

Extending Dry Storage Of Spent LWR Fuel For Up To 100 Years*

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EXTENDING DRY STORAGE OF SPENT LWR FUEL FOR 100 YEARS¹

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Abstract

Because of delays in closing the back end of the fuel cycle in the U.S., there is a need to extend dry inert storage of spent fuel beyond its originally anticipated 20-year duration. Many of the methodologies developed to support initial licensing for 20-year storage should be able to support the longer storage periods envisioned. This paper evaluates the applicability of existing information and methodologies to support dry storage up to 100 years. The thrust of the analysis is the potential behavior of the spent fuel.

In the USA, the criteria for dry storage of LWR spent fuel are delineated in 10 CFR 72 [1]. The criteria fall into four general categories: maintain subcriticality, prevent the release of radioactive material above acceptable limits, ensure that radiation rates and doses do not exceed acceptable levels, and maintain retrievability of the stored radioactive material. These criteria need to be considered for normal, off-normal, and postulated accident conditions. The initial safety analysis report submitted for licensing evaluated the fuel's ability to meet the requirements for 20 years. It is not the intent to repeat these calculations, but to look at expected behavior over the additional 80 years, during which the temperatures and radiation fields are lower.

During the first 20 years, the properties of the components may change because of elevated temperatures, presence of moisture, effects of radiation, etc. During normal storage in an inert atmosphere, there is potential for the cladding mechanical properties to change due to annealing or interaction with cask materials. The emissivity of the cladding could also change due to storage conditions. If there is air leakage into the cask, additional degradation could occur through oxidation in breached rods, which could lead to additional fission gas release and enlargement of cladding breaches. Air in-leakage could also affect cover gas conductivity, cladding oxidation, emissivity changes, and excessive creep and mechanical property changes. Postulated accident scenarios would be the same for 20-year or 100-year storage, because they are mostly governed by operational or outside events, and not by the cask or fuel. Analyses of accident scenarios during extended dry storage could be impacted by fuel and cask changes that would result from the extended period of storage.

Overall, the results of this work indicate that, based on fuel behavior, spent fuel at burnups below ~45 GWd/MTU can be dry stored for 100 years. Long-term storage of higher burnup fuel or fuels with newer cladding will require the determination of temperature limits based on evaluation of stress-driven degradation mechanisms of the cladding.

¹ Work conducted at the Pacific Northwest National Laboratory under contract with EPRI, Palo Alto, CA. The results of this work are reported in *Data Needs for Long-Term Dry Storage of LWR Fuel*, EPRI TR-108757 (August 1998)

1. EXPERIENCE BASE FROM EXISTING STORAGE WHERE APPLICABLE TO LIFE EXTENSION

Several countries are storing UO_2 spent fuel. Detailed performance data are generally not available from these programs, but no adverse or unexpected performance has been reported. The two major foreign dry storage programs are in Canada and Germany.

CANDU fuel has been dry stored in over 200 concrete canisters using an air atmosphere starting in 1985 (see Refs [2] and [3]). No adverse or unexpected performance has been reported. Both intact and intentional defected spent CANDU fuel have been stored for the last 18 years in a saturated air or helium atmosphere at 150°C . No degradation or additional failure of the intact rods has been detected. The water in the casks became very acidic due to radiolysis of the air and water [4]. Germany has been routinely putting LWR fuel in dry inert storage since 1992 [5] with no problem [6]. No surveillance data are available about the condition of the fuel in these casks. In addition, Germany has been conducting full-scale dry-storage cask demonstrations with irradiated PWR and BWR fuel since 1982. Tests lasted up to two years, at which time cladding creep stopped due to pressure, temperature, and decay heat drops in the fuel rod. The United States has over a dozen of licensed dry-storage cask systems [7]. The longest operating dry storage has been in 29 casks at the Surry plant and eight at the HB Robinson plant. Cask loading at these sites was started in late 1985. Monitoring of the operational casks has not indicated any storage operational problems such as leaking casks.

Since 1984, a monitored dry-storage demonstration program has been conducted by the U.S. Department of Energy (DOE) at the Idaho National Environmental and Engineering Laboratory (INEEL). Krypton-85 indications were found during the three-month testing of the TN-24P cask, when it held unconsolidated fuel. It is thought that the leak in the TN-24P cask occurred when the cask was rotated. A similar finding of Krypton-85 was found in the REA 2023 cask tested by DOE with Cooper BWR fuel at Morris, Illinois. In neither case was the leaker able to be visually identified at the end of the test. The rate of the ^{85}Kr release along with the lack of visible identification indicates that the breaches were very small. While the tests tell little about the defect mechanisms and the potential for failures in long-term storage, it is important to remember that no gross breaches occurred, and fuel assemblies/canisters were able to be pulled out of the casks with no adverse effects, signs that longer-term storage is feasible.

