Aquarius Services Corp.
17 Pokahoe Drive
Sleepy Hollow, NY 10591
USA

Phone: +1 (914) 366-8875
Fax: +1 (914) 366-8876
e-mail: Stras69@att.net

Advanced Nuclear Technology
Uppsala Science Park
SE-751 83, Uppsala
SWEDEN

Phone: +46-(0) 18-50 66 80
Fax: +46-(0) 18-50 66 85
e-mail: ant@ant.se

THE ANNUAL REVIEW OF ZIRCONIUM ALLOY TECHNOLOGY FOR 2000

(ZIRAT-5)

Ron Adamson
Brian Cox
Peter Rudling
Alfred Strasser
Gunnar Wikmark

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This document is produced by Aquarius Services and Advanced Nuclear Technology for the ZIRAT-5 membership.

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EXECUTIVE SUMMARY

2-FUEL PERFORMANCE GOALS AND ACHIVEMENTS

The deregulated market means that the nuclear utilities need to needs to reduce maintenance and fuel cycle costs to remain competitive. Also reactor safety needs to be improved while the plant radiation buildup must be limited to cope with the change in political environment towards nuclear power. To achieve the above mentioned goals the following changes have been and are being introduced:

- Longer cycles
- Higher discharge burnups,
- Modified water chemistries
- Plant power uprates
- Increased coolant temperatures
- More aggressive fuel management methods

However there are a number of different issues that needs resolution to be able to successfully implement the above mentioned changes. The most important issues are:

- Oxidation levels higher than predicted,
- Excessive internal gas pressure in burnable poison rods,
- Incomplete control rod insertion (IRI) events,
- Large axial offsets or axial offset anomalies (AOA),
- Fuel failures due to high fuel duty,
- Adverse effects of water chemistry,
- High crud buildup, and
- Accelerated growth of rods and assemblies
- Fretting
- Criteria and Analysis for Reactivity Accidents
- Criteria and Analysis for Loss-of-Coolant Accidents
- Criteria and Analysis for BWR Power Oscillations

The fuel vendors on their part are developing new fuel designs, including more advanced materials to resolve some of the issues.

5-DIMENSIONAL STABILITY

- In terms of dimensional stability issues, highlights include the following:
  - Many new PWR alloys being developed, primarily for corrosion resistance at high burnup. These alloys have creep and growth properties, which are equivalent to or better than for SR Zircaloy-4. Many have been tested to 50-60 MWd/kgU.
  - The new PWR alloys have less irradiation growth and creep than standard SR Zircaloy-4.
  - For fuel rod growth, the new PWR alloys are all about equivalent to 50 MWd/kgU. The real benefit or difference may come at higher burnups not yet reported.
  - Promising “new” PWR alloys include M5, Duplex Zircaloys like DX ELS, Zirlo, MDA, “modified” Zircaloy. At the moment, there is no clear winner, but higher burnup data will likely clarify the situation.
- Promising approaches to reducing guide tube elongation include:
  - Decrease corrosion hydrogen
  - Use new alloys
  - use “high Sn” Zircaloy-4
  - use beta-treated Zircaloy-4
  - use high S M5
- All of the above require further testing, which appears to be in progress.
- RX Zircaloy-2 in BWRs has about the same fuel rod growth as the new PWR alloys under PWR conditions.
- There is a heightened awareness of the effect of corrosion hydrogen on dimensional stability of all reactor components. Decreasing hydrogen will decrease fuel and guide tube/water rod elongation.
- In unirradiated SR Zircaloy-4, hydrides are shown to increase the creep strength. It is yet to be shown if this is so far RX Zircaloy, or in-reactor.
- Firm data show that breakaway irradiation growth of RX Zircaloy-2 at BWR temperature will remain liner with fluence to fluences well above projected fuel exposure limits. Accompanying data show that the c-dislocations, which drive growth also, increase linearly with fluence.
- The temperature dependencies of creep rod growth have been confirmed. Growth goes down and creep goes up with increasing temperature.

- It has been demonstrated that instrumented in-reactor creep experiments are possible. Such experiments have shown that primary creep reoccurs after any reversal of stress on RX Zircaloy-2, that there is a tendency for creep strength to be higher in compression than tension, and that steady state creep (if ever truly achieved in-reactor) has a stress dependency near unity.

- SPP stability in the neutron flux is being shown to vary between SPP types. This may be an important guide to alloy development.

- No new data has clarified the uncertainties, which exist about the Russian data concerning fluence-magnitude and experimental details.

6-CORROSION & HYDRIDING

An introductory summary of current views on the mechanisms of zirconium alloy corrosion and hydrogen uptake demonstrates the importance of the oxide structure (and how this develops and degrades) to both these processes. The impact of alloy composition, irradiation, and water chemistry on oxide growth and breakdown is discussed. Possible oxide degradation processes such as mechanical cracking; shear cracking during tetragonal to monoclinic ZrO2 phase transformation; and local oxide dissolution in aggressive aqueous environments are the main mechanisms considered. The hydrogen uptake mechanism is now thought to be controlled by the nature (size, shape and frequency) of the pores and cracks generated in the oxide film by these mechanisms. These processes are discussed in terms of their relevance to behaviour in PWR (VVER) or BWR water conditions.

During the past year, there has been little improvement in our understanding of the formation and degradation of ZrO2 films as there has not been a major conference that dealt with such basic topics. Some published papers in the zirconia ceramics and zirconia optical coatings fields have provided evidence that is of some help in this understanding. For instance, it appears that the solubility of hydrogen in the ZrO2 lattice is less than 1ppm(wt.), so that all the hydrogen measured in corrosion films on zirconium alloys must be present as OH or H2O on crack or pore surfaces.

Additional studies of the effects of second phase particle size on the corrosion behaviour of various alloys were reported, but do not change our understanding of these effects, merely extend the knowledge to different precipitate compositions.

New results on the behaviour of advanced alloys in PWRs and BWRs were presented at the Park City ANS meeting and extend the database to higher burnups. However, anecdotal evidence of the poor performance of some of the advanced alloys has yet to be translated into published reports on these situations. "Shadow Corrosion" in BWRs continues to attract much attention, and appears to be more certainly an example of galvanically enhanced corrosion under irradiation, with the cathodic (H uptake) part of the process transferred elsewhere.

7-WATER CHEMISTRY EFFECTS

Noble Metal Chemical Addition, to enhance the oxygen suppression by HWC application in BWRs, has been applied in a number of plants for a few years. The experience gained is not yet indicating any problems for the fuel cladding, despite some concerns in relation to previous experience. The application of NMCA has, however, been found to produce a more global effect on the water chemistry. One such effect is the general suppression of iodine during HWC and the appearance of occasional transients of iodine. This effect could make fuel failure supervision more difficult in NMCA plants. Some other water chemistry effects have also been experienced, and the proper monitoring and control of water chemistry is hence becoming more complex with such chemistry regimes.

The data released on the enhanced spacer shadow corrosion incident in BWR Leibstadt (KKL) in 1997 and the crud induced failures in the BWR River Bend in 2000 stress the need for a better understanding and control of the water chemistry. The KKL incidence was reportedly due to iron deficiency, i. e. too little iron in the deposited crud, but the River Bend incidence was instead due to excessive, and non-monitored, amounts of iron. Both plants operated within the water chemistry guidelines. The understanding for the interaction between water chemistry, crud, and cladding corrosion is still no sufficiently good to avoid this kind of incidences, and the water chemistry guidelines are hence insufficient for today’s plant operation.

8-PRIMARY DEFECTS

Fretting is by the far the most common primary failure cause for both PWRs and BWRs today. Grid-to-rod fretting is also still an important failure mechanism in the PWR and considerable efforts are spent on developing test loops and advanced fluid dynamics codes today to cope with the problem. Very little is, however, still known of the real conditions during operation producing the fretting. Development of new spacer designs is consequently still largely empirical. Yet, using best knowledge and considering other factors, such as the PWR assembly inflow turbulence, seems to be a viable way to mitigate grid-to-rod fretting.

A German analysis of the recent dry-out experiments in the Halden BWR has resulted in potentially much larger margins towards dry-out than used for licensing today. The possibility to release some of the dry-out margins could be of great importance for the core loading possibilities, as the target burn-ups are forcing more rods close to the maximum available uranium enrichment of 5%. The approach has to be accepted by authorities first, which is probably not a simple task.

The development of the Zr-Fe liner by Siemens has provided good corrosion properties, but the relative high alloying element concentration has raised some doubt about the actual PCI behaviour. Recent ramp tests show that the PCI properties for this cladding are in reasonably good agreement with other liners.

9-DEGRADATION

An interesting finding this year is the difficulty involved in designing an in-pile experiment to verify the better secondary degradation resistance of a specific fuel cladding design. In the Siemens test in Halden Zr-sponge liner and Zr0.4Fe liner rods behaved equally good. This means that one has to be cautious in interpreting results related to secondary degradation mechanisms obtained in such test reactors.

In previous years a large number of different theories were put forward to explain the axial split degradation mechanism of failed BWR fuel. However, today it appears that there is a consensus in the industry that Delayed Hydride Cracking, DHC, is the mechanism responsible for these long cracks. There are three prerequisites for this failure mechanism to occur:

- A sharp crack, could either be a primary defect such as a PCI or cladding manufacturing defect, or cracks in a secondary degradation hydride blister
- A hydrogen concentration in the cladding above TSS_p
- A large enough tensile stress in the cladding that will drive the hydrogen to the crack tip and eventually fracture the hydride; the stress results from a power ramp and the stress level increases if the liner oxidises fast

Due to the current trend in PWRs to replace Zircaloy with Zr-Nb alloys and since it is well-known that Zr-Nb alloys are more susceptible to DHC than Zircaloy, DHC may be a potential degradation mechanism in failed PWR rods. This mechanism could also result in failures in sound BWR/PWR rods with significant hydrogen contents subjected to a power ramp and with a significant oxide and/or hydride layer at the clad outer surface. The oxide/hydride layer may crack and form a sharp crack that can propagate through the cladding during the power ramp. DHC may also result in failures in fuel rods during dry storage.

10-CLADDING PERFORMANCE UNDER ACCIDENT CONDITIONS

LOCA

The applicability of LOCA criteria to high burnup cladding is being evaluated in the US, France and Japan. Some of the conclusions to date by industry (not the regulators) are:

- The 17% ECR (Equivalent Cladding Reacted) criterion is considered too conservative based on recent as well as past results.
- Pre-transition (in-reactor) corrosion level does not impact fuel rod behavior during LOCA conditions. Cladding irradiated up to 55 GWD/MT with 44μ oxide had slightly lower oxidation rates and equal or better ductility than unirradiated cladding in Japanese tests.
- Irradiation damage is completely annealed during a typical LOCA transient.

- The ductility decreases with increasing H content, however, the hydrides are redistributed and dissolved during the transient but could affect mechanical property limits if they precipitate during the reflood cooldown.
- The oxidation rates of unirradiated, pre-hydrided cladding and cladding irradiated to 55 GWD/MT were more conservative than Baker-Just rates. French claims are that Baker-Just is too conservative.

RIA

The RIA simulation test programs are continuing in the French CABRI, Japanese NSRR and Russian IGR/BGR reactors. Since none of these reactors can truly represent LWR RIAs, a water loop is being planned for CABRI by an international consortium. Unfortunately, this will not be ready for testing until 2004.

The French industry recommended and their regulatory body accepted the following RIA criteria for UO_2 fueled rods up to 64 GWD/MT:

- 100μ ZrO_2 corrosion limit,
- <60 cal/g enthalpy increase,
- >30 ms pulse at mid height,
- >700^{0}C cladding temperature.

The criteria for MOX had not been set yet. Tests to 55 GWD/MT indicated that internal rod pressure generated by high fission gas from the heterogeneous fuel increases clad stresses in addition to the PCMI and these still need investigation.

A new high burnup Japanese PWR rod test indicated that differences in pellet manufacturing methods might affect fuel rod performance in a RIA.

Two Japanese 61 GWD/MT BWR rods failed at 130 cal/g, whereas previous tests with 56 GWD/MT rods did not fail up to 140 cal/g. No definite cause for the difference has been established, but it is believed that pellet-clad bonding in the higher burnup rods vs. an open gap in the lower burnup ones played a role in the failures.

Russian tests of Zr-1%Nb clad VVER fuel rods with very long (630-850 ms) and very short (2-3 ms) pulses both showed no failures to 160 cal/g. The similar performance for the widely different pulse widths indicates a significant difference from French and Japanese tests with Zircaloy. The difference must be due to differences in clad properties, the VVER annular pellet design or test conditions.

Intensive model improvement and modeling efforts are in progress in all the countries involved: US, France, Japan, and Russia.

11- FUEL PERFORMANCE CRITERIA AND EXPERIENCE DURING DRY STORAGE

By the end of the year 2001 nearly 40 US reactors are expected to lose full core discharge capability. Without DOE's capability to fulfill its requirement to take possession and store spent fuel, the ability to license fuel for the dry storage option becomes a critical issue.

Numerous licensed commercial casks are available for dry storage and/or transport.

Fuel performance conditions in dry storage are different from those during reactor operation as follows:

- storage time is up to 40 years or longer,
- storage atmosphere is inert gas (He) at slightly higher than atmospheric pressure resulting in a high pressure differential and clad stresses due to fuel rod internal pressure,
- inert gas instead of high pressure water or steam during storage will eliminate further water corrosion and hydrogen pickup by the cladding,
- lack of radiation during storage will discontinue further radiation damage to the cladding,
- decay heat – dependent on burnup and cool-down time prior to dry storage – will raise the cladding temperature (up to as much as 400^0C) causing creep deformation and potentially eventual rupture.

Key parameters for estimating margins to failure by creep rupture, needed to license the fuel for dry storage, are therefore:

- cladding hoop stress (function of fuel rod internal pressure),
- cladding temperature (function of decay heat),
- cladding creep properties and cladding condition,
- storage time.

The NRC has established a model to calculate the acceptable combination of clad stresses, temperatures and storage times for a conservative margin to rupture of Zircaloy (advanced alloys have not been considered). The model has a creep deformation and a diffusion controlled cavity growth fracture modeling component. Based on this the NRC stated that there are sufficient experimental data to support transport and storage of fuel with an average assembly burnup up to 45 GWD/MT. Failed fuel, defined as having more than a pin hole or thin crack cladding, breach must be canned in a separate container within the storage cask. The major concern is fuel dispersal from large cracks in a rod that could create a criticality hazard, an unexpected heat load or additional radiation hazards.

Licensing of fuel in the range of 45 – 60 GWD/MT has been a recent effort. The lack of adequate cladding data for this exposure level and dry storage conditions has led to very conservative licensing criteria:

A1. No more than 1% of the rods in the assembly have peak cladding oxide thickness >80μ.

A2. No more than 3% of the rods in the assembly have a peak cladding oxide thickness of >70μ.

For fuel meeting criteria A1 and A2 the applicant should employ and acceptable methodology for calculating cladding temperature limit using a 1% strain limit.

A high burnup fuel assembly should be treated as potentially failed fuel if either of the following conditions is met:

B1. The fuel assembly does not meet both criteria A1 and A2; or

B2. The fuel rods with oxide that has become detached or spalled from the cladding."

Spalled cladding is associated with hydrogen concentrations at the cooler spalled locations that could reduce the margin to creep rupture.

