EXAMINATION OF PWR SPENT FUEL RODS AFTER 15 YEARS IN DRY STORAGE

BACKGROUND

The original license and the certificate of compliance (CoC) for spent nuclear fuel dry storage casks are due to expire for many commercial nuclear power plants in the next few years. The Spent Fuel Project Office (SFPO) of the United States Nuclear Regulatory Commission (NRC) is developing revisions to interim staff guidance (ISG) for review of technical documents to be submitted by licensees in support of their applications for renewal of licenses and CoCs. The renewal of licenses and CoCs for the existing casks would cover an additional period of 20 to 100 years and would require development of a technical basis for continued safe performance under the extended service conditions. Consistent with the above and the regulatory requirements of 10 CFR Part 72 for storage and handling of spent fuel, the acceptance criteria for spent fuel in storage casks in ISG-11 are being revised. Revision 2 of ISG-11 (Ref. 1) recognizes creep to be the dominant mechanism for cladding deformation under normal conditions of storage and recommends that the maximum cladding temperature during storage and for short-term vacuum drying and cask backfilling operations be limited to 400°C to ensure that the extended storage does not result in gross rupture of fuel cladding. The research program reported herein provides a technical basis for the revision.

OVERVIEW OF THE PROGRAM

The research program, initiated by the NRC Office of Nuclear Regulatory Research (RES) in November 2000 in response to the user need letter from the SFPO (NMSS-1998-005), involved post-storage characterization and creep testing of representative pressurized water reactor (PWR) fuel rods, stored for over 15 years in a single assembly (T11) in a CASTOR V/21 dry storage cask. These rods came from the Surry nuclear power plant, had an initial enrichment of 3.11%, and were irradiated for three fuel cycles to an average burnup of 35.7 GWd/MTU and a fast (E>1 MeV) neutron fluence of 6.38×10^{25} n/m^2. The fuel cladding was made from cold-worked/stress-relieved Zircaloy-4. The rods had cooled for ~1300 days before they were stored for ~15 years, during which the peak cladding temperature decreased from 350°C to 150°C.

The post-storage characterization consisted of: (1) visual inspection of rods for crud, rod bow, rod bulge, spallation, cracking, or other deleterious rod behavior, (2) profilometry of rods to determine diametral changes as a result of creep deformation during storage, (3) fission gas analysis and void volume determination of rods to estimate rod internal pressure and isotopic composition of fission gas, (4) metallography of rod segments to determine the fuel-cladding gap and the extent of cladding corrosion (oxide thickness), (5) hydrogen analysis of rod segments to determine possible deleterious effects of hydrides on cladding integrity, and (6) microhardness measurements to estimate cladding resistance to deformation. Table 1 below summarizes the post-storage characterization matrix.

Visual inspection and profilometry were performed on 12 rods from a single assembly that had the highest assembly-average burnup and was stored in the hottest part of the cask. The profilometry data exhibited very similar cladding diametral profiles for all 12 rods. The differences found in the diametral profiles were insignificant relative to experimental uncertainty. Four rods with marginally higher diametral changes were selected for fission gas analysis.

Measured gas pressures in all four rods were too close to allow further discrimination for the remaining characterization work. Thus, two rods with the largest diameters and highest gas pressures were selected for segmentation and further analysis that included metallography, hydrogen analysis, and microhardness measurements. In selecting specific locations for rod segmentation, the factors considered included (a) uniformity of burnup and fast fluence, (b) proximity to the axial regions from which the creep samples were to be prepared, (c) significant clad oxidation, and (d) highest temperatures experienced during thermal performance testing and storage.