2. NOMINAL STORAGE CONDITIONS

The maximum temperature in the cask occurs after loading and dryout. This maximum temperature is limited by licensing consideration. Thereafter, except for fluctuations due to the external temperature, the temperature in the cask decreases because of the drop in decay heat. For this analysis, the decay curve reported in [8] is used. During a 100-year storage period, the fuel will experience two temperature regimes. The first occurs during the first 10 years when the fuel temperature drops from a maximum of about 380°C , depending on the fuel, to approximately 100°C . The second regime is for the remaining storage time when the temperature is decreasing slightly; it can generally be given a constant upper bound of approximately 100°C .

During dry storage, the spent fuel will be subjected to gamma fields of $\sim 10^5$ R/h and a neutron flux (>1 MeV) of $\sim 10^4$ to 10^6 n/cm²-s [9]. The total dose during 100-year dry storage is anywhere from four (gamma) to seven (neutron) orders of magnitude less than for two- to four-year residence in the reactor. The gamma field decreases with the age of the fuel and increases proportionately to the burnup. At the maximum expected burnup for LWR fuel (60 GWd/MTU), these dose rates would no more than double those calculated for current cask.

3. ANALYSIS OF 100-YEAR FUEL ASSEMBLY BEHAVIOR

There are potentially four different atmospheres in the cask: (1) inert fill atmosphere with some residual water depending on level of drying achieved (this is the most likely since experience to date has shown that in-leakage is highly unlikely [normal condition]); (2) inert fill plus a very small ingress of air due to a small leak in the cask (off-normal); (3) a mistaken air backfill (off-normal); and (4) an unlimited air atmosphere due to a large breach in the cask (off-normal, accident). In the first case where the inert atmosphere stays intact, no oxidative corrosion will take place. Malinauskus [10] has shown that for a small leak in a cask, the supply of oxygen will limit the amount of fuel or cladding oxidation. Although operational history indicates that a large cask leak early in storage is highly unlikely, the potential for degradation of the fuel or cladding in the remaining two cases (mistaken air backfill or large leak) can be evaluated for their potential ramifications.

The only identified potential fuel problem is oxidation of the fuel and consequences of this oxidation, such as fission gas release if the cask leaks during off-normal conditions. As long as the fuel cladding stays intact or the cask does not leak, there is little chance of detrimental fuel oxidation under normal conditions in a well-dried storage cask. Should a rod with a breach be put in storage, there is the potential for fuel oxidation should the cask leak.

With regard to cladding performance, the following aspects have been considered: (1) the maximum additional cladding oxidation that can occur during the additional 80 years of residence in the dry storage canister and its ramifications on emissivity and breach; and (2) the potential for cladding breach due to a stress-driven mechanism such as creep, stress rupture, etc. This includes the potential effect of any cladding annealing due to the initial higher storage temperatures or off-normal temperature excursions. In addition, changes in the mechanical properties of the structural components of the fuel assembly that would affect its ability to maintain critical configuration or impair retrievability are evaluated.

3.1. Fuel oxidation

3.1.1. Mistaken air backfill, limited air at high temperatures

Both the exposed fuel and cladding are competing for the available oxygen in a cask, each with a different rate law. The partitioning of the oxygen take-up depends on the number of rods with breaches and the surface area of the total number of rods in the cask. The oxidation of both the fuel and the cladding depends strongly on temperature, so only those parts of the rods whose temperatures are within about 20°C of the maximum temperature will participate in the oxidation.

The amount of oxidation depends on the amount of available oxygen. The total amount of oxygen from the backfilled air depends on the free volume in the cask. For the Castor type cask, this is about 6600 liters or 60 moles of oxygen [11].

Based on the rod dimensions, number of rods per assembly, and number of assemblies in the cask, the rod surface area was calculated. The oxygen uptake as a function of temperature measured by Suzuki [12] was multiplied by the surface area to determine the oxygen usage rate for cladding oxidation.

Due to the different activation energies and rates of oxidation of the fuel and the cladding, the maximum oxidation of the fuel in a limited air environment occurs if the peak cladding temperature is about 250 to 275°C. As the fuel oxidizes to U_3O_8 , it swells and splits the cladding. The rate at which the fuel is being oxidized is equal to the rate of splitting of the cladding. There appears to be a temperature-dependent incubation period before this splitting begins. The incubation time and cladding splitting rates measured by Einziger and Strain (see Refs [13] and [14]) were used. In any of the large casks containing many assemblies, the fuel oxidation is equivalent to one or less rods independent of the number of failed

rods in the cask. Oxidation takes place in two years or less and results in a decrease of internal cask pressure of about 20%.

