UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555-0001

MEMORANDUM TO: Gary M. Holahan, Director
Division of Systems Safety and Analysis
Office of Nuclear Reactor Regulation

FROM: Farouk Eltawila, Acting Director
Division of Systems Analysis and Regulatory Effectiveness
Office of Nuclear Regulatory Research

SUBJECT: RES REVIEW OF AND RESPONSE TO ACRS COMMENTS ON SPENT FUEL CLADDING BEHAVIOR FOLLOWING A LOSS-OF-WATER ACCIDENT DURING POOL STORAGE

I am attaching to this memorandum a draft report for review and comments. The report was prepared by RES and its contractor, ANL, to address the subject of spent fuel cladding behavior following a loss-of-water accident during pool storage. As you are aware, in a letter to Chairman Meserve, dated April 13, 2000, the Advisory Committee on Reactor Safeguards (ACRS) provided its comments on the staff report, "Draft Final Technical Study of Spent Fuel Pool Accident Risk at Decommissioning Nuclear Power Plants." The comments indicated that the staff needed to strengthen the technical basis for addressing the subject of spent fuel cladding behavior at high temperatures in air. The attached report has been prepared in response to ACRS comments and recommendations. Highlights of the report findings regarding breakaway oxidation, clad ballooning and burst, cladding creep, and autoignition are stated below.

- Based on available data on air oxidation of fresh cladding in the temperature range of 800-1200°C, breakaway oxidation of Zircaloy in air at temperatures below 800°C is not an issue. For lack of required oxidation kinetics information on pre-oxidized cladding, accelerated oxidation of spent fuel cladding at temperatures above 800°C cannot be ruled out.

- Cladding is likely to fail by ballooning and burst in the temperature range of 700-850°C. The exact burst temperature is influenced by many factors such as fuel burnup, internal pressure, pre-accident oxide layer thickness, heating rate, and pellet-cladding bonding. Given the inherent uncertainty in burst temperature, potential for cladding failure by ballooning and burst at a peak cladding temperature above ~700°C, cannot be ruled out.

- Breakaway oxidation alone does not necessarily lead to runaway autocatalytic reaction, a precursor to autoignition. Hence, unless some type of impact force is applied to the fuel rod accidentally or intentionally leading to opening of fresh cladding surface,

autoignition is extremely unlikely prior to ballooning and burst. At burst (temperature range of 700-850°C), clean i.e., oxide-free and hot Zircaloy metal, will be abruptly exposed to air and autoignition may occur depending on surface area exposed to air. For cladding burst in simulated LOCA-like heatup tests in steam, autoignition has not been observed up to 1300°C temperature. However, for air oxidation situation, higher oxidation rate and significantly larger heat of oxidation may promote the susceptibility of fresh hot metal to autoignition. Also, by the time the cladding temperature reaches 700°C, most of the hydrides will be dissolved in the metal. Thus, Zircaloy with dissolved hydrides will be abruptly exposed to air if cladding burst occurs at 700°C. Therefore, the potential for autoignition of spent fuel cladding at minimum burst temperature (~700°C) cannot be ruled out, although the extent of material involved in such a process may be somewhat limited.

- Accelerated creep failure of the cladding at temperatures above 550°C is also a possibility. Limited creep data on low to medium burnup fuel rods indicate that the calculated 10-hour creep failure occurs at a cladding temperature of ~600°C. To exclude creep failure, the cladding temperature should be further limited to no higher than ~600°C.

Many of the above findings are based on experimental data on fresh cladding or on irradiated cladding in steam environment. As such, some confirmatory work is recommended to verify the robustness of the findings. Specifically, RES recommends air oxidation experiments with spent fuel cladding to better define the temperature limits for runaway reaction, cladding burst, and autoignition. RES further recommends, as an extension of its planned program at the Argonne National Laboratory on the high burnup dry storage issue, that creep tests be performed under conditions prototypic of spent fuel pool loss-of-water accident situation to determine the creep failure temperature.

We plan to issue the final report upon receipt of your comments.

Attachment: As stated

Spent Fuel Cladding Behavior Following Loss-of-Water Accident During Pool Storage

H. M. Chung
Energy Technology Division
Argonne National Laboratory
Argonne, Illinois

and

S. Basu
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC

1. Introduction

After discharge from an operating light water reactor (LWR), used fuel assemblies are stored and cooled in water in a spent fuel pool. Under postulated accident situations during the storage, such as an inadvertent drainage of water or a seismic event, spent fuel rods will heat up slowly following the loss of water from the pool. This document analyzes the sequence of metallurgical processes that occur in a fuel rod during a spent fuel pool loss-of-water accident, and examines the issues pertaining to potential for autoignition of fuel cladding, loss of cladding structural integrity, and release of gaseous and solid fission products. The information on fuel behavior is essential in determining temperature limits for spent fuel assemblies that will assure coolable geometry.

