

# ZIRAT-9 Annual Report

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## 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 objective is met through a review and evaluation of the most recent data on zirconium alloys and to identify the most important new information and discuss its significance in relation to fuel performance now and in the future. Included in the review are topics on materials research and development, fabrication, component design, and in-reactor performance.

Within the ZIRAT-9 Program, the following technical meetings were covered:

- Int. Seminar on *PCI* in Water Reactor Fuels, March 9-11, 2004, Cadarache
- Pacific Basin Nuclear Conference, March 21-25, 2004, Honolulu, Hawaii
- KTG conference, March 30 -31, 2004, Karlsruhe
- Enlarged Halden Programme Group Meeting, May 9-14, 2004, Sandefjord
- Jahrestagung Kerntechnik, May 25-27, 2004, Dusseldorf
- 14th Int. Conf. On Zirconium, Stockholm, June 14-17, 2004
- Utility conference 2004 in US, June 21-23, 2004, Birmingham, AL
- 2004 ANS conference on *LWR* fuel performance, Sept. 19-22, 2004, Orlando, FL
- Water Chemistry of Nuclear Reactor Systems, October 11-14, 2004, San Francisco, CA
- Nuclear Safety Conference, Washington DC, Oct. 2004

The extensive, continuous flow of journal publications is being monitored by several literature searches of world-wide publications and the important papers are summarised and critically evaluated. This includes the following journals:

- Journal of Nuclear Materials,
- Nuclear Engineering and Design,
- Kerntechnik
- Metallurgical and Materials Transactions A
- Journal of Alloys and Compounds
- Canadian Metallurgical Quarterly
- Journal de Physique IV
- Journal of Nuclear Science and Technology
- Nuclear Science & Engineering
- Nuclear Technology



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:

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

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

The information within the ZIRAT-9 Program is either retrieved from the open literature or from proprietary information that ANT International has received the OK from the respective organisation to provide this information within the ZIRAT-program.

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

The work reported herein will be presented in three Seminars: one in Barcelona on January 11-13, 2005 and one in Marco Island, Florida, on January 25-27, 2005 and one in Japan in October, 2005.

The Term of ZIRAT-9 started on February 1, 2004 and ends on January 31, 2005.

## 2 UTILITY AND REGULATORY BODY PERSPECTIVES (PETER RUDLING)

### 2.1 INTRODUCTION

#### 2.1.1 Regulatory perspective

The objective of nuclear reactor safety is to ensure that the operation of commercial nuclear power plants does not contribute significantly to individual as well as societal health risks. Reactor safety is thus primarily concerned with the prevention of radiation-related damage to the public from the operation of commercial nuclear reactors. Safety limits are introduced to avoid fuel failures during normal operation, or to mitigate the consequences of reactor accidents in which substantial damage is done to the reactor core.

In most countries the dose rate limits are defined for a possible off-site radiological release following a reactor accident. Fuel safety criteria that relate to fuel damage are then specified to ensure that these limits are not exceeded. Numerous criteria related to fuel damage are used in safety analyses. These criteria, however, may differ from country to country. Some criteria are used to minimize cladding degradation during normal operation and some are used to maintain cladding integrity during anticipated transients, thus avoiding fission product release. Others are used to limit fuel damage and ensure core coolability during design-basis accidents, or to limit the public risk from low probability severe accidents.

The fuel issues that concern the regulatory bodies today during normal operation (class I) and anticipated operational occurrences (class II) 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 build-up, and
- Accelerated growth of rods and assemblies.

Also performance of high burnup fuel is a concern since the current *LOCA* and *RIA* limits are based upon non-irradiated fuel or fuel with very low burnup. In addition, the current criteria were based upon tests on Zircaloy while today other materials are used such as ZIRLO and M5, that may show different material behaviour during such accident conditions compared to that of Zircaloy.

### 2.1.2 Utility perspective

The fuel discharge burnup has increased steadily over time, Figure 2-1 and Figure 2-2. Figure 2-3 shows that also cycle length and enrichment level in US have increased over time.

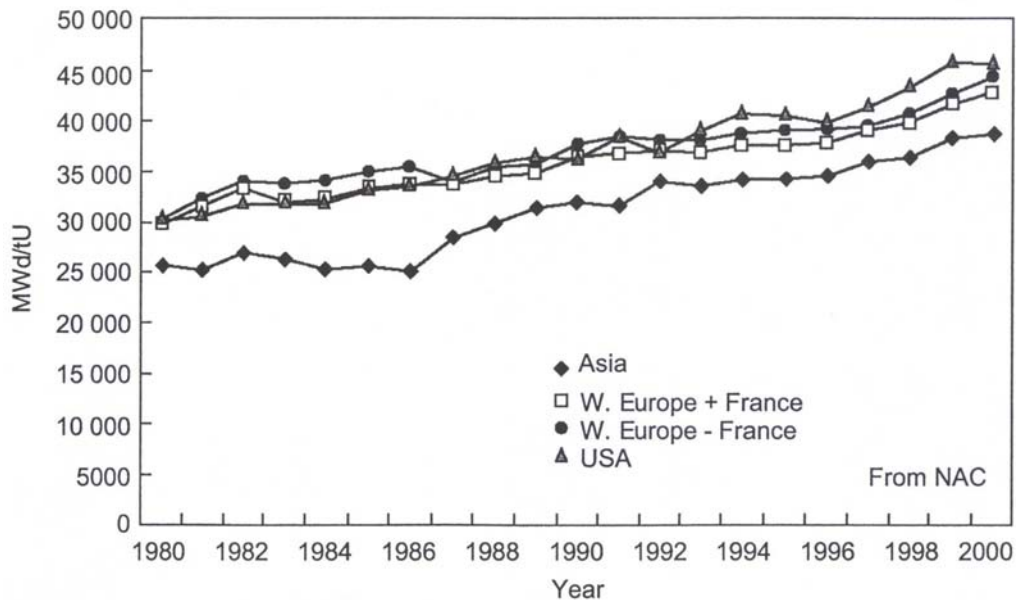
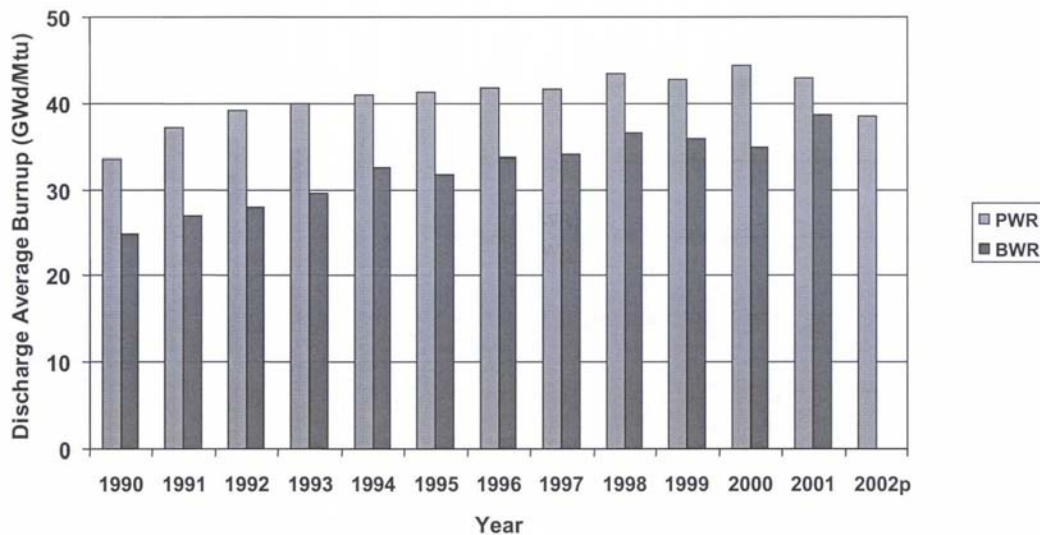
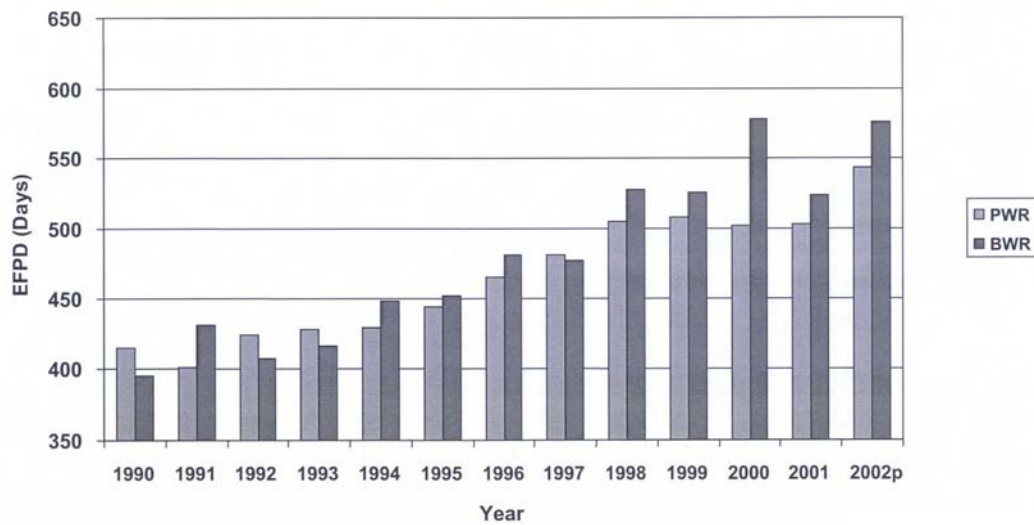


Figure 2-1: Historical Trend of Burnup, Estève, et al., 2002.

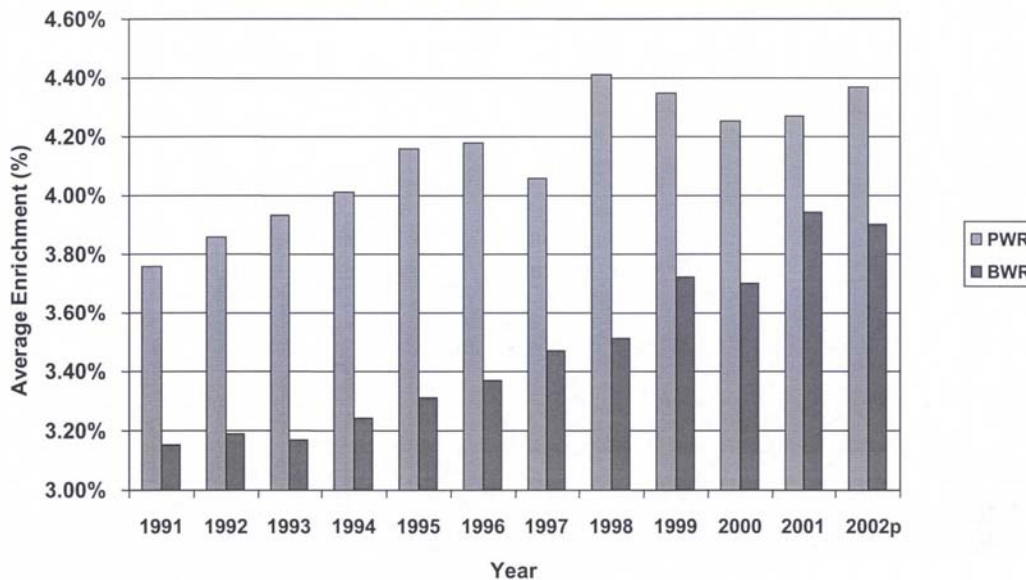


c) Trends In Discharge Batch Average Burnup - U.S.

Figure 2-2: High Burnup History in the US, Yang, 2003(b).



a) Trends In Cycle Length - U.S.



b) Trends In Average Initial Enrichment - U.S.

Figure 2-3: Cycle Length and Enrichment History in the US, Yang, 2003(b).

The burnup levels of operating plants for different countries are shown for *BWRs* in Table 2-1 and for *PWRs* in Table 2-2 for batch, assembly and rod burnups achieved in standard reload batches. The table also includes the current licensing limits for each country as well as the burnup levels that the industry hope to be able to license in the future.

Table 2-1: Current *BWR* Burnup Status and Limits (GWD/MTU) Adamson, et al., 2003.

| Country     | Status                    | Current Licensing Limit<br>(Expected Extension)  |
|-------------|---------------------------|--|
| USA         | 35-40 batch<br>42-44 assy | 62 rod ave.<br>(70 for 24 mo. cycles)  |
| Finland     |                           | 36 assy.<br>max. 10 assy. > 34   |
| Germany     | 40 batch<br>50 assy       | 53 assy  |
| Sweden      | 45-48 batch               | 55 batch<br>61 rod<br>(Intermediate storage same as <i>PWR</i> )                         |
| Switzerland | 48-50 batch               | 55 (KKL) assy. ave.<br>70 (KKM) 75 (KKL) peak pellet                                     |
| Japan       | 39 batch (Step II, 8x8)   | 50 assy. (Step II, 8x8)<br>45 batch, 55 assy (Step III, 9x9)<br>(peak rod not regulated) |

Table 2-2: Current *PWR* Burnup Status and Limits (GWD/MTU), Adamson, et al., 2003.

| Country     | Status                                | Current Licensing Limit<br>(Expected Extension)   |
|-------------|---------------------------------------|---|
| USA         | 45-50 batch<br>50-55 assy.<br>57 rod  | 62 rod ave.<br>(75 for 18 mo. cycles)   |
| Belgium     | 45-55                                 | 55 assy. UO <sub>2</sub><br>50 assy. MOX  |
| Finland     |                                       | 40 assy.<br>48 rod<br>max. 12 assy. with a rod > 44   |
| France      | 45-50 assy.                           | 52 assy. UO <sub>2</sub><br>(60 assy. in 2005)<br>(70 assy. in 2010)<br>42 MOX assy. for 3/12 mo. cycles                |
| Germany     | 50-65 F/A                             | 65 assy.  |
| Hungary     |                                       | 60 rod ave. (BNFL)<br>55 rod ave. (TVEL)  |
| Netherlands | 33.6 batch<br>55 assy.<br>59 peak rod | 60 rod<br>66 peak pellet  |
| Sweden      | 50 batch                              | 60 assy.<br>64 rod<br>(50 assy. for cask, to be raised to 55)<br>(Analyses in process to allow 4.5% enrichment in pool) |
| Switzerland | 40-43 (KKB) 53 (KKG) batch            | 50 (KKB) 60 (KKG) assy. ave.<br>55 (KKB) 65 (KKG) peak rod ave.<br>59 (KKB) 76 (KKG) peak pellet                        |
| Japan       |                                       | 48 assy. UO <sub>2</sub><br>45 assy. MOX  |
| Korea       |                                       | 60 (W) rod ave.<br>58 (CE) rod ave.   |

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, utilities are looking into the possibilities to:

- A. Increase the burnup
- B. Longer fuel cycles
- C. To use high duty fuel management schemes
- D. Reactor power uprating
- E. Achieve clean cores, i.e., absence of defects and low system radiation buildup (may be obtained by water chemistry changes such as high pH in *PWRs* and Zn additions in *BWRs* and *PWRs*)
- F. Decrease tendency for stress corrosion cracking in reactor components such as piping (may be achieved by water chemistry changes such as Hydrogen Water Chemistry and Noble Metal Addition in *BWRs* and Zn additions in *PWRs*)
- G. Increase operating flexibility (load following)
- H. Continuously improve or maintain operating safety margins.

