C, the time of expected first breach was considerably exceeded. with no breaches. To investigate why breach time hadn't been predicted, an interim examination was performed on the test at 482°C prior to the time of expected first breach.

RESULTS AND DISCUSSION

After shutdown of the test at 482°C, the cover gas surrounding the fuel rods in the capsules was analyzed for xenon and krypton to see if there had been any small undetected leaks. Two of the rods were then removed, visually inspected, profilometered, eddy current and gamma scanned and punctured for fission gas analysis.

No outstanding differences were noticed between the pre-test and post-test visual examinations. This was expected since the visual examination would only reveal very gross changes in the rod condition. The eddy current examination showed no indication of any crack formation during the test, but the limit of detection of the eddy current technique for an inner wall crack is about 15% of the wall thickness and tends to be somewhat qualitative. There was no detectable change in the gamma activity profile which indicates that there was no axial transport of ^{137}Cs or other fission products. There were no detectable length changes of the rods during the test indicating a 2:1 biaxial stress state which was consistent with the calculational assumptions.

Comparison of pre- and post-test profilometry revealed between 1-1/2 and 2% cladding creep. In addition to the creep, the cladding showed a significant reduction in ovality and ridging. Rod puncturing, gas analysis and internal void volume determinations revealed an increased final internal void volume as would be expected due to the significant cladding creep, but the total amount of gas in the rods and the volume of fission gas in the rods remained the same as before the test started. Except for an increase in the water content of the internal gas, the composition of the gas remained approximately constant indicating no apparent fission gas release during the test. These results also show that the cladding stress dropped 31% during the course of the test.

The single most significant observation is the unexpectedly high cladding creep. This was a factor of 6 higher than predicted by the MATPRO creep equation[3] used in the original analysis and largely explains the lack of breach as predicted by calculations. The difference between the measured creep strain and the creep strain predicted by the MATPRO equation results either because the MATPRO equation is not suitable for the test conditions or the hoop stress in the rods was higher than calculated. This calculated stress was determined using nominal diameters and wall thicknesses and an internal pressure of approximately 6.9 MPa. The actual stress could be higher if the actual cladding diameter was larger, the wall was thinner or the pressure higher than used in the calculation. In actuality, when all these variations are assessed, the calculations were done at approximately 92% of the actual stress which implies that the problem lies with the creep equation.

Figure 2 is a plot of expected time to breach based on the stress rupture mechanism as a function of constant operating temperatures determined by a number of different methods. Blackburn's method[1] which was used to develop the temperature limit uses a fixed room temperature internal pressure of 8 MPa and a stress rupture correlation which is 2/3 of the lower 95% confidence interval for unirradiated stress rupture properties. The design lifetimes resulting from this formulization are shown as the bottom line in this figure. Using the same formulization as

Blackburn except using the actual pressure range determined from the companion rods, one gets a range of breach times indicated by the cross hatched region. Neither of these calculations take into account the cladding creep and subsequent reduction in hoop stress that was observed. If one goes further and uses the nominal expected internal pressures, the ±95% confidence band on stress rupture properties and allows creep as calculated by the MATPRO equation, the predicted breach times are indicated by the arrowed bands. If the measured creep rate predicted by Merckx creep equation[4] is factored into the calculation of lifetimes, then breach is not expected within a reasonable laboratory time frame via a stress rupture mechanism. For all three tests, the predicted design recipe breach time using nominal pressures and indicated by the cross hatch region is exceeded. With respect to the breach time, Blackburn's isothermal formulization is conservative by factors of at least approximately 10^6 at 571°C, 10^5 at 510°C and 10^4 at 482°C as determined by comparing predictions with operating experience and recognizing that no breaches have occurred.

It is significant to note, however, that these tests were conducted at temperatures which were above the temperature where in-reactor radiation damage quickly anneals.[5] This means that the material condition of the cladding and test temperatures may be significantly different than at anticipated geologic disposal temperatures where the damage may never be completely annealed. Based on comparisons with other post irradiation mechanical properties such as ultimate and yield strength[6] which show that the strength increases and ductility decreases, one can hypothesize that compared to unirradiated material, the irradiated material should have a longer time to stress rupture since it is stronger but the breach should come at a lower strain. While this appears to be a favorable situation from the point of view of disposal, it must be confirmed by testing.

CONCLUSIONS

These tests were conducted over a narrow temperature range considerably above anticipated disposal conditions and utilized only one set of rods from a single reactor. Therefore, the extent to which these results apply to disposal conditions for the entire population of fuel rods which would be subject to disposal is uncertain. With these qualifications, the following conclusions are made relative to the results of these particular tests. The measured cladding strain was sufficiently large so that failure mechanism verification by inducing breaches in unmodified rods heated to elevated temperatures for short periods of time does not appear to be practical based on a stress rupture mechanism. At the elevated test temperatures though, Blackburn's formulization based on stress rupture gives very conservative estimates of breach times. In addition to the high cladding strain, the fuel exhibited no additional gas release or axial fission product migration at 482°C. The nondestructive examination gave no additional indication of internal deterioration of the fuel rod.