Table 1. Spent Fuel Post-Storage Characterization Matrix

Characterization Type	Number of Rods	Rod Segment Identification
Visual inspection	12 rods	Full rod
Profilometry	12 rods	Full rod
Fission gas analysis	Rods H7, H9, G6, G9	Full rod
Metallography and oxide thickness	Rod H9	3 rod segments (at fuel centerline, 500 mm above, and 1000 mm above centerline)
	Rod G6	2 rod segments (at fuel centerline, 500 mm above centerline)
Hydrogen analysis	Rod H9	3 rod segments (at fuel centerline, 500 mm above, and 1000 mm above centerline)
Hydride orientation	Rod H9	3 rod segments (at fuel centerline, 500 mm above, and 1000 mm above centerline)
	Rod G6	2 rod segments (at fuel centerline, 500 mm above centerline)
Microhardness	Rod H9	3 rod segments (at fuel centerline, 500 mm above, and 1000 mm above centerline)

Aside from the extensive post-storage characterization work indicated above, the research program focused on creep testing of fuel rods that had been in dry storage for ~15 years. The purpose of creep testing was to determine the residual creep capacity of stored fuel rods in order to assess if the rods can be safely re-stored in a dry cask for an extended period of 20 to 100 years and safely transferred to a permanent repository. The creep data would also provide valuable information for the analysts to develop creep correlations for incorporation into the codes used for licensing review.

Five creep tests were conducted spanning a temperature range of 360°C to 400°C and a stress range of 190 MPa to 250 MPa. As mentioned previously, specimens for creep testing came from locations in close proximity to rod segments used for post-storage characterization. Table 2 summarizes the creep test matrix. The temperature range is prototypic of spent fuel rod temperatures at the beginning of dry storage. The rod temperature decreases with time

during the storage so the temperatures selected for experiments are expected to provide a conservative estimate of residual creep capacity. The stress range is not prototypic. Fuel rods in dry storage are expected to encounter much lower stresses (~60 to 70 MPa at a burnup level of 45 GWd/MTU). However, creep tests at this low stress level are not expected to yield any measurable creep deformation within the test time frame (<<10,000 hours). Thus, accelerated testing at higher stress levels was pursued, keeping in mind that the results from such tests would be translated to stress levels appropriate for dry cask storage by analytical means.

Table 2. Spent Fuel Creep Test Matrix

Test #	Temp. (°C)	Stress (MPa)	Test Time (hr.)	Test Purpose
1	380	220	2180	residual creep capacity; primary-secondary creep data
2	380	190	2348	residual creep capacity; primary-secondary creep data
3	400	190	1873	residual creep capacity; primary-secondary creep data
4	400	250	693	residual creep capacity; primary-secondary creep data
5	360	220	3305	residual creep capacity; primary-secondary creep data

RESULTS

The results of post-storage characterization and creep testing are discussed in detail in NUREG/CR-6831 (Ref. 2) and summarized below under each characterization category. Note that the visual examinations of the 12 selected rods did not show any appreciable degradation such as rod bow, spalled cladding, or cladding creep, even after 15 years of dry storage. There was evidence of crud and oxide buildup from reactor operation but no crud or oxide appeared to have fallen off. The selected rods are those that came from a 15x15 fuel assembly stored close to the hottest part of a Castor V/21 dry storage cask (Ref. 3). The assembly was irradiated in the Surry PWR for three fuel cycles and had the highest assembly average burnup.

Profilometry

Linear profilometry traces of each of the 12 rods were obtained at 0, 45, 90, and 135° relative orientations and at ~2.5 mm intervals along the length, starting at ~330 mm from the top of the rod. Averaging the four linear traces at a given axial location generated the cladding outer diameter profile at that location. An example profile for one of the 12 rods, marked H9, is shown in Figure 1. Noting that the nominal as-fabricated rod diameter is 10.71 mm, it is seen that the maximum decrease in cladding diameter of rod H9 is ~0.06 mm, which is equivalent to a creepdown of ~0.6% that is typical of PWR rods in the burnup range of 36 GWd/MTU.

Profilometry traces of other rods were very similar to that of rod H9 and showed no discernible differences.