Clearly, if a cask has a breached rod with a pinhole, which in-reactor breach statistics indicate will happen, and the cask is mistakenly backfilled with air instead of helium, there will be oxidation of the fuel pellets early in the storage history. The fuel will form grain-sized particulates, splitting the cladding in the process. The consequence of this oxidation is a split in the cladding, which now contains grain-sized oxidized fuel instead of pellets. As the fuel is moved, some of this powder will fall from the cladding into the cask and be available for dispersal. The effect of this oxidized particulate on removing the fuel from the cask is the same whether the fuel is stored for 20 or 100 years; it is contamination that will have to be addressed. There is no apparent reason for this oxidation early in storage to impair continued storage in the same cask.

3.1.2. Cask leaks after 10 years (low temperatures)

It should take fuel about 17 years to reach the U_4O_9 plateau at 150°C and about 100 years at 125°C. The fuel remains on the plateau until sufficient U_3O_8 forms to start splitting the cladding. Even for the most conservative estimator, fuel oxidation should not be a problem during dry storage after 10 years of storage.

3.1.3. Radiolysis

As much as 65g of water or water vapor might be left in a cask after vacuum drying [15]. Upon radiolysis, this water could produce 1.8 moles of oxygen. In the larger casks, this could result in ~10 to 15 g of oxidized fuel, which is probably not enough to split the cladding open. The radiolysis of nitrogen, should a leak occur in the cask, and/or moisture in the canister can lead to the creation of some very aggressive oxidants such as nitric acid and hydrogen peroxide. In stagnant systems with a prototypical radiation field, Canadian tests indicate accelerated oxidation and a significant drop in the pH of the canister water. U.S. tests have had similar effects in unirradiated fuel where 1% nitric acid was added to the system.

3.2. Cladding oxidation

In general, the rods attain a transition oxide layer while in the reactor so the post-transition linear kinetics will be operative [16].

The data below 350°C are bounded by a model developed for corrosion in water by Dalgaard [17]:

$$h \text{ (mm)} = 8.26 \times 10^2 \times \exp(-93 \text{ kJ/mol/RT}) \times t \quad (1)$$

where, in all the above relationships, T is temperature in degree Kelvin, and t is time in days.

Even though the equation was developed for water corrosion of Zircaloy, for lack of an adequate database for air oxidation of Zircaloy, the Dalgaard equation was used to predict the amount of oxidation for extended storage.

3.2.1. Limited air

In a sealed cask that is potentially accidentally backfilled with air, the thickness of any additionally oxidized zircaloy is determined by the volume of air present. A maximum additional 6% of the cladding thickness would oxidize and would not be detrimental to performance. The oxidation would be slightly less if there were breached rods in the cask also using oxygen. Of course, the rate of oxidation

may be too low to use all the oxygen and achieve these oxide thicknesses. At 200°C, the time required would be about 200 years

3.2.2. Large cask leaks (unlimited air)

During the first 10 years, the temperature drops from 380 to ~100°C. The amount of cladding oxidation can be calculated by integrating Dalgaard's equation. Less than 5×10^{-2} mm of the cladding is expected to be oxidized. At an expected temperature of 100°C for the last 80 years of storage, no more than an additional 7.6×10^{-5} mm of cladding is expected to oxidize. The thinnest cladding used is 0.42 mm, so the oxidation during the first 20 years is approximately 13% of the thinnest cladding, and oxidation at 100°C represents less than 0.01% wastage.

3.2.3. Radiolysis

Radiolysis will produce 7.2 g of hydrogen, equivalent to 54 ppmw. Even assuming that the original hydrogen level as the rod came out of the reactor was ~130 ppmw, the additional hydrogen would not bring the hydrides to the level where they would cause deterioration of the mechanical properties of the cladding (see Refs [18] and [19]). Radiolysis of any remaining water in the cask after drying should not cause any problems for the extended storage of the fuel. Because there have been very few measurements of the mechanical properties of zircaloy as a function of hydrogen content, this is still an area of controversy.

3.3. Potential cladding rupture

There is a requirement to set the storage atmosphere such that gross degradation of the fuel rod is prevented. The NRC does not define gross degradation, but they have shown concern for the ability to recover the fuel from dry storage for further movement or disposal without handling problems or gross contamination. Subsequently, the NRC determined that the most probable mechanism of cladding failure was diffusion-controlled cavity growth (DCCG) in spite of the fact that voids, necessary for void growth, have never been observed in Zircaloy.