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TABLE 1

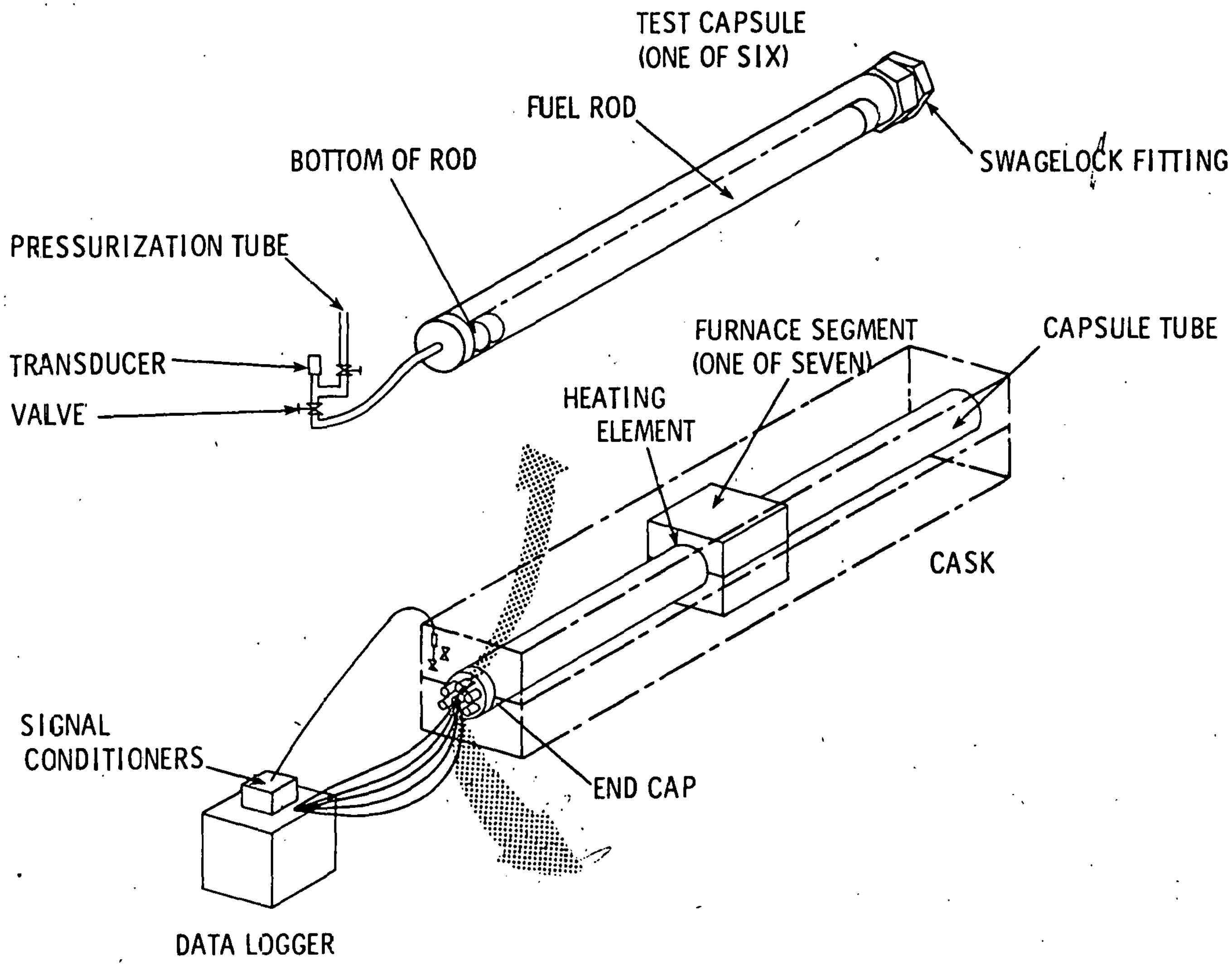
CHARACTERISTICS OF FUEL RODS USED IN THE WHOLE ROD TESTS

- Westinghouse Prepressurized PWR Rods from Turkey Point Unit 3
- 2 Cycles near core center with ∿28,000 MWd/MTu BU
- 3-2/3 Years in H_2O Pool Prior to Testing
- Cold Worked Zircaloy-4 Cladding with 10.7 mm OD and 0.62 mm Wall
- 2.6% Enriched UO_2

TABLE 2

OPERATING CONDITIONS, EXPECTED BREACH TIMES,
AND
STATUS OF THE HIGH TEMPERATURE TESTS

TEST	TEMPERATURE (°C)	EXPECTED 1ST BREACH (HRS)	HOURS OPERATED	STATUS
2a	482	1×10^4	4.6×10^3	Stopped
1	510	2.4×10^3	7.8×10^3	Continuing
2b	571	20	6.7×10^2	Stopped