The US industry represented by NEI/EPRI is countering with technical arguments to increase the clad oxidation limit, the limiting creep strain and to eliminate spalled cladding as "failed fuel". In addition the industry is developing one or more strain limited creep models that employ empirical creep correlations, rather than the NRC model that depends more on basic physical phenomena and microstructural data. Both the NRC and the industry lack all the desirable data, but the data requirements for the industry creep model can probably be obtained more rapidly, at lower cost, than the basic metallurgical data for the NRC models.

A well-planned and organized R&D program has been underway in France for some time and is producing pertinent results that could resolve some of the current questions. A program in the US is just getting off the ground. Additional mechanical testing and metallurgical characterization of cladding typical of cladding to be stored is urgently needed.

Fuel in US dry storage appears to have performed satisfactorily. Some inconsistent storage experiments exist as well. Most tests to date lack good pre-characterization of the stored fuel; combined with very limited post-storage examinations lead to a slim database. Well planned and monitored "lead storage casks" are needed to evaluate behavior of a variety of fuel designs, advanced alloys, of a range of exposures/cooling times during dry storage prior to large scale storage of similar fuel. The development of a dry storage fuel performance data base should also be started at this time to assist future licensing actions for dry storage as well as the ultimate final disposal.

12- POTENTIAL BURNUP LIMITATIONS

The potential fuel assembly burnup limitations related to zirconium alloy components are summarised in this Section. The burnup limitations that have actually been reached, but have been or are being extended, are:

- Corrosion of Zry-4 in high power PWRs, extended by improved cladding alloys, but not yet finally eliminated,
- Bowing of PWR fuel assemblies contributed in part by growth of Zry-4, extended by improved guide tube materials, guide tube design changes and reduced assembly holddown forces, but not yet finally eliminated
- Bowing of BWR channels, extended by improved manufacturing processes, and in-core channel management programs,
- RIA and LOCA related burnup licensing limits, in the process of being extended by additional experimental data and analyses.

CONTENTS

1 INTRODUCTION

The objective of the Annual Review of Zirconium Alloy Technology (ZIRAT) is to review and evaluate the latest developments in zirconium alloy technology as they apply to nuclear fuel design and performance. The large amount of data presented at technical meetings, published in the literature, and provided through discussion with the vendors in the year 2000 are evaluated and discussed to highlight the significance of the results and their potential effect on fuel performance and reliability. The review is a joint effort of Aquarius Services Corporation (Aquarius) and Advanced Nuclear Technology Sweden AB (ANT).

The primary issues addressed in the review and this report are zirconium alloy research and development, fabrication, component design, ex- and in-reactor performance including:

1. Regulatory bodies and utility perspectives related to fuel performance issues, fuel vendor developments of new fuel design to meet the fuel performance issues
2. Fabrication and quality control of zirconium manufacturing, zirconium alloy systems
3. Mechanical properties and their test methods (that are not covered in any other section in the report)
4. Dimensional stability (growth and creep)
5. Primary coolant chemistry and its effect on zirconium alloy component performance
6. Corrosion and hydriding mechanisms and performance of commercial alloys
7. Cladding primary failures
8. Post-failure degradation of failed fuel
9. Cladding performance in postulated accidents (LOCA, RIA)
10. Dry storage
11. Potential burnup limitations

Current uncertainties and issues needing solution are identified throughout the report.

Background data from prior periods have been included wherever needed. All the data are from non-proprietary sources; however, their compilation, evaluations, and conclusions in the report are proprietary to Aquarius, ANT and ZIRAT members as noted on the title page.

The authors of the report are Dr. Ron Adamson, Dr. Brian Cox, Professor Emeritus, University of Toronto; Al Strasser, President of Aquarius; Dr. Gunnar Wikmark, President of ANT, and Peter Rudling, Vice-President of ANT.

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The term of ZIRAT-5 is January 1, 2000 to January 31, 2001.

2 UTILITY PERSPECTIVE/EXPERIENCE, REGULATORY BODIES, AND FUEL VENDOR BURNUP EXPERIENCE

2.1 REGULATORY PERSPECTIVE

Meyer, *Ref. 2-1*, provided the historical background for the burnup limits and the fuel design criteria used by the US industry today. By the mid 1980s, NRC had de-emphasized its work on reactor fuels and shifted most of those resources to severe accidents in the wake of the accident at Three Mile Island. Yet this was a time when the industry was moving into new operating regimes with fuel and developing new fuel designs. In response to a request from NRC, all five fuel vendors in the U.S. submitted licensing topical reports requesting approval to apply their safety analysis methods to high-burnup fuel. These reports were given NRC approval in 1985 and 1986 for various burnup levels up to 60 GWd/t average burnup for the peak rod. As a result of the NRC review of these reports, the regulatory burnup limit was later raised to 62 GWd/t. Meyer, *Ref. 2-1*, reported that some of the findings of those early NRC reviews in the area of the postulated accidents were as follows:

- The embrittlement criteria used for analyzing a loss-of-coolant accident (17% cladding oxidation and 2200°F peak cladding temperature) were believed to be unaffected by extended burnup operation because beginning-of-life conditions were expected to be limiting.

- The fuel enthalpy limit for reactivity accidents (280 cal/g) was said to be acceptable for extended burnup application although at that time there were only four related tests above 6 GWd/t (two around 13 GWd/t and two around 32 GWd/t).

- Cladding ballooning models used for LOCA analysis with fresh fuel were believed to be more conservative at high burnup because it was thought that cladding oxidation at extended burnup levels may result in reduced cladding strains.

- Fuel assembly structural analysis using properties for unirradiated material was thought to apply for seismic and LOCA loads at high burnup because yield strength would increase with irradiation and the corresponding decrease in cladding ductility was believed to be small or negligible. This was based on the conclusion that the ductility decreases that occur with increasing fluence will saturate around 8-12 GWd/t burnup.

- Steady-state fuel rod codes, which are used by the vendors for LOCA input (and other calculations related to normal operation) were updated to include burnup-dependent properties such as stored energy and fission gas release.

- Source terms, which are used to assess the radiological release to the environment from an accident with core melting, were not discussed.

However, results generated during the 15 years since those approvals were given indicate that the design criteria related to postulated accidents may not be conservative. Meyer, *Ref. 2-1*, gave the following examples:

- Cladding oxidation can approach the 17% LOCA limit during normal operation in this burnup range and that the associated hydride accumulation can embrittle the underlying metal. High concentrations of burnable poisons, which are needed to achieve high burnups, delay the occurrence of peak power such that the worst case is no longer at beginning of life. Therefore, oxidation and hydrogen pickup raise questions about the adequacy of the LOCA embrittlement criteria at higher burnups.

- Recent testing under reactivity accident conditions of fuel with burnups around 60 GWd/the has demonstrated that cladding failure and fuel dispersal can occur well below the 280 cal/g level, thus calling into question the fuel enthalpy criteria being used for these events.

- Changes in pellet properties at high burnup, along with the rim structure that develops, also alter the thermal performance of fuel rods. Radial power profiles are altered and centerline temperature is increased, thus increasing the important stored energy input for LOCA analysis. Changes such as the reduction in the delayed neutron fraction, which results from the buildup of plutonium isotopes, also affect neutron kinetics codes. These changes necessitated the modification of some fuel rod and kinetics codes used in licensing.

Recognizing these changes in fuel behavior at high burnup, the NRC developed an Agency Program Plan for High-Burnup Fuel in 1998 stated Meyer, *Ref. 2-1*. The program plan identified nine issues related to high-burnup fuel.

- Cladding Integrity and Fuel Design Limits
- Control Rod Insertion Problems
- Criteria and Analysis for Reactivity Accidents
- Criteria and Analysis for Loss-of-Coolant Accidents
- Criteria and Analysis for BWR Power Oscillations
- Fuel Rod & Neutronic Computer Codes for Analysis
- Source Term and Core Melt Progression
- Transportation and Dry Storage
- High Enrichments (>5%)

It was concluded that the first two issues were being satisfactorily addressed by industry activities and that the last two were related to future actions argued Meyer, *Ref. 2-1*. Attention was thus focused on the remaining five issues (Nos. 3-7) related to the postulated accidents. The regulatory criteria for reactivity accidents were found to be non-conservative in light of the test data from France and Japan. Nevertheless, it was concluded that no action was necessary pending the outcome of confirmatory research because of the low probability of the accident and because of generic calculations that implied that energy inputs would remain below the relevant test data failure levels. The regulatory criteria for loss-of-coolant accidents would be affected by enhanced cladding oxidation and related effects (hydriding), according to the program plan, but current criteria are conservative for fresh fuel and may be adequate at high burnup provided that the initial oxide accumulation is taken into account. Thus no action was thought to be needed unless the confirmatory research demonstrated a need for change. The same 280 cal/g enthalpy criterion that was used for reactivity accidents was also being used for BWR power oscillations. Based on the test results for the reactivity accidents, the conservatism in this application was also questioned. However, it was believed that the power oscillations would be slower and probably less damaging than the sharp pulses in the tests and that this did not necessarily imply unacceptable fuel damage for the BWR power oscillations. Again, it was concluded that there was no need to change the approved burnup levels unless the confirmatory research demonstrated a need for change. Meyer, *Ref. 2-1* mentioned that the attention was given to BWR power oscillations that are related to anticipated transients without scram (ATWS) rather than to the BWR rod drop accident because the perceived risk from the oscillations was greater. The need for modifications to NRC's steady-state fuel rod code (FRAPCON) was recognized earlier and this had been rectified by the time the program plan was issued. Similar changes were in progress for NRC's transient fuel rod code (FRAPTRAN), and needed modifications were also underway in Purdue University's PARCS kinetics code, which the NRC is using. The staff did not originally plan research on source terms at high burnup because they believed that it was unlikely that high burnup would have a significant effect on source terms or core melt progression. This belief was based on the facts that there would be less unoxidized metal in the core, that gap activity which might be increased is only a small fraction of a source term, that fragments from small grain sizes would not get into the atmosphere as aerosols, and that the release fractions would not be affected by the isotope shifts. Upon review, the Advisory Committee on Reactor Safeguards did not agree with the staff, and consequently the NRC decided to pursue an understanding of burnup effects on source terms in its research program. Finally, an important licensing and research strategy was described in the program plan and was stated as follows. In the past, the NRC has always performed the research needed to define regulatory criteria, and the industry has performed research to develop methods of demonstrating compliance with those criteria. In recent years, NRC's research budget has declined to a level that the NRC can no longer support such research. Thus, if the industry wants further burnup extensions, it will have to develop a data base for revised (or confirmed) regulatory criteria. The staff will make it clear to the industry that such research must be non-proprietary, to ensure that resulting criteria are fully scrutable, and the NRC staff must have full access to those research programs. If NRC resources are available, the NRC will actively participate in those research programs; however, the industry will be expected to take the lead in this work. Meyer, *Ref. 2-1*, lists the mix of home-grown and international programs that comprise NRC's research effort as follows:

- Argonne National Laboratory (NRC program): hot cell LOCA tests of fuel rods and mechanical properties of cladding
- Pacific Northwest National Laboratory (NRC program): steady-state and transient fuel rod codes and analysis
- Brookhaven National Laboratory (NRC program): neutron kinetic codes and analysis of plant transients
- Halden Reactor Project (Norway): tests of fuel rods in steady state and mild transients
- Cabri Test Reactor (France): reactivity accident tests of fuel rods and related programs
- Nuclear Safety Research Reactor (Japan): reactivity accident tests of fuel rods and related programs
- Impulse Graphite Reactor (Russia): reactivity accident tests of fuel rods and related programs
- Grenoble Research Center (France): high temperature fission product release tests

Meyer, *Ref. 2-1*, also stated that there are a large number of regulatory criteria that address normal operation, and these are the so-called specified acceptable fuel design limits or SAFDLs. These fuel design limits arise from the General Design Criteria No. 10 whose purpose is to ensure integrity of the first fission product barrier -- the fuel cladding – during normal operation, including the effects of anticipated operational occurrences. These regulatory criteria are now being left for the industry to address because of their lower risk significance. Recently, a suggestion was made by Meyer, *Ref. 2-1*, to eliminate most of the specified acceptable fuel design limits (except for those related to critical heat flux) without changing the intention of the General Design Criteria to ensure cladding integrity. The basis for this suggestion was that (a) low cladding failure rates of 1-2 rods per core are being maintained by aggressive industry action and clearly meet the intention of the regulation, and (b) few if any of the recent failure causes bear a relationship to the specified acceptable fuel design limits such that those limits are no longer contributing to this record of compliance. According to Meyer, *Ref. 2-1*, NRC has licensed two new cladding materials so far, as follows. In 1990, Westinghouse submitted a licensing topical report on their Vantage+ fuel assembly in which they introduced ZIRLO cladding. ZIRLO was approved by the NRC for burnups to 60 GWd/t about a year later. The issues that were discussed related to the ZIRLO approval were the following mentioned Meyer, *Ref. 2-1*.

- The Zircaloy embrittlement criteria used for analyzing a loss-of-coolant accident (17% cladding oxidation and 2200°F peak cladding temperature) were believed to be applicable to ZIRLO because oxidation rates are lower in ZIRLO and the same mechanism for embrittlement would occur in ZIRLO as in Zircaloy. About a year after the approval of the Westinghouse report, the regulations were amended to include ZIRLO along with Zircaloy.

- Westinghouse was using more conservative fuel enthalpy limits than the regulatory limit of 280 cal/g for reactivity accidents. Because these values (225 cal/g for unirradiated fuel and 200 cal/g for irradiated fuel) were significantly below the NRC limit, it was concluded that the ZIRLO fuel design changes would not impact the fuel enthalpy limit.

- Cladding ballooning and rupture models that are used for LOCA analysis were modified for ZIRLO based on single-rod burst tests performed by Westinghouse with unirradiated ZIRLO tubes. Beginning-of-life conditions were thought to remain limiting.

- Fuel assembly structural analysis for seismic and LOCA loads was concluded to be the same as for similar Westinghouse Zircaloy fuel. This is related to the finding that irradiation hardening and ductility of ZIRLO were similar to Zircaloy.

- The Westinghouse steady-state fuel rod code, PAD, which had already been upgraded for high-burnup operation, was further modified to account for the different creep behavior of ZIRLO. Comparisons were made with data taken from lead test assemblies to validate the code modification.

- Source terms should not be affected by the cladding alloy and were not addressed explicitly.

In 1997, Framatome Cogema Fuels submitted a licensing topical report on their cladding and structural material, M5. The NRC safety evaluation of this report and its conclusions were quite similar to those for ZIRLO according to Meyer, *Ref. 2-1*, with the following exceptions.

- The Zircaloy embrittlement criteria used for analyzing a loss-of-coolant accident (17% cladding oxidation and 2200°F peak cladding temperature) were concluded to be applicable to M5 based on quench tests with M5 cladding. Although the test specimens were unirradiated and were not subjected to ballooning deformation, the criteria were thought to be acceptable up to currently approved burnup levels (i.e., 62 GWd/t).

- The safety evaluation report noted that recent testing on the fuel enthalpy limit (280 cal/g) for reactivity accidents has indicated that fuel expulsion and fuel failure may occur before the 280 cal/g limit and the onset of departure from nucleate boiling (DNB), respectively. It was concluded that further testing and evaluation are needed and the limits may decrease in the future, but the current limits will continue to be accepted. It was also concluded that there would be little impact on the use of M5 cladding on fuel expulsion and failure (compared to the use of Zircaloy-4) as long as the cladding remains ductile.