2. Initial Metallurgical State of LWR Fuel Rod

The initial state of an LWR fuel rod at the onset of a postulated loss-of-water accident is influenced by many factors, such as reactor type (BWR or PWR), fuel burnup, linear heat generation rate, reactor coolant temperature during operation, irradiation history, cladding material type (e.g., standard Zircaloy-4, low-Sn Zircaloy-4, standard Zircaloy-2, liner Zircaloy-2, Zirlo, or M5), cladding fabrication variables, and the time of storage in the pool.

The spent fuel pools at power plants scheduled for decommissioning will have an inventory of fuel cladding fabricated mostly from conventional Zircaloys, i.e., standard Zircaloy-4 for PWR and Zircaloy-2 for BWR. Of these, Zircaloy-4 cladding exhibits more pronounced degradation in microstructure (i.e., thicker oxide layer, higher hydrogen uptake, and higher volume fraction of hydrides) under conditions of otherwise similar burnup and irradiation history. Furthermore, the degradation in standard Zircaloy-4 cladding becomes significantly more pronounced as fuel burnup increases to >55 MWd/kgU.

The degradation of a BWR fuel rod is relatively less than that of a PWR rod for similar burnup and fluence level. However, the nature of irradiation-induced damages in Zircaloy-2 cladding could differ from those of Zircaloy-4, primarily because irradiation temperature is lower in a BWR than in a PWR (i.e., ~290°C vs. ~325°C). As a result, depending on exact burnup and oxidation, irradiation-induced increase in yield strength and decrease in ductility and fracture toughness could be more pronounced for standard Zircaloy-2 cladding than for standard Zircaloy-4 cladding at lower temperatures. Nonetheless, for the purpose of the present evaluation in which potential for autoignition, ballooning and burst, and breakaway oxidation at temperatures well above the ones mentioned are the issues, standard Zircaloy-4 cladding will be considered as a bounding case as it exhibits more oxidation, more hydriding, and thinner wall.

Standard Zircaloy-4 PWR fuel cladding is characterized by the following microstructural features (see illustration in Figure 1):

(1) The thickness of the inner diameter (ID) oxide layer varies nominally up to ~10 μm. For high burnup (>55 MWd/kgU), the ID of the cladding and fuel are bonded tightly through interdiffusion of Zr and U, which forms a compact layer of monoclinic $(Zr,U)O_2$. The bonding appears to be stronger and more prevalent at higher burnups.

(2) The total thickness of the oxide layer at the cladding outer diameter (OD) ranges from ~20 μm to ~120 μm. Of this, the innermost layer could be 2 to 20 μm thick whereas the outer layer is much thicker. The thickness also varies strongly as a function of the grid span position (see Fig. 2).

(3) The oxygen stoichiometry and structure of the oxide layer, produced in steam or oxygen, vary as function of its location relative to phase boundaries, i.e., from the coolant-oxide boundary to the oxide-metal boundary. Under equilibrium condition, Zr oxide retains monoclinic structure at <1000°C, tetragonal structure at 1000°C-1480°C, and cubic structure at [illegible]°C. However, under transient oxidation and irradiation conditions that prevail during reactor operation, all three forms of oxides can be present.

(4) The OD oxide layer adjacent to the coolant boundary has a higher oxygen stoichiometry and a monoclinic crystal structure which has relatively lower fracture toughness of ~1-2 MPa√m. The outer oxide layer is usually porous, contains dense interconnected microcracks, and has low fracture toughness. As a result, the OD oxide layer has high susceptibility to breakaway oxidation that may occur during in-reactor service. Within the outer oxide layer, steam or air molecules can penetrate and migrate freely through the interconnected microcracks.

(5) In contrast, the inner oxide layer adjacent to the metal has a lower oxygen stoichiometry, higher volume fraction of tetragonal phase, and relatively higher fracture toughness (because the tetragonal oxide has relatively high fracture