Fission Gas Analysis

The void volume, gas composition and inventory, and fission gas isotopic composition were measured on four of the 12 rods to determine internal rod pressure and to estimate the extent of maximum possible fission gas release during the storage period. The void volumes ranged from 19.5×10^{-6} to 20.4×10^{-6} m^3, which is typical for 15x15 PWR rods. The gas composition measured in all four rods were essentially the same, i.e., 96-98% helium fill gas with small amounts of oxygen (<0.01%) and nitrogen (<0.03%). The fission gas release values ranged from 0.4 to 1.1%, well within the range reported in the literature for the rod type and burnup range, indicating that no significant amount of fission gas was released from the fuel during the dry storage period. The Xenon fraction in the fission gas was between 1.5 to 3.2 mol%, and the Krypton fraction was between 0.1 to 0.4 mol%. The Xenon to Krypton ration ranged from 9.0 to 11.4 for the four rods, again typical of values reported for PWR fuel rods.

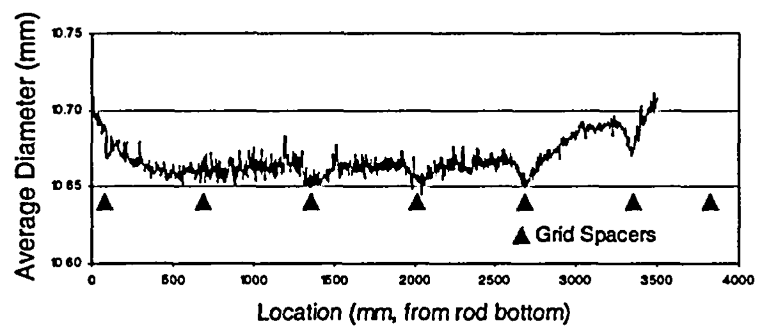


Figure 1. Average Outer Diameter Profile of Rod H9 (from Surry-2 Assembly T11) After 15 Years of Dry Cask Storage

Metallography and Oxide Thickness

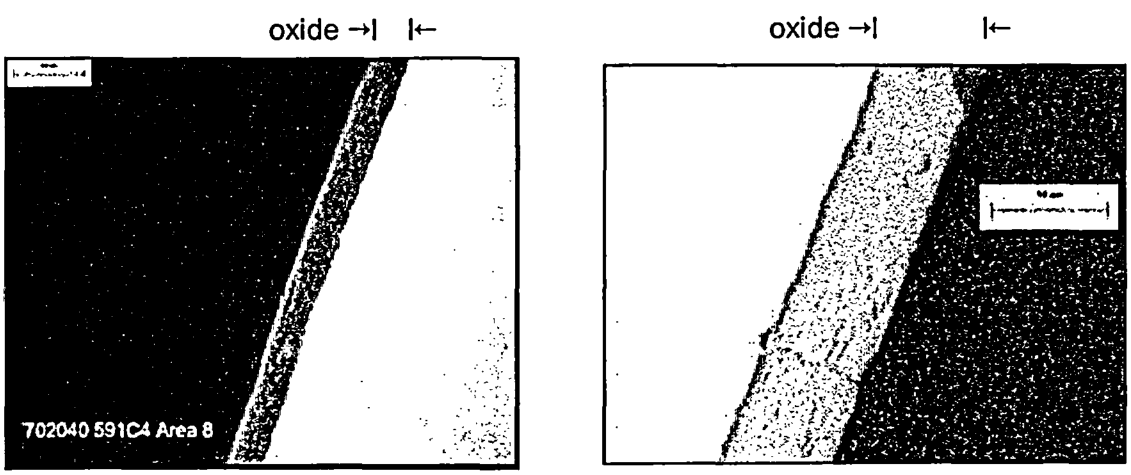
The thickness of the oxide layer on the cladding outer surface was measured from micrographs (metallographs) taken at eight azimuthal locations, spaced evenly around the cladding and covering ~45% of the cladding circumference. For rod H9, the oxide layer thickness was measured at three axial locations: fuel centerline, 500 mm above centerline, and 1000 mm above centerline. For another rod G6, the thickness was measured at two axial locations: fuel centerline and 500 mm above centerline. The oxide thickness measurements are shown in Table 3.