The situation for the nuclear industry is complicated since implementing A. through G. in the list above would normally decrease the operating safety margins (item H.). However, possibilities to implement all items A. through H. may be given by introduction of new advanced fuel designs (with new materials) as well as better models to predict fuel behaviour during class I through IV operation. However, other situations that may make it more difficult to improve operating safety margins are the following:

- There is a drainage of competence in the nuclear industry, very knowledgeable experts are retiring while very few “bright” young people are attracted to the nuclear industry
- Fuel vendors have less resources to solve fuel related issues for utility customers. This situation is driven by the deregulation of the electrical market that has reduced the price for nuclear fuel.

Figure 2-4 tries to summarise the above.

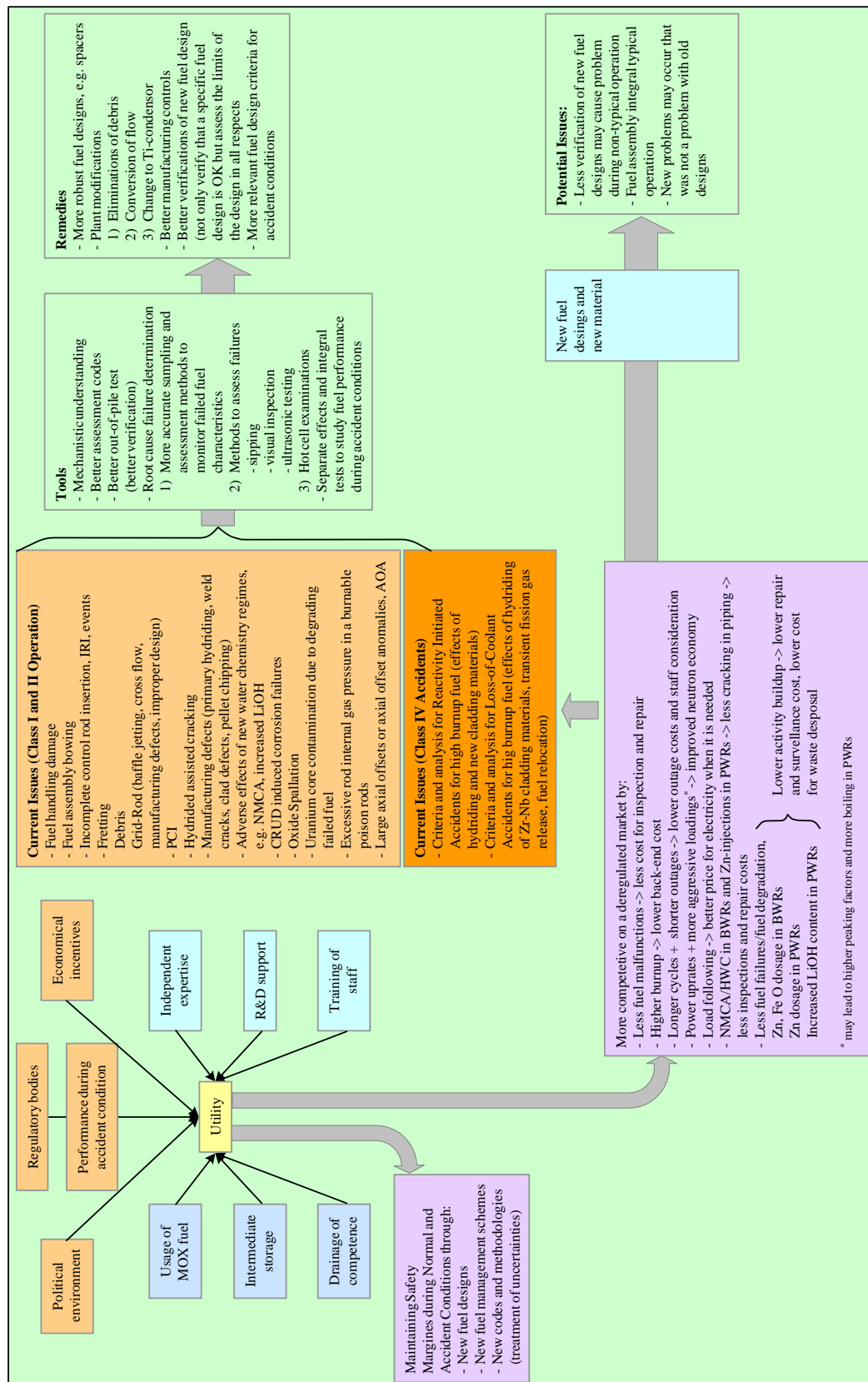


Figure 2-4: Utility environment.

The potential consequences of increased burnup on fuel are shown in Figure 2-5 and Figure 2-6.

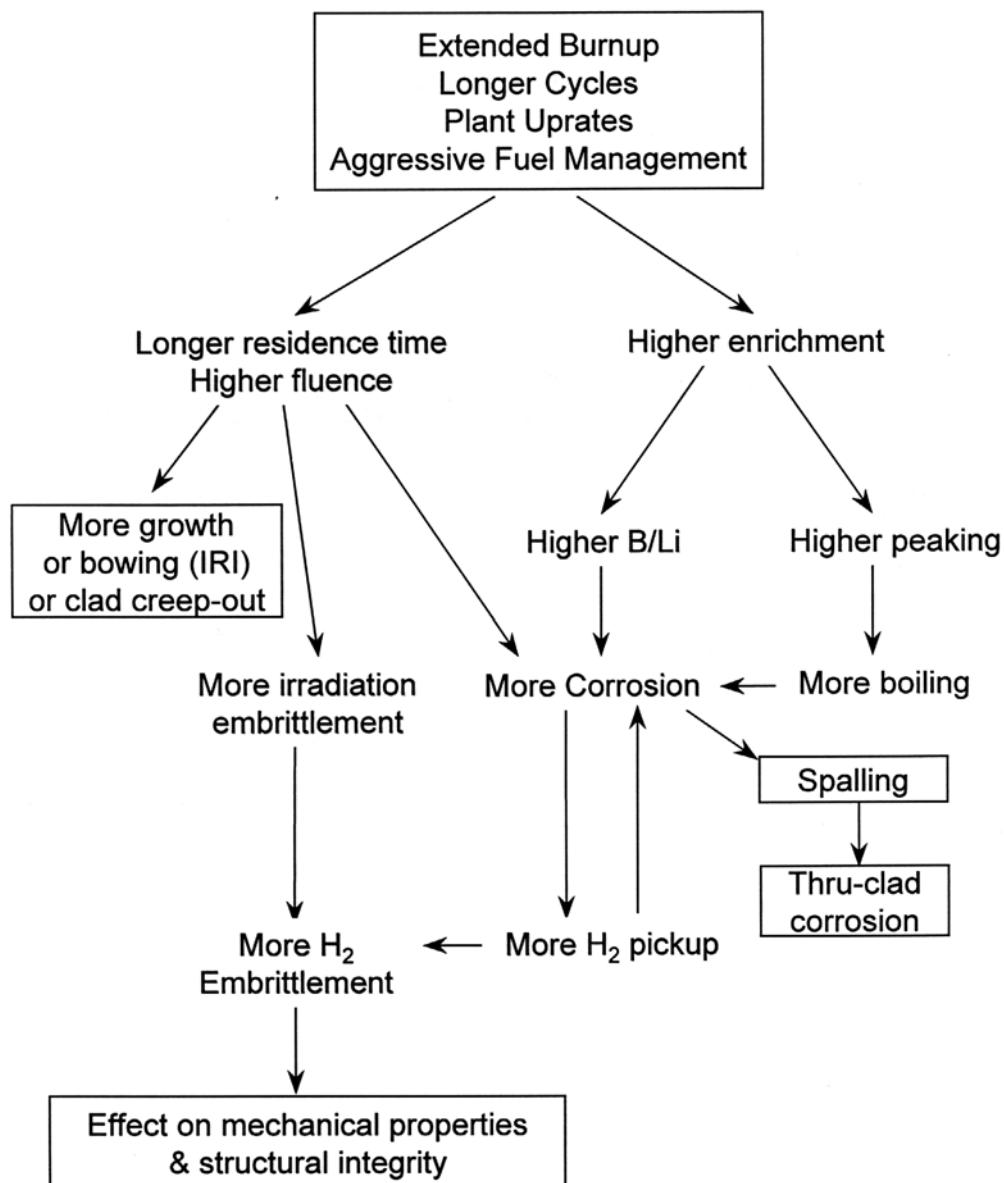


Figure 2-5: The effect of extended burnup on fuel performance – PWRs.



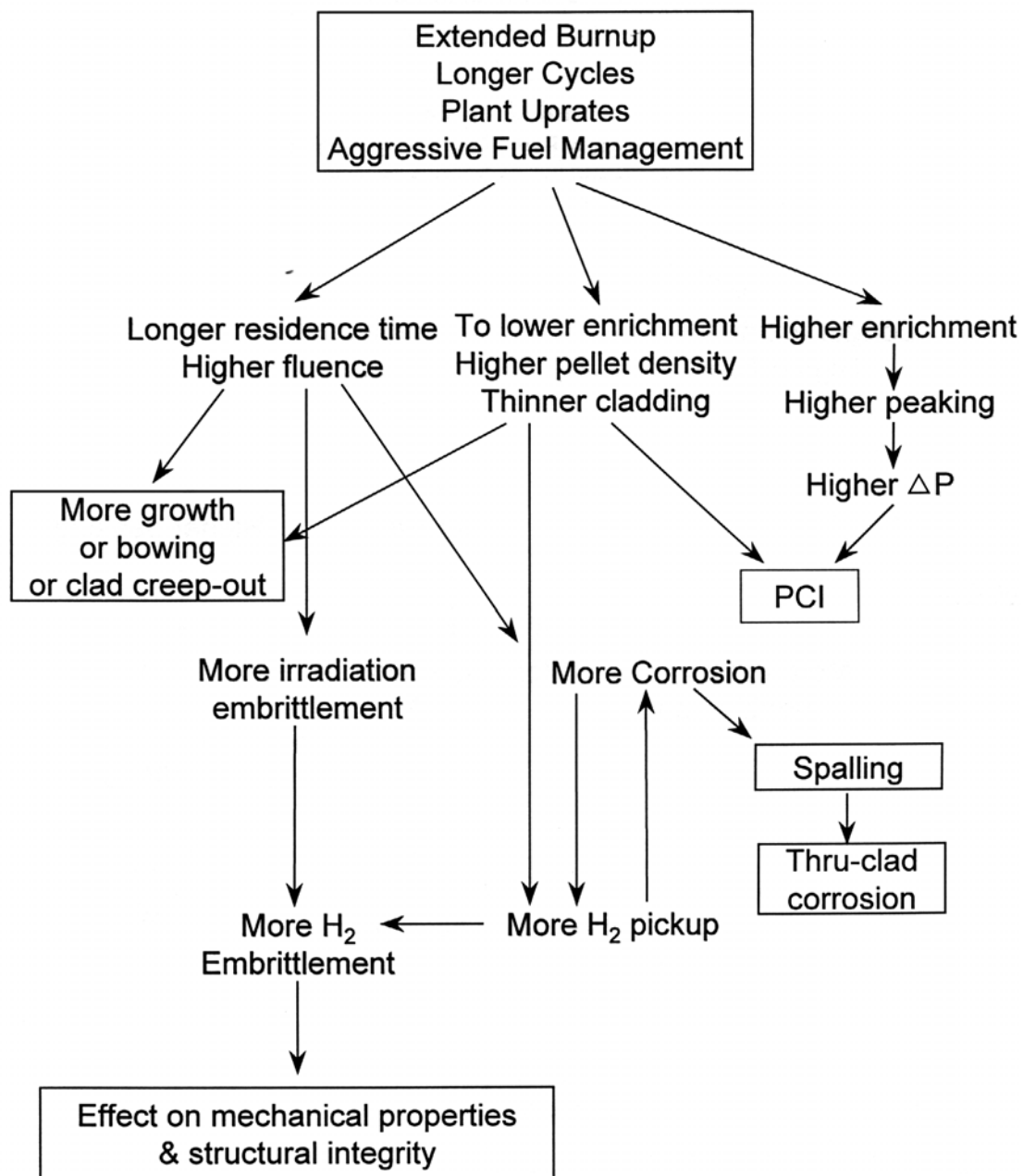


Figure 2-6: The effect of extended burnup on fuel performance – BWRs.

## 2.2 NEW DATA

### 2.2.1 Regulatory perspective

*NRC* are now planning to make a revision of the *LOCA* cladding embrittlement regulations and they will sample data up to September 2005 after which they intend to start to develop the new criteria, Meyer, 2004. *For more details, see section 10.3.3.3.* *NRC* envisages that there will be two options for the licensing procedure:

- 1) Grandfather procedure (only possible for Zry-4, ZIRLO and M5 up to burnup of 62 MWd/kgU) assuming that there are the same effects of burnup on M5 and ZIRLO as for Zry-4. This due to lack of *LOCA* relevant high burnup data on M5 and ZIRLO.
- 2) Performance based criteria for other alloys than Zry-4, M5 and ZIRLO or for Zry-4, M5 and ZIRLO at burnups >62 MWd/kgU. These criteria will allow a somewhat higher *PCT* than 2200°F (perhaps up to 2307°F) and the possibility to use the best-estimate Cathewell-Powel correlation (instead of the conservative Baker-Just correlation).