3.3.1. Diffusion-controlled cavity growth

The mechanism was first proposed and analytically developed [20] to determine the time-to-failure. It was reformulated [21] so that for any given temperature profile and duration, the percent of decohesion could be calculated iteratively. In addition, Schwartz and Witte arbitrarily set the acceptable amount of decohesion at 15%. A number of properties of the fuel rod, such as the grain boundary thickness of the cladding, stress on the cladding, and grain boundary diffusion rate, which might change with prolonged service, are required for the calculation.

The maximum temperature limit was set, so at 20 years the maximum decohesion would be 15%. The integral equation can be rewritten so that the limits on time run from 20 to 100 years, and the limits on decohesion run from 0.15 to the new calculated maximum. Since the temperature changes very little after 20 years, a constant storage temperature of 100°C was chosen. Even though there are 80 additional years of storage time, the grain boundary diffusion rate at 100°C is so low that the additional decohesion is <0.00001 .

3.3.2. Strain limited rupture

Cladding breach was arbitrarily deemed by Peehs [22] to occur when a rod strain of 1% had been reached. Based on stress due to internal rod pressure, the creep strain was calculated parametrically with the initial maximum fuel rod temperature for 40 years of storage. Due to the cladding strain and a decreasing temperature, the cladding stress decreases to a level where, by 40 years, the additional strain

with time is infinitesimal. Hence, based on the creep-rupture mechanism, if the temperature is set to allow 40-year storage, additional storage time without cladding breach would also be permissible. A major uncertainty in this approach is the choice of the appropriate constitutive equation for the creep and thus the actual anticipated creep strain, which can range over an order of magnitude and may or may not cease after sufficient internal pressure drop has occurred [23]. The creep equations need to be resolved properly before the Peehs methodology can be used with confidence for long times. The 1% strain limit also seems excessively conservative. Numerous tests have resulted in spent fuel cladding strain to over 2% without breach (see Refs [24] and [25]). These strain limits were measured at temperatures above 350°C and would have to be confirmed for cooler storage. The Peehs calculations for maximum storage temperature would need to be redone for high-burnup fuel where the temperature profile would be expected to be much flatter with time.

Another concern for long-term storage is the release of helium from decay products in the fuel pellet to the plenum, thus increasing the internal rod pressure. The additional pressure was calculated using the production and release models presented by Cunningham et al. [8]. After 100 years for a rod with 72 GWd/MTU burnup, assuming a linear production rate with burnup, the pressure would increase <1 psi, which is negligible.

3.3.3. Cladding annealing effects

The properties of zircaloy cladding are affected by irradiation. However, the radiation fields to which the cladding is exposed during storage are many orders of magnitude lower than those already encountered in-reactor and do not change the effects that take place in the reactor, which usually saturate during the first or second irradiation cycle. For this reason, changes of yield strength, elastic modulus, and fracture toughness due to the radiation field from the fuel are expected to be small during the storage period (see Refs [26] and [27]). Although the strength of the cladding may be increased by irradiation, the temperature may be high enough during the initial part of the storage period to cause some annealing to occur. The result of annealing is to decrease the strength and increase the ductility of the material. It may also assist in relaxing internal stresses (see Refs [26] and [27]). The temperature for 50% recovery of irradiation damage in Zircaloy-2 was measured [28] and determined to be 380°C. Extrapolation of Kemper's [29] data indicated an approximate 40 to 50% recovery in six months at temperatures between 325 and 350°C. Below 200°C, there is virtually no annealing effect on the mechanical properties (see Refs [28] and [30]).

3.4. Transfer to a new cask after 20-year storage

3.4.1 Dry-to-Dry Transition

During reactor operation, a coating of crud developed from corrosion of the materials in the primary system forms on the fuel rod. The main constituent is ^{60}Co . The crud tends to be very tenacious on PWR rods and much more flaking and easily dislodged on BWR rods. A definitive compilation on the properties of crud is given in Sandoval, et al. [31].