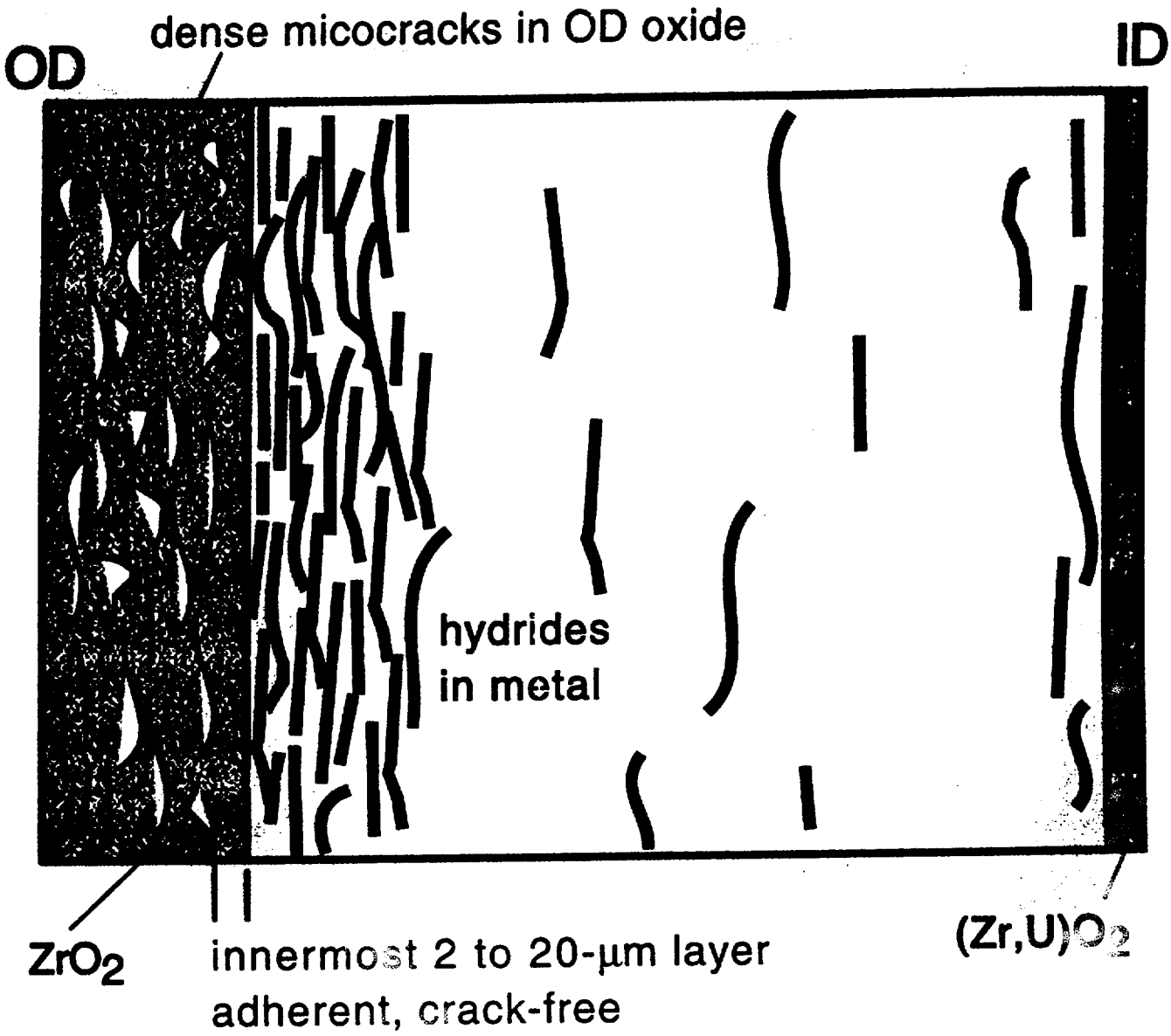


Figure 1. Schematic Illustration of Microstructural Characterization of High Burnup Fuel Cladding