Meyer, 2004 further stated that details of both options are unknown at this time and will depend on the results of ongoing tests at ANL. A schedule has been established to develop the technical basis for the performance-based criteria by September, 2005, and this schedule is being tracked at a high level within the *NRC*. *NRC* rulemaking should begin shortly thereafter reported Meyer, 2004.

### 2.2.2 Utility perspective

#### 2.2.2.1 Japan

The paper by Ishiguma, 2004, discusses the various fuel design improvements and R&D efforts to develop high burnup fuel *PWR* assemblies. Some of the R&D efforts to increase fuel burnup includes: introduction of Zr-Nb corrosion resistant cladding material, reducing fuel rod inner pressure, using large grain size pellet. Ishiguma, 2004 further reported that today, 10 *PWR* plants have been approved to increase the discharge burnup from 48 MWd/kgU (assembly average) 55GWd/t. by METI (Ministry of Economy, Trade and Industry). It is envisioned that longer operation cycle, even higher discharge burnups (60-65 GWd/t), and reactor up-rating will be adopted to further improve economical efficiency and flexibility.

Fuel discharge burnup of the 29 *BWRs* in Japan plants has increased by about 50% over the last 20 years, Otsuka & Kitamura, 2004. Figure 2-7 shows the past trend of batch average discharge burnup in Japanese *BWRs*. Fuel designs with 9x9 geometry by four fuel vendors (two domestic and two foreign) and a 45 GWd/t batch average discharge burnup (55 GWd/t assembly burnup is the current licensing limit) are now employed in Japanese *BWRs*. The authors reported that TEPCO is considering the possible introduction of 10x10 fuel geometry (today only 8x8 and 9x9 *BWR* lattices are used). This would allow for increased operating flexibility in terms of higher burnup or plant up-rate.

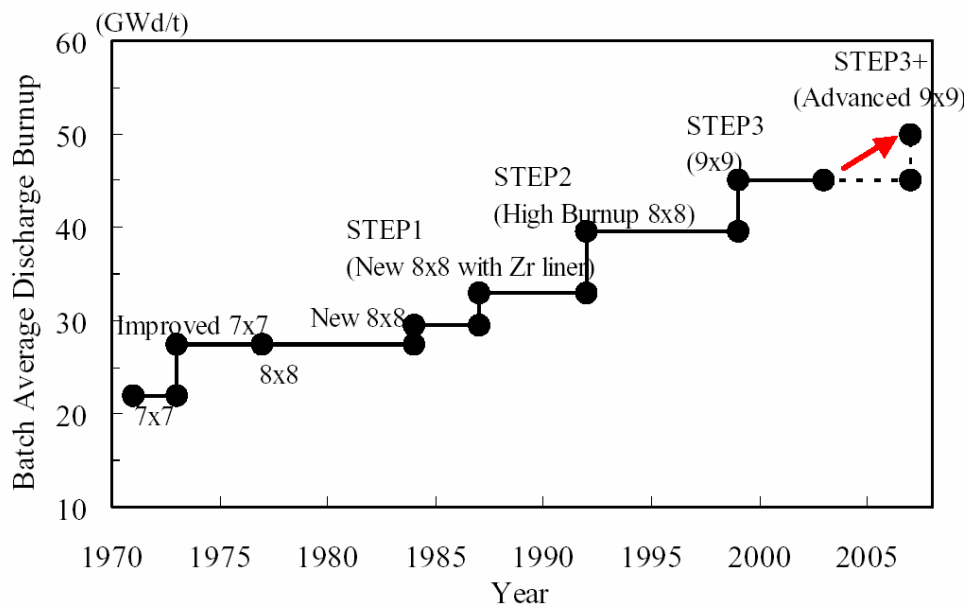


Figure 2-7: Burnup Trend in Japanese BWR, Otsuka & Kitamura, 2004.

#### 2.2.2.2 France

There are now 58 PWR reactors (34x900MW, 20x1300MW and 4x1500MW) operating in France Figure 2-8, Debes, 2004, Provost & Debes, 2004 and Dangouleme 2004.

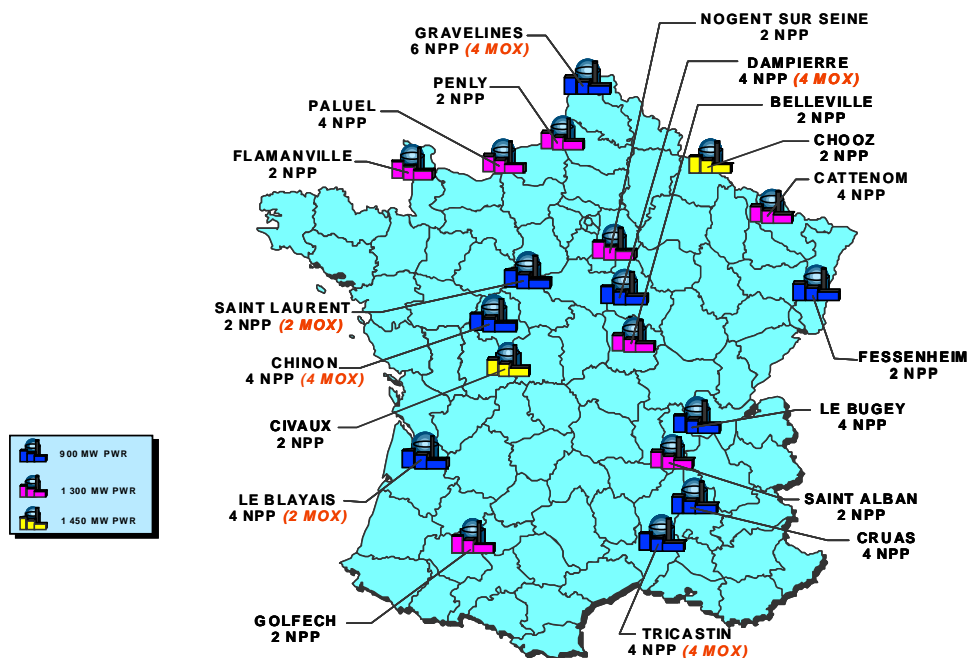


Figure 2-8: EDF nuclear power plants, Dangouleme, 2004.

The current fuel management modes, resulting in an average burn up of 44 MWd/kgU (licensing assembly average burnup is 52 MWd/kgU, are the following:

- For the 900 MW units: 4.2% per third and 18 months cycle on CP0 units; 3.7% per quarter and 13 months cycle on standardised CPY units;
- For the 1300 MW series: 4% per third and 15/18 months cycle;

and for the N4 1500 MW units: 3.4% per quarter and 11 months cycle.

The objective for EDF is now to increase competitiveness both by optimizing operating cycle length and flexibility and by improving nuclear fuel efficiency by increasing the current assembly average burnup of 44 to 55 MWd/kgU in the next decade. In the years to come, the following design and operation changes are envisioned:

- For the twenty 900 units using MOX fuel, the new "MOX parity" management will be implemented to increase the plutonium content (from 7% today up to 8.65%).
- For the twenty 1300 units the UO<sub>2</sub> enrichment will be increased from current 4% up to 4.5%, with 18 months cycle, to reach an average assembly discharge burnup of 55 MWd/kgU.
- For the four 1450 MW N4 series, the operating cycle length will be increased from 11 to 17 months, and the enrichment will be increased from 3.4% (1/4) to 4% (1/3), to reach an assembly average burn-up of 47 MWd/kgU.

### **2.2.3 Fuel vendor perspective**

#### **2.2.3.1 GNF**

Schneider et al. 2004a reported on the GNF fuel performance experience of the most recent GNF fuel designs. The maximum burnups reached for GE11-14 fuel are as follows:

- Fifty-six (56) GE11/GE13 (9x9 lattice) LUAs have been irradiated in 11 different reactors, to lead exposures of up to 58,000 MWd/MTU bundle average.
- Eighty-eight GE14/GE12 LUAs have been operated in 16 different reactors, achieving lead bundle average exposures of 68,000 MWd/MTU. The GE14 fuel design extends the earlier GE12 10x10 fuel design with a higher linear heat rating, optimized spacer positioning for improved critical power performance, and standardization to a Zircaloy ferrule spacer and debris filter lower tie plate. The earlier GE12 designs included either Inconel X-750 or Zircaloy-2 spacers with the debris filter lower tie plate as a design option.

The authors also summarized the GE/GNF fuel clad development over the years that primarily have been focusing on obtaining optimum corrosion performance. The GE/GNF Processes 3 and 4 applied a heat treatment to the high  $\alpha+\beta$  temperature range at the incoming tubeshell stage. Process 5 applied the heat treatment prior to the last two pilgering steps; termed In-Process Heat Treatment (IPHT)) in the  $\beta$  temperature range. *Process 5 is thus quite similar to the Westinghouse 1k II BWR cladding.* Process 6 and 7 similarly applied an in-process heat treatment, but to the high  $\alpha+\beta$  temperature range. Process 8 retains the same liner Fe content as Process 7 (1000-1200 wtppm Fe), to retain PCI resistance and degradation resistance, but returns to a Process 5 type IPHT heat treatment, a change made specifically to increase nodular corrosion resistance.

### 2.2.3.2 TVEL

TVEL Corporation, a Russian fuel vendor presented some new design features of their WWER 1000 designs, Figure 2-9 and, Figure 2-10, Troyanov, et al., 2004 and Dragunov, et al., 2004.

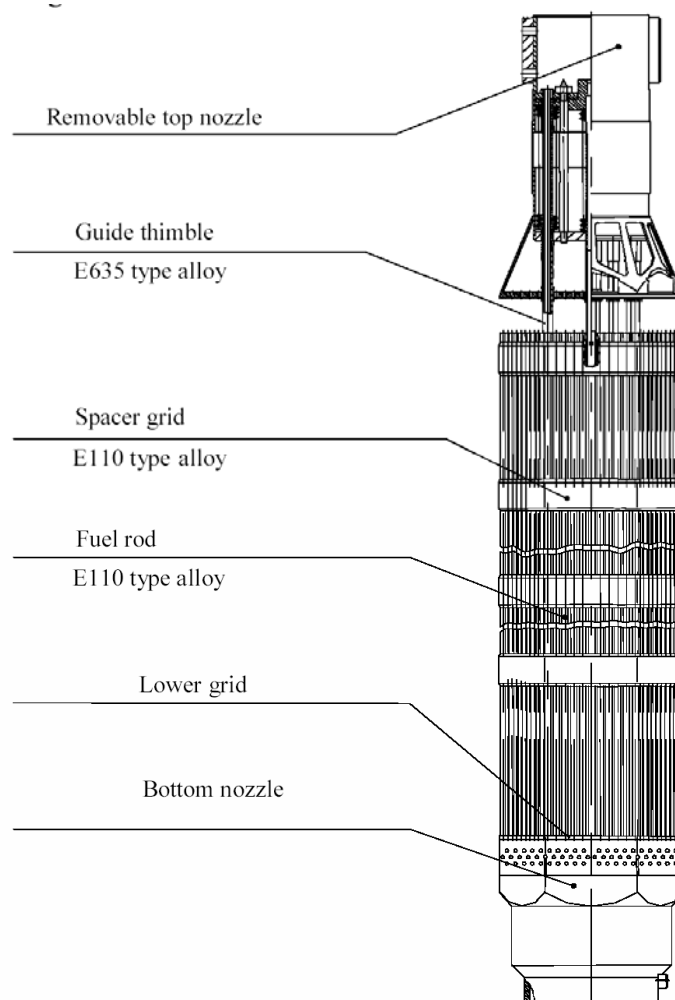


Figure 2-9: The TVS-2 fuel assembly design, Troyanov, et al., 2004.

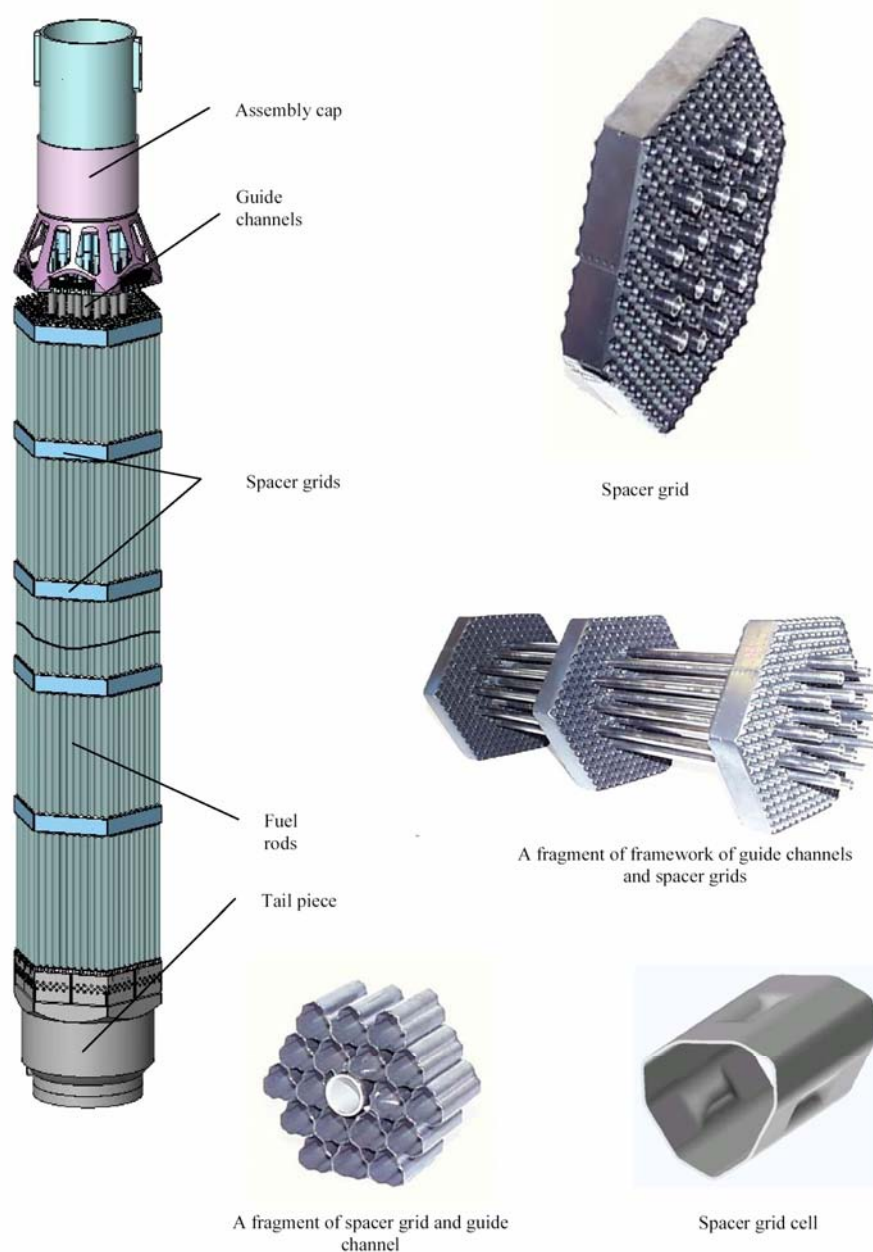


Figure 2-10: WWER-1000 fuel assembly and its components, Dragunov, et al., 2004.