There is the potential that some fuel assemblies will have to go through a dry-to-dry transfer in order to be stored for more than the licensed 20 to 40 years. During that transfer, there is the possibility that with the lid not yet secure, the cask could tip over and release spalled crud to the atmosphere. The extent of the release will depend on the amount of activity released, density of crud on the surface of the rods, amount of the surface that is actually covered with crud, amount of crud that spalls when the cask tips over, and crud that becomes airborne. NUREG-1536 [32] specifies the surface density for ^{60}Co on fuel rods as 140 $\mu\text{Ci}/\text{cm}^2$ for PWR rods and 600 $\mu\text{Ci}/\text{cm}^2$ for BWR rods. The maximum spot density is the maximum concentration of radionuclides found anywhere on the rod. The average crud density on a rod is at least factor of 10 less. Cobalt-60 has a half-life of 5.26 years. For fuel put in dry storage, five or more half-lives will have passed when a dry transfer is made with a consequent reduction in the ^{60}Co

activity of a factor of 30. The densities quoted by NRC are the high end points in the distribution. Any particular assembly might have a substantially lower maximum spot density. Accounting for half-life decay or the use of a realistic crud density for any particular set of assemblies to be stored does not decrease conservatism, but reduces the activity by a factor of ~ 300.

Estimates of crud spallation from rods stored at 230°C ranged from 1.6 to 4.8%. These measurements are probably reasonable for static dryout up to a temperature where rod ballooning occurs. Once ballooning over 6% starts to occur at 400°C, then additional crud will spall. In any case, the measurements are all within the 15% spallation figure for static storage given by the NRC. When a cask tips over, the rods are subjected to an impact. There are no measurements of the adherence of crud to the rods during an impact. For lack of data, the Sandoval evaluation used a spallation factor of one for accidents. Once the crud spalls, only a small fraction will be available for gaseous transport. The remainder will settle quickly on other surfaces within the cask. The degree of settling will depend on the particle size distribution of the crud. No data are available for crud that has been removed from a rod as a result of impact.

3.4.2. *Wet transfer through the pool*

Cask reflood must be managed in a manner that prevents adverse thermal shock of the fuel and adverse pressurization of the cask.

3.4.2.1. Effect of wet transfer on pinhole breaches

When a failed rod is placed back in the reactor pool, the water is at atmospheric pressure and ~50°C, while the uncooled rod temperature is between 100 and 150°C. As the rod cools, its internal pressure will drop, which might fill up to 25% of the internal rod void volume with water to equilibrate the pressures. If this does happen, steam could form, and a mixture of water and steam could be in contact with the fuel. The steam would soon subside as the rod cooled down, leaving the fuel in contact with water at ~50°C. At these temperatures, many studies [33] have shown that the leaching of spent fuel is very slow. Considering that the fuel is in the pool for transfer purposes only, the duration would be short; i.e., <30 hours, and leaching of the fuel would be negligible. Even if the fuel stays hot for a short time, Canadian studies of fuel oxidation at 150°C in a saturated humidity atmosphere show limited surface oxidation of the grain boundaries to U₄O₉, and only after many years.

3.4.2.2. Effect of transfer on intact cladding

When the cladding at 125°C is submersed in the pool water at 25°C, the thermal stress on the cladding is given by

$$\sigma = E \times \Delta T \times \alpha \quad (2)$$

where

σ is the thermal stress across the cladding,
 E is the elastic modulus equal to 14×10^6 [34],
 α is the coefficient of thermal expansion, and
 ΔT is the temperature differential across the cladding.

The largest value for the thermal expansion coefficient for either Zircaloy-2 or -4 in the range of 0 to 1000°C is $6.48 \times 10^{-6}/^\circ\text{C}$. Using the conservative assumption that one side of the cladding has completely cooled and the other side not at all, the temperature differential is 100°C. The thermal stress

is calculated to be 66 MPa. This stress is small compared with the yield stress, so no permanent deformation is expected as a result of exposure to cold water.

3.4.2.3. Effect on Crud Spallation

Although the thermal shock induced by exposure of cladding to cold water is insufficient to cause permanent deformation in the cladding, it is possible that a significant amount of crud spallation could occur. The amount of spallation depends on the thermal transfer characteristics of the crud, its mechanical properties (strength, bulk modulus, and coefficient of thermal expansion), and its adhesion to the cladding. Sufficient data are not available at this time to determine the propensity for crud spallation to occur, either for PWR fuel or BWR fuel, which will have to be separately evaluated due to the difference in their crud adhesion properties.

3.5. Accident evaluation

If conditions are set within a cask so that fuel rods do not fail under normal operating conditions, an additional 80 years of storage either within the same cask or after transfer to a new cask should not further degrade the fuel rods. The two major concerns for the fuel are oxidation due to the formation of a cask leak or further fracturing of the fuel pellet fragments into a fine powder due to the impact. The considerations for the cladding are breach due to impact, oxidation due to a large leak, crud spallation, development of incipient cladding cracks that could lead to breach at a later time, breach due to rapid cooling during a flood, and eutectic formation, melting, and breach due to rapid heating during a fire.