Table 3.Cladding Oxide Thickness (μm) at 0, 500, and 1000 mm Above the Fuel Centerline

Rod ID	0 mm (fuel centerline)	500 mm	1000 mm
H9	24 ± 4	33 ± 8	40 ± 5
G6	22 ± 6	26 ± 3	not measured
TP-D[a] range	4 - 7	5 - 8	8 - 12

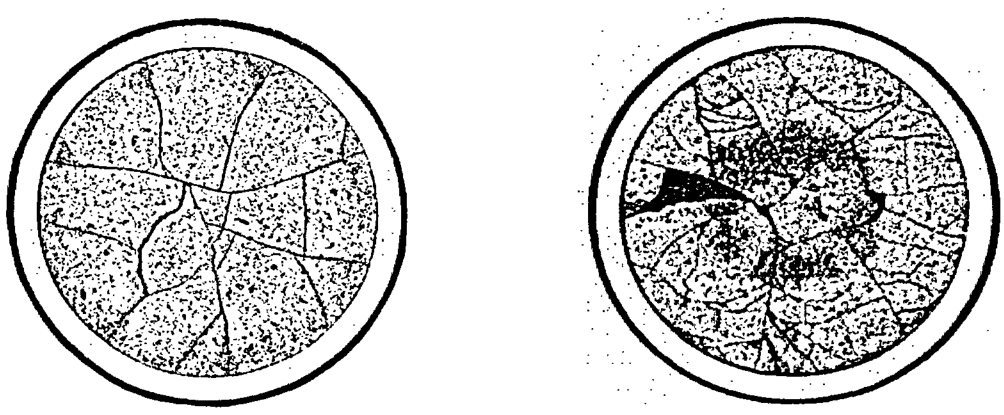
[a]Turkey Point - D assemblies data provided for comparison. The fuel in D assemblies had lower burnup than the Surry fuel and hence, smaller oxide thickness.

It is seen from the table that at higher elevations, the oxide layer is thicker because of the higher in-reactor temperatures at these elevations. Figure 2 shows the micrographs of oxide layer at the fuel centerline and 1000 mm above the centerline at one azimuthal location in rod H9. The oxide layer at the centerline is thin and adherent whereas the oxide layer at 1000 mm above the centerline is thicker and has isolated cracks and/or microvoids. Again, this is consistent with oxide growth morphology whereby thicker oxide layers are more prone to cracking. Figure 3 shows the cross-sectional views of rod H9 at the fuel centerline and 1000 mm above the centerline, depicting a fuel pellet with multiple cracks and voids that are prototypic of this type of medium burnup PWR fuel.



(A) (B)

Figure 2. Cladding Oxide Layer of Surry-2 Rod H9 from Assembly T11 at:
(A) Fuel Centerline, and (B) 1000 mm Above Centerline



(A) (B)

Figure 3. Cross-Sectional Views of Surry-2 Rod H9 from Assembly T11 at:
(A) Fuel Centerline, and (B) 1000 mm Above Centerline

Hydrogen Analysis and Hydride Orientation

The hydrogen content of samples from eight azimuthal locations at each axial elevation (fuel centerline, 500 mm above, and 1000 mm above) was measured using a Leco analyzer. The hydrogen content was also calculated from the oxide thickness measurements. Both measured and calculated values are shown in Table 4 for rods H9 and G6. Note that the calculated values are based on a pickup factor (fraction of available hydrogen migrating into cladding) in the range between 16 and 24%, typical of medium burnup PWR fuel rods, and assuming no axial migration within the cladding.