TEST CAPSULE
(ONE OF SIX)
FUEL ROD
BOTTOM OF ROD
SWAGELOCK FITTING
PRESSURIZATION TUBE
TRANSDUCER
VALVE
FURNACE SEGMENT
(ONE OF SEVEN)
CAPSULE TUBE
HEATING
ELEMENT
CASK
SIGNAL
CONDITIONERS
END CAP
DATA LOGGER

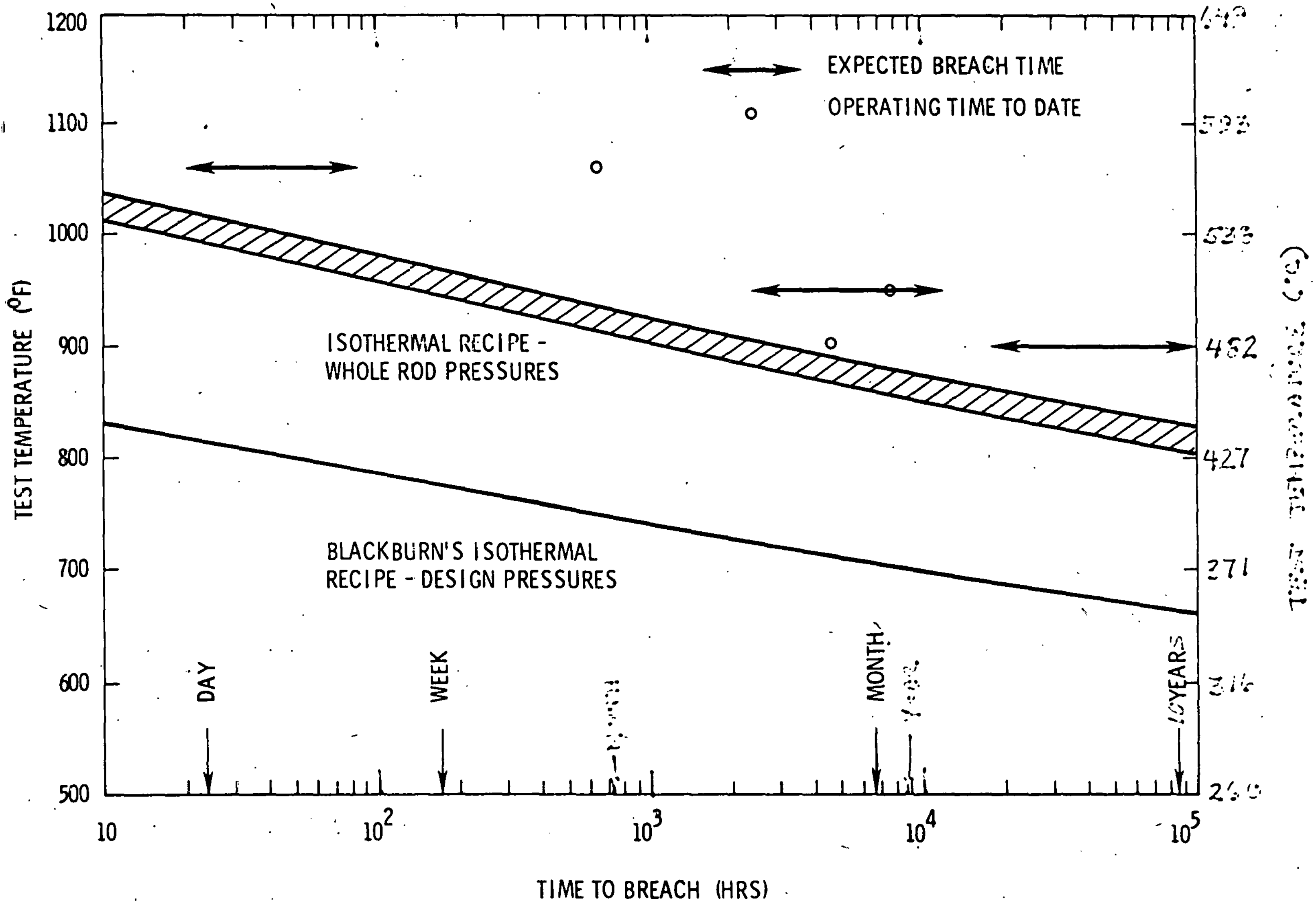


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FIGURE CAPTIONS

FIGURE 1 - High Temperature Whole Rod Tests Apparatus Detailing the Furnace Construction and Rod Encapsulation.

FIGURE 2 - Fuel Rod Predicted Time to Breach as a Function of Operating Temperature Based on the Stress Rupture Mechanism Determined by a Number of Different Formulations.