An accident is a short-duration event. As such, many of the models of fuel rod and fuel assembly responses developed by Rashid [10] for transportation are applicable. The assumptions made in that analysis and the basis of the assumptions appear plausible for both transportation and storage accident events. In some instances, the conditions for the evaluations such as for flood, fire, and cask tipover due to impact are directly applicable. Other calculations, such as for the puncture drop and 9-m fall, will have to be related to the actual energy impacted onto the cask during storage-related events such as a tornado, wind storm, or missile attack. The g-forces during transportation appear to be considerably greater than the seismic standard earthquake's 0.12g, and thus the results of the analysis may also be transferable. For fire, such phenomena as rod burst, cladding melting, and eutectic formation between the cladding and the assembly support structure are considered and found not to be possible. For the impact events, degradation phenomena such as rod fracture, rod rupture, and structural analysis of the assembly are considered. For the rod evaluation, the three main materials or rod inputs to the evaluations are the stress in the rod, cladding flaw state, and fracture toughness of the materials. Only fracture toughness data from Zircaloy-2 over a temperature range of 25 to 250°C was used. Calculations using data for irradiated Zircaloy-4 should be conducted.

3.6. Transportability of the fuel

After interim storage, be it 20 or 100 years, the fuel will have to be transported to a repository for final disposal. At the time the storage system was licensed for 20-year storage, the fuel needed to be suitable for transport after 20 years. As discussed, the fuel or cladding damage does not increase appreciably over the last 80 years of storage. Hence, transportation requirements can be met after either 20 or 100 years.

3.7. Burnup effects

For operational efficiency, the utilities are driving their fuel to higher burnups. The current average in the neighborhood of 35 to 45 GWd/MTU could go up to 55 to 65 GWd/MTU. This is being approached in two ways: 1) extending the burnup of current designs until reactor performance is no longer acceptable, and 2) development of newer designs and use of materials that are more corrosion resistant, have greater dimensional stability, and increased margins of performance. The impact of higher

fission gas inventory, rim-effect on oxidation, more hydrides in the cladding, and significantly larger oxide layers in higher burnup fuels on extended dry storage are being evaluated. Interestingly, the breach rate of the fuel has not increased appreciably with the increased burnup [35]. The three areas where higher burnup has the potential to affect the long-term dry storage of spent fuel are stress-driven cladding-failure lifetimes, thermal performance of the rods, and radionuclide inventory of the source term.

Higher enrichments combined with longer irradiation times will result in the fuel containing a higher radioisotope inventory. This will reflect in cask licenses to accommodate increases in heat load, partial cask loadings, or longer cooldown times so the cask can handle the heat load and shielding requirements.

Many of the degradation mechanisms for the cladding are stress driven. Extra corrosion of the Zircaloy cladding results in cladding thinning and a higher cladding stress. Manufacturers have countered this trend by introducing newer types of more corrosion-resistant claddings, such as Zirlo and Zircaloy-2p.

The internal pressure in the rod can occur from increased production of the noble gases in the fuel due to the higher burnup and increased release of the gas from the fuel matrix to the plenum. The production of xenon, which is the main fission gas, is proportional to the burnup [36] and thus the amount of fission gas produced by a PWR rod at 50 GWd/MTU will result in a greater rod pressure at higher burnups even with the same gas release. In addition, there are strong indications that the gas release increases at the higher burnups. This increased release is observed in high-burnup fuels that have formed a rim of fine grains near the pellet surface. This region of the fuel tends to exhibit much higher gas release.

The mechanical properties of the Zircaloy saturate after approximately one-cycle of irradiation, so these properties should not change with the higher burnup if other changes in the cladding did not occur. Extending the burnup of standard Zircaloy cladding results in additional oxide formation. About 10 to 20% of the concurrently produced hydrogen dissolves in the cladding that precipitates predominately as circumferential hydrides upon cooling. The ultimate and yield strengths are not affected strongly by the hydrogen content until above 1500 ppm (see Refs [37] and [38]). As the hydrogen content increases, the Zircaloy becomes progressively more brittle at higher temperatures [39]. A high hydrogen content could reduce the allowable strain before a cladding breach or reduce the ability of the cladding to survive an accident impact. The mechanical properties of the new cladding types are still proprietary. The ability of these new alloys to withstand accidents and behavior under dry storage conditions has to be evaluated.