Axial Profiles, Burnup and Oxidation

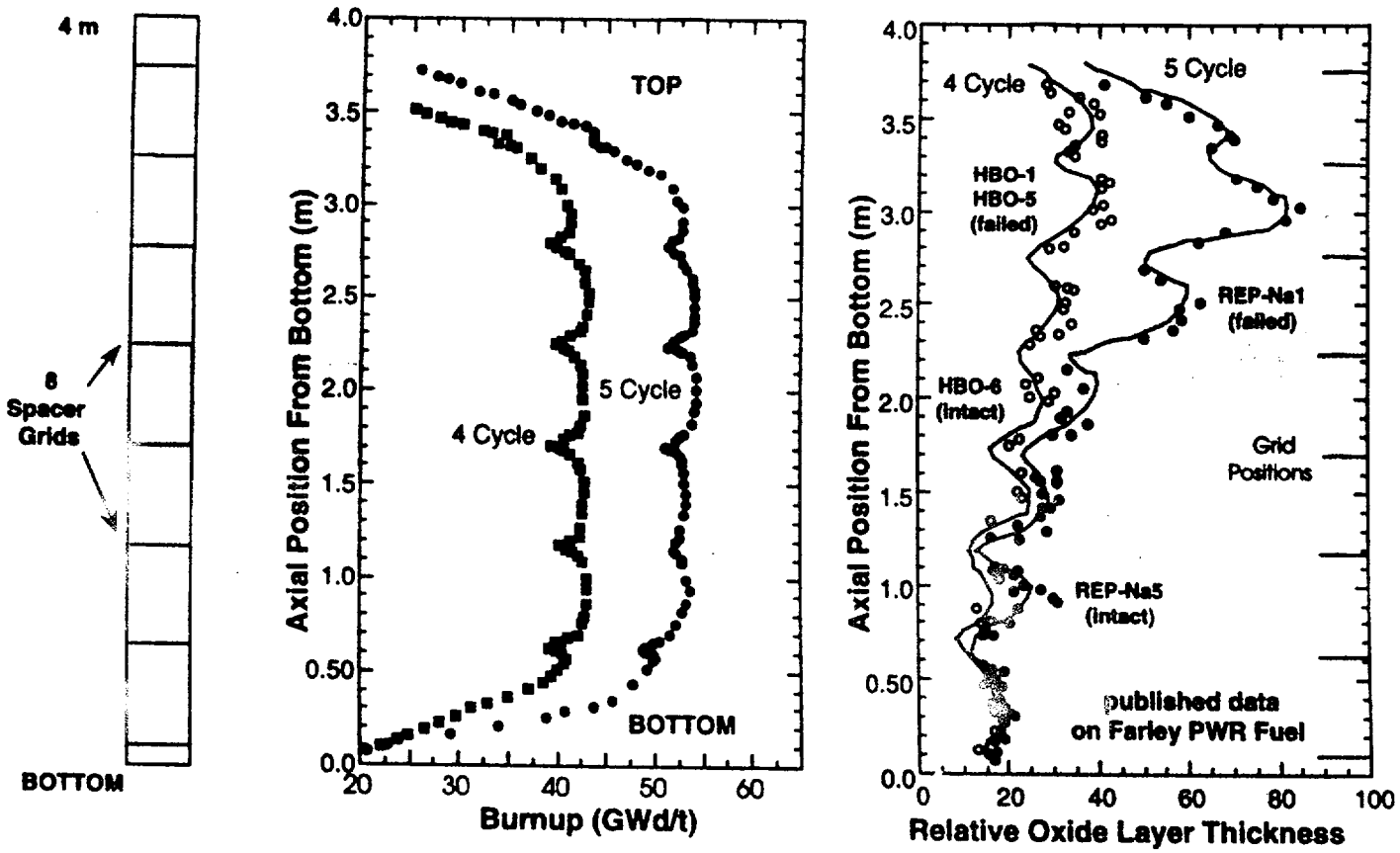


Figure 2. Typical Axial Profiles of Fuel Burnup and Waterside Oxide Layer Thickness in High Burnup Fuel Cladding

toughness of ~4-8 MPa $\sqrt{m}$). Most of all, the inner oxide layer is virtually free of microcracks, and provides a tight adherent cover to the OD-sidemetal (or metal-hydride composite in high-burnup fuel cladding). Thus, steam or air molecules cannot make a direct contact with the hydride-rich metal rim.

(6) The metal in contact with the OD-side oxide contains δ-phase Zr hydrides (stoichiometry close to $ZrH_{1.66}$) in high density. Volume fraction of the hydrides in the rim can be as high as ~70% at high burnup. However, hydride volume fraction in the cladding midwall and the ID-side metal is insignificant even at high burnup.

(7) Most hydrides are aligned predominantly in the circumferential direction of the cladding. In this orientation, hydrides are not deleterious to the mechanical properties of the cladding under tensile or hoop stress. The volume fraction of deleterious radial hydrides in as-irradiated state is insignificant.

(8) A significant level of residual stress, produced in association with oxidation, rod bowing, and pellet-cladding mechanical interaction (PCMI), may be present in the oxide and the metal.

The above features play important roles in influencing the behavior of fuel cladding during a loss-of-water accident.

3. Reaction of Zircaloy Cladding in Air

After the onset of a loss-of-water accident, pool water is assumed to be fully drained in a relatively short period of time, e.g., within several minutes, although the exact time will depend on the type of accident and the size of the drainage opening. The fuel cladding will then be exposed initially to steam-water mixture, then to steam, steam-air mixture, and eventually to air, under near atmospheric pressure.

In air or oxygen-nitrogen mixture, reaction of fresh (i.e., unirradiated oxide-free) Zr or Zircaloy cladding has been reported to be faster than the reaction in steam [1-6]. A mixture of Zr oxide and Zr nitride, or probably Zr oxynitride, is formed in air as a reaction product. Corrosion (oxide) layer of Zr formed in air is characterized by distinctive gold or yellowish color which is in contrast to black or greyish-black color of Zr oxide formed in steam or oxygen.

The reaction rate (measured in terms of weight gain) of Zr or Zircaloy varies strongly with temperature. At or above ~700°C, oxidation kinetics in steam is represented by parabolic rate equations. Accelerated oxidation (i.e., linear kinetics) in steam at high temperatures occurs only under certain conditions, e.g., in hydrogen-rich steam-hydrogen mixture at ~1100°C [7]. In an unpublished work [8], reaction of fresh Zircaloy-4 was investigated at 1000-1450°C in air and steam environments. The results from this work show that at 1100°C, the rate of oxide layer growth is ~1.8 times faster in air than in steam within the range of parabolic regime of reaction.

Susceptibility to breakaway oxidation of Zircaloy is greatly promoted by exposure to air. For example, accelerated oxidation of fresh cladding was observed to occur in air at 800-1200°C

[4,6,9]. In long exposure tests (e.g., >43 minutes at 1100°C), breakaway oxidation occurred and, as a result, oxidation was greatly accelerated.