- Fuel assembly structural analysis methodology for seismic and LOCA loads was unchanged, but would require strength values for M5, should M5 be used for guide tubes or thimble tubes.

Doesburg, Ref. 2-2, reported on the formation of the OECD/CSNI/PWG2 Task Force on Fuel Safety Criteria, TFFSC. The objective of this group consisting of staff from different regulatory agencies from US and western Europe are to assess if the development of new fuel and core designs, and the replacement of statistical instead of deterministic analysis methods have decreased the fuel safety margines. The different safety related criteria being reviewed are listed in *Table 2-1*.

Table 2-1: ***Fuel safety criteria, and task force review basis, the numbers designate the following: I – normal operation, II – anticipated operational occurrences, III and IV – accidents, Ref. 2-1.***

Safety Related Criteria	**"New" Elements Affecting Criteria**										
	New Fuel Design	*New Core Design*	*New Cladding Materials*	*New Manu Methods*	*Longer Fuel Cycles*	*Uprated Power*	*High Burnup*	*MOX*	*Mixed Core*	*Water Chem Changes*	*Current/New Operating Pract*
CPR/DNBR	I-IV	I-IV			I-IV	I-IV	I-IV		I-IV		
Reactivity Coefficient		II-IV			II-IV	II-IV	II-IV	II-IV	II-IV		
Shutdown Margin	I-IV	I-IV			I-IV	I-IV	I-IV	I-IV			I-IV
Enrichment	I-IV	I-IV			I-IV						
CRUD deposition	I	I	I	I	I		I			I	
Strain Level	I-II						I-II	I-II			
Oxidation			I-IV	I-IV			I-IV	I-IV		I-IV	
Hydride Concentration			I-IV	I-IV			I-IV	I-IV		I-IV	
Internal Gas Pressure	I-IV				I-IV	I-IV	I-IV	I-IV			
PCMI	I-II		I-II	I-II			I-II				
PCI	I-IV	I-IV	I-IV	I-IV		I-IV	I-IV	I-IV			I-IV
Fragmentation (RIA)							III-IV	III-IV			
Fuel Failue (RIA)	III-IV		III-IV	III-IV			III-IV	III-IV			
Peak Cladding Temperature			III-IV	III-IV			III-IV	III-IV			
Cladding Embrittlement/Oxidation			III-IV	III-IV			III-IV	III-IV			
Blowdown/Seismic Load			III-IV				III-IV				
Assembly Holddown Force	I-IV										I-IV
Coolant Activity					I-IV	I-IV	I-IV	I-IV			
Gap Activity					III-IV	III-IV	III-IV	III-IV			
Source Term					III-IV	III-IV	III-IV	III-IV			

Chatterton, Ref. 2-3discussed the requirements to be met to increase the burnup levels beyond those approved today and focused on a series of fuel issues related to normal operation (class I) and anticipated operational occurrences (class II) that were reported during the last years, as follows:

- oxidation levels higher than predicted,
- excessive internal gas pressure in burnable poison rods,
- incomplete control rod insertion (IRI) events,
- large axial offsets or axial offset anomalies (AOA),
- fuel failures due to high fuel duty,
- adverse effects of water chemistry,
- high crud buildup, and
- accelerated growth of rods and assemblies.

All of these problems have been at least partially associated with high burnup fuel or aggressive fuel duty according to Chatterton. To date, these events have not raised safety questions serious enough to require regulatory action such as a restriction of a burnup limit. However, to allow a burnup extension from that existing today in US of 62 GWd/T rod average the industry must address all of the above-mentioned issues argues Chatterton. The nuclear industry in the United States has already indicated that it would like to increase the peak rod average of 75 GWd/T for pressurized-water reactors (PWRs) and 70GWd/T for boiling-water reactors (BWRs) and that this will be done with an enrichment of 5 weight percent U-235 or less, Chatterton.

The author further state that to allow a burnup extension the following is required:

- A prototypical lead test assembly (LTA) program up to the proposed limit addressing all points in the current licensing basis (Standard Review Plan (NUREG-0800), fuel design criteria, and General Design Criteria(GDC)). In the past, the NRC has restricted the LTAs to non-limiting locations. This has resulted in burnup histories that were not aggressive and in many cases not typical. Fuel performance in the last few years has led NRC to conclude that LTAs must be prototypical in order to be of the maximum value. Because of the large number of variables that affect fuel behavior, it may be necessary to have a sizeable number of LTAs in order to cover the range of operation expected by a large number of plants. Alternatively, if only a small amount of LTA data is available, operation may have to be restricted to those conditions that closely duplicate operation of the LTAs. The LTAs should further be well characterized before irradiation.

- A risk-informed approach. Risk-informed analysis considers both the probability and the consequences of the event.

- Addressing RIA, loss-of coolant accidents (LOCAs) and anticipated transients without scram (ATWS); and

- A fuel performance monitoring program that includes collection of poolside data such as:

    - Oxide and Crud Thickness for Clad and Spacer Grid and Guide Tube
    - Cladding diameter
    - Fuel Rod and Assembly Growth
    - Fuel Rod Bow
    - Guide Tube Distortion
    - Fuel Assembly Bow

- Hotcell examination data including:

    - Rod Internal Pressure
    - Clad Ductility
    - Oxide thickness
    - Hydride Content
    - Fuel Microstructure

The data from the LTA program would be needed as input for analysis of all types of operation (normal operation, transients and accidents) and the methodologies and codes used in the analysis must be examined to verify that they are appropriate for higher burnups, argues Chatterton.

2.2 UTILITY PERSPECTIVE

West and Gautier,Ref. 2-4 stated that one of the major present challenges to nuclear energy lies in its competitiveness. To stay competitive the industry needs to reduce maintenance and fuel cycle costs, while enhancing safety features. To meet these objectives, EDF is looking into the possibilities to:

- Increase the burnup to 60 to 70 MWd/kg from the present 52 GWd/t licensing limit (max. fuel assembly),
- To use high duty fuel management schemes, and
- Achieve "Parity" in terms of fuel performances, between enriched UO_2 and MOX fuels.
- Achieve clean cores (absence of defects),
- Increase operating flexibility (load following) and
- Continuously improve operating margins.

West and Gautier, argue that the current issues for improving fuel performances are:

- Cladding corrosion. The current cladding material in Zircaloy 4 reaches the limit of 100 µm beyond which there is a risk of spalling, for burnups in the range of 55-60 GWd/t and for the current normal reactor operating conditions. To be able to use new more aggressive fuel management schemes (18 months cycles, low leakage loading patterns), new alloys with better corrosion resistance are needed.
- Rod internal pressure. The current limit based on the non re-opening of the pellet cladding gap is practically reached for the licensed burnup of 52 GWd/t (if the uncertainty and the potential effect of load following is included). Two routes are pursued to resolve this issue: (i) development of new microstructures able to better retain fission gas in the matrix and, (ii) relaxation of the non re-opening of the pellet cladding gap criterion. Also, increasing the initial void volume could be used to resolve this issue but this would of course reduce the amount of energy coming from the fuel rod.
- Assembly bow and incomplete rod insertion. For the present cores and fuel assemblies, the gap between the fuel assembly and the core plates is still sufficient to accommodate the fuel assembly growth at discharge burnup, but the margins are very close to zero. It is the same situation for the gap between the fuel rods and the fuel assembly nozzles. New guide tube and fuel cladding materials with lower growth rates may resolve this issue,
- fretting and

- PCI limitations. The licensing requirement in France states that no fuel rod failures during a class 2 condition, such as an uncontrolled withdrawal of a control rod may occur. During such a situation PCI failures may occur. More aggressive fuel management schemes may increase the risk for PCI failures also ramp tests of high burnup fuel needs to be done to assess the PCI performance at higher burnups.

According to West and Gautier, also licensing aspects become more and more crucial and have to cover a wider range of accidental situations, such as: RIA and LOCA. This is presently a big issue in France since current fuel had some difficulties to satisfy the criteria for the reactivity initiated accidents. The REP Na tests in CABRI showed that a fuel cladding with excessive corrosion and hydriding would fail at enthalpies relatively close to those which could be encountered in a reactor during an rod ejection accident. That is why to go beyond the current 52 GWd/t, there is a need to get a more corrosion/hydriding resistant cladding argues West and Gautier, Ref. 2-4. In addition EDF focus on the importance of validation and licensing issues for modelling codes and methodologies. Finally, the back end of the fuel cycle also has to be taken into consideration especially with a view to intermediate storage.

Also in US, recent trends are for increased burnup and cycle lengths for both PWRs and BWRs, Figure 2-5 stated Yang et al. Ref. 2-5. Cycle lengths of 18 months are now common for BWRs and PWRs; 24-month cycles have been implemented by some BWRs as well as some PWRs. EPRI is now co-operating with NRC and ANL by providing fuel for the test program to assess appropriate fuel design criteria related to design basis accidents, see section 2.1. There is also co-operation in the planning of the tests. Therefore, BWR fuel from Limerick with 57MWd/kgU will be shipped to ANL for those tests. There are also plans to provide high burnup PWR fuel from H.B.Robinson at 72MWd/kgU.

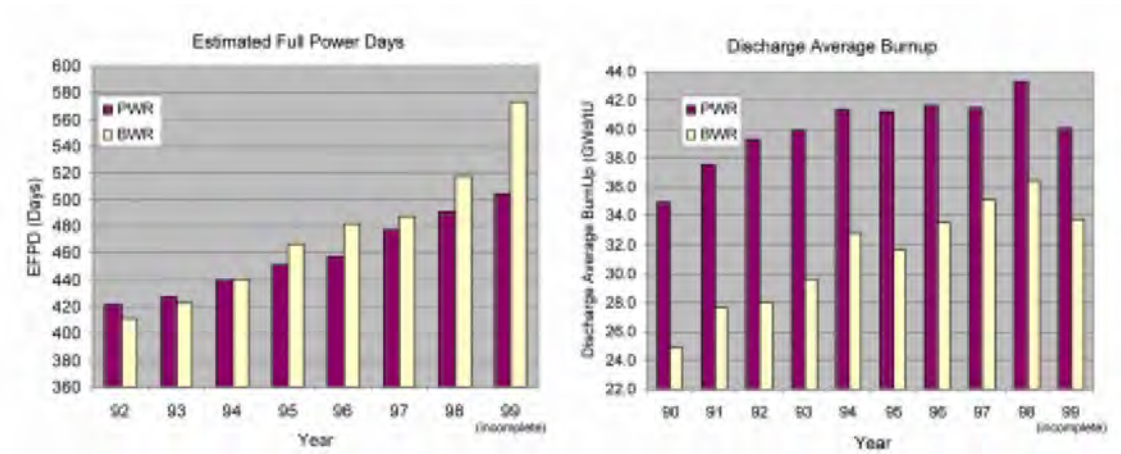


Figure 2-1: Trends in burnup and cycle lengths, Ref. 2-5

2.3 VENDOR DEVELOPMENT

2.3.1 Siemens

Seibold et al., Ref. 2-6 reported on Siemens PWR and BWR material development programs. Severe fuel duty, connected with improved core designs, can lead to local steam formation and might result in new phenomena such as H-induced accelerated corrosion, Li-corrosion, and O-induced nodular corrosion, Figure 2-2. This situation calls for more corrosion resistant materials and the Siemens PWR program has yielded the materials shown in *Table 2-2*.

Table 2-2: Material development for PWR fuel assemblies **Ref. 2-6.**

Boundary Condition	Cladding (year of introduction of full reloads)	Structural Parts
Meet ASTM specification*)	**Optimization of Zry-4**	
	Low-Sn Zry-4 (1987) Optimized Zry-4 (1989)	Low-Sn Zry-4 (1989)
In total, meet ASTM specification*)	**Implementation of the duplex concept (DX) for cladding**	
	Duplex Extra Low-Sn (0.5-0.8%) /2/ (DX ELS, DX D4) (1989/90)	
Keep close to ASTM specification*)	**Insertion of an advanced Zry-type alloy**	
	Modified Zry-4 (1995)	Modified Zry-4 (1997)
High burnup, severe fuel duty, alloy composition not limited to ASTM specification	**Development of high performance alloys (since 1985)**	
	Zr1Nb, DX Zr2.5Nb Zr0.6SnFeV (HPA-4, DX and throughwall)	Zr0.6SnFeV(HPA-4)

*) licensing reasons

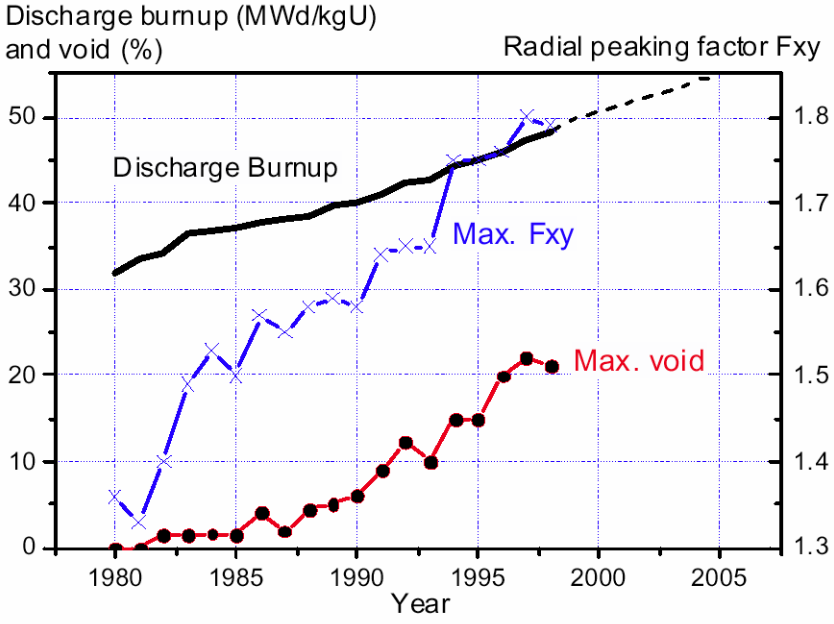


Figure 2-2. Evolution of operating conditions (discharge burnup, local power, void) for Siemens PWR fuel assemblies, Ref. 2-6.

Garzarolli et al, Ref. 2-7 reported on the burnup experience on the PWR DUPLEX alloys. DUPLEX cladding, consisting of a Zircaloy-4 tube with a thin outer layer of an ELS (extra low Sn) alloys. The first commercial DUPLEX ELS material used for reloads was DX ELS 0.8a inserted in 1991 in PWR A, Ref. 2-7This cladding material had a Sn content of 0.8%, the other alloying not very different from Zry-4. The reduction in Sn reduced corrosion significantly. However, low Sn alloys with 0.3% Fe+Cr (as Zry-4) showed an increased sensitivity to Li. An increase of the Fe+Cr content above 0.3% over-compensated the Li sensitivity and further improved the in-reactor corrosion resistance. When the effect of Fe+Cr was fully understood, a new DUPLEX ELS cladding material with an enhanced Fe+Cr content (0.45%) was specified (DX ELS 0.8b) and used for reloads from 1992 onward according to Garzarolli et al., Ref. 2-7. In parallel, the development of a DX ELS material with a further reduced Sn content and a further enhanced Fe+Cr content (DX D4) was pursued. Operating experience with DX D4 demonstrated that this alloy with 0.5% Sn and 0.7% Fe+Cr had an even higher burnup capability and therefore contributed to higher fuel cycle efficiency than DX ELS 0.8b, even at quite high heat fluxes.