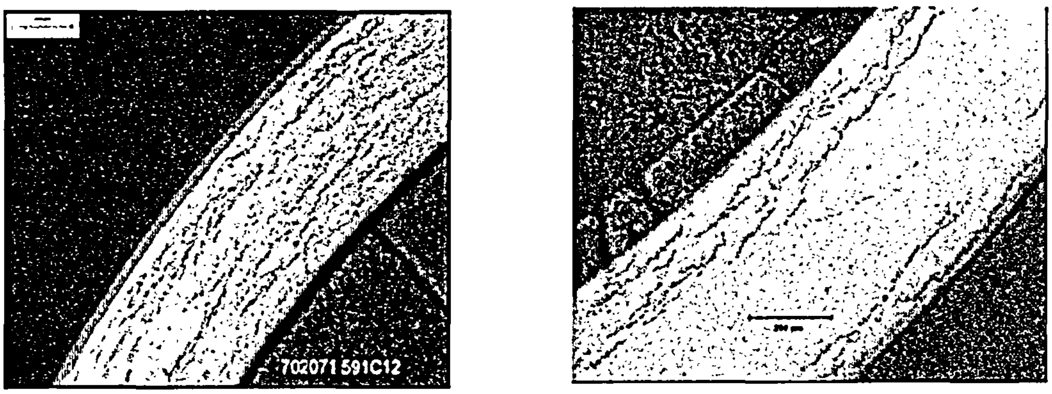
Table 4. Hydrogen Content (wppm) at 0, 500, and 1000 mm Above the Fuel Centerline

Rod ID	0 mm (fuel centerline)		500 mm		1000 mm	
	Measured	Calculated	Measured	Calculated	Measured	Calculated
H9	250 ± 40	200 ± 70	300 ± 25	270 ± 100	255± 14	330 ± 100
G6	not measured	180 ± 70	not measured	210 ± 60	not measured	not calculated
TP-D01/04[a]	40-60	40 -85	50-70	40 - 100	75-90	65 – 150

[a]Turkey Point - D assemblies data provided for comparison. The fuel in D assemblies had lower burnup and the cladding had smaller oxide growth than the Surry fuel and, hence, lower hydrogen content.

A hydrogen content of 250 to 300 wppm in cladding is typical of medium burnup fuel. Almost all of the hydrogen (200 to 250 wppm) stays in solid solution in the zircaloy matrix at temperatures 400°C (pre-storage temperature) and above. As the cladding temperature decreases during the storage period, the hydrogen in solid solution eventually precipitates as hydrides. At about 150°C corresponding to the end-of-storage temperature of fuel rods after 15 years in dry storage, essentially all of the hydrogen would have precipitated as hydrides. Figure 4 shows hydride

structure in rods H9 and G6, at 500 mm above the fuel centerline and at the fuel centerline, respectively.



(A) (B)

Figure 4. Hydride Structure in Surry-2 Fuel Rod Cladding:
(A) 500 mm Above Fuel Centerline of Rod H9; (B) Fuel Centerline of Rod G6

As seen in the above figure, hydrides are oriented circumferentially and though very few smaller hydrides in some samples may be oriented at angles between 0 and 90°, no radial hydrides were observed at the cladding/oxide interface. Hydrides, which result from clad oxidation during in-reactor operation, contribute to a loss of clad ductility if oriented radially in large quantities in cladding.

Microhardness Measurements

Cladding microhardness was measured at three axial locations of rod H9 and at four azimuthal points (~90° apart) per each axial location. The hardness numbers at three locations were calculated to be 234 ± 18, 239 ± 5, and 236 ± 10, respectively. The microhardness is a measure of cladding resistance to permanent or plastic deformation. During reactor operation, the cladding undergoes radiation hardening. When out of the reactor, the same cladding experiences thermal excursions at 400°C and above during pre-storage operation. Most of the irradiation damage (hardening) is annealed out if the cladding is exposed to such temperatures for a reasonable length of time. At temperatures ~350°C and below, hardly any annealing takes place. Little, if any, annealing occurred during the pre-storage operation or during storage of the Surry cladding under consideration. This is supported by the uniformity of the calculated hardness values as above at three different locations and also by a comparison of the Surry hardness data with the Turkey Point hardness data.

Post-Storage Thermal Creep Tests

As mentioned previously, five creep tests were conducted under conditions summarized in Table 2 above. In these tests, changes in cladding outer diameter were measured periodically at several locations (9 axial locations and 5 azimuthal points at each axial location) in a 76-mm long creep test specimen as a function of time. The hoop strain was determined from these measurements as a function of time (see, for example, Figure 5 for Test #1).