### 2.2.3.3 MHI

Shimomura, et al., 2004, presented some of the major changes in the Mitsubishi *PWR* fuel design.

As a countermeasure towards debris fretting failures, long bottom end plugs, Figure 2-11, and a Debris Filter Bottom Nozzle (DFBN), Figure 2-12, was introduced already in 1991 and the authors claim that since these fuel design feature were introduced no debris fretting failure has occurred.

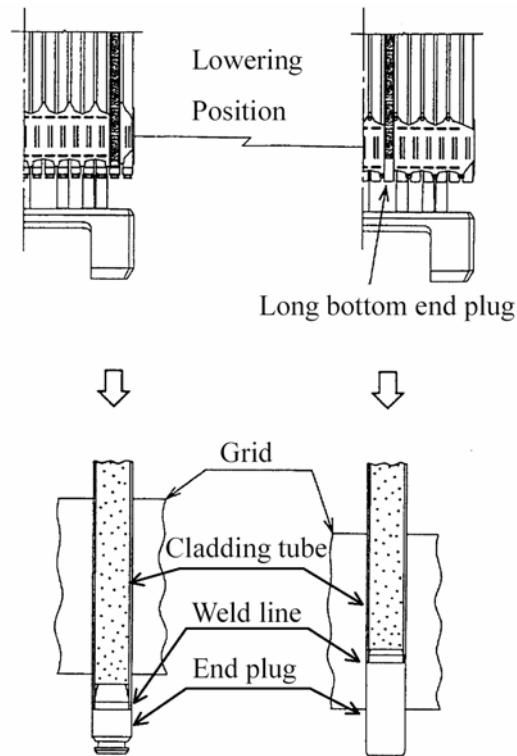


Figure 2-11: Debris resistant design fuel Shimomura, et al., 2004.

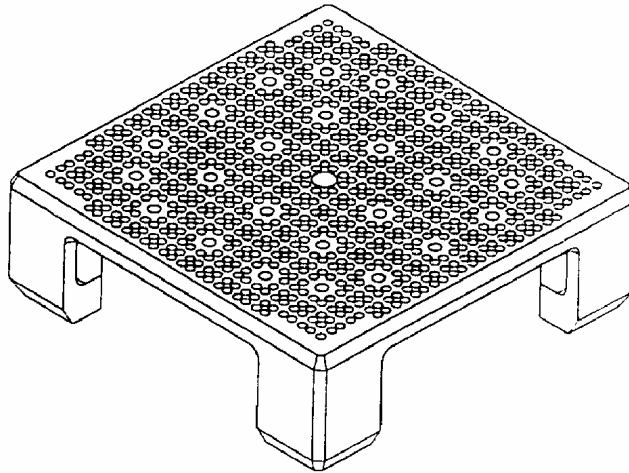


Figure 2-12: Debris filter bottom nozzle Shimomura, et al., 2004.

For the 17x17 type fuel assembly design, top and bottom low-pressure drop nozzles (LPBN or LPTN) with enlarging flow area holes were introduced. Figure 2-13 shows the low-pressure bottom nozzle. A debris filter (DF) is installed right on LPBN, Figure 2-14. Moreover, a skirt is installed in LPBN to prevent debris passing through from the bottom nozzle to the gap between fuel assemblies.

The integrated debris filter bottom nozzle and long thin Inconel straps are specific design features in the 14x14 type and a 15x15 type fuel assembly, Figure 2-15. This type debris filter can trap smaller debris than the DFBN according to the authors. The taper of the bottom end plug of the rod is enlarged in order to compensate pressure drop increased due to the built-in debris filter. Also, a skirt is installed in the bottom nozzle to improve the debris trap performance similarly as the 17x17 design mentioned above.



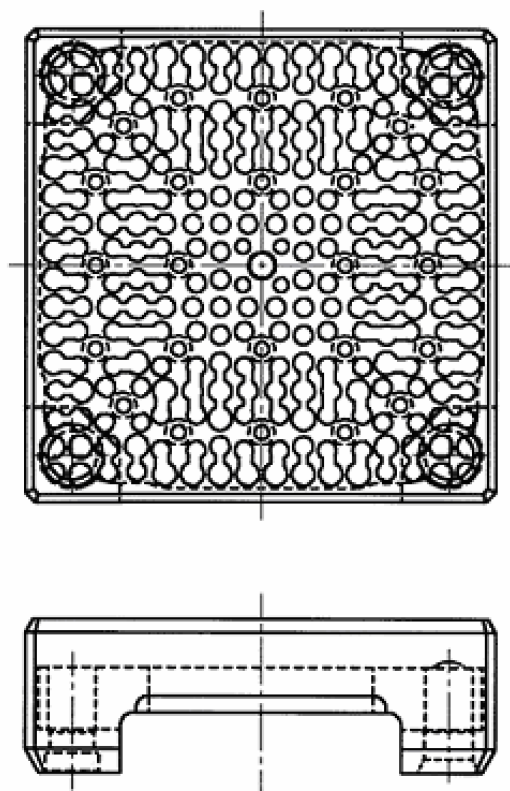


Figure 2-13: Low pressure bottom nozzle, Shimomura, et al., 2004.

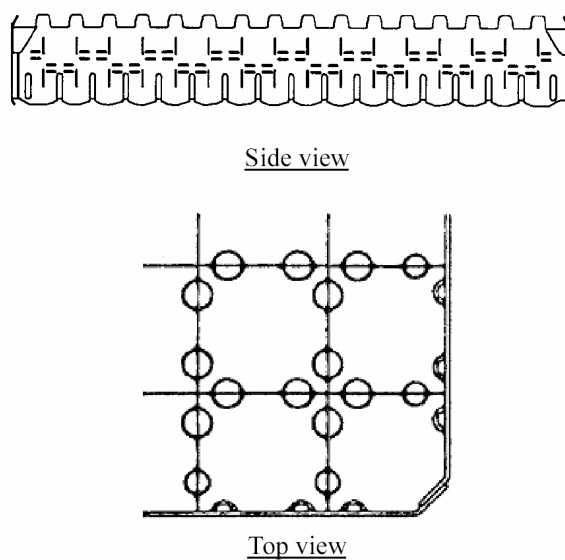


Figure 2-14: Debris filter Shimomura, et al., 2004.

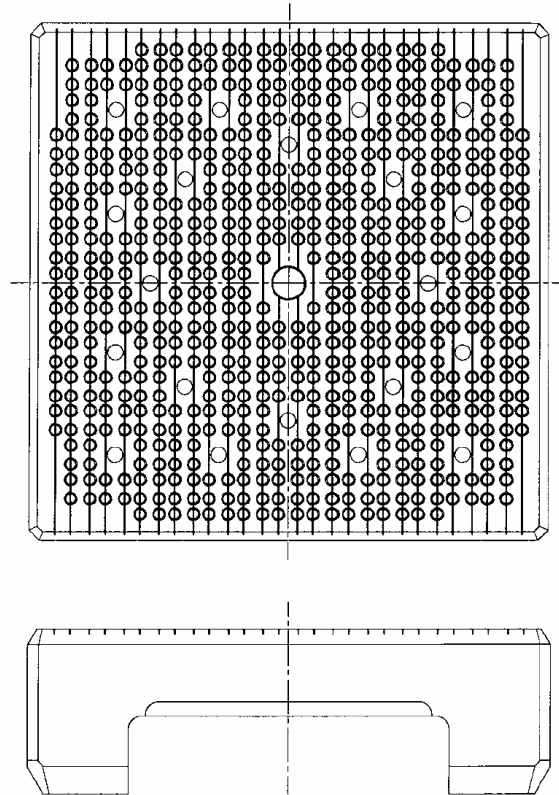


Figure 2-15: Debris filter integrated in the bottom nozzle (15x15), Shimomura, et al., 2004.

To reduce the tendency for grid-rod fretting failures, the following fuel design modifications were introduced already in 1988, and since then no grid-rod-fretting failure has occurred, Shimomura, et al., 2004:

- 1) Eliminations of the flow hole on the guide vane and the guide tab which original function was to decrease the pressure difference between outside and inside of the grid, Figure 2-16.
- 2) Structural damping of the fuel assembly was increased by decreasing the number of bulge forming from three to one, Figure 2-17.

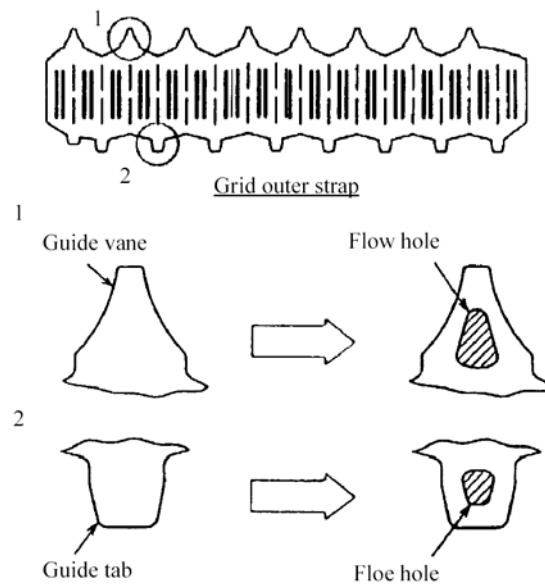


Figure 2-16: The countermeasure against vibration of a grid Shimomura, et al., 2004.

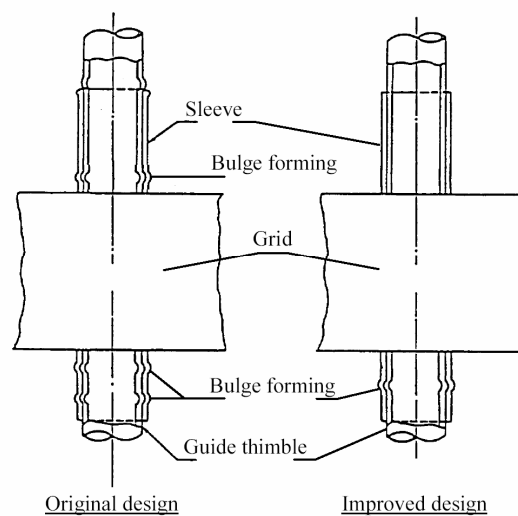


Figure 2-17: The countermeasure to increase structural damping of a fuel assembly. Shimomura, et al., 2004.

The newly development type *I* grid is assumed to further improve the resistance towards grid-to-rod fretting failures according to the authors, Figure 2-18. Figure 2-19 shows the improved spring characteristics compared to the previous design. It can be seen that the new design provides an increased spring force over a larger spring deflection distance. Also, the new *I* grid design allows rods to be loaded into the fuel assembly without any contact with the grid springs and dimples. This feature may prevent the grid spring damage that could otherwise result in grid-rod fretting failures.

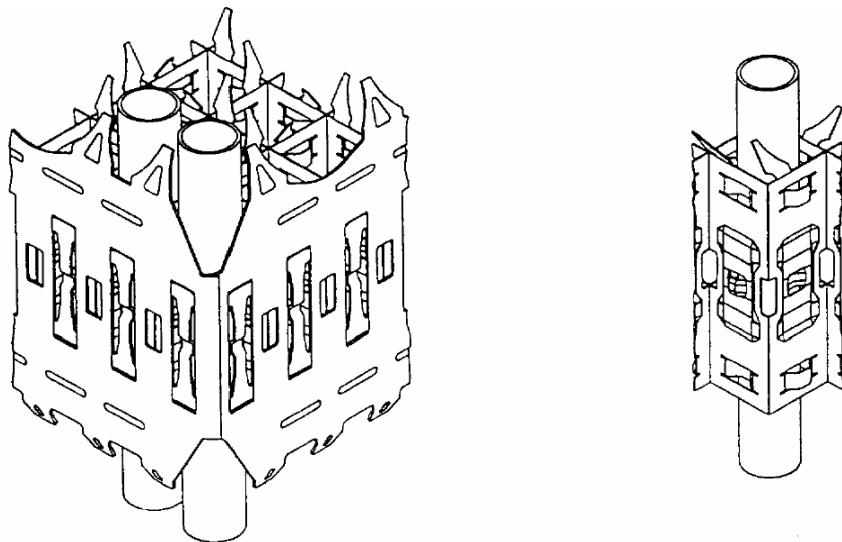


Figure 2-18: I type grid Shimomura, et al., 2004.

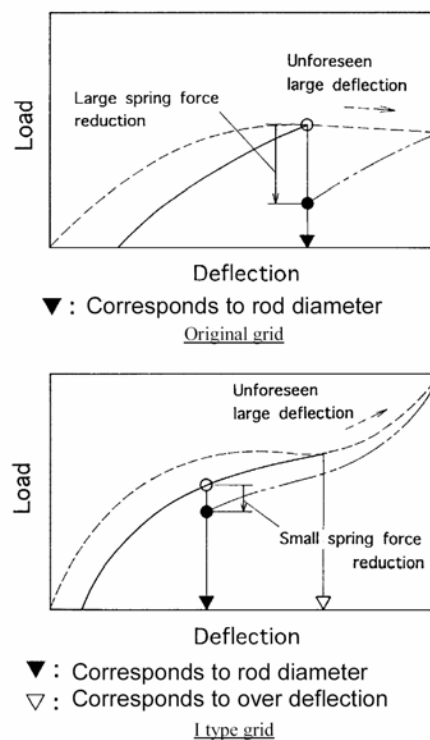


Figure 2-19: The spring characteristic Shimomura, et al., 2004.