Table 2-3: ***Irradiation of DUPLEX ELS reloads,*** **Ref. 2-7.**

PWR	First use	Material	Total no. fuel rods	Max FA burnup MWd/kgU	No. of rods PIE pool	PIE HC
A(15*15)	1991	DX ELS0.8a	14,437	56	153	1
B(16*16)	1992	DX ELS0.8a	13,452	60	12	
D(16*16)	1993	DX ELS0.8a	3,776	55	0	
A(15*15)	1992	DX ELS0.8b	56,440	59	376	13
B(16*16)	1994	DX ELS0.8b	46,256	49		
C(16*16)	1995	DX ELS0.8b	30,208	47	70	
D(16*16)	1993	DX ELS0.8b	50,964	56		
E(16*16)	1993	DX ELS0.8b	55,224	41	10	
F(16*16)	1994	DX ELS0.8b	45,312	51	?	
G(18*18)	1993	DX ELS0.8b	94,800	48	87	
H(18*18)	1996	DX ELS0.8b	34,800	47	30	
I(18*18)	1992	DX ELS0.8b	85,879	53	181	3
J(17*17)	1998	DX ELS0.8b	716	24	50	
L(14*14)	1998	DX ELS0.8b	448	7		
H(18*18)	1992	DX D4	18,000	41	12	
I(18*18)	1992	DX D4	2,400	52	48	4
M(17*17)	1994	DX D4	1,056	52	265	

Seibold et al. Ref. 2-6 also reported on the Siemens BWR development program, *Table 2-4.* For high burn-up applications the intermetallic particle size was increased in the Zircaloy PGP+ variant compared to that of Zry-2 LTP. The larger particles in the Zry PGP+ variant will compensate for particle dissolution tendency by irradiation. Also a Zr1SnNb alloy without any intermetallic particles have been tested and may be used for very high burnup levels.

***Table 2-4: Material development for BWR fuel assemblies,* Ref. 2-4.**

Objective	Measure	Introduction
Avoid severe nodular corrosion	ß -quenching	early 70s
Avoid PCI	Zr-liner cladding	1985
Minimize nodular and late accelerated uniform corrosion	Low Temperature Processed (LTP) cladding and sheet material with optimized A-parameter	1987
Avoid PCI and long splits in defective rods	Fe-enhanced Zr-liner cladding	1993
Provide PCI resistant throughwall cladding with improved corrosion and hydrogen pickup behavior	Fine grain size, highly texturized throughwall Zry-type-cladding (Fe, Cr, outside ASTM), alloy used also for structural parts (Zry-BWR)	1995*
Provide resistance against late accelerated uniform corrosion at high burnup	Particle size adjusted to high burnup needs for structural materials and cladding (Zry PGP+)	1998
High burnup, high strength alloy for thin-walled parts with irradiation resistant microstructure	Zr1SnNb for BWR application	1999*

* lead fuel assemblies, pathfinder rods

Urban et al., Ref. 2-8reported on the development of the BWR ATRIUM 10 fuel design, Figure 2-3. The ULTRAFLOW spacer was introduced along with eight part length fuel rods and a small hole design lower tie plate. As of August 1999, more than 2000 ATRIUM 10 fuel assemblies have been inserted in 17 different boiling water reactors in Europe, the United States and the Far East, according to Urban et al., Ref. 2-8. The maximum fuel assembly average discharge burnup is now 59 MWd/kgU and may increase to 63 MWd/kgU in early 2000. The FUELGUARD lower tie plate is now introduced with increasing frequency in ATRIUM 10 reloads to reduce the risk of debris fretting failures. Lead assemblies for the demonstration of high burnup fuel and cladding properties have been inserted in 1999. The enrichment of these lead assemblies amounts to 4.6 w/o U-235 in the enriched axial part and thus is close to the maximum achievable as a lattice average with the present enrichment limit of 4.95 w/o U-235. A special version, ATRIUM 10P, was developed with about 15 % lower pressure drop by applying e.g. Inconel spacers, larger inside channel width and increased number of part length fuel rods, Figure 2-4. Lead assemblies of ATRIUM 10P have been inserted in a German BWR in 1996. An "ATRIUM 10C" fuel assembly design is being developed to increase MCPR margin and thermal hydraulic stability by optimizing number and axial positions of spacers as well as number and radial position of part length fuel rods.

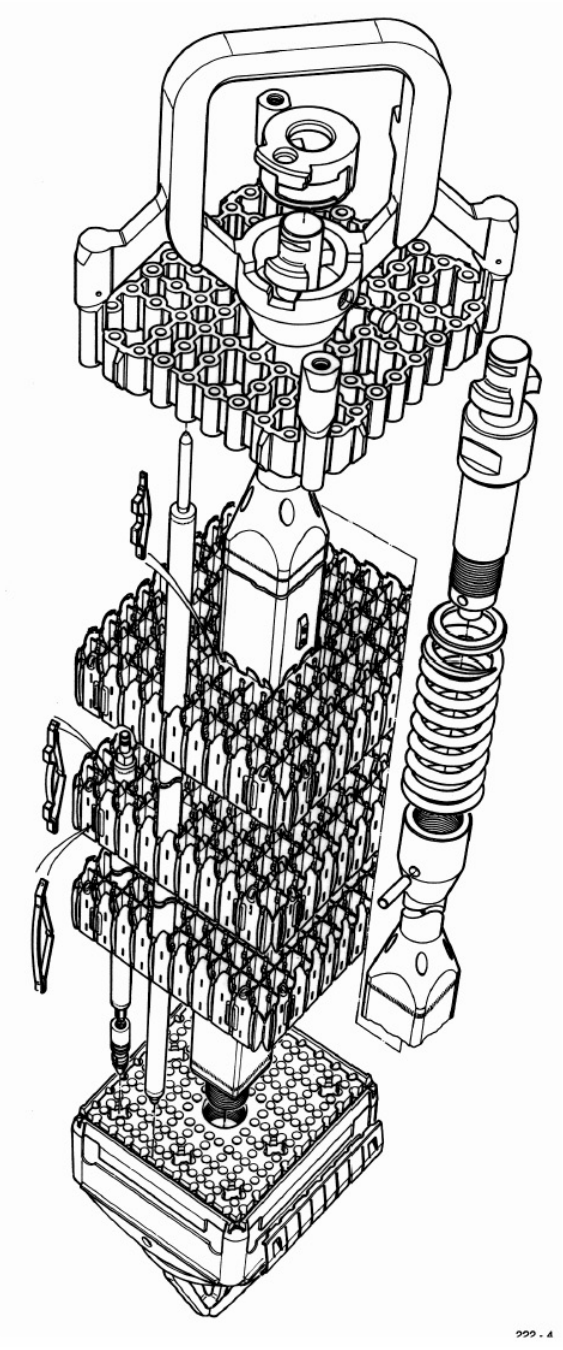


Figure 2-3: The ATRIUM 10 fuel assembly, Ref. 2-8

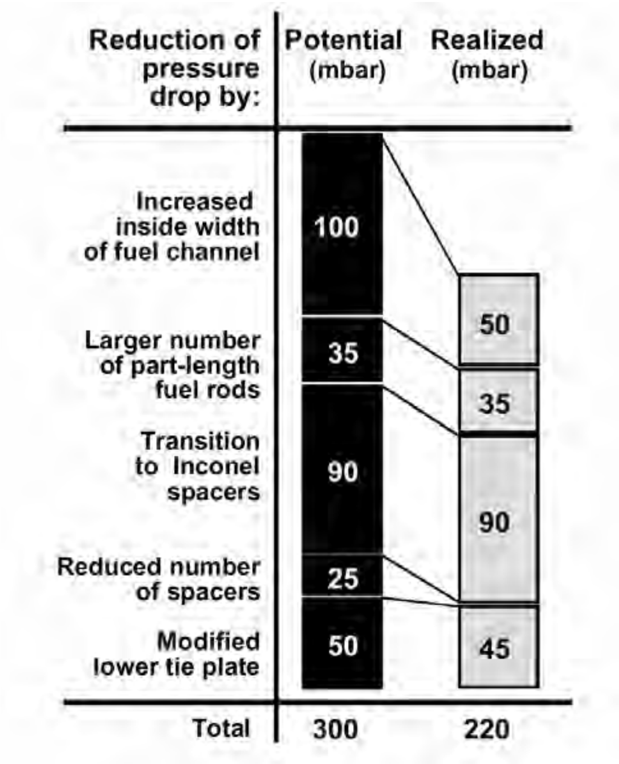


Figure 2-4: Comparison between the identified potential and the realized pressure drop decrease of the ATRIUM 10P design, Ref. 2-8

2.3.2 Mitsubishi

Up to Oct. 1999, more than 13000 fuel assemblies with Zry-4 fuel cladding material were loaded into 23 PWR plants in Japan according to Doi et al. Ref. 2-9. As a first step, high burnup fuel with a 4wt % enrichment and a maximum discharged assembly burnup limit of 48GWd/t has been utilized since 1990. The highest achieved discharged assembly burnup so far is 47GWd/the, Figure 2-5. To reduce the number of spent fuel assemblies and to improve fuel management, Mitsubishi has developed the advanced fuel cladding material *MDA* intended for a discharged assembly burnup of 55GWd/t in collaboration with Japanese PWR utilities, *Table 2-5.*

3 ZIRCONIUM ALLOY MANUFACTURING AND ALLOY SYSTEMS

3.1 INTRODUCTION

The development of manufacturing of the zirconium alloys has, during the last years, been characterised by decreased variation in process parameters, to cope with higher demands on the materials; increase efficiency, in order to reduce manufacturing costs; and introduction of new materials, again to comply with higher demands on the cladding.

In all these cases, it is important to be aware of the possible influence of the changes on materials properties not in focus for the change of the process or the new alloy introduced. For instance, a change to improve the corrosion properties by alloying with niobium may change the mechanical properties during power ramp, i. e. that the same PCI rules should not apply.

3.2 NEW ZIRCONIUM ALLOY PHASE INFORMATION

The zirconium is always in the α phase at operating temperatures. At elevated temperatures, the zirconium will transform to a β phase. The β phase has generally much higher alloying element solubility and a also a different crystal structure (bcc, vs. hcp in the α phase). The transformation by heat treatment of zirconium alloys to the β phase and a following quick cooling, hence "β quenching", during manufacturing at least once in a process route, is very important in order to obtain a well-defined and reproducible material for cladding or other zirconium alloy components. A new study has been made in order to determine the transition temperature from the α phase to the range of the mixed phases $\alpha + \beta$, as well as the transition temperature for $\alpha + \beta \rightarrow \beta$ for Zr-Nb-Sn-Fe materials, such as the Zirlo and E635 ("Anikkuloy") materials. The important finding by Canay et al [Ref. 3-1] is that the tin concentration, as well as the iron concentration, will have an effect on the transition temperatures for $\alpha \rightarrow \alpha + \beta$ as well as the $\alpha + \beta \rightarrow \beta$ transitions, as shown in Figure 3-1 and Figure 3-2, respectively,

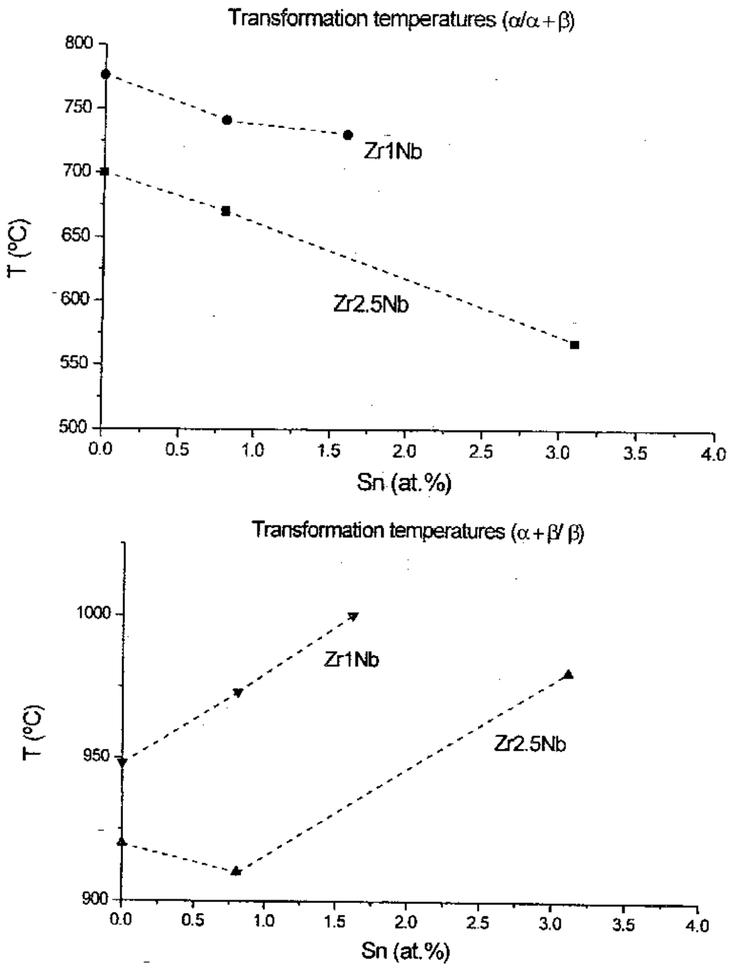


Figure 3-1. The $\alpha \rightarrow \alpha + \beta$ (upper) and the $\alpha + \beta \rightarrow \beta$ (lower) transitions temperature for Zr-1Nb-Sn and Zr-2.5Nb-Sn alloys as a function of varied tin contents [Ref. 3-1]

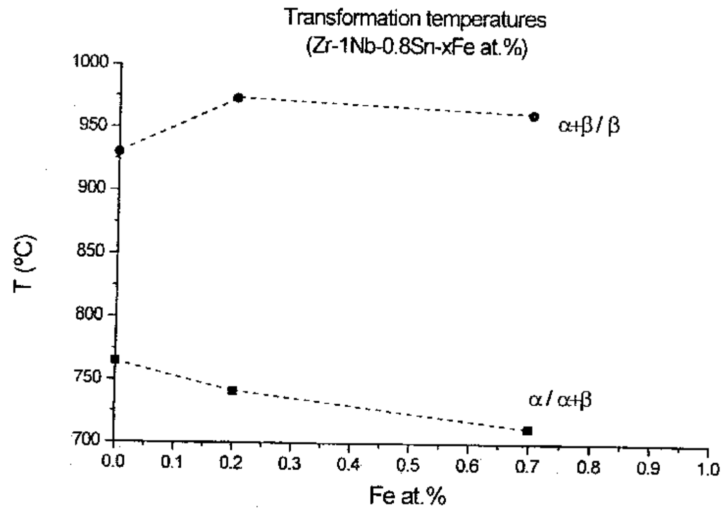


Figure 3-2. The α → α + β (lower curve) and the α + β → β (upper curve) transition temperatures for Zr-1Nb-0.8Sn-Fe alloys as a function of varied iron contents [Ref. 3-1]

The implication of this is that variation of the tin or iron content will change the allowable ranges during heat treatments in the zirconium alloy components manufacturing. Hence, a proven successful process route for a Zr-1Nb-0.8Sn-0.1Fe alloy (like Zirlo) could be much less successful if the tin, or iron, concentration is changed 10 - 20%.