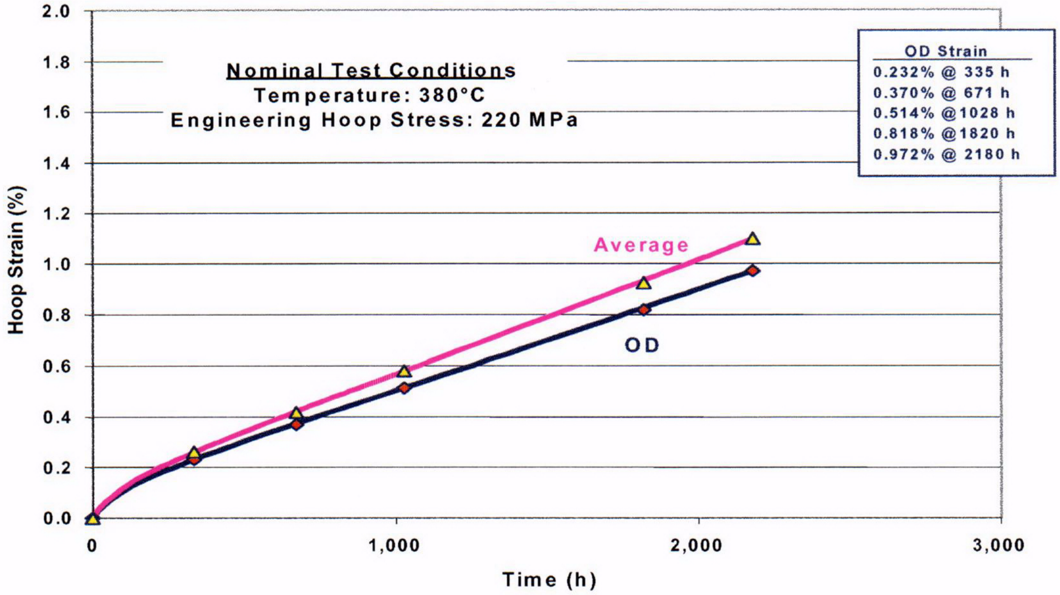


Figure 5. Thermal Creep Data (Test #1) for Irradiated, Dry-Cask-Stored Surry Cladding
Diamond: hoop strain from measurements; Triangle: wall-averaged strain

Figure 6 shows the hoop strain data for tests #2 and 3 combined. Note that Test #3 was a continuation of Test #2 beyond 1873 hours when the stress level was increased from 190 MPa to 250 MPa. The hoop strain increased significantly at the elevated stress level and at the conclusion of Test #3, the total creep strain was ~6%. The creep sample appeared to be still within the secondary (steady-state) creep regime and no creep failure was observed. Table 5 summarizes the creep data from all 5 tests.