Reaction of oxide-covered spent fuel cladding in air could be different from that of oxide-free fresh Zircaloy cladding because of the existing oxide layer (20-120 μm thick) produced in water during operation. If free of interconnected microcracks, this pre-existing oxide layer will provide a degree of protection from the air attack. However, if the oxide layer contains pores and interconnected microcracks as in Figure 1, it will promote breakaway oxidation and enhance oxidation rate even at a relatively low temperature. Currently, no data base exists on air oxidation tests of oxide-covered spent fuel or pre-oxidized unirradiated cladding. For lack of such information, it is reasonable to assume that following a loss-of-water accident, a breakaway oxidation in air of pre-oxidized cladding is possible practically at all temperatures of interest. However, reaction at lower temperatures (<800°C) is expected to be slow in air, as it is in steam. Consequently, breakaway oxidation of Zircaloy in air at temperatures below 800°C is not a serious concern.

In high-burnup (>55 MWd/kgU) cladding, the outer i.e., the OD-side oxide layer will very likely contain pores and microcracks (see Figure 1). As such, this layer or a fraction of it is likely to peel off under stress thus promoting breakaway oxidation. However, the crack-free innermost oxide layer (2-20 μm thick) will prevent a direct contact of air with the metal or metal-hydride composite. Once again, no data base currently exists to confirm this hypothesis and, as such, it is reasonable to assume that breakaway oxidation in high burnup cladding is likely at all temperatures of interest, in particular, above 800°C. Air oxidation tests of pre-oxidized cladding are recommended to validate this assumption and to determine air-oxidation kinetics that are applicable to pre-oxidized spent-fuel cladding.

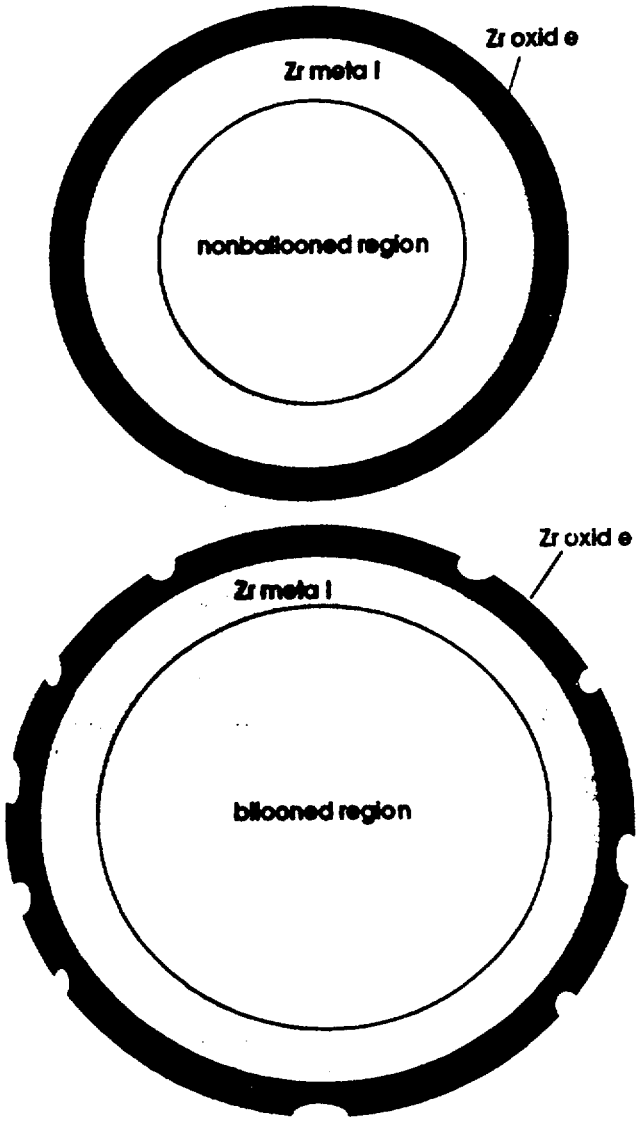
In addition to reaction rates being relatively high, reaction of Zircaloy in air is also more exothermic than in steam. The exothermic heats (per mole of Zr or ZrH_2 reacted) of reaction of Zr and Zr hydride in steam, oxygen, and nitrogen environments are listed below:

$Zr + O_2 = ZrO_2$	ΔH_0 = -1100.6 kJ (263 kcal)
$Zr + 2H_2O$ (steam) $= ZrO_2 + 2H_2$	ΔH_0 = -617 kJ (147 kcal)
$Zr + H_2 = ZrH_2$	ΔH_0 = -169 kJ (40.3 kcal)
$Zr + 1/2N_2 = ZrN$	ΔH_0 = -87.2 kJ (20.8 kcal)
$ZrH_2 + O_2 = ZrO_2 + H_2$	ΔH_0 = -931.6 kJ (223 kcal)
$ZrH_2 + 2H_2O$ (steam) $= ZrO_2 + 3H_2$	ΔH_0 = -448 kJ (107 kcal)

This information indicates that, under otherwise identical conditions, high temperature runaway reaction is more likely to occur for Zr or Zr hydride in oxygen (and hence, in air) than for Zr (or fresh clean Zircaloy) in steam.