Some new information related to the high temperature behaviour has been reported on the Zircaloy-4 - oxygen thermodynamic behaviour. By applying a pyrometer-based differential thermal analysis method, lower solidus-to-liquidus phase transition temperatures for Zircaloy has been established [Ref. 3-2]. For instance, the new value for β Zry ↔ liquidus transition was found to be 2035±20 K at 0.74 at.% oxygen, as opposed to 2128 K for the corresponding transition for Zr. Normally, Zr transition data have been used in accident simulations due to lack of appropriate Zry data.

3.3 NEW ALLOYS

3.3.1 Burn-ups Reached with New Alloys

Several new alloys have been introduced and have been tested during the 1990's. By far, there are more PWR than BWR alloys developed, which is explained by the fact that the common Zircaloy-4, even in the low tin variant, will generally not be sufficiently good for the burn-ups utilities today are aiming for. The same situation has yet not been reached for Zircaloy-2 in the BWRs. In order to provide a more structured information, data from in-pile exposure of these alloys that has been presented this year have been compiled in Table 3-1.

4 MECHANICAL PROPERTIES

4.1 INTRODUCTION

The mechanical properties of essentially two different components are treated in this section. Firstly, it is the *LWR* fuel assembly and, secondly, it is *Pressure tubes*, e.g., in *CANDU* reactors. The difference between these two components is that the fuel is reloaded after some time in-reactor while the *Pressure tube* is a part of the reactor design and must consequently performance satisfactory during the lifetime of the reactor.

The mechanical properties of the fuel assembly are crucial for its satisfactory performance in-reactor. *Standard Review Plan, SRP,* section 4.2, lists different mechanical failure modes of the *LWR* fuel components and also the corresponding design criterion to ensure that the fuel assembly behaviour is satisfactory. These design criteria are set to ensure that:

- the fuel assembly will not *fail* during normal operation (class I) and anticipated operational occurrences (class II). *Failing* in this sense has a broader meaning, namely that the fuel rod may not be breached and that the dimensional changes of the assembly during irradiation must be limited. The latter requirement is to ensure that control rods can be inserted and that the fuel can be handled during shutdown. Also the BWR fuel outer channel cross section must not have increased to such an extent that it is impossible to pass it through the upper core grid during reloading.

- the fuel remains coolable during an accident (class III and IV). Class IV design basis accident are *LOCA*, *RIA* and earthquake. During class III and IV situations limited fuel failures are however accepted. Another criterion that must be fulfilled in these situations is that it should be possible to insert the control rods.

- During class I and II operation, the following mechanical *failure* mechanisms and corresponding design criterion for the fuel assembly, including its components, are listed in *SRP* section 4.2:

- Plastic deformation – the component is regarded as *failed* if it is plastically deformed and the appropriate criterion is that the stresses must be lower than the yield stress. *SRP* section 4.2 also state what type of methodology should be used when calculating these stresses. In these calculations the stress in the assembly location subjected to maximum stresses is calculated. In calculating this stress, all types of stresses are taking into account, such as welding residual stress, thermal stress, stress imposed by rod-system differential pressure, etc. It is interesting to note that the criterion on maximum allowable oxide thickness on fuel rods is related to this criterion. If the oxide thickness becomes too large in a *PWR*, the oxide thickness will increase the cladding temperature due to its lower thermal conductivity and would then increase corrosion rate. The oxide thickness would increase further, raising the clad temperature and corrosion rate, resulting in thermal feedback. Since increasing temperature decreases the yield strength of the material, the material would eventually mechanical fail, i.e. plastically deform, provided that the cladding stresses are large enough.

- Excessive creep deformation that could either result in creep fracture or too large plastic deformations that could e.g. lead to *dryout* due to excessive outward creep of the fuel cladding diameter that would limit coolant flow. Creep occurs at a stress level lower than the yield stress. The corresponding criterion is very general and just specify that the creep deformation must be limited.

- Fatigue failure – Most fuel assembly components are subjected to fatigue stresses and *SRP* section 4.2 provides the maximum allowable fatigue stress level.

- *PCI* – The criterion to eliminate this type of failure is by limiting the elastic and uniform plastic deformation in the cladding circumference during a class I and II transient to 1%. This value is of course not sufficiently to ensure that *PCI* failures do not occur. However, the fuel vendors are still designing their fuel so this 1 % limit is achieved in their design.

- Hydride embrittlement – The criterion just mentions that the hydrogen content in the material must be limited so the fuel assembly component will not fail.

During accident conditions such as *LOCA* and *RIA*, the mechanical performance of the fuel cladding is crucial to meet the objective that the fuel must remain coolable during these types of accidents. In both situations, it is important that the fuel cladding may not fail in a brittle fashion during the *reflooding*[1] phase during *LOCA* and due to *PCMI* during the *RIA* transient.

Fuel vendors have developed codes to model the fuel assembly mechanical performance during class I, II, III and IV situations and to be able to do this modelling correctly, data on mechanical performance of the fuel assembly must exist. The data are generated in two types of tests, either separate effect tests or integral tests. The former test studies only the impact of one parameter at a time on the mechanical performance. This could e.g., be the impact of hydrogen content on ductility. The data from these separate effect tests are then used by the fuel vendor to develop adequate models in their fuel performance codes. To then verify that the code comes up with the correct prediction on fuel assembly mechanical performance e.g. during a *LOCA*, the code predictions are benchmarked towards integral tests. In the integral test, the fuel assembly design and environment is as similar as possible as is existing in the situation that is simulated in the test, e.g. a *LOCA*.

In this report, most mechanical performance data from either separate effect or integral tests are presented in the appropriate section, e.g., mechanical tests to study *PCI* performance is presented in the section that treats *PCI*. However, the mechanical test data that are not treated elsewhere in this report is provided in this section in the following.

[1] This is the last phase during a *LOCA* situation when the core is reflooded with water that cools the fuel cladding surface imposing very large thermal stresses that may fracture the fuel cladding

4.2 NEW RESULTS

Ishimoto et al., Ref. 4-5 reported on some new alloys being irradiated in the Japanese Kashiwazaki-Kariwa-5 commercial reactor, *Table 4-1*. Figure 4-1 shows that UTSs of all the alloys showed a saturation tendency up to the fluence of 10^{26} n/m^2. UTSs of High Fe/Zry-2 and High FeNi/Zry-2 were slightly higher than that of Zry-2 at all the temperatures and fluences. Elongations of High Fe/Zry-2, High FeNi/Zry-2 and Zry-2s are shown in Figure 4-2shows that the Elongation of all the alloys also showed a saturation tendency. Elongations of High Fe/Zry-2 and High FeNi/Zry-2 were almost the same as that of Zry-2s at 300 K, but were slightly larger than that of Zry-2s at 561 K and 616 K. Vickers micro-hardnesses of High Fe/Zry-2 and High FeNi/Zry-2 also showed a saturation tendency and the values were slightly higher than that of the Zry-2s, Figure 4-3. Creep strains increased linearly with irradiation time, Figure 4-4. High Fe/Zry-2 and High FeNi/Zry-2 showed lower creep strains than Zry-2 after two cycles. The cycles to fatigue failure increased by irradiation for all the alloys, Figure 4-5. There was no significant difference in the cycles to failure between High Fe/Zry-2, High FeNi/Zry-2 and Zry-2.

***Table 4-1: Composition of different alloys*, Ishimoto et al. Ref. 4-5.**

Alloy	Element (wt%)						
	Sn	Fe	Cr	Ni	Nb	Mo	Bi
TSHT/Zry-2	1.61	0.14	0.11	0.06	-	-	-
TSHT/CC/Zry-2	1.31	0.18	0.10	0.07	-	-	-
IPHT/CC/Zry-2	1.31	0.20	0.10	0.07	-	-	-
High Fe/Zry-2	1.46	0.26	0.10	0.05	-	-	-
High FeNi/Zry-2	1.43	0.26	0.10	0.10	-	-	-
0.5Nb/Zry-2	1.34	0.16	0.09	0.05	0.50	-	-
XLL	1.24	-	-	-	0.30	0.29	-
BAG	-	-	-	-	0.49	-	1.03

5 DIMENSIONAL INSTABILITY

5.1 BACKGROUND

One of the most unique aspects of material behavior in a nuclear power plant is the effect of radiation (mainly neutrons) on the dimensional stability of the reactor components. In fast breeder reactors the Fe and Ni-based alloys creep and swell, that is, they change dimensions in response to a stress and change their volume in response to radiation damage. In light water reactors, zirconium alloy structural components creep, do not swell, but do change their dimensions through the well-known constant volume process called irradiation growth. Radiation effects are not unexpected since during the lifetime of a typical component every atom is displaced from its normal lattice position at least 20 times! With the possible exception of elastic properties like Young's Modulus, the properties needed for reliable fuel assembly performance are affected by irradiation. A straightforward summary of such effects is given in Ref. **5-1**.

Practical effects of dimensional instabilities are well known and it is a rare technical conference in the reactor performance field that does not include discussions on the topic. Because of the difference in pressure inside and outside the fuel rod, cladding creeps down on the fuel early in life, and then creeps out again later in life as the fuel begins to swell. A major issue is to have creep strength sufficient to resist outward movement of the cladding if fission gas pressure becomes high at high burnups. PWR guide tubes can creep downward or laterally due to forces imposed by fuel assembly hold down forces or cross flow hydraulic forces – both leading to assembly bow which can interfere with smooth control rod motion. BWR channels can creep out or budge in response to differential water pressures across the channel wall, again leading toward control blade interference. Fuel rods, water rods or boxes, guide tubes, and tie rods can lengthen, potentially leading to bowing problems. (For calibration, a recrystallized (RX) Zircaloy water rod or guide tube could lengthen due to irradiation growth more than 2 cm. during service; a cold worked/stress relieved (SRA) component could lengthen more than 6 cm.) Even RX spacer/grids could widen enough due to irradiation growth (if texture or heat treatment was not optimized) to cause uncomfortable interference with the channel.

In addition, corrosion leading to hydrogen absorption in Zircaloy can contribute to component dimensional instability due to the fact that the volume of zirconium hydride is 16% larger than zirconium.

The above discussion leads to the concept that understanding the mechanisms of dimensional instability in the aggressive environment of the nuclear core is important for more than just academic reasons. Reliability of materials and structure performance can depend on such understanding.

5.1.1 Irradiation Growth

Irradiation growth is a change in dimensions of a reactor component even though the applied stress is nominally zero. It is roughly a constant volume process, so if there is an increase in, say, length of a component, the width or thickness must decrease to maintain constant volume. The detailed mechanism of growth is still uncertain even after 35 years of observation. However, it is due in some way to the aniosotropic flow of irradiation-induced vacancy and interstitials (or complexes) to sinks in the Zr matrix like grain boundaries and irradiation induced dislocations and dislocation loops. The inherent anisotroy of the Zr crystal structure plays a strong role in the mechanism, as materials with an isotropic crystal structure (like stainless steel, copper, Inconel, etc.) do not undergo irradiation growth. Several factors that influence irradiation growth are reviewed here.

5.1.1.1 Fluence and Microstructure

Growth increases with fluence. The early experiments determined that growth of cold worked (CW or CWSR) Zircaloy appeared linear with fluence starting in the first cycle of irradiation but that growth of crystallized (RX) Zircaloy appeared to level off to a low rate at low fluences. It is now certain that both RX and CW materials have a similar high growth rate at high fluences. Figure 5-1 indicates the general shape of the growth vs. fluence curves. In practical terms, the growth rate of RX material becomes high above a fluence of about $5x10^{21}$ n/cm2 (E>1MeV), or about 25 MWd/kgU burnup). The growth rate of cold worked material is roughly linear with fluence from startup and the rate depends on the amount of cold work experienced after the last recrystallization anneal. For Zircaloy tubes the last cold work step gives generally around 70% deformation, but smaller amounts of cold work can be introduced in nominally RX material by tube straightening operations, by channel sizing operations, or by a number of possibilities for handling damage. Even small amounts of residual cold work will influence local irradiation growth, so understanding the sources and consequences of such is important. In microstructure terms, the source of high growth rates appears to be the presence of a particular type of dislocation called a <c> type or c-dislocation. In cold worked material c-dislocations are present in the as-fabricated microstructure, but in RX material they are not. However, irradiation produces c-dislocations in RX Zircaloy (by an admittedly mysterious process) starting at about $4x10^{21}$ n/cm^2, the same fluence at which the growth rate starts to accelerate. In CW Zircaloy the c-dislocations multiply with increasing irradiation.

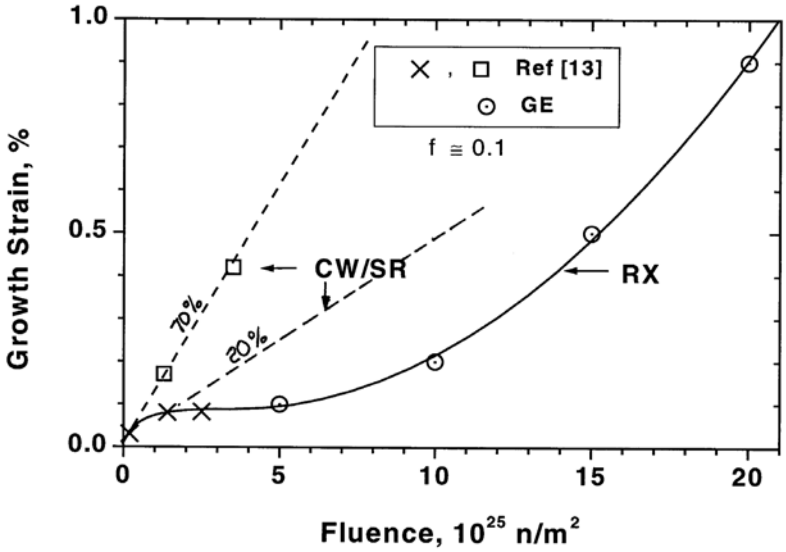


Figure 5-1 **Irradiation growth of Zry-2 at about 565 K, Ref. 5-1**

The above discussion applies to Zircaloy-type alloys and to Zr2.5Nb. For some of the other Nb-containing alloys, like Zr1Nb or Zr1Sn1NbXFe, there is evidence that c-dislocations do not form during irradiation at PWR-type temperatures. This gives good promise that irradiation growth of such alloys will maintain a low rate to high fluences.

5.1.1.2 Texture

The unit cell for zirconium is shown in Figure 5-2. As opposed to most of the metals or alloys that we deal with, the cell is anisotropic; i.e., the dimensions of the cell are larger in the c-directions than in the a-directions.