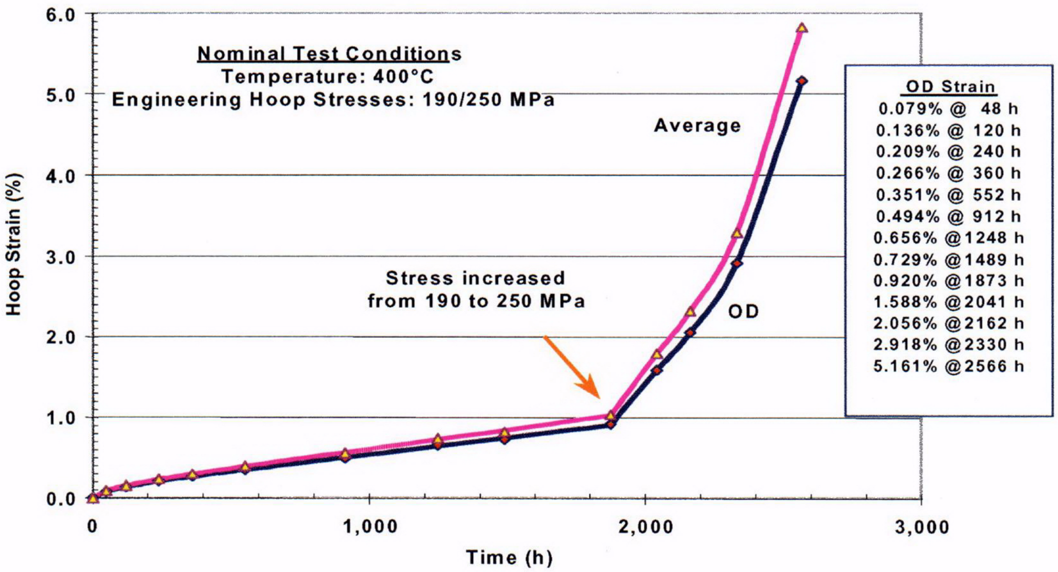


Figure 6. Thermal Creep Data (Tests #2 & 3) for Irradiated, Dry-Cask-Stored Surry Cladding
Diamond: hoop strain from measurements; Triangle: wall-averaged strain

C01

Table 5. Hoop Strain and Secondary Creep Rate Data from Five Creep Tests

Test #	Temp. (°C)	Stress (MPa)	Test Time (hr.)	Hoop Strain (%)	Secondary Creep Rate (%/hr.)
1	380	220	2180	1.10	4.5×10^{-4}
2	380	190	2348	0.35	8.8×10^{-5}
3	400	190	1873	1.04	4.9×10^{-4}
4	400	250	693	5.83	4.9×10^{-3}
5	360	220	3305	0.22	4.2×10^{-5}

Inasmuch as no bulging or asymmetric wall thinning, a precursor to creep failure, was observed in any of the creep specimens, the results indicate that significant residual creep capacity remains in fuel cladding after the initial storage period of 15 years. With decreasing temperature and stress during the extended storage beyond the initial 15 years, the cladding is not likely to experience a significant amount of additional creep. Noting that two of the five tests were conducted at 400°C, the results support the recommendation of 400°C as the maximum allowable beginning-of-storage cladding temperature for dry cask storage of spent fuel.

REGULATORY IMPLICATIONS

The research results reported herein suggest that the PWR spent fuel cladding after 15 years in a dry storage environment retained sufficient structural integrity to remain as a barrier to fission product release. There was no evidence of hydrogen pickup or hydride reorientation during the storage period. Little, if any, cladding annealing occurred during the pre-storage operation and 15-year dry storage period. Post-storage creep data indicate significant creep capacity remaining in the cladding, thus suggesting that very little thermal creep occurred during the storage period. These observations suggest the viability of spent fuel cladding to remain as a barrier to fission product release during extended storage up to 100 years in a dry cask environment. Further, these suggest the likelihood of spent fuel cladding to retain sufficient structural integrity after the extended storage period for safe transfer to a final repository. The results provide a sound technical basis in support of Revision 2 of ISG-11, which recommends that the beginning-of-storage cladding temperature be limited to 400°C. The results also provide a sound technical basis for granting an extension of cask licenses and CoCs, thereby reducing regulatory burden on licensees while maintaining the safety of spent fuel storage. Note that the results are valid for spent fuel with a burnup limit of 45 GWd/MTU. RES is currently conducting a similar experimental program for high burnup spent fuel cladding (burnup in excess of 45 GWd/MTU). The results from the high burnup program will be reported separately in the future.

REFERENCES

1. "Interim Staff Guidance 11 - Revision 2," Spent Fuel Project Office, Office of Nuclear Materials Safety and Safeguard, July 30, 2002.

2. R.E. Einziger, H. Tsai, M.C. Billone, and B.A. Hilton, "Examination of Spent PWR Fuel After 15 Years in Dry Storage," NUREG/CR-6831, USNRC (Prepared by Argonne National Laboratory), September 2003.

3. W.C. Bare and L.D. Torgerson, "Dry Cask Storage Characterization Project - Phase 1: CASTOR V/21 Cask Opening and Examination," NUREG/CR-6745, USNRC (Prepared by Idaho National Engineering and Environmental Laboratory), September 2001.