4. Potential for Cladding Failure by Ballooning and Burst

As cladding temperature increases following a spent fuel pool loss-of-water accident, irradiation damages start to anneal out, and cladding is virtually restored to that of its unirradiated state at temperatures higher than 700°C. The situation is similar to that of unirradiated cladding exposed



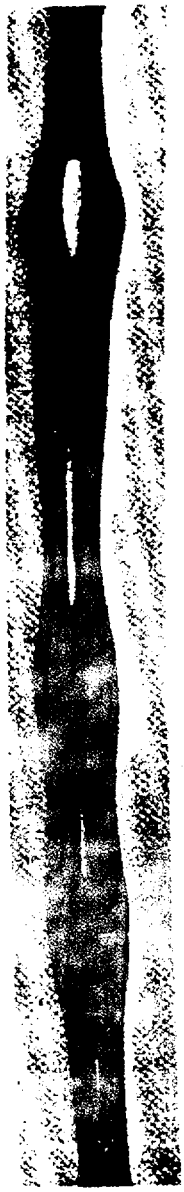


Figure 3. Schematic Illustration of Ballooning and Burst of Fuel Cladding

to small-break LOCA-like conditions. Hence, ballooning and burst will likely occur at temperatures between ~700°C and ~850°C [9]. Figure 3 shows a typical ballooned shape and cross section of a fuel rod. Wall thinning occurs in the ballooned regions which ultimately leads to cladding burst. The exact burst temperature is influenced by many factors such as fuel burnup, internal pressure, pre-accident oxide layer thickness, heating rate, and pellet-cladding bonding. Given the inherent uncertainty in burst temperature, potential for cladding failure by ballooning and burst at a peak cladding temperature above ~700°C cannot be ruled out.

After burst, the ID surface of the burst cladding will be exposed continuously to air-fission-gas mixture and then to air. As a result, high-temperature oxidation will occur at the boundary between the ID-side metal and pellet (or pellet-cladding bonded layer in high burnup fuel). By this time, steam will be virtually absent in the environment; therefore, generation of hydrogen gas from the reaction of the ID surface of the burst cladding will be negligible. Hence, potential for ignition of any air-steam-hydrogen or air-fission-gas-hydrogen mixture is not a concern.

For high burnup fuels, the tight cladding-pellet-bonded layer, formed after operation to burnups >55 MWd/kgU, is not likely to be separated during burst. Thus, the ID surface of a burst cladding is likely to remain covered with a relatively inert layer of $(Zr,U)O_2$, which may limit the exposure of fresh cladding to air.

5. Potential for Autoignition of Cladding

Autoignition can occur in Zr-base alloys and Zr hydride driven by the unusually large exothermic heat of oxidation and fast oxidation rate, which produce exceedingly high rate of heat input than the rate of heat loss, thereby increasing the material temperature rapidly. As the temperature increases, the reaction rate increases further, thus accelerating heat generation further, leading to a classical autocatalytic reaction. For autoignition of cladding to occur, however, three conditions must be met at the same time: (1) sufficiently high temperature of the material, (2) sufficiently high surface-to-volume ratio of the material, and (3) clean surface of the material (i.e., metallic Zircaloy or Zr hydride) in contact with the environment.

A pile of fine freshly machined Zr turnings or powders, exposed to moist air, is one of the worst cases; autoignition can occur in such a pile even at room temperature. Another worst case is when hot clean metal (e.g., fresh oxide-free Zircaloy preheated to ~800°C in vacuum) is abruptly exposed to oxygen or air. Sheperd et al.[10] reported that oxidation of pre-existing ZrN layer led to violent superheating of cladding specimen and destruction of test tube. It is not clear whether this was due to autoignition, or more likely, due to temperature escalation from an excessive heat input, which resulted in a breakaway oxidation and further superheating.

The situation and potential for autoignition of spent fuel cladding under conditions of loss-of-water accident are different from the above idealized scenarios. For one thing, the spent fuel cladding in the storage pool is covered with a relatively thick oxide layer. Also, zirconium hydrides in spent fuel cladding are present in the metallic Zircaloy as dispersed second-phase material. This metal-hydride composite is covered with either Zr oxide, Zr oxynitride, or a mixture of Zr oxide and Zr nitride [10]. Therefore, it is unlikely that significant amount of oxygen or air can come into direct contact with clean metal or metal-hydride composite. Hence, unless some type of impact force is applied to the fuel rod accidentally or intentionally leading to

opening of fresh cladding surface, autoignition is extremely unlikely prior to ballooning and burst failure. For transient oxidation tests of oxide-free unirradiated cladding in air, no case of autoignition has been mentioned in Refs. 5, 6, and 10.

If there is cladding failure by ballooning and burst (in the temperature range of 700-850°C), clean (i.e., oxide-free) and hot Zircaloy metal will be abruptly exposed to air. Also, by the time the cladding temperature reaches ~700°C, most Zr hydrides will be dissolved in the metal because the solubility limit of hydrogen in Zircaloy metal increases significantly with increasing temperature. At the heatup rates reported by Benjamin et al. [11], for spent fuel following a loss-of-water accident, hydride dissolution will be significant. As a result, clean and hot material (consisting of Zr metal with dilute hydrides) will be abruptly exposed to air. However, the surface-to-volume ratio of the air-exposed material will be low, because at most only the circumferential cross section of the cladding metal will be exposed to air and the rest of the metal or metal-hydride composite will remain covered with oxide, oxynitride, or a mixture of oxide and nitride.