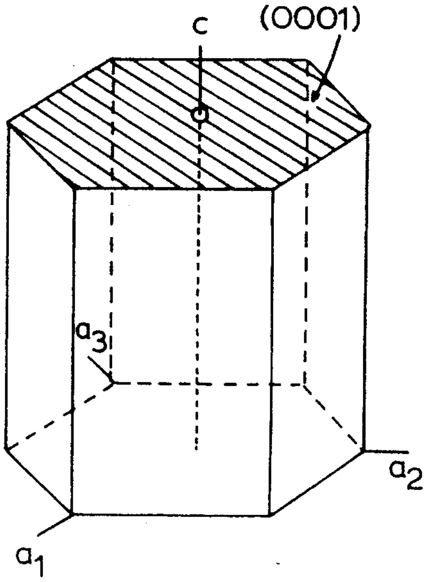


Figure 5-2: The Zirconium HCP unit cell. The basal plane is shaded

6 CORROSION AND HYDROGEN UPTAKE

6.1 INTRODUCTION

The oxidation kinetics of zirconium and its alloys were measured in the early days [*Ref. 6-1*] in a number of oxidizing gaseous and aqueous environments. In nearly all these environments the kinetics were very similar [*Ref. 6-2*] and consisted of two basic periods. The initial period (pre-transition) where the oxidation rate decreased approximately parabolically with time at high temperatures, but was close to a cubic (or even quartic) kinetics at temperatures in the water reactor range (250-350°C). When the oxide reached a thickness of about 2 μm a transition to an approximately linear kinetic rate (post-transition) occurs. This post-transition period may start off with a series of cycles (Figure 6-1). The pre-transition kinetics are regarded as being

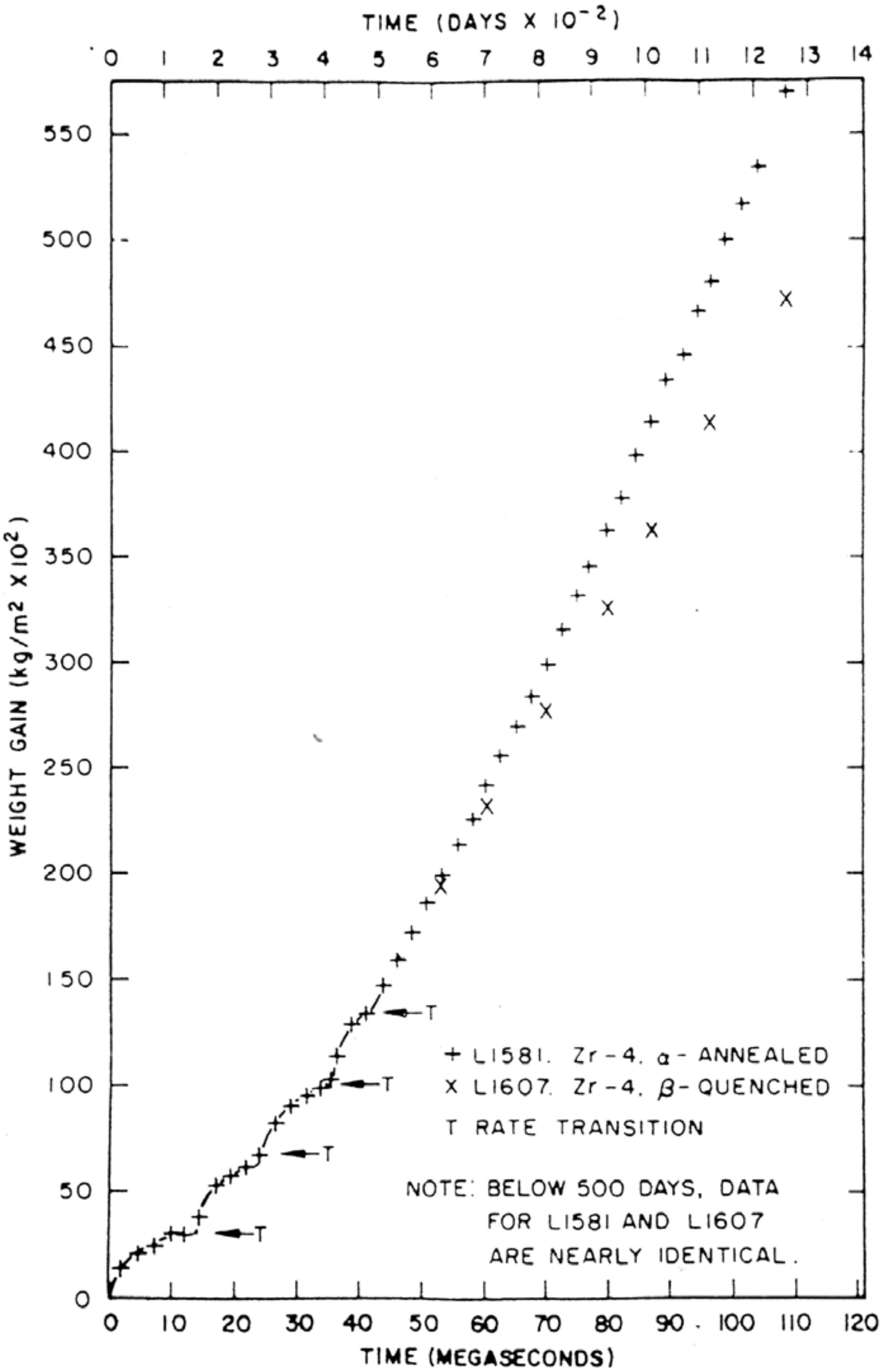


Figure 6-1: Typical Corrosion Kinetics in High Temperature Water. Weight gains on Zry-4 during corrosion tests in 633K water.

controlled by the diffusion of oxygen through a protective oxide layer, and the transition represents the breakdown of this film to leave an approximately constant diffusion barrier. When the reaction environment contains water molecules, a situation that covers anything from moist air to supercritical steam, some hydrogen from the reduction of water molecules enters the metal. The amount of hydrogen entering the metal is usually related to the amount of hydrogen released by the oxidation reaction (number of water molecules reacted) and expressed as a percentage of this quantity.

Research on the corrosion and hydrogen uptake process in the last 20 years have concentrated on understanding how the oxide film grows on a microscopic scale (the individual oxide crystallites formed are often only a few nanometers in cross-section, but may be much longer (0.2-0.5μm). giving a columnar structure), and how it breaks down at transition [*Ref. 6-3,6-4*]. One of the results of these studies has been a continually changing understanding of the growth and breakdown processes so that readers should be cautioned that some of the ideas expressed in early publications have changed as new evidence appeared. The changes over the past 10 years can be readily seen by comparing the relevant chapters of the two IAEA-TECDOC's. The latest references in the first one [*Ref. 6-3*] are up to about the end of 1989, while the references used for the second one [*Ref. 6-4*] only go to the end of 1995 (despite the respective publication dates), so there have been further changes that are needed to bring the story up to date.

This understanding of the structure and properties (chemical and physical) of the oxide film is essential because these are the factors that control both the rate of corrosion and hydrogen uptake. Under irradiation in a PWR the kinetics are generally similar, although the transition to a linear (or accelerating) rate appears to be at a much greater oxide thickness (about 10μm) than in the laboratory (Figure 6-2), and so may be the result of a different breakdown phenomenon [*Ref. 6-5*]. In

7 EFFECTS OF WATER CHEMISTRY

7.1 INTRODUCTION

The coolant water chemistry, and the crud depositions formed by the matter transported by the coolant, are becoming increasingly crucial for the nuclear fuel behaviour. Two important reasons are the higher heat fluxes and longer exposures in core, that are coming with elevated burn-ups, higher power densities, and longer cycles developing for all LWR designs. Another important factor is the strive for lower activity build-ups and suppressed stress corrosion cracking. Observations and development of theories has lead to the use of water chemistry control to reduce the rate of cracking and the activity build-up. New chemical additions to the coolant and modified water chemistries are consequently introduced at an increasing rate.

The higher demand on the fuel cladding from both the operation and the environment is occasionally causing elevated corrosion rates, sometimes quite unanticipated. The number of cases with surprising and fast fuel cladding corrosion accelerations is today increasing. It is hence increasingly important for the fuel engineers to keep track of the development on the water chemistry side in order to be able to make a full technical and economical assessment of the impact of changed water chemistry regimes.

7.2 GENERAL

7.2.1 Fuel Cleaning

Especially driven by the problems with Axial Offset Anomalies (AOA), see Section 7.3.1, EPRI has developed a fuel crud cleaning method by use of ultrasound [Ref. 7-1, Ref. 7-2]. The main objects for the method are one cycle assemblies that are expected to be less prone to have AOA once the crud from one cycle has been removed. It is stated that the crud on “high-duty assemblies”, i. e. assemblies that will be affected by AOA, is more adherent and does not dissolve despite the change of chemistry during shut-down [Ref. 7-1, Ref. 7-3]. A second advantage with the fuel cleaning is that the activity will also be removed from the fuel cladding, mitigating the activity distribution during operation. *There will however be a waste problem with the fílters collecting the removed crud.* The ultrasonic equipment is applied to a fuel bundle without dissembling. The equipment has been applied to 16 fuel bundles in Callaway (which has been affected by AOA, see below) that are currently under irradiation.

One consideration during the development of the method was the risk for the pellets in the rods to disintegrate. Frattini [Ref. 7-1], however, claims that the energy absorbed by the fuel during the ultrasonic cleaning was determined to be “benign” (?) relative to other anticipated vibration to the fuel during operation in core.

7.3 PWRS

7.3.1 Axial Offset Anomalies (AOA)

More than 20 cores in the US have been affected by AOA [Ref. 7-4], see Table 7-1. The Callaway plant had to decrease power to 70% for 8 months of the 9th cycle due to that the AOA (up to -14%) decreased the shut-down margin significantly [Ref. 7-4, Ref. 7-5, Ref. 7-3].

Table 7-1
Representative Cores in the US with AOA [Ref. 7-4]

Plant	Cycles with AOA
Callaway	4-6 and 8-10 (indications BOC 10)
Catawba-1	8
Commanche Peak-1	5, 6 (only individual ass.)
Commanche Peak-2	3
Millstone-3	4,5
Palo Verde-2	9
Seabrook	5,6
TMI	10
Vogtle-1	4,6
Vogtle-2	4,5
Wolf Creek	8-10

It is generally assumed that the AOA effect occurs due to boron absorption in the thicker crud deposits in the upper part of the PWR core, possibly due to boiling. Support for this is that AOA is found only for the high duty young fuel at higher boron concentrations.

It has generally been claimed that the most probable form whereby the lithium is retained in the crud is as lithium metaborate, $LiBO_2$ [Ref. 7-5]. Some recent investigations have, however, indicated that a nickel-iron oxyborate, Ni_2FeBO_5, has actually been identified on crud deposits from Callaway in fuel scrapings from EOC 9 [Ref. 7-3, Ref. 7-4]. Another possibility is that the boron is physically absorbed in the crud [Ref. 7-4]. Calculations have been performed within EPRI's Robust Fuel program, in order to try to quantitatively model the impact of various forms of boron absorption. It is inferred from the modeling that the solubility of lithium borate is too high to be of importance. Physical absorption in the crud did, however, give a rather good correlation with the boron expected to be required to form the actually produced AOA in a number of Callaway assemblies during cycle 6 [Ref. 7-4]. A good correlation was, however, only attained if it was assumed that the locally formed crud was not only produced due to local supersaturation of the dissolved iron and nickel in the coolant, but also included some 10% redistribution of old crud in the core, mainly in the form of particles. In this modeling, extrapolation from old chemistry data and obviously assumed chemical behavior has been employed.

There are two ways devised by EPRI to mitigate the AOA problem [Ref. 7-4]. One is to try to employ a higher and more constant pH during the whole cycle ("Coordinated Chemistry at Elevated pH"), which is actually recommended by EPRI in the EPRI primary coolant chemistry guidelines. There is yet no evidence that such operation will be successful. Another approach is to use boron acid enriched in boron-10 (EBA) (see also Section 7.3.3). The idea is to have less boron in the coolant during operation and hence have a smaller risk of exceeding the solubility products for boron species. On the other hand, if the boron is physically absorbed, and if the physical absorption is little sensitive to the coolant boron concentration and instead controlled by, for instance, the formed crud, EBA application could make the situation worse. *There seems to be a lack of proper evaluation of the absence of AOA in the hottest European PWRs (15x15). A deeper analysis of their very high duty assemblies and environment could possibly advance the understanding considerably.*

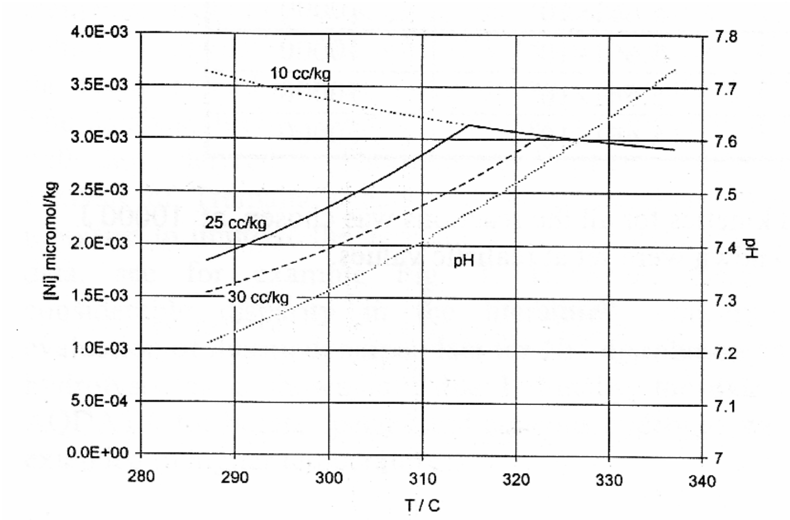


Figure 7-1. The calculated nickel solubility as a function of temperature and hydrogen content [Ref. 7-6].

One essential aspect when it comes to AOA is the influence of nickel. Generally, it is considered that a rather high nickel concentration, in relation to the iron concentration, in the coolant is a prerequisite for AOA formation. The local oxidizing conditions due to radiolysis in a PWR and the thermodynamics of nickel has therefore been revisited [Ref. 7-6]. Dickinson et al. find that the nickel solubility depends on a combination of temperature and hydrogen concentration, as shown in Figure 7-1. More importantly, their conclusion is that boiling will not produce oxidising conditions in the top of the core (due to change of the radiolysis yields), and hence no dramatic influence on the nickel solubility should be expected due to boiling. If there was an influence due to boiling, they imply that the deposits should be due to iron and chromium, but not nickel. A higher and more constant pH is what they indicate should be mitigating AOA. *They also claim that reduction of boiling would have an effect, a recommendation that seems to be contradicting their conclusions on the influence of boiling.*

They also conclude that the hydrogen concentration will not have a dramatic effect on nickel solubility, since the solubility of nickel and NiO was found to be similar under the realistic range of PWR conditions.

8 PRIMARY FAILURES

8.1 INTRODUCTION

A number of different modes of primary failures are possible and many of these modes have also been experienced in real operation. The more recently experienced primary failures are listed in Table 8-1.