To reduce the tendency for incomplete rod insertion (IRI) events, fuel design features were introduced to reduce fuel assembly bowing by increasing the stiffness of the fuel assembly by increasing the diameter of the lower span guide thimble, Figure 2-20.

To ensure appropriate damping of the RCCA during its insertion despite the increased diameter of the dashpot region, a new type of thimble screw was designed, Figure 2-21. The new screw design has small diameter of the flow hole, resulting in a larger pressure drop of the downward flow that is generated by the insertion of the RCCA at the shutdown event. The larger pressure drop would result in increased damping.

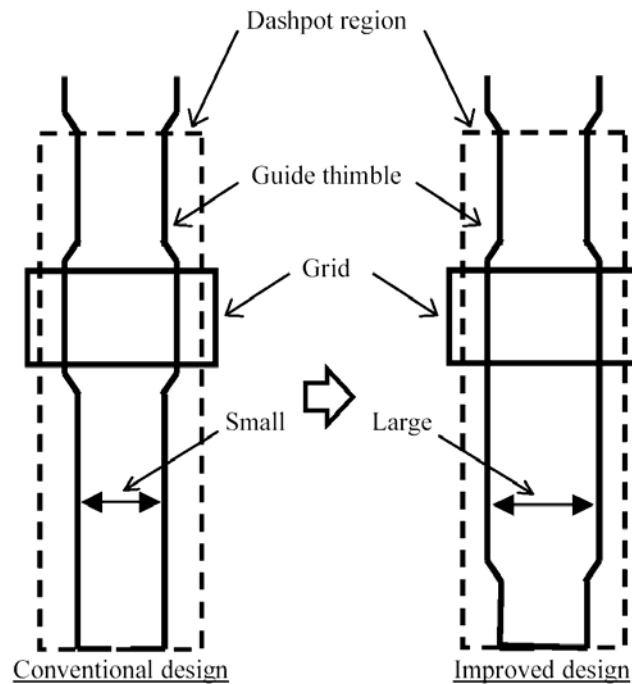


Figure 2-20: The outline of the improved guide thimble Shimomura, et al., 2004.

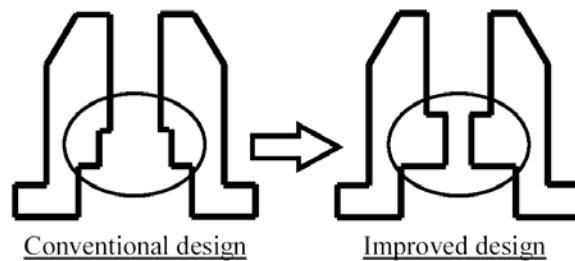


Figure 2-21: the outline of the improved thimble screw Shimomura, et al., 2004.

A high thermal performance grid, Figure 2-22, has also been introduced, with the following characteristics:

- Higher DNB performance and lower pressure-drop by the design improvement of the mixing vanes.
- Improved seismic performance by the design improvement of the grid strap.
- Reduction of the stress at the springs by the design improvement of the spring shape.

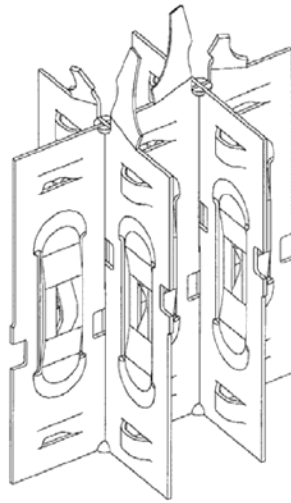


Figure 2-22: High thermal performance grid Shimomura, et al., 2004.

To reduce the risk of getting SCC(PWSCC) failures in the top nozzle leaf spring (LS) a new spring is being developed with increased thickness which reduces the spring stress level, Figure 2-23.

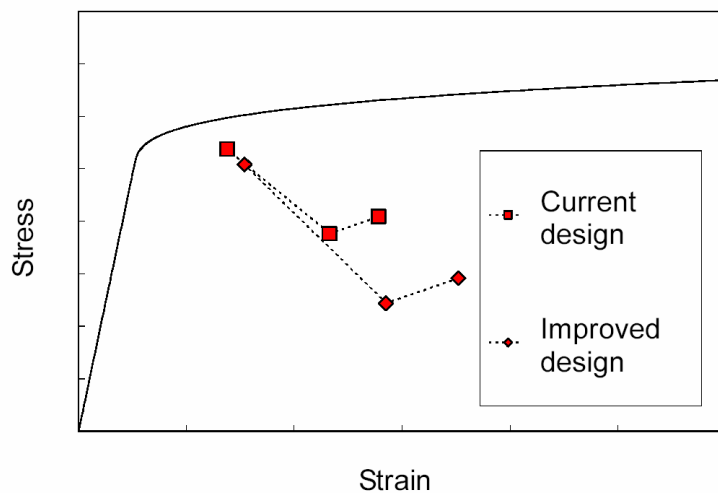


Figure 2-23: Improved and Current Leaf Spring stress Shimomura, et al., 2004.

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### 3 ZIRCONIUM ALLOY SYSTEMS (PETER RUDLING)

#### 3.1 INTRODUCTION

The material performance in-reactor is a function of the reactor environment as well as the material microstructure. The microstructure depends on the chemical composition and the manufacturing process of the alloy. Characterisation of the material microstructure in relation to the material performance can elucidate the mechanisms behind the material performance. This knowledge can be used by fuel vendors to modify the manufacturing process to get optimum material performance in-reactor. Table 3-1 provides a summary of commercial and experimental alloys.

Table 3-1: Zirconium base material currently used.

| Alloy                         | Sn<br>% | Nb<br>% | Fe<br>%    | Cr<br>%   | Ni<br>%   | O<br>%    | Others<br>alloy elem. | %   | Ref. |
|-------------------------------|---------|---------|------------|-----------|-----------|-----------|-----------------------|-----|------|
| <b>1. Commercial alloys</b>   |         |         |            |           |           |           |                       |     |      |
| Zircaloy-2                    | 1.2-1.7 | -       | 0.07-0.2   | 0.05-0.15 | 0.03-0.08 | 0.1-0.14  | -                     | -   | 1)   |
| Zircaloy-4                    | 1.2-1.7 | -       | 0.18-0.24  | 0.07-0.13 | -         | 0.1-0.14  | -                     | -   | 1)   |
| E-110                         | -       | 0.9-1.1 | 0.014      | <0.003    | 0.0035    | 0.05-0.07 | -                     | -   | 2)   |
| Alloy E125                    | -       | 2.5     | -          | -         | -         | 0.06      | -                     | -   | 3)   |
| Zr2.5Nb                       | -       | 2.4-2.8 | <0.15      | -         | -         | 0.09-0.13 | -                     | -   | 1)   |
| ZIRLO                         | 1       | 1       | 0.1        | -         | -         | -         | -                     | -   | 4)   |
| DX-ELS-Liner                  | 0.5/0.8 | -       | 0.3/0.5    | 0.2       | -         | 0.12      | -                     | -   | 5)   |
| PCA-2b                        | 1.3     | -       | 0.3        | 0.2       | -         | -         | -                     | -   | 5)   |
| M5                            | -       | 0.8-1.2 | 0.015-0.06 | -         | -         | 0.09-0.12 | -                     | -   | 6)   |
| <b>2. Experimental alloys</b> |         |         |            |           |           |           |                       |     |      |
| Zircaloy-1                    | -       | 2.5     | -          | -         | -         | -         | -                     | -   | 7)   |
| Zircaloy-3A                   | 0.25    | -       | 0.25       | -         | -         | -         | -                     | -   | 7)   |
| Zircaloy-3B                   | 0.5     | -       | 0.4        | -         | -         | -         | -                     | -   | 7)   |
| Zircaloy-3C                   | 0.5     | -       | 0.2        | -         | 0.2       | -         | -                     | -   | 7)   |
| Zr1Sn0.5Fe                    | 1       | -       | 0.5        | -         | -         | 0.1       | -                     | -   | 8)   |
| ZrSnFe                        | 1.3-1.5 | -       | 0.26-0.3   | <0.05     | -         | -         | -                     | -   | 9)   |
| M4                            | 0.5     | -       | 0.6        | -         | -         | 0.12      | V                     | 0.3 | 10)  |
| D2                            | 0.5     | -       | 0.4        | -         | -         | 0.1       | -                     | -   | 11)  |
| High Fe Zry-2                 | 1.5     | -       | 0.26       | 0.10      | 0.05      | -         | -                     | -   | 12)  |
| HighFeNi Zry-2                | 1.4     | -       | 0.26       | 0.10      | 0.10      | -         | -                     | -   | 12)  |
| HIFI                          | 1.5     | -       | 0.4        | 0.10      | 0.08      | -         | -                     | -   | 17)  |

Table 3-1: Zirconium base material currently used, Adamson, et al., 2002. Cont'd.

| Alloy                                 | Sn<br>% | Nb<br>% | Fe<br>% | Cr<br>% | Ni<br>% | O<br>%    | Others<br>alloy elem. | %       | Ref. |
|---------------------------------------|---------|---------|---------|---------|---------|-----------|-----------------------|---------|------|
| <u>2. Experimental alloys, Cont'd</u> |         |         |         |         |         |           |                       |         |      |
| ZrSnFeO                               | 0.5     | -       | 0.4     | -       | -       | 0.22      | -                     |         | 13)  |
| Alloy-C                               | 0.4     | -       | 0.5     | 0.24    | -       | 0.18      | -                     |         | 13)  |
| HPA-4                                 | 0.4-0.6 | -       | Fe      | -       | -       |           | V                     |         | 14)  |
| Valloy                                | -       | -       | 0.15    | 1.2     | -       | -         | -                     |         | 7)   |
| Zr.7Fe0.7Ni                           | -       | -       | 0.7     | -       | 0.7     | 0.03      | -                     |         | 8)   |
| Zr0.25Fe0.2V                          | -       | -       | 0.25    | -       | -       |           | -                     |         | 7)   |
| E635                                  | 1.1-1.4 | 0.9-1.1 | 0.3-0.5 | -       | -       | 0.05-0.07 | -                     |         | 3)   |
| Alloy-A                               | 0.5     | 0.3     | 0.35    | 0.25    | -       | 0.15      | -                     |         | 13)  |
| Alloy-E                               | 0.7     | 0.4     | 0.45    | 0.24    | -       | 0.13      | -                     |         | 13)  |
| NSF 0.2                               | 1       | 1       | 0.2     | -       | -       | 0.1       | -                     |         | 7)   |
| T18/I18                               | 1       | 0.6     | 0.2     | -       | 0.05    |           | -                     |         | 15)  |
| NSF 0.5                               | 1       | 1       | 0.5     | -       | -       | 0.1       | -                     |         | 15)  |
| Zr3Nb1Sn                              | 1       | 2-3     | -       | -       | -       |           | -                     |         | 9)   |
| Ozhenite 0.5                          | 0.2     | 0.1     | 0.1     | -       | 0.1     |           | -                     |         | 7)   |
| M3                                    | 0.5     | 0.5     | 0.25    | -       | -       | 0.12      | -                     |         | 10)  |
| 0.2 Nb Zry-2                          | 1.5     | 0.2     | 0.15    | 0.10    | 0.06    |           | -                     |         | 15)  |
| 0.5 Nb Zry-2                          | 1.5     | 0.5     | 0.15    | 0.1     | 0.05    |           | -                     |         | 12)  |
| D3                                    | 1.4     | 1       | 0.2     | 0.1     | -       |           | -                     |         | 11)  |
| EXCEL                                 | 3.5     | 0.8     | -       | -       | -       |           | Mo                    | 0.8     | 7)   |
| XXL                                   | 1.2     | 0.3     | -       | -       | -       |           | Mo                    | 0.3     | 12)  |
| T12-15/I12-15                         | 1       | 1-2     | -       | -       | -       |           | Mo                    | 0.2-0.5 | 15)  |
| T19/I19                               | 1.4     | 0.4     | -       | -       | -       |           | Te                    | 0.2     | 15)  |
| T20/I20                               | 1.2     | -       | -       | -       | -       |           | Te                    | 0.6     | 15)  |
| BAG                                   | -       | 0.5     | -       | -       | -       |           | Bi                    | 1       | 12)  |
| T68                                   | 0.8     | -       | 0.3     | 0.1     | 0.1     |           | Cu/Ta                 | 0.2/0.2 | 16)  |
| T40                                   | 1       | -       | 0.25    | 0.1     | -       |           | Cu/Ta                 | 0.1/0.2 | 16)  |
| MDA                                   | 0.8     | 0.5     | 0.2     | 0.1     | -       |           |                       |         | 18)  |
| NDA                                   | 1       | 0.1     | 0.27    | 0.16    | 0.01    |           |                       |         | 19)  |

<sup>1)</sup> ASTM; <sup>2)</sup> Shebaldov, et al., 2000; <sup>3)</sup> Solonin, et al., 1999; <sup>4)</sup> Comstock, et al., 1996; <sup>5)</sup> Garzarolli, 2001;

<sup>6)</sup> Mardon, et. al., 2000; <sup>7)</sup> Cox, et al., 1998; <sup>8)</sup> Amaev, 1971; <sup>9)</sup> Garzarolli, et al., 2001; <sup>10)</sup> Mardon, et. al., 1994(b);

<sup>11)</sup> Besch, et al., 1996; <sup>12)</sup> Ishimoto, et. al., 2000; <sup>13)</sup> Garde, et al., 2001; <sup>14)</sup> Seibold, 2001; <sup>15)</sup> Etoh, et al., 1996;

<sup>16)</sup> Takeda and Anada, 2001, 17) Ishimoto et al., 2003, 18) Tsukada et al., 2003, 19) Tsukada et al., 2003.