4. CONCLUSION

Overall these results indicate that, based on fuel behavior, spent fuel at burnups below ~45 GWd/MTU can be dry stored for 100 years. This length of storage will not adversely affect normal transfers and transport activities during, or at the end of, the storage period. Long-term storage of higher burnup fuel or fuels with newer cladding will require the determination of temperature limits based on evaluation of stress-driven degradation mechanisms of the cladding. Post-irradiation mechanical properties of the newer high-burnup claddings and fuels may need to be taken into account.

5. REFERENCES

- [1] U.S. NUCLEAR REGULATORY COMMISSION, Licensing Requirements for the Independent Storage of Spent Nuclear Fuel and High-Level Radioactive Waste, U.S. Code of Federal Regulations, 10 CFR 72 (1985).

- [2] PARE, F.E., JOUBERT, W.M., "Evolution of the Macstor Dry Spent Fuel Storage System," (Proc. of IAEA Symp., Vienna, 1994) Safety and Engineering Aspects of Spent Fuel Storage. IAEA, p. 247 (1995).
- [3] WASYWICH K.M., et al., "Canadian Spent Fuel Storage Experience," Safety and Engineering Aspects of Spent Fuel Storage (Proc. of IAEA Symp., Vienna 1994), IAEA, p. 41. (1995).
- [4] WASYWICH K.M., et al., "The Canadian Long-Term Experimental Used Fuel Storage Program," Nuclear Waste Management and Environmental Remediation (Proc. of Int'l. Conf., Prague, Czech Republic 1993), Vol. 1. (1993).
- [5] INTERNATIONAL ATOMIC ENERGY AGENCY, Extended Storage of Spent Fuel, IAEA-TECDOC-673, Vienna (1992).
- [6] DREISVOGT H., et al., "Dry Interim Spent Fuel Storage Casks," Safety and Engineering Aspects of Spent Fuel Storage (Proc. of IAEA Symp. Vienna, 1994), p. 355 IAEA (1995).
- [7] RADDATZ M.G., WATERS, M.D., Information Handbook on Independent Spent Fuel Storage Installations, NUREG-1571, U.S. Nuclear Regulatory Commission, Washington, D.C. (1996).
- [8] CUNNINGHAM M.E., et al., Control of Degradation of Spent LWR Fuel During Dry Storage in an Inert Atmosphere, PNL-6364, Pacific Northwest Laboratory, Richland, Washington (1987).
- [9] JOHNSON A.B., GILBERT, E.R., Technical Basis for Storage of Zircaloy-Clad Spent Fuel in Inert Gases, PNL-4835, Pacific Northwest Laboratory, Richland, Washington (1983).
- [10] SANDERS T.L., et al., A Method for Determining the Spent-Fuel Contribution to Transport Cask Containment Requirements. SAND-2406, Sandia National Laboratory, Albuquerque, New Mexico (1992).
- [11] GENERAL NUCLEAR SYSTEMS, INC., Topical Safety Analysis Report for the Castor X Cask for an Independent Spent Fuel Installation (Dry Storage), Rev. 4, Docket 72-1018 (1990), GNS.
- [12] SUZUKI, M., KAWASAKI, S., "Oxidation of Zircaloy Cladding in Air," J. Nucl. Mater., Vol. 140 (1986), pp. 32-43.
- [13] EINZIGER, R.E., STRAIN, R., "Behavior of Breached PWR Spent-Fuel Rods in an Air Atmosphere Between 250 and 360°C," Nuc. Tech, Vol. 75 (1986).
- [14] EINZIGER, R.E., 1991. "Effects of an Oxidizing Atmosphere in a Spent Fuel Packaging Facility," Nuclear Waste Packaging, Focus'91 (Proc. of Topical Meeting, Las Vegas, Nevada, 1991) 88 pp.
- [15] HOWE, L.M., THOMAS, W.R., "The Effect of Neutron Irradiation on the Tensile Properties of Zircaloy-2," J. Nucl. Mater., Vol. 2 (1960) 248-260.
- [16] HILLER, E., "Corrosion of Zirconium-Based Alloys: An Overview," Zirconium in the Nuclear Industry, American Society for Testing and Materials, ASTM STP-633, Philadelphia, Pennsylvania (1976), 211 pp.
- [17] GARZAROLLI, F., et al., Review of PWR Fuel Rod Waterside Corrosion Behavior, EPRI NP-1472, Electric Power Research Institute, Palo Alto, California (1980).
- [18] HINDLE, E.D., "Effect of Circumferentially Aligned Hydrides on the Ductility of Zircaloy-2 Tubing under Biaxial Stress," J. Inst. Metals, Vol. 95 (1967) 359 pp.