For similar burst conditions during simulated LOCA-like heatup tests in steam, it has been well established that autoignition of Zircaloy cladding does not occur. In numerous LOCA tests conducted in 1970s and 1980s in many laboratories on unirradiated Zircaloy cladding in steam, no autoignition has been reported. For example, no autoignition was observed during burst tests of unirradiated cladding tubes in stagnant steam at 650-1300°C, which is documented in Ref. 9. In that study, the ballooning and burst sequences in numerous experiments were recorded on high-speed color videotapes. The video records show that only a localized hot spot is developed at the moment of burst at the burst location, which is followed by prompt disappearance of the hot spot (within several seconds) without an ignition of the cladding.

However, for the air-oxidation situation in a spent-fuel-pool accident, it is necessary to take into account several factors: (1) the effects of the higher oxidation rates in air, (2) significantly larger heat of oxidation (i.e., 1[illegible] kJ per mole of Zr in air vs. 617 kJ in steam), and (3) sudden exposure of the clean surface to oxygen. Presence of a large amount of oxygen molecules could play a key role in promoting susceptibility of the freshly exposed hot metal in the burst region to autoignition. It would be necessary to conduct actual ignition tests on either spent fuel or pre-oxidized and hydrided cladding to generate experimental data to understand these various effects and to determine unambiguously the potential for autoignition. For lack of such experimental data, the potential for autoignition after ballooning and burst cannot be ruled out at this time.

6. Potential for Creep Failure

At cladding temperature >550°C, irradiation damages are virtually annealed out and irradiation-induced hardening starts to be recovered. At the same time, hydrides start to dissolve in the metal. As a result, yield and rupture strengths of the cladding decrease significantly. Therefore, cladding deformation and failure by thermal creep is a distinct possibility at temperatures above 550°C. For a given spent fuel cladding of a given burnup and operating history, potential for creep failure is strongly influenced by peak cladding temperature and stress (i.e., residual stress plus the hoop stress due to the rod internal pressure). Old data [12] on low to medium burnup

fuel rods indicates that the calculated 10-hour creep failure occurs at a peak cladding temperature of ~600°C.

Long-term creep failure at even lower temperatures is not unlikely, however, it is more relevant to the dry storage issue. In the experiment of Einziger and Kohli [13], PWR cladding irradiated to burnup of 30-32 MWd/kgU remained intact under an applied stress of ~143 MPa at 323°C for ~87 days and during subsequent cooling, despite the fact that extensive reorientation of hydrides occurred in the cladding. This level of stress is higher than or comparable to the maximum stress expected for PWR spent-fuel cladding irradiated to <60 MWd/kgU. In Ref. 14, failure of PWR fuel cladding (burnup 22-28 MWd/kgU) was reported to occur in 10-13 days at 325°C under unrealistically high applied stress in the range of 315-469 MPa,. A more recent work [15], soon to be published in an open journal, focused on accelerated creep tests of high burnup PWR cladding (burnup ~64 MWd/kgU) under even higher stress conditions (400-620 MPa as opposed to an expected stress of ~140 MPa). At 300°C and 620 MPa, no failure was observed in 8 days whereas at 370°C and 400MPa, failure occurred in 3-5 days.

There is a planned RES-sponsored program at the Argonne National Laboratory which will generate accelerated creep data to address the high burnup dry storage issue. As an extension to this program, it is recommended that one or two creep tests be performed at temperature and stress conditions prototypic of spent pool loss-of-water accident situation.

7. Summary

In summary, the spent fuel cladding behavior following a loss-of-water accident was examined from a sequence of metallurgical processes based on different regimes of maximum temperature that can be reached in such an accident. Specifically, breakaway oxidation, ballooning and burst, creep failure, and potential for autoignition were examined, and temperature regimes for these events were identified (see Figure 4).

The breakaway oxidation of cladding in air can occur at all temperatures relevant to spent fuel loss-of-water accident. However, the breakaway oxidation alone is not a concern as long as it does not lead to accelerated oxidation and runaway autocatalytic reaction (precursor to autoignition). Accelerated oxidation of Zircaloy cladding in hydrogen-rich steam environment has been reported to occur at ~1100°C. Accelerated oxidation of fresh cladding in air was observed at 800-1200°C. Reaction of oxide-covered spent fuel in air is likely to be different from that of fresh fuel. However, no data base currently exists to confirm this hypothesis. As such, it is assumed here that breakaway oxidation of spent fuel leading possibly to autoignition is not likely to occur as long as cladding temperature is limited to no higher than ~800°C. Air oxidation experiments with spent fuel cladding are recommended to verify the assumption.