### 3.2 NEW RESULTS

#### 3.2.1 Zr

Dubinko, et al., 2004, presented a model of the homogeneous nucleation and growth of vacancy and interstitial loops in irradiated hcp metals. The model is based on a hierarchy of coupled ordinary differential equations. The first two equations are the rate equations for vacancy and interstitial concentrations. Other equations describe random walks of interstitial and vacancy clusters in a size space, i.e. the time dependence of loop densities. As an input, the model contains the capture efficiencies of point defects by loops, which depend self-consistently on the loop size and dislocation density. The model results are qualitatively consistent with experimental observations of a coexistence of interstitial and vacancy dislocation loops on the same habit planes in Zr and other hcp metals.

#### 3.2.2 Zircalloys

##### 3.2.2.1 Second phase particles

TEM investigations in Zr-Fe and Zr-Fe-Ni, Zr-Ni-Cr and Zr-Cr-Fe alloys, Table 3-2 were performed to assess the second particles present in these alloys, either in quasi equilibrium conditions, or after quenching and annealing experiments, Barberis, et al., 2004(a). The study showed that  $Zr_3Fe$  dissolved Ni up to 12at% at equilibrium conditions. Ab initio computations further provided the formation enthalpies for several Zr-Fe compounds, and showed that the  $Zr_2Fe$  is metastable at low temperature. In addition, quenching and annealing experiments suggested that  $Zr_2Fe$  is formed for low quenching rates, which can be attributed to the oxygen segregation during the beta to alpha phase transformation. The results from the TEM studies were compared to the thermodynamic database ZIRCOBASE and the results were in good agreement, Figure 3-1.

Table 3-2: Chemical composition of the ternary alloys, Barberis, et al., 2004(a).

| Mark    | 1     | 2      | 3      | 41    | 5      | 61    | 71    | 91    | 101   | 111   | 12    | 13    |
|---------|-------|--------|--------|-------|--------|-------|-------|-------|-------|-------|-------|-------|
| Fe (%)  | 0.103 | 0.120  | 0.450  | 0.155 | 0.045  | 0.104 | 0.120 | 0.150 | 0.016 | 0.013 | 0.016 | 0.013 |
| Cr (%)  | 0.095 | 0.070  | 0.435  | 0.043 | 0.150  | 0.002 | 0.002 | 0.002 | 0.105 | 0.12  | 0.472 | 0.149 |
| Ni (%)  | 0.001 | <0.001 | <0.001 | 0.001 | <0.001 | 0.102 | 0.071 | 0.038 | 0.100 | 0.069 | 0.502 | 0.043 |
| O (ppm) | 630   | 610    | 640    | 570   | 600    | 680   | 650   | 650   | 670   | 690   | 740   | 740   |

## 4 MECHANICAL PROPERTIES

### 4.1 INTRODUCTION (PETER RUDLING)

The mechanical properties of essentially two different components are treated in this section. First, the *LWR* fuel assembly and, second, the *Pressure tubes* 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 perform satisfactorily during the lifetime of the reactor.

Delayed hydride cracking, *DHC*, is a failure mechanism that may limit the lifetime of *CANDU* and *RBMK* pressure tubes, and this mechanism is therefore treated in the pressure tube section. Delayed Hydride Cracking, *DHC*, is a fracture mechanism that may result in pressure tube failures as well as degradation of failed *LWR* fuel. A better understanding of the relation of the *DHC* mechanism to the material properties may e.g. assist the fuel vendors to develop products with enhanced resistance towards *DHC*.

In a *CANDU* reactor the cold-worked Zr-2.5Nb pressure tubes operate at temperatures between about 250 and 310°C and at coolant pressures of about 10 MPa corresponding to hoop stresses of about 130 MPa. The maximum flux of fast neutrons from the fuel is about  $4 \cdot 10^{17} \text{ n m}^{-2} \text{ s}^{-1}$ .

The pressure tubes used in a *CANDU* reactor are made from Zr-2.5Nb. The tubes are extruded at 815°C cold worked 27% and stress relieved at 400°C for 24 hours, resulting in a structure consisting of elongated grains of hexagonal-close-packed (HCP)  $\alpha$ -Zr, partially surrounded by a thin network of filaments of body-centered-cubic  $\beta$ -Zr. These  $\beta$ -Zr filaments are metastable and initially contain about 20% Nb. The stress-relief treatment results in partial decomposition of the  $\beta$ -Zr filaments with the formation of hexagonal-close-packed  $\omega$ -phase particles that are low in Nb, surrounded by an Nb-enriched  $\beta$ -Zr matrix. The hcp  $\alpha$ -Zr grains are oriented with their unique c-axes aligned in the radial-transverse plane, mostly tilted towards the transverse direction.

The mechanical properties of the *LWR* fuel assembly is 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 behavior 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 accidents 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 taken 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 mechanically 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 sufficient 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<sup>1</sup> phase during *LOCA* and due to *PCMI*<sup>2</sup> during a *RIA* transient.

Fuel vendors have developed codes to model the fuel assembly mechanical performance during class I, II, III and IV situations. To be able to do the modeling correctly, data on mechanical performance of the fuel assembly must exist. The data are generated in two types of tests, either separate effect tests and integral tests. The former test studies only the impact of one parameter at a time on the mechanical performance, see e.g. Adamson & Rudling, 2001, 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*.

#### 4.2 EFFECTS OF HYDRIDES (RON ADAMSON)

Earlier work (reviewed in Adamson & Rudling, 2001 and Strasser, et al., 2003 has shown:

- The effect of hydrides is strongly temperature dependent. Material at reactor operating temperature (300-360°C (573-633K)) retains much more ductility than at 20°C (293K).
- At least for unirradiated material, *SRA* Zircaloy retains more ductility than *RXA* material.
- Hydride distribution is very important. Dense layers of hydrides (for instance, at fuel cladding outer surface) retain little ductility at any temperature, and are susceptible to crack formation. Whether or not the crack will be arrested by the relatively ductile Zircaloy under-layer depends on the layer thickness.
- Irradiated material at 300°C (573K) retains adequate ductility to at least 1200 ppm hydrogen.

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<sup>1</sup>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.

<sup>2</sup> Pellet Cladding Mechanical Interaction, i.e., interaction without the influence of iodine (that would instead result in *PCI* (Pellet Cladding Interaction)).

## 5 DIMENSIONAL STABILITY (RON ADAMSON)

### 5.1 INTRODUCTION

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 approximately 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 by Adamson, 2000, and the ZIRAT Special Topical Report on Mechanical Properties, Adamson & Rudling, 2001.

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, possibly leading to bowing problems. (For calibration, a recrystallized (RX or *RXA*) 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, at least in part, to the fact that the volume of zirconium hydride is about 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.

A comprehensive review of dimensional stability has been given in the ZIRAT 7 Special Topical Report; Adamson, et al., 2002. The sources of dimensional changes of reactor components (in addition to changes caused by conventional thermal expansion and contraction) are: irradiation growth, irradiation creep, thermal creep, stress relaxation (which is a combination of thermal and irradiation creep), and hydrogen and hydride formation.

Irradiation effects are primarily related to the flow of irradiation-produced point defects to sinks such as grain boundaries, deformation-produced dislocations, irradiation-produced dislocation loops, and alloying and impurity element complexes. In zirconium alloys, crystallographic and diffusional anisotropy are key elements in producing dimensional changes.

In the past, hydrogen effects have been considered to be additive to and independent of irradiation; however, recent data have brought this assumption into question. It is certain that corrosion-produced hydrogen does cause significant dimensional changes simply due to the 16-17% difference in density between zirconium hydride and zirconium. A length change of on the order of 0.25% can be induced by 1000 ppm hydrogen in an unirradiated material. Whether or not the presence of hydrides contributes to the mechanisms of irradiation creep and growth is yet to be determined.

Fuel rod diametral changes are caused by stress dependent creep processes. Fuel rod length changes are caused by several phenomena:

- Stress free axial elongation due to irradiation growth.
- Anisotropic creep (before pellet/cladding contact) due to external reactor system pressure. Because of the tubing texture, axial elongation results from creep down of the cladding diameter; however for heavily cold worked material, it has been reported that some shrinkage may occur. In a non-textured material such as stainless steel, creep down of the cladding would only result in an increase in cladding thickness, with no change in length.
- Creep due to pellet-cladding mechanical interaction (*PCMI*) after hard contact between the cladding and fuel. This occurs in mid-life, depending on the cladding creep properties and the stability of the fuel.
- Hydriding of the cladding due to corrosion.

Bow of a component such as a *BWR* channel or *PWR* control rod assembly can occur if one side of the component changes length more than the other side. Such differential length changes occur due to differential stress and creep, to relaxation of differential residual stresses, or to differential growth due to differences in flux-induced fluence, texture, material cold work, and hydrogen content (and, although not usually present, differences in temperature or alloying content).

In the ZIRAT 8 Annual Report the following areas were listed as requiring more data and understanding to assure optimum component performance at the high fluences and burnups currently achieved or proposed for modern fuel designs:

- Empirical understanding of most component dimensional stability issues is good. Areas where more data is required include:
  - detailed temperature dependence of creep and growth in the 300-400°C (573–675K) range
  - possible synergistic effects between hydrogen and growth/creep mechanisms.
  - data out to projected end-of-life burnups of newer alloys such as ZIRLO and M5
  - factors which affect bow and lean of components (*BWR* channels, *PWR* assemblies), including growth, hydrogen and residual stresses.
  - high burnup and high fluence effects in the “constant volume” assumption of irradiation growth.
  - effects of Sn on in-reactor creep and growth.
- Fundamental mechanism understanding is still lacking in several areas
  - critical factors affecting formation of c-component dislocations
  - chemistry and microchemistry effects on growth (for example, why are the growth behaviors of M5 and E110 (both basically Zr-1Nb alloys) so different?)
  - the role and relative importance of various point defect sinks in determining creep and growth characteristics.

In the recent past, the main fuel performance issue in the dimensional stability area has been *BWR* channel bowing, which interferes with control blade insertion in specific plants, Cheng, 2002. The recent literature indicates that also of importance are *PWR* assembly bow, performance of newer alloys to moderate burnups, and performance of all alloys at high burnup (a topic which is addressed in detail in the ZIRAT 8 Special Topics Report, High Burnup Issues, 2003). This review examines what the most recent literature says about these and other issues.

## **6 ZIRCONIUM ALLOY MANUFACTURING (PETER RUDLING)**

### **6.1 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; increased 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 may not apply.

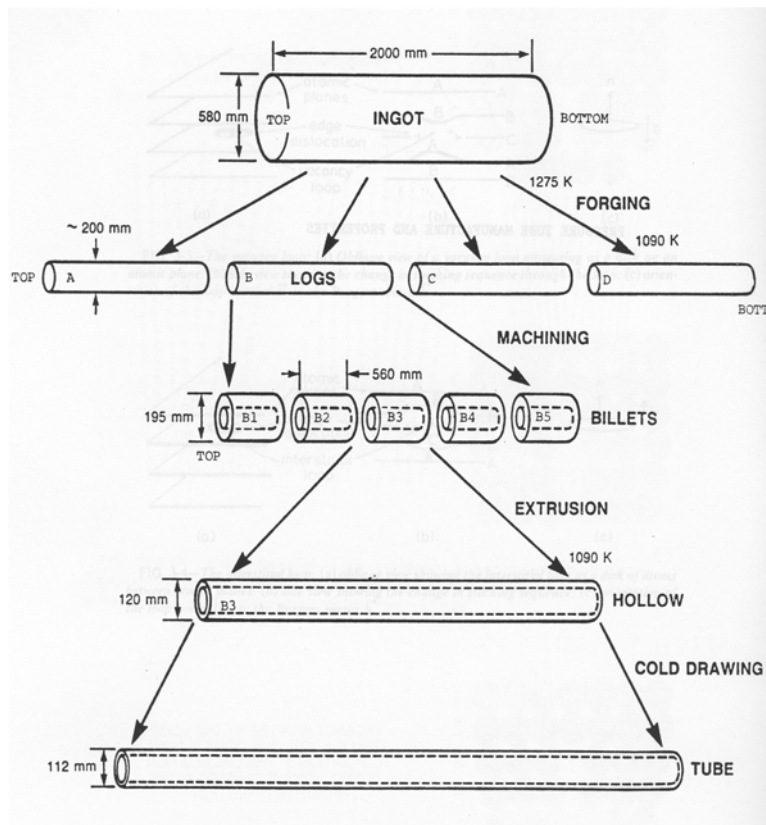
### **6.2 NEW RESULTS**

#### **6.2.1 Pressure tubes**

Li, et al., 2004 studied the correlation between texture and grain morphology, dislocation substructure using TEM/SAD, SEM/EBSD in Zr-2.5 Nb. The study showed that a variety of microstructures were developed in extruded Zr-2.5Nb tubes depending upon the extrusion temperature, extrusion ratio and billet microstructure, Figure 6-1 to Figure 6-2. More specifically, three components of texture were developed dependant on extrusion temperature, Figure 6-3:

- Radial component (low in  $\alpha + \beta$  regime, mostly  $\alpha$  present)
- Transverse component (middle of  $\alpha + \beta$  regime)
- Axial component (high in  $\alpha + \beta$  regime, large fraction of  $\beta$  present)





**vacuum arc melt**

**press forge, rotary forge**

**machine billet**

**beta-quench optional**

**extrude at ~ 815 °C**

**cold draw 25-30%**

Figure 6-1: Manufacturing outline, Li, et al., 2004.