Table 8-1
Recently Experienced Primary Failure Causes for LWR Fuel

Primary Failure Cause	Short Description
Debris Fretting	Debris transported by the coolant frets a hole in the cladding. This is the most common primary failure mode in both PWRs and BWRs today.
Excessive Corrosion	An accelerated general or local cladding corrosion causes cladding penetration.
Primary or Reactor Water Chemistry Influenced Corrosion	The water chemistry accelerates the corrosion as found for CILC (Crud Induced Localised Corrosion) and heavy crud deposits. This has by far been most common primary failure mode in BWRs (CILC) until recently.
Dry-out	The coolant flow is too limited, causing the cladding to be overheated. The overheating causes (local) corrosion penetration of the cladding.
PCI	Pellet Clad Interaction, a stress corrosion crack penetrates the cladding. Fission products, mainly iodine, are producing an aggressive chemical environment. The pellet swelling due to a power increase causes a tensile cladding stress.
Grid-Rod Fretting	The spacer grid frets a hole on the cladding (only experienced in PWRs, where it is still the second most common primary failure cause).
Manufacturing Defects	Manufacturing failures can occur in many ways, the most common have been primary hydriding (due to moisture or organic material in the pellets), welding defects around top and bottom end plugs, and cladding tube cracks.

Since the listed failure modes have been experienced rather recently, most of the work performed and reported is mainly on the listed primary failure modes. Additional information specifically on water chemistry influenced primary failures is given in ZIRAT-5 Section 9, Water Chemistry.

A compilation of the fuel failures in the US has recently been presented [Ref. 8-1], as shown by Table 8-2 and Table 8-3.

Table 8-2

BWR Fuel Cladding Failures in the US 1989 - 1999 [Ref. 8-1]

Failure Cause	**1989**	**1990**	**1991**	**1992**	**1993**	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**	**Total**	
Debris fretting	2	2	17	2	6	4		2	3	5	3	**46**	*20%*
Grid fretting												0	*0%*
Fabrication failures	3	3	1	1	1	2						11	*5%*
PCI		1			2		2	2	1	1		9	*4%*
Crudding/Corrosion											7	7	*3%*
CILC	**52**	5	3						**3**	**46**		**109**	*48%*
Unknown/Uninspected		4	3	9	7	9	2	10	1	1	1	**47**	*21%*
Total	***57***	***15***	***24***	***12***	***16***	***15***	***4***	***14***	***8***	***53***	***11***	*229*	*100%*

Table 8-3

PWR Fuel Cladding Failures in the US 1989 - 1999 [Ref. 8-1]

Failure Cause	**1989**	**1990**	**1991**	**1992**	**1993**	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**	**Total**	
Handling failures		6	2			1	1		2			12	*1%*
Debris fretting	146	11	67	20	13	6	10	1	10	3		**287**	*29%*
Grid fretting	14	18	9	33	36	9	33	52	21	57	5	**287**	*29%*
Primary hydriding		1		4								5	*1%*
Crudding/Corrosion							4		4			8	*1%*
Cladding creep collapse							1					1	*0%*
Other fabrication failures	1	15	1	5	3	1	15	5			1	47	*5%*
Other hydraulic failures					1							1	*0%*
Unknown					36	36	13	9	10	2	1	**107**	*11%*
Uninspected	43	58	35	61	14	3	12	3	8		3	**240**	*24%*
Total	***204***	***109***	***114***	***123***	***103***	***56***	***89***	***70***	***55***	***62***	***10***	*995*	*100%*

8.2 FRETTING FAILURES

As indicated in the Tables above, fretting is today the most important primary fuel failure cause both in the PWRs and the BWRs. Especially for PWRs, fretting has caused a large number of failures, as shown also by the data reported from Siemens (both Siemens in Germany and Siemens Power Corporation) in Table 8-4.

Table 8-4

Siemens Fretting Failures on BWR and PWR Fuel with Bi-metallic Spacers [Ref. 8-2]

Failure Cause		**PWR**			**BWR**	
	#	*%*	*per 100000*	*#*	*%*	*per 100000*
Handling failure	10	*4%*	0,5	0	*0%*	0,0
Debris fretting	109	*40%*	5,0	16	*6%*	1,0
Grid fretting	21	*8%*	1,0	0	*0%*	0,0
Baffle jetting	48	*18%*	2,2		*0%*	0,0
Manufacturing/Other	71	*26%*	3,3	0	*0%*	0,0
Shroud	12	*4%*	0,6		*0%*	0,0
Total	***271***	***100%***	***12,4***	***16***	***100%***	***1,0***

The explanations generally proposed for the higher fretting rates in the PWRs are the faster creep-down of the cladding, due to higher system pressure and higher temperature, which enhanced the risk for grid-to-rod fretting (compare BWR and PWR in the Tables above); higher flow rate, enhancing the vibrations; and more cross flows, since there is no flow channel as in the BWR.

Significant efforts have been spent recently in developing countermeasures, especially debris filters at the bottom of the PWR and BWR fuel assemblies), which is today almost standard for all fuel designs. Much work has also been done in order to develop flow fretting tests and mathematical models to simulate fretting, especially grid-to-rod fretting in PWRs.

Vallory et al. [Ref. 8-3] have reviewed the literature on such fretting and summarise that there is little quantitative information available in the literature, especially regarding actual extent of fretting and conditions of assembly operation when fretting has been recorded in a commercial reactor. The grid-to-rod fretting phenomenon is a result of the combination of the rubbing and impacting forces between the cladding and the grid springs or other contact surfaces. Vallory et al. explain that there is a large number of factors that will affect the impact of fretting between a grid and a rod, the more important listed below.

the nature of the cladding material (chemical composition, manufacturing, and surface condition)

9 SECONDARY DEGRADATION OF FAILED FUEL

9.1 INTRODUCTION

During reactor operation, the fuel rod may fail due to a primary cause such as fretting, *PCI*, manufacturing defects, corrosion, etc. A failed BWR fuel rod may degrade either by developing long axial cracks, splits and/or transversal breaks or may not degrade at all. During the period 1992-1993 six plants in US and in Europe were actually forced into unscheduled outages because of concerns about failed Zr sponge liner fuel degradation leading to excessively high off-gas activities. Both long axial cracks and significant loss of fuel pellet material were observed. In all these cases, the very high off-gas activities resulted from only one or two failed rods. Such a situation may result in very large utility costs. Failed rods in PWRs may also degrade, but long axial cracks does not exist and the activity release rate is much lower than that in a BWR. The rationale for the less severe behaviour in PWRs is probably related to that the smoother power regulation in a PWR compared to that in a BWR.

The secondary degradation scenario is described as follows. Once a fuel rod is perforated, water and steam can enter. The penetration of the fuel clad will subsequently allow water to enter the gap between the cladding and the fuel pellet stack, since the fuel rod pressure normally is much lower than the reactor system pressure. The water vaporises and hydrogen is mainly produced through the oxidation of the fuel cladding inner surface. However, two additional reactions may also contribute to the hydrogen production, namely:

- fuel oxidation, and
- radiolysis of the steam.

The oxide formed at the clad inner surface is uniform and its thickness decreases with increasing distance from the primary defect. This oxide is crucial for resistance against localised hydrogen pickup. Also, due to steam consumption the hydrogen to steam partial pressure will increase with increasing distance from the primary defect. Provided that, at a certain distance from the primary defect, the following conditions are met:

- 1) the hydrogen to steam partial pressure ratio is above a critical ratio and
- 2) the clad inner surface oxide thickness is smaller than a critical thickness,

localised secondary hydriding may occur forming *hydride blisters* or *sunbursts*. Since the specific volume of the hydride is larger than that of the zirconium alloy, a large local stress field will build up in and just outside the *hydride blister.* Also, since the hydride is brittle, many sharp cracks may form within the *hydride blister*. These sharp cracks or other cracks such as *PCI* defects or manufacturing cladding defects may propagate during a large enough power ramp and form an axial split.

If all the secondary hydrides concentrate to only one axial elevation, the massive hydrides may grow throughout the cladding thickness along the cladding whole circumference. This hydride location will be very brittle and easily fracture during reactor operation and it appears that a transversal break in this location may result even without a power ramp. It may be that the stresses generated by the hydrides, due to its larger specific volume, are sufficient to cause a secondary failure. It is not known today what parameters impacts the tendency for hydriding only at a single axial elevation.

9.2 NEW RESULTS

Edsinger, 2000, reviewed the axial crack formation mechanism of failed BWR fuel.

He states that the key characteristics of axial splits are:

- Crack initiation at sharp flaws (massive hydrides, PCI defects, etc.)
- Crack advance by a combination of axial and radial propagation with crack fronts leading near the outside surface, Figure 9-1
- Distinctive fractography characterized by chevron pattern, Figure 9-1
- Macroscopically brittle crack propagation as indicated by radially oriented fracture surfaces and little measurable plastic deformation, Figure 9-2
- Microscopically brittle crack propagation with ductility in Zircaloy only along vertical steps joining regions of radial fracture
- Completely brittle propagation through massively hydrided regions with flat (almost featureless) fractography
- Ductile separation in the zirconium liner (slight ductility also observed at inner surface of nonbarrier cladding), Figure 9-2
- Brittle crack propagation in cladding often well away from massively hydrided regions, with hydrogen as low as ~150 wtppm

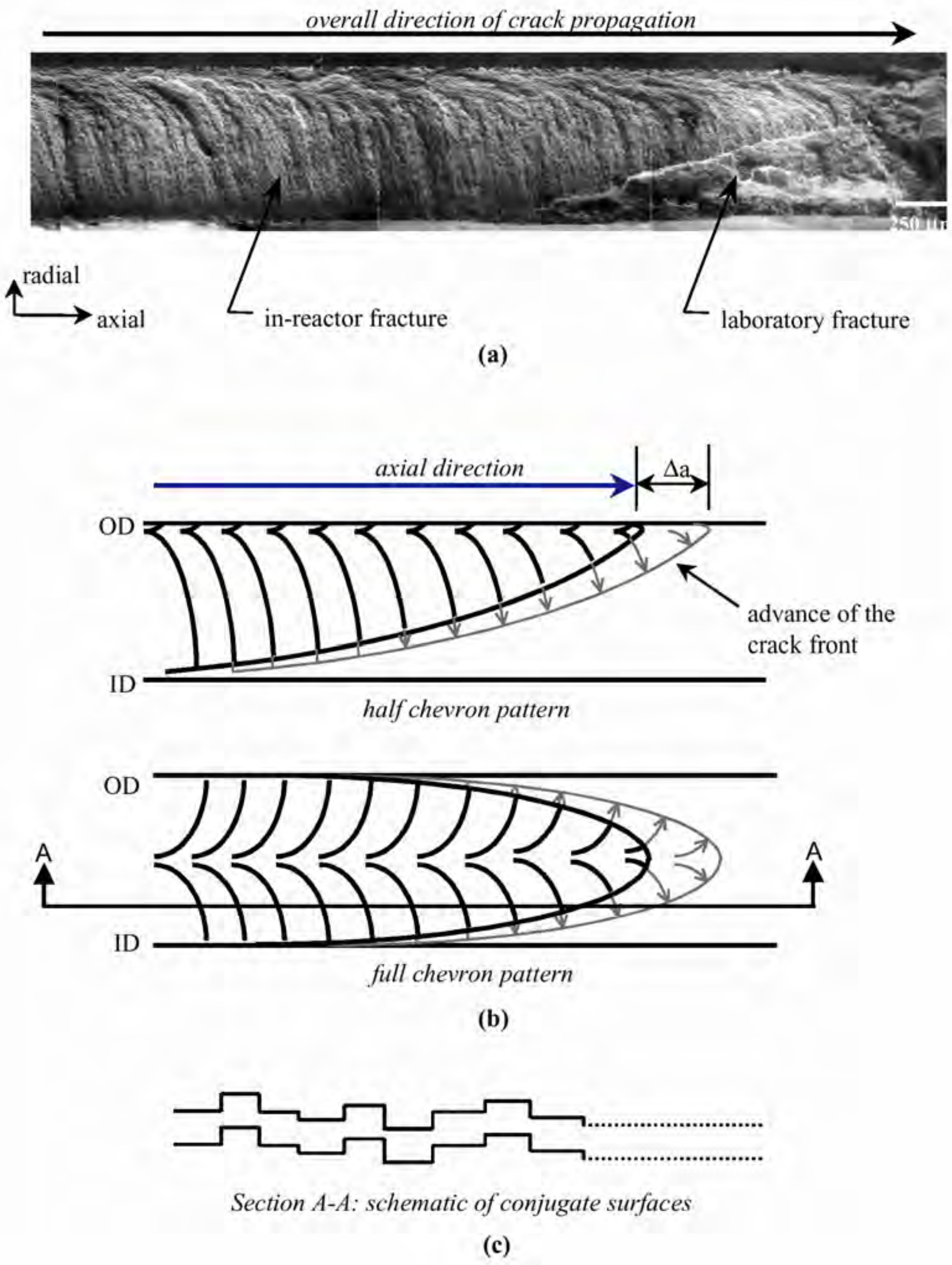


Figure 9-1: *(a) SEM fractography of crack tip in better preserved splits with schematics of crack fracture patterns in (b) and (c),* Edsinger et al., Ref. 9-2 and Lysell et al,.Ref. 9-4

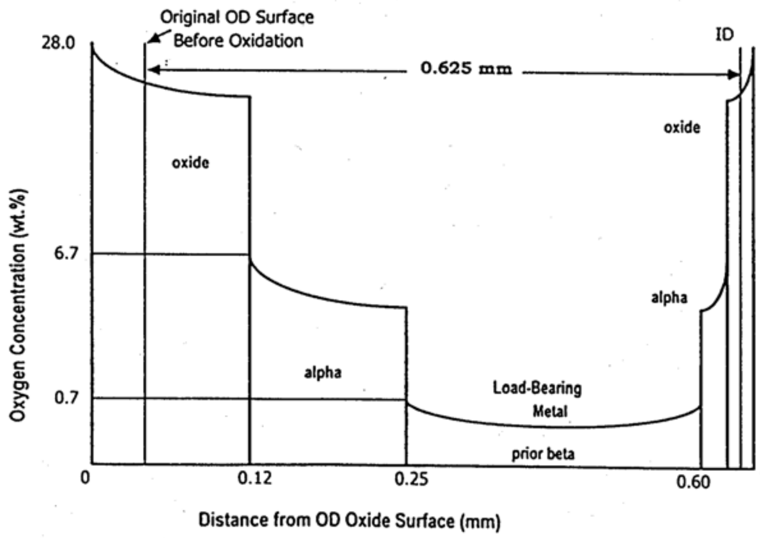
10 CLADDING PERFORMANCE UNDER ACCIDENT CONDITIONS

10.1 LOCA

10.1.1 LOCA Criteria

The objectives of the Loss Of Coolant Accident (LOCA) criteria are to maintain core coolability and preserve heat transfer area and coolant flow geometry during the quench phase and post-quench phase of a LOCA. The criteria resulted from a 1973 rule-making hearing based on unirradiated cladding data. Current efforts are to review the applicability of the data to high burnup cladding. A useful review of the bases for the original criteria was presented recently (***Ref. 10.1***) and summarized here. For a summary of the history and experimental results that led to current criteria the reader is referred to ***Ref. 10.6***.

Cladding is embrittled above the α/β transformation temperature by the formation of ZrO_2, oxygen stabilized α Zr(O), and further diffusion of oxygen into the underlying β phase, resulting in a structure shown on ***Figure 10-1***. Embrittlement by hydrogen pickup during irradiation and high temperature steam oxidation occurs as well. The oxide and the O stabilized α phases are brittle and the integrity of the cladding in a LOCA depends on the remaining ductility of the former β phase cladding section.