However, if the cladding temperature exceeds 700°C, ballooning and burst will likely occur. If there is cladding failure by ballooning and burst (in the temperature range of 700-850°C), clean (i.e., oxide-free) and hot Zircaloy metal will be abruptly exposed to air. Because of lack of experimental data with oxide-covered spent fuel, the potential for autoignition at burst, though not confirmed at this time, cannot be ruled out. At any rate, following a burst event, fission gas (gap activity) will be released to the pool containment. For high burnup fuel, a portion of loose fuel particles contained in the rim zone could also be released through the burst opening. This

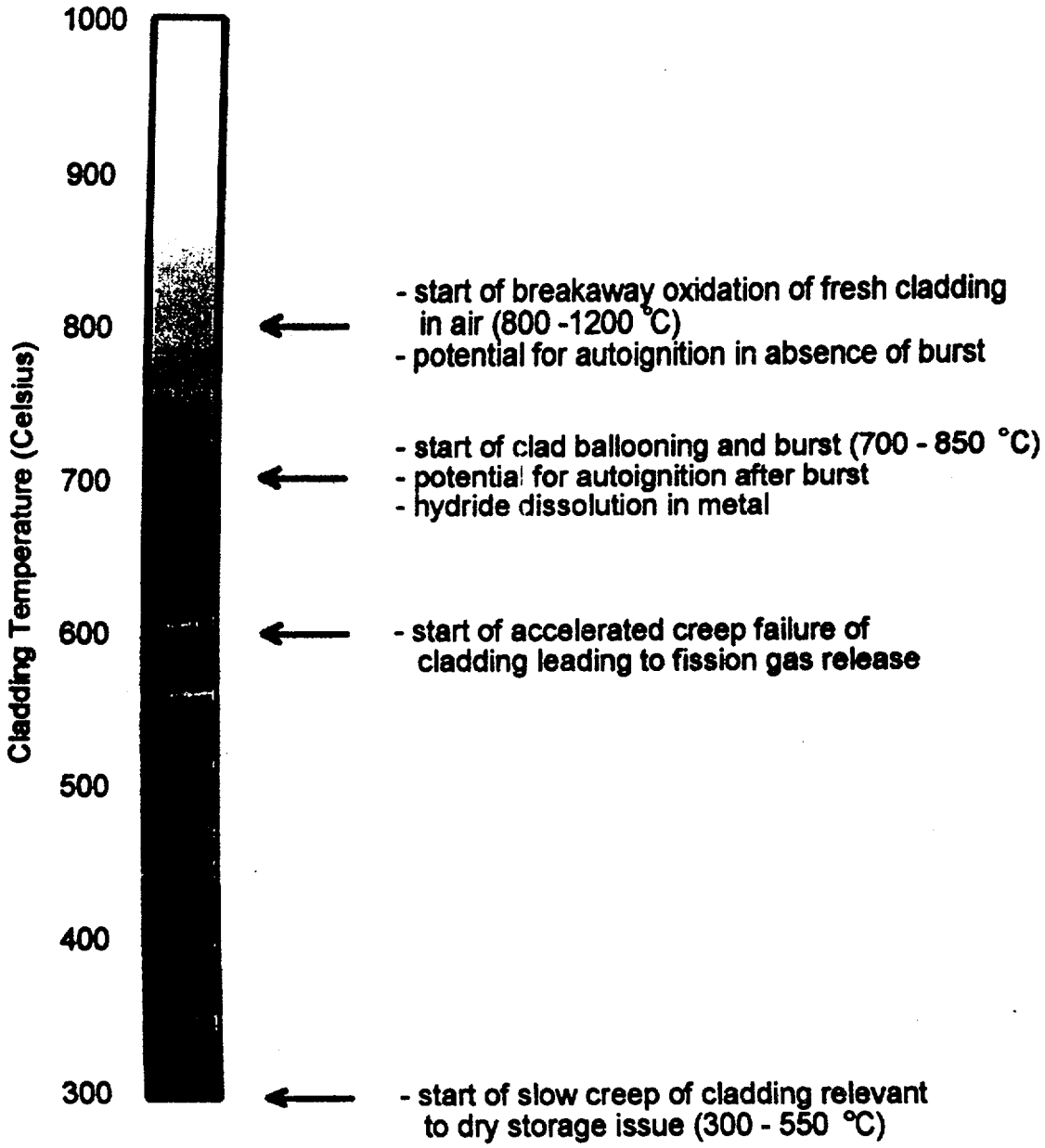


Figure 4. Temperature Regimes for Various Phenomena Associated With Spent Fuel Cladding

can be prevented by limiting the cladding temperature to no higher than ~700°C, and thus avoiding cladding burst.

Creep failure of spent fuel cladding can occur at temperatures significantly lower than the burst temperature. However, at low temperatures (300-400°C) and under stress conditions prototypic of spent fuel pool loss-of-water accident scenario, the creep process is rather slow. At ~600°C, however, the creep process may be accelerated as evidenced from the limited data (i.e., calculated 10-hour creep failure). The potential for autoignition due to creep failure is likely to be very small. Nevertheless to exclude creep failure and hence, release of fission products, the temperature should be further limited to no higher than ~600°C. It is recommended that creep tests be performed at conditions prototypic of spent pool loss-of-water accident situation, to confirm the creep failure temperature.

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