- [19] HUANG, J.H., HUANG, S.P., "Effect Of Hydrogen Content On The Mechanical Properties Of Zircaloy-4," J. Nucl. Mater., Vol. 208 (1994), 166 pp.
- [20] RAJ, R., ASHBY, M.F., "Intergranular Fracture at Elevated Temperatures," Acta Metallurgica, Vol. 23 (1975), 653 pp.
- [21] SCHWARTZ, M.C., WITTE, M.C., Spent Fuel Cladding Integrity During Dry Storage, UCID-21181, Lawrence Livermore National Laboratory, Livermore, California (1987).
- [22] PEEHS, M., et al., Spent Fuel Storage Performance in Inert Atmosphere, Third International Spent Fuel Storage Technology Symposium/Workshop. Seattle, Washington (1986) p. S-215.
- [23] ROSEN, R.S., O'CONNELL, W.J., "Creep Strains Predicted from Constitutive Equations for Zircaloy-Clad Spent Fuel Rods," Sixth International Conference on High Level Radioactive Waste Management, (Proc. Las Vegas, 1995) 621 pp.
- [24] NOVAK, J., et al., "Post-Irradiation Behavior of UO₂ Fuel I: Elements at 220-250°C in Air," Nucl. Tech., Vol. 63 (1983), 254 pp.
- [25] EINZIGER, R.E., et al., "High-Temperature Post-Irradiation Materials Performance of Spent Pressurized Water Reactor Fuel Rods Under Dry Storage Conditions," Nucl. Tech., Vol. 57 (1982), 65 pp.
- [26] SANDERS, T.L., et al., A Methodology for Estimating the Residual Contamination to the Source Term in a Spent-Fuel Transport Cask, SAND90-2407, Sandia National Laboratory, Albuquerque, New Mexico (1991a).
- [27] SANDERS, T.L., et al., Considerations Applicable to the Transportability of a Transportable Storage Cask at the End of the Storage Period, SAND88-2481, Sandia National Laboratory, Albuquerque, New Mexico (1991b).
- [28] CARPENTER, G.J.C., WATTERS, J.F., "Irradiation Damage Recovery in Some Zirconium Alloys," *Zirconium in the Nuclear Industry*. ASM STP 551, American Society for Testing and Materials (1973), 400-415.
- [29] KEMPER, R.S., ZIMMERMAN, D.L., Neutron Irradiation Effects on the Tensile Properties of Zircaloy-2, HW-52323, General Electric, Hanford Atomic Products Operation, Richland, Washington (1957).
- [30] EINZIGER, R.E., KAHLI, R. "Low-Temperature Rupture Behavior of Zircaloy-clad Pressurized Water Reactor Spent Fuel Rods Under Dry Storage Conditions," Nuc. Tech., Vol. 67 (1984), 107 pp.
- [31] SANDOVAL, R.P., et al., Estimate of CRUD Contribution to Shipping Cask Containment Requirements, SAND-1358, Sandia National Laboratory, Albuquerque, New Mexico (1991).
- [32] U.S. NUCLEAR REGULATORY COMMISSION, Standard Review Plan for Dry Cask Storage System, NUREG-1536, NRC, Washington, D.C (1997).
- [33] GRAY, W.J., WILSON, C.N., Spent Fuel Dissolution Studies FY 1991 to 1994, PNL-10540, Pacific Northwest Laboratory, Richland, Washington (1995).

- [34] SCHEMEL, J.H., ASTM Manual on Zirconium and Hafnium, ASTM STP 639, American Society for Testing and Materials, Philadelphia (1977).
- [35] WILSON, A.W., et al., "Westinghouse Fuel Performance in Today's Aggressive Plant Operating Environment," International Meeting on LWR Fuel Performance (Proc. Mtg. Portland, Oregon 1997), 23 pp.
- [36] U.S. NUCLEAR REGULATORY COMMISSION, Point Beach Augmented Inspection Team Report, EA No. 96-215, NRC, Washington, D.C. (1996c).
- [37] MARSHALL, .RP., LOUTHAN, M.R., "Tensile Properties of Zircaloy with Oriented Hydrides" Trans. of ASM, Vol. 56 (1963), 693 pp.
- [38] NORTHWOOD, D.O., KOSASIH, U., "Hydride And Delayed Hydrogen Cracking In Zirconium And Its Alloys," Internat. Metals Rev., 28(2):92 (1983).
- [39] COX, B., Effect of Hydrogen Injection on Hydrogen Uptake by BWR Fuel Cladding. NP-3146, Electric Power Research Institute, Palo Alto, California (1983).