Table 6-1: Extrusion Conditions, Tubes 10 and 20, Li, et al., 2004.

| tube | billet prep | preheat temp, K | preheat time, min | ext. ratio | Q   |
|------|-------------|-----------------|-------------------|------------|-----|
| 10   | quench      | 923             | 180               | 4:1        | 2.6 |
| 20   | slow cool   | 1123            | 30                | 4:1        | 2.6 |

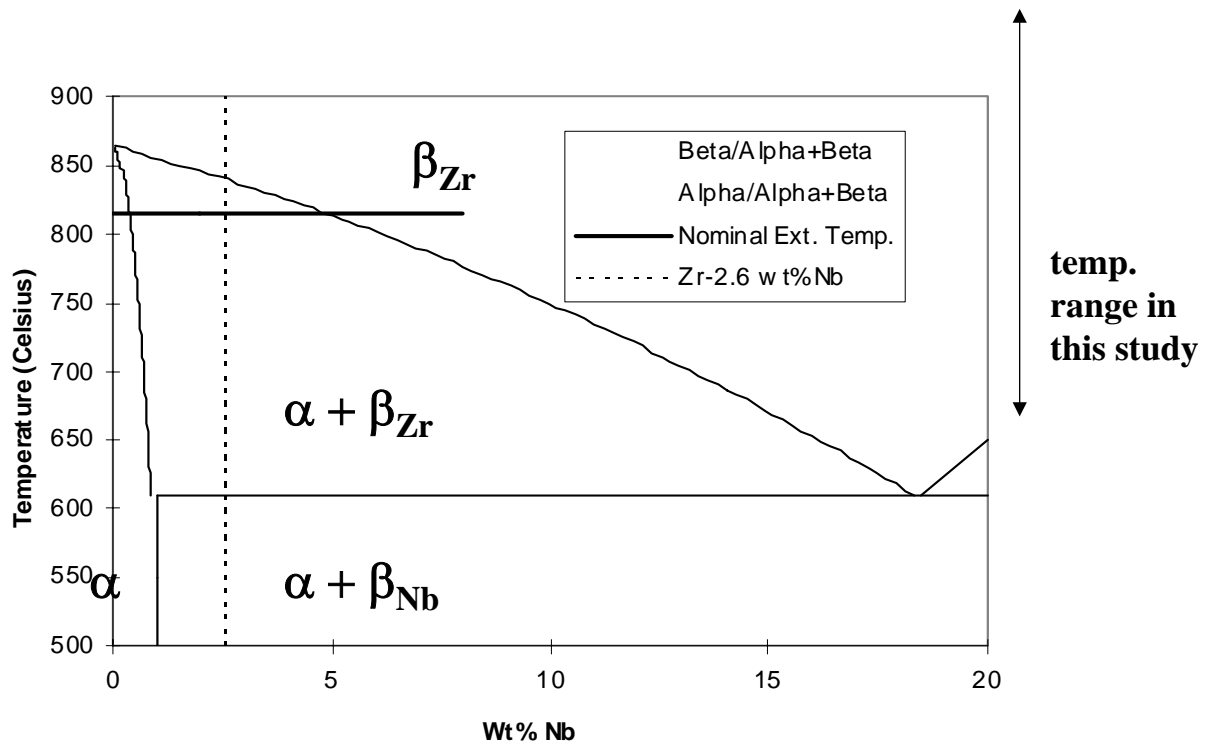


Figure 6-2: Zr-Nb Phase Diagram, Li, et al., 2004.

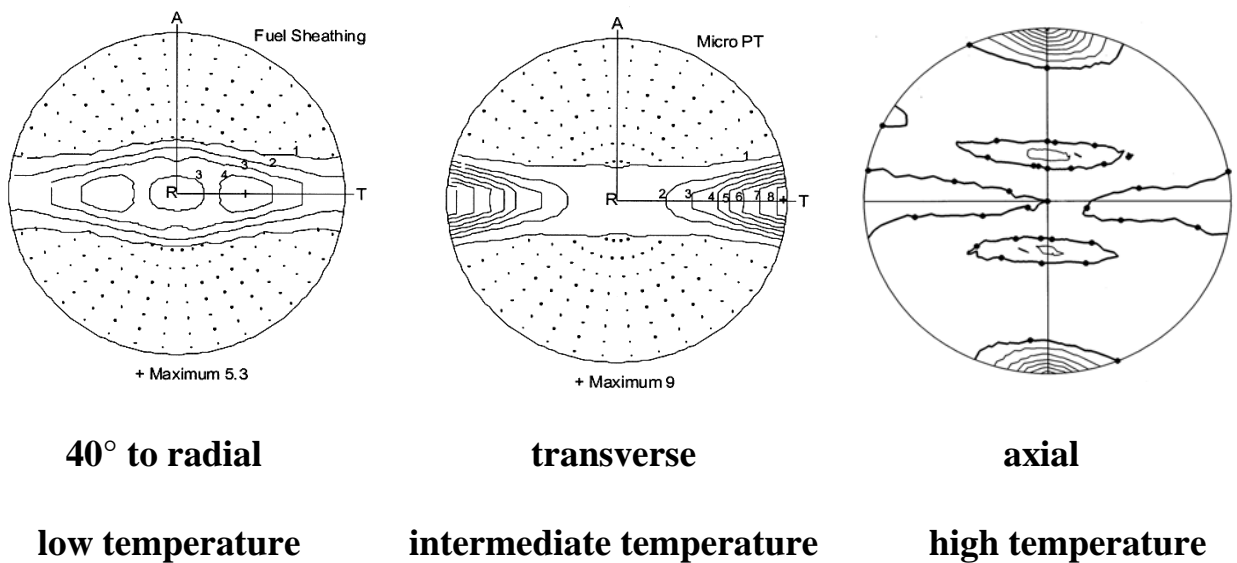


Figure 6-3: Three texture components seen, Li, et al., 2004.

## 7 CORROSION AND HYDRIDING

### 7.1 OUT OF PILE STUDIES (BRIAN COX)

Studies published over the past year can elucidate corrosion mechanisms of zirconium alloys, even if not performed on alloy samples corroded in high temperature water or steam. Thus, studies of the properties of bulk zirconia can tell us something about the behaviour of impurities that could be relevant to the behaviour of the same impurities in oxide corrosion films. Studies of the widely investigated use of thin zirconia films in electronic devices may help us to understand the conduction of electrons through oxide films formed in water or steam, and information about the degradation of such films in electronic devices by water may be relevant to the degradation of oxide films on zirconium alloy fuel cladding. This year's search of the general scientific literature has revealed a number of interesting papers that will be discussed according to the form of zirconia being studied.

#### 7.1.1 Bulk Zirconium Oxide

Tetragonal and cubic zirconias are stabilized by the vacancy concentration induced by the aliovalent ions (Ca, Y, etc.) that are added. As an approximation, about 3% of anion vacancies are necessary to stabilise the tetragonal form, and ~10% are necessary to stabilize the cubic form. It is not necessary for the added element to be cationic; aliovalent anionic additions (e.g. N) are equally capable of stabilizing isotropic zirconias. The questions of how the anionic vacancies are ordered in the stabilized zirconia structures is of interest because of the different stabilities of the different ordering arrangements. The formation and degradation of tetragonal zirconia in zirconium alloy corrosion films is thought to be a potential contributor to oxide film breakdown. Cubic zirconia can also be stabilized by a very small crystallite size (<40 nm) without the necessity for aliovalent additions, and pure cubic zirconia can also be stabilized in multilayer films (with different materials as intermediate layers) if the zirconia layer thickness is <40 nm, Wang et al., 2004, obtained multilayer cubic zirconia films (alternating with ceria layers) in which the P-43m space group oxygen sublattice was displaced diagonally, rather than along the c-axis as is believed to be the case with bulk yttria stabilised zirconia (YSZ). Since there appears to be at least three forms of tetragonal zirconia, which depend on their formation method, and have different axial ratios. This is obviously a complicated system. It is not clear that we know precisely which structural form the tetragonal zirconia crystallites found in zirconia corrosion films belong to, since in neither of the two recent studies by Raman Spectroscopy, Barberis, et al., 2001 and Masler, et al., 2001, do the authors appear to be aware of the different forms of *t*-ZrO<sub>2</sub>. The differences that results from the small displacement of the oxygen atoms in the "pseudo-fluorite" type lattice can be seen in Figure 7-1, Wang et al., 2004.

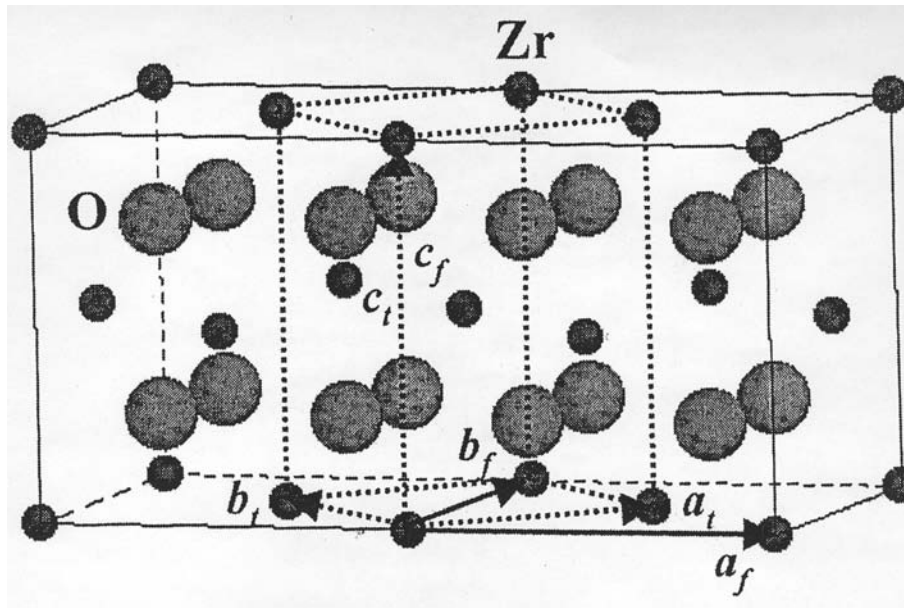


Figure 7-1: Structure of tetragonal  $\text{ZrO}_2$  drawn in the primitive tetragonal (subscripted with  $t$ ) and pseudo-fluorite (subscripted with  $f$ ) unit cell.

Nevertheless, the bulk of the zirconia corrosion film is in the monoclinic form, and we still need better knowledge of this phase if we are to understand the growth and breakdown of these films. Most of the mechanical properties studies on bulk  $m\text{-ZrO}_2$  are conducted on material with a much larger grain size than is typical of zirconia corrosion films, and either at much higher temperatures than are relevant to reactor operation, or at room temperature. The recent study of the fracture toughness of  $m\text{-ZrO}_2$ , Eichler, et al., 2004, was done on material with a grain size of  $<200$  nm (130-160 nm), and is, therefore, giving results, Table 7-1, that

Table 7-1: Characterization and Mechanical Properties of Monoclinic Zirconia.

| Sample            | Relative Density (%) | Grain Size (nm) | $E$ (GPa)   | $\sigma_f$ (MPa) | $K_{Ic}$ (MPa $\text{m}^{1/2}$ )* |
|-------------------|----------------------|-----------------|-------------|------------------|-----------------------------------|
| A (1100°C/(10 h)) | $92.2 \pm 0.4$       | $133 \pm 14$    | $199 \pm 2$ | $234 \pm 47$     | $2.06 \pm 0.04$                   |
| B (1105°C/(5 h))  | $91.1 \pm 0.4$       | $163 \pm 22$    | $190 \pm 2$ | $220 \pm 35$     | $2.00 \pm 0.13$                   |

\*  $E$  = Young's modulus,  $\sigma_f$  = fracture strength,  $K_{Ic}$  = fracture toughness

may be relevant to corrosion films. Note, however, that the grains were equiaxed, and not columnar as is typically the case for corrosion films. Another measurement of Young's Modulus of  $\text{ZrO}_2$  thermal barrier coatings, which were YSZ, Guo & Kagawa, 2004, gave values from 130-170 GPa (the range is related to the heating times up to 200 hr. at 1150°C), in good agreement with the Eichler et al. results, Figure 7-2.

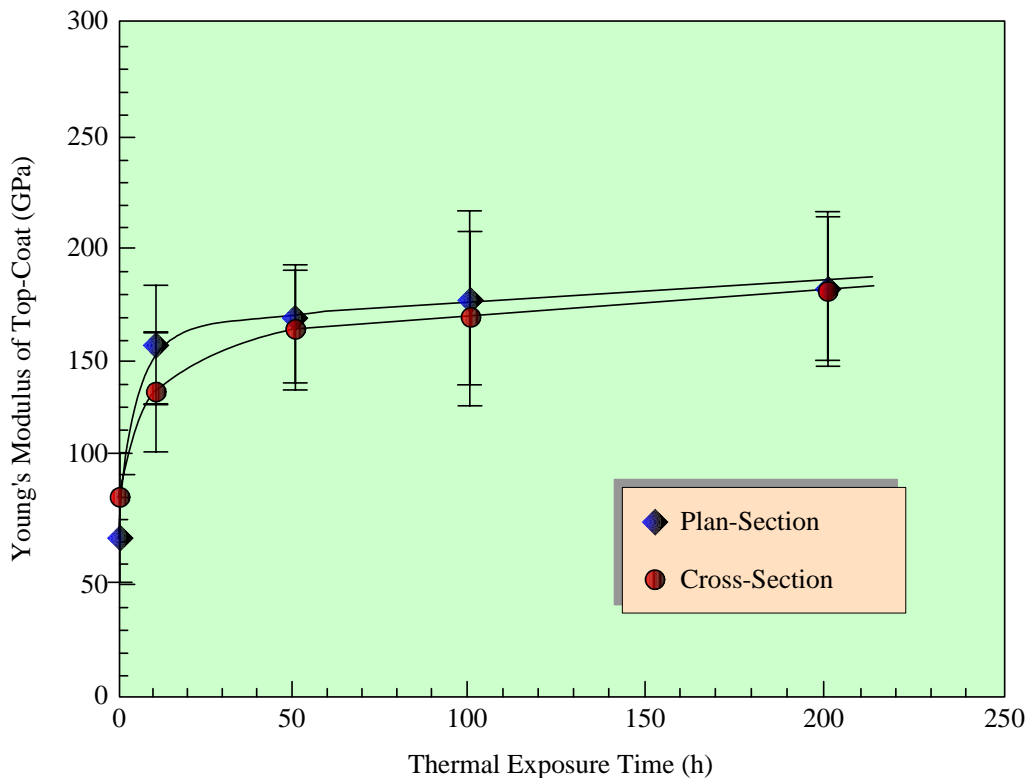


Figure 7-2: Plot of Yong's moduli measured on the plan-section and the cross-section of the top-coat as a function of the thermal exposure time at 1150°C.