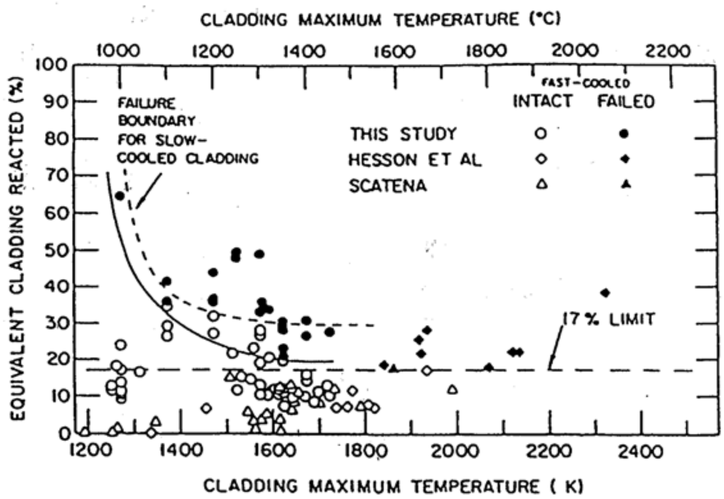


Resistance to thermal-shock during reflood and post-quench ductility and fracture toughness are largely determined by the thickness of and the oxygen concentration in the prior-beta layer.

Figure 10-1: OXYGEN DISTRIBUTION ACROSS CLADDING WALL DURING LOCA (*Ref. 10.1*)

The decision of the LOCA hearings was that retention of ductility was the best assurance against potential fragmentation of the cladding under the loading conditions proposed. The two criteria proposed for maintenance of adequate ductility were 17% Equivalent Cladding Reacted (ECR) and 2200^{0}F (1204^{o}C) (1477^0 K) Peak Clad Temperature (PCT) based on slow ring compression tests of samples from simulated LOCA tests. The ECR is defined as the total thickness of cladding that would be converted to stoichiometric ZrO_2 and this includes the oxide layer existing from the steady state operation. The potentially excessive conservatism of this criterion is being questioned currently.

A summary of some of the test results is shown on ***Figure 10-2***. The 17% ECR was derived from the Baker-Just Zircaloy oxidation rate correlation; at the PCT the ECR also coincides with the thermal shock failure boundary derived from other tests. The compression tests also established a zero ductility temperature of <275^{0}F (135^{o}C) during reflood, equivalent to ξ/w=<0.44, where ξ is the combined thickness of ZrO_2+αZr layer and w is the pre-oxidation cladding thickness. Samples oxidized at 2200^{0}F were significantly less brittle than those at 2400^{0}F in spite of comparable oxidation levels; this was attributed to solid solution hardening of the prior β phase at O concentrations of >0.7%.



Thermal-shock Failure Map for Zircaloy-4 Cladding (Bottom-flooded with water at the oxidation temperature) Relative to the Equivalent-cladding-reacted Parameter and Maximum Oxidation Temperature after Rupture in Steam. The best-estimate failure boundary for cladding that was slow-cooled through the $\beta \rightarrow \alpha'$ transformation before flooding with water and the data of Hesson et al. and Scatena are shown for comparison

Figure 10-2: ZIRCALOY-4 BEHAVIOR IN SIMULATED LOCA TESTS (*Ref. 10.1*)

Numerous tests, subsequent to the hearings, at ANL, ORNL and AECL (***Ref. 10.1***) confirmed the validity of both the PCT and ECR criteria, including an account for H pickup during steam oxidation. The effect of H pickup on embrittlement at high burnup has not been evaluated, how ever, and that is one of the objectives of the current NRC programs described in ***Ref. 10.2*** and summarized below.

10.1.2 USNRC LOCA Test Program

A test program was initiated in late '98 to evaluate the effect of simulated LOCA conditions on high burnup cladding >50 GWD/MT, with a completion planned in 2002. The program is carried out at Argonne National Lab.'s (ANL) Illinois and Idaho facilities.

High exposure cladding may have oxide films as thick as 100µ which is equivalent to 13% ECR, leaving a small margin to the 17% LOCA limit. For that reason oxidation studies are being carried out to evaluate the behavior of high exposure cladding relative to this limit. The test matrix planned is given in ***Table 10-1***.

Table 10-1
TEST MATRIX FOR INVESTIGATING THE OXIDATION KINETICS OF IRRADIATED AND NONIRRADIATED CLADDING (*Ref. 10.2*)

Material	Grid Span Location	Oxidation Temperature, °C	Oxidation Time, min.	Number of Tests
High-Burnup Zr-2	6	900	40, 300	2
	6	1000	20, 60, 150, 300	4
	6	1200	5, 10, 20, 30	4
	6	1300	3, 10	2
	2	1000	20, 60, 150, 300	4
	2	1200	5, 10, 20, 30	4
	2	1300	3, 10	2
	Archive	900	40, 300	4
	Archive	1000	20, 60, 150, 300	8
	Archive	1200	5, 10, 20, 30	8
	Archive	1300	3, 10	4
Intermediate Burnup Low-Sn Zr-4	4	1000	20, 300	2
	4	1200	5, 30	2
High-Burnup Low-Sn Zr-4	6	900	40, 300	2
	6	1000	20, 60, 150, 300	4
	6	1200	5, 10, 20, 30	4
	6	1300	3, 10	2
	2	1000	20, 60, 150, 300	4
	2	1200	5, 10, 20, 30	4
	2	1300	3, 10	2
	Archive	900	40, 300	4
	Archive	1000	20, 60, 150, 300	8
	Archive	1200	5, 10, 20, 30	8
	Archive	1300	3, 10	4

Tests under LOCA conditions are being made to determine the ballooning, rupture characteristics on heat-up and fragmentation characteristics during reflood of high burnup Zircaloy cladding. The results will assess the applicability of current embrittlement criteria to high burnup fuel and provide for any modifications if necessary. Internally pressurized irradiated fueled rod segments will be used so that potential fuel-clad bonding effects will be included in the test. The test sequence expected is:

- Slow heat-up in flowing steam (5°C/s) to rupture at 750-1000°C,

11 FUEL PERFORMANCE CRITERIA AND EXPERIENCE DURING DRY STORAGE

11.1 INTRODUCTION

The US Department of Energy (DOE) failed to meet the 1998 deadline, as required by law, to take possession of and store spent nuclear fuel assemblies; the date has been moved to 2007 by the DOE. The delay has left numerous reactor sites with insufficient wet storage capacity for their spent fuel and to meet the requirement of additional capacity to accommodate a full core unload. The numbers of reactors affected are shown as a function of time in Figure 11-1. The alternative dry storage of spent fuel assemblies has become of critical importance to assure the continued operation of the nuclear plants.

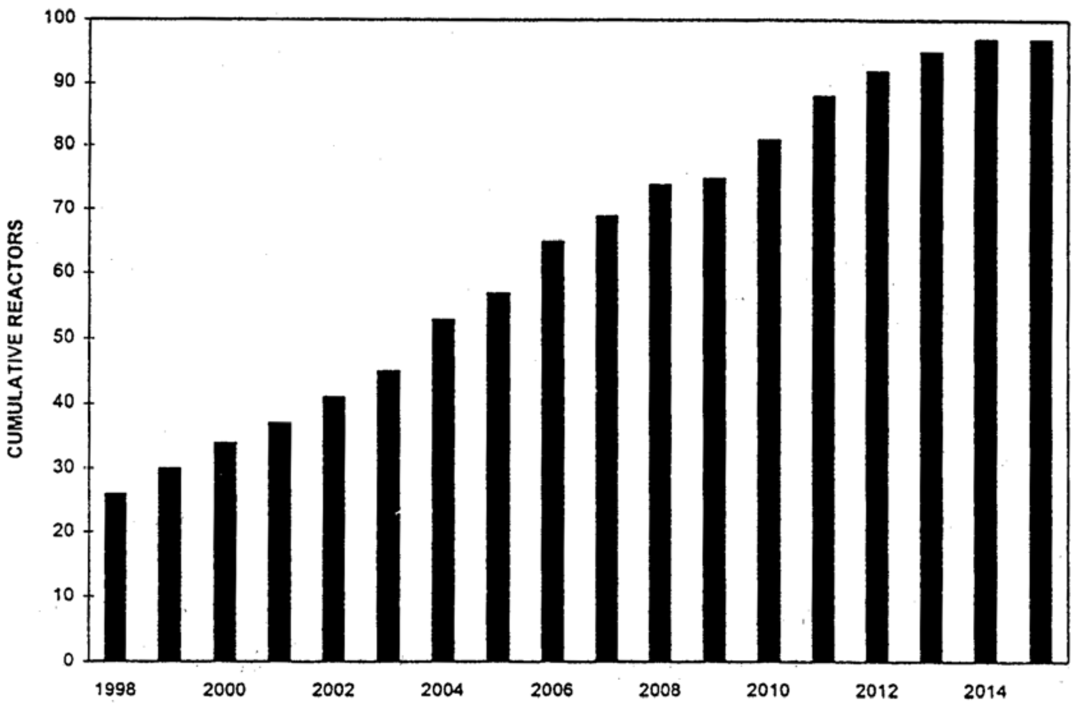


Figure 11-1: Cumulative reactors projected to lose full core discharge capability by 2015 in the US, *Ref. 11-1*.

- Countries outside of the US still have the option of reprocessing or have adequate, off-site intermediate storage facilities such as CLAB in Sweden. With the balance of economics shifting from reprocessing to permanent storage, the interest in dry storage is also increasing in countries that reprocess their fuel.

- The objective of this report is to

- summarize the US licensing criteria the fuel has to meet during dry storage with specific emphasis on the maintenance of cladding integrity,

- indicate the degree to which the criteria have been met,

- summarize future licensing goals and data needed to support them,
- discuss current differences between the USNRC and industry on the criteria and approaches to be used,
- summarize r&d programs to provide additional data,
- discuss fuel performance experience in dry storage to date,
- Valuable input to the report was provided by discussions with utilities in the process of licensing dry cask storage (Duke, Exelon), a utility with a major related research program (Electricité de France), the US Nuclear Regulatory Commission (NRC) Research and Regulatory Branches, Pacific Northwest Laboratories (PNL) consultants to the NRC, the Electric Power Research Institute (EPRI), the Nuclear Energy Institute (NEI), and Transnuclear (TN) a dry cask storage designer/manufacturer. Applicable regulatory documents and literature were reviewed.

11.2 CASK DESIGN AND LICENSING STATUS

- The design of the casks differ in detail depending on the vendor, but have generic similarities. They consist of a core with a structural basket made of stainless steel and either Boral or borated steel sheets for criticality control. The basket is sized for either BWR or PWR assemblies. There are similarities to a wet storage rack.
- The body of the cask is usually a thick carbon or low alloy steel, cast iron or sometimes stainless steel and serves as a gamma and neutron shield in addition to its function as a container. Additional lead shielding is added in some casks for gamma shielding. Most casks have neutron shielding in the form of borated polymers embedded in the walls of the cask. Metallic fins or convective air passages (outside the cask containment) serve as heat rejection devices. Some casks have penetrations for thermocouples to measure the internal temperatures – critical to licensing and performance. Others have pressure sensing devices to assure maintenance of a slight positive internal pressure.
- Schematics of a variety of vertical storage cask designs are shown in Figures 11-2 through 11-5 and a horizontal one in Figure 11-6. Most casks are transportable or have transportable canisters eliminating the necessity of reloading from a storage to a transport container – clearly a desirable feature.

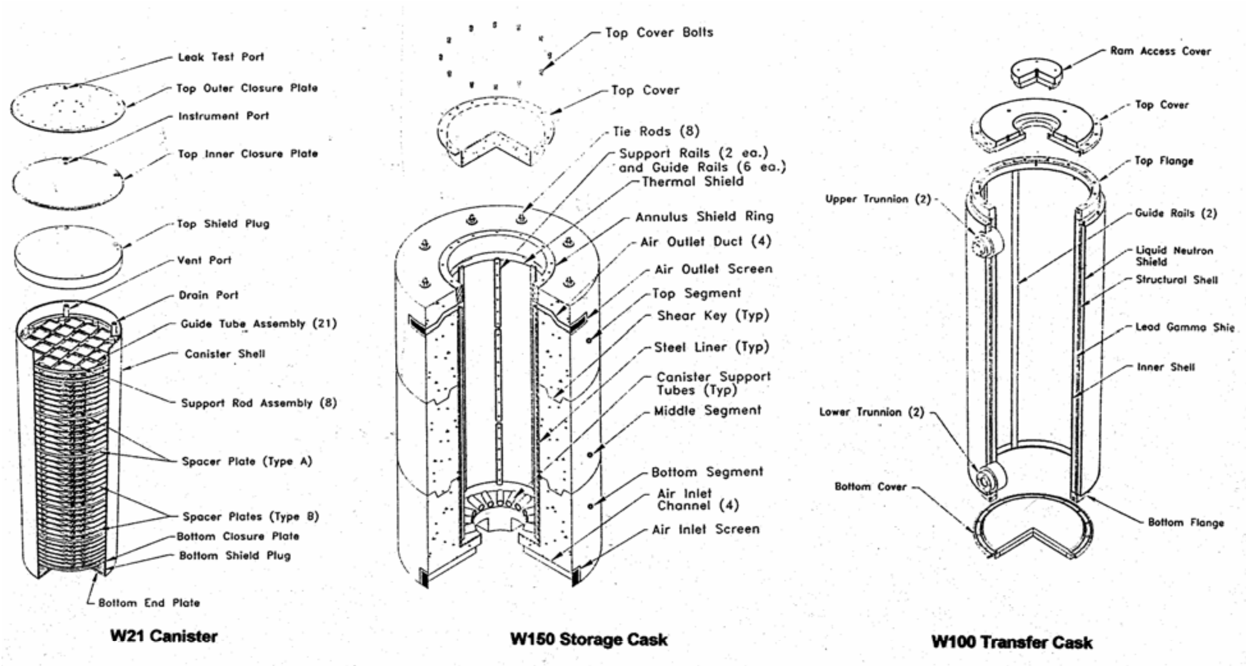


Figure 11-2: WESFLEX TRANSPORTABLE STORAGE SYSTEM (Now BNFL's Fuel Solutions Storage System)

Figure 11-2: Wesflex transportable storage system (now BNFL's fuel solution storage system)

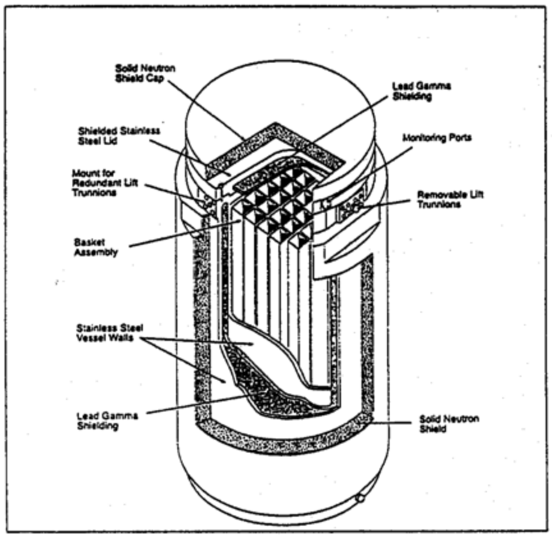


Figure 11-3: NAC S/The metal cask, *Ref. 11-1*