The electrical properties of bulk stabilised  $\text{ZrO}_2$  again received attention this year, Li et al., 2003. In the bulk zirconia the grain boundaries are usually barriers to electronic conduction. They can reduce the electronic conductivity of the material by several orders of magnitude. This "grain boundary effect" is usually attributed to the accumulation of some common impurities (e.g. Si, Al, Mg, Ca) at the grain boundaries. Since the negative potential on the metal during oxidation shows that the oxidation of zirconium alloys is controlled by the electronic conductivity of the oxide, rather than by oxygen diffusion, Cox, 2003, we need to know more about the accumulation of impurities in the crystallite boundaries of thermally formed oxides on Zr alloys, and their effect on electronic conductivity. Sn and Si are known to accumulate at crystallite boundaries (see ZIRAT-8, Annual Report) in oxide films and Fe also segregates in crystallites away from the original intermetallic particle, Cox & Sheikh, 1997 and Li et al., 2003, varied the grain size in cubic  $\text{ZrO}_2$  stabilised by a mixture of Ca and Y and with constant, but unspecified, Si + Fe impurity level. They measured the grain boundary resistance using a Frequency Response Analyser (Solartron?) in the temperature range 600-1100 K and the frequency range  $10^2$ – $10^7$  Hz. From the plots of  $\sigma_{gb}$  versus inverse temperature the constants affecting grain boundary conduction could be plotted versus grain size, Figure 7-3. A rough extrapolation of these constants to typical grain sizes

## 8 EFFECTS OF WATER CHEMISTRY

### 8.1 *PWR WATER CHEMISTRY (FRIEDRICH GARZAROLLI)*

The water chemistry of *PWRs* is governed by following additions:

- Addition of hydrogen (2-4 ppm). In the VVERs  $\text{NH}_3$  is added which decays to  $\text{H}_2$  by radiolysis.
- Addition of boric acid for reactivity control (1000-1600 ppm B at BOC).
- Addition of LiOH (2-5 ppm at BOC) to counterbalance the pH decrease by boric acid. The strategy of Li additions (the pH strategy) has changed over the years and is still changing. In the VVERs KOH is used instead of LiOH.
- An increasing number of *PWRs* add Zn (5-40 ppb) to reduce (1) the plant activation, (2) ISCC of SG Inconel 600 tubing, and (3) to reduce the metal release from structural parts by corrosion.

As far as the fuel elements are concerned the most important issues are:

- Deposition of corrosion products from the coolant and formation of degrading CRUD especially at positions with sub-cooled boiling. Heavy CRUD can cause accelerated corrosion defects locally and axial power shift by boron precipitation (AOA).
- For the pH strategy, believed to be best, rather high LiOH concentrations are necessary at least at BOC. LiOH however can reduce the corrosion resistance if it concentrates above a critical value. Thus new strategies have to be explored carefully.
- Zn addition reduce the corrosion product release from the structural materials, but increases the Fe, Ni, and Co concentration of the coolant what may lead to a more degrading CRUD at positions with high steaming rates at least in the first cycles after introduction. Thus surveillance programs are usually applied after introduction of this strategy for high duty *PWRs*.
- Changes of water chemistry guidelines.

Fruzzetti, et al., 2004, reviewed the EPRI water chemistry guidelines and described the recent modifications. The most important changes for fuel rod considerations are:

- The new guidelines provide increased emphasis on the desirability of using constant elevated pH(T), such as constant pH(T) between 7.1 and 7.3, at all plants, but especially at those with high duty cores. Constant elevated pH(T) provides benefit in CRUD management, fuel deposits, AOA, and shut down dose rates. The Li level at which consideration of fuel vendor review is recommended is increased from >2.2 to >3.5 ppm.

- With regard to the core duty, a new parameter called the High Duty Core Index (HDCI) is proposed to be applied e.g. for AOA considerations. HDCI is calculated from the average heat flux of the peak fuel assembly, the coolant flow, and the coolant exit temperature of the plant.
- The guidelines recommend for all plants to consider the use of zinc due to the dose reduction benefit. The increased experience with use of zinc additions to the primary coolant is reviewed and it is pointed out that use of zinc still needs successful completion of demonstration program for high duty cores.

### 8.1.1 PWR Fuel CRUD and AOA

Several papers reported on the effects that influence CRUD formation and on observed consequences of heavy CRUD layers. Historically, U.S. PWRs have experienced less cladding defects due to corrosion than U.S. BWRs. Only three cases of CRUD induced fuel defects occurred since 1995, Turnage, et al., 2004.

Tropasso, 2004 reported on the post irradiation examinations of the TMI-1/cycle 10 failures. TMI-1 experienced CRUD induced failures during Cycle 10 in 1995. The TMI-1 Cycle 10 core was designed for a 24-month cycle, 661 EFPD (692 calendar days). The reload consisted of 80 fresh fuel assemblies arranged in a modified checkerboard array that included four adjacent fresh fuel assemblies near the core periphery in a “T” arrangement. Power distribution was controlled by burnable poison assemblies and gadolinia contained in selected fuel rods. The BOC steady state full power boron concentration was very high (1851 ppm). Lithium (limited to  $\leq 2.2$  ppm) was not added at BOC, Li was allowed to burn in from the  $B^{10}+n \rightarrow \alpha+Li^7$  reaction. Thus the pH(T) at BOC was rather low ( $\sim 6.7$  after some days, and 6.9 after 24 weeks). After 121 calendar days an increase in fission product activity indicated a leaking fuel rod. Transient cesium ratios pointed out that the leaking rod was from the feed fuel batch. Several additional increases occurred periodically throughout the remainder of the cycle. Visual inspection during the refueling outage revealed an unusual CRUD pattern on the outer face of upper spans of peripheral rods of many of the feed assemblies. Nine failed rods were found during the poolside inspection, all rods having the distinctive CRUD pattern. Localized cladding penetration due to CRUD induced localized corrosion was identified as the failure mechanism as a consequence of the high corrosion product entering due to the low pH(T) at BOC. For confirmation, a hot cell investigation was performed. PIE was performed on four fuel rods that failed or showed increased EC indications after cycle 10. The hot cell PIE results confirmed that the defects were due to locally enhanced corrosion, Figure 8-1.



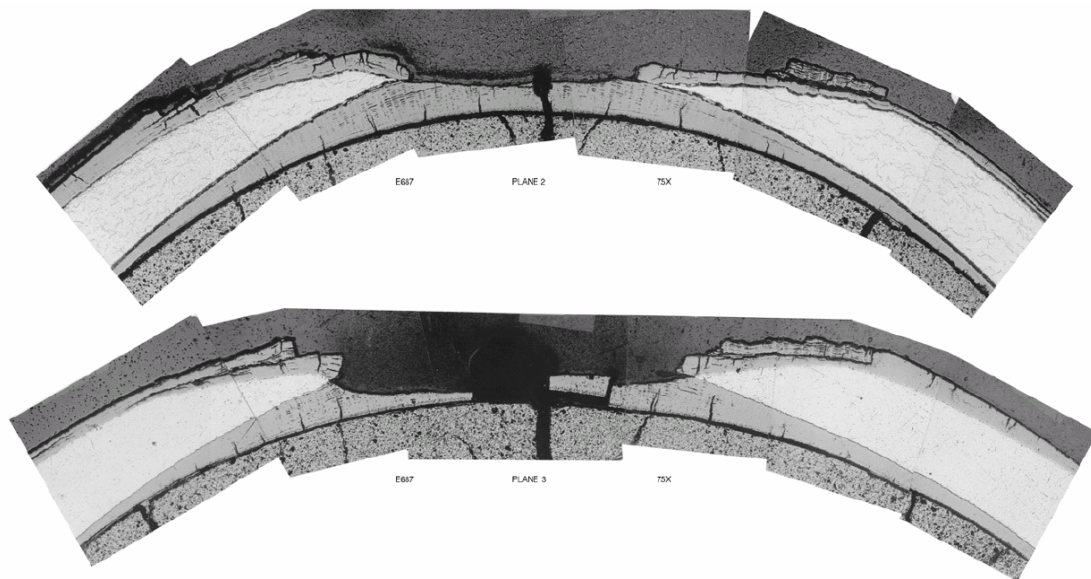


Figure 8-1: Cladding corrosion of Rod O11 at 3000 mm elevation, Tropasso, 2004.

The burnup distribution did not reveal any abnormalities, suggesting that the failure was unlikely to result from an abnormal power condition at the outer positions of two adjacent fresh fuel assemblies. The most notable result was that local regions of the exterior surfaces of the fuel rods were subjected to temperatures well above normal operating temperatures, Figure 8-2. Cladding grain recrystallization indicates that local regions of the cladding had experienced temperatures of 450-500°C. It is believed that only degradation of heat transfer due to trapping of steam within a thick CRUD layer can result in such high temperatures. Redistribution of hydrides away from the damaged side of the fuel rod indicates that the damaged side was the hotter of the two sides. The fact that corrosion penetration at the failure sites was extremely localized could not have resulted from abnormal power distribution. Thus PIE confirmed that the TMI-1 cycle 10 failures were due to localized cladding corrosion induced by CRUD. The results suggest a certain thermal-hydraulic (T-H) condition could have existed at the outer face of peripheral rods in a fresh high power assembly facing another fresh high power assembly to cause the localized CRUD deposition, although T-H analyses could not predict the presence of sub-cooled boiling at the fresh-to-fresh gaps. This root cause investigation concluded that increases of fuel duty beyond the prior experience base should be implemented only in small increments and supported by monitoring and subsequent fuel inspections. Another key recommendation for avoiding recurrence of the CRUD-related localized corrosion failure is control of the pH in the primary coolant system as proposed by the EPRI primary water chemistry guidelines to minimize the total CRUD inventory from beginning of the cycle. *These failures are also treated in Section 9.*



## 9 PRIMARY FAILURE AND SECONDARY DEGRADATION (PETER RUDLING)

### 9.1 INTRODUCTION

#### 9.1.1 Primary Failures

During reactor operation, the fuel rod may fail due to a primary cause such as fretting, *PCI*, manufacturing defects, corrosion, etc., Table 9-1.

Table 9-1: Primary failure causes for *LWR* fuel during normal operation and anticipated operational occurrences.

| Primary Failure Cause | Short Description   |
|-----------------------|---|
| Excessive Corrosion   | An accelerated corrosion process results in cladding perforation.<br>This corrosion acceleration can be generated by e.g. CRUD deposition (CILC <sup>5</sup> ), enhanced spacer shadow corrosion, ESSC, <sup>6</sup> (in <i>BWRs</i> ), dry-out due to excessive fuel rod bowing.   |
| Manufacturing defects | Non-through-wall cracks in the fuel cladding developed during the cladding manufacturing process.<br>Defects in bottom and/or top end plug welds.<br>Primary hydriding due to moisture in fuel pellets and or contamination of clad inner surface by moisture or organics.<br>Too large gap between the fuel rod and the spacer grid supports (poor spacer grid manufacturing process) leading to excessive vibrations in the <i>PWR</i> fuel leading to fretting failures.<br>Chipped pellets may result in <i>PCI</i> failures both in liner and non-liner fuel                     |
| <i>PCI</i>            | Pellet Cladding Interaction-an iodine assisted stress corrosion cracking phenomenon that may result in fuel failures during rapid power increases in a fuel rod. There are three components that must occur simultaneously to induce <i>PCI</i> and they are: 1) tensile stresses- induced by the power ramp, 2) access to freshly released iodine-occurs during the power ramp, provided that the fuel pellet temperature becomes large enough and 3) a sensitised material – Zircaloy is normally sensitive enough for iodine stress corrosion cracking even in unirradiated state. |
| Cladding collapse     | This failure mechanism occurred due to pellet densification. This failure mode has today been eliminated by fuel design changes and improved manufacturing control.   |
| Fretting              | This failure mode has occurred due to:<br>Debris fretting in <i>BWR</i> and <i>PWR</i><br>Grid-rod fretting - Excessive vibrations in the <i>PWR</i> fuel rod causing fuel failures. This situation may e.g. occur due to different pressure drops in adjacent fuel assemblies causing cross-flow.<br>Baffle jetting failures - Related to unexpectedly high coolant cross-flows close to baffle joints   |

<sup>5</sup> Crud Induced Localised Corrosion – an accelerated form of corrosion that have historically resulted in large number of failures in *BWRs*. Three parameters are involved in this corrosion phenomenon, namely: 1) Large Cu coolant concentrations- as a result of e.g. aluminium brass condenser tubes, 2) Low initial fuel rod surface heat flux – occurs in Gd rods and 3) Fuel cladding that shows large initial corrosion rates- occurs in cladding with low resistance towards nodular corrosion.

<sup>6</sup> This corrosion phenomenon resulted recently in a few failed rods. The mechanism is not clear but seems to be related to galvanic corrosion. This corrosion type may occur on the fuel cladding in contact or adjacent to a dissimilar material such as Inconel. Thus, this accelerated type of corrosion occurred on the fuel cladding material at spacer locations (the spacer springs in alloy *BWR* fuel vendors fuel are made of Inconel). Water chemistry seems also play a role if the fuel cladding material microstructure is such that the corrosion performance is poor. Specifically coolant chemistry with low Fe/(Ni-Zn) ratio seems to be aggressive (provided that the cladding material shows poor corrosion performance. A fuel cladding material with good corrosion resistance does not result in ESSC, enhanced spacer shadow corrosion, even in aggressive water chemistry.

*The failure statistics up to 2001-2002 were presented in the ZIRAT-8 Annual Report, Strasser, et al., 2003, Table 9-2 to Table 9-4. For US BWRs, about 30 % of the failures during the time period of 1991 to 2001 were due to corrosion (CILC/ Crud) and 26 % were due to debris fretting failures. For US PWRs, 40 % of the failed assemblies during the same time period were due to grid-rod fretting, and 17 % were related to debris fretting. In the year 2001, 40 out of 48 failed assemblies (83 %) in the 58 EDF PWRs were related to grid-rod fretting,*

Table 9-2: BWR failure root causes in US plants. The table shows the number of failed rods, Strasser, et al., 2003.

| Cause          | 1991      | 1992      | 1993      | 1994      | 1995     | 1996      | 1997      | 1998      | 1999      | 2000      | 2001     | 2002       | Total      |
|----------------|-----------|-----------|-----------|-----------|----------|-----------|-----------|-----------|-----------|-----------|----------|------------|------------|
| CILC           |           |           |           |           |          |           | 6         | 46        |           |           |          |            | 52         |
| Crud/Corrosion |           |           |           |           |          |           |           |           | 14        |           |          | X          | 14         |
| Fabrication    | 1         | 2         | 1         | 2         |          |           |           |           |           |           |          |            | 6          |
| PCI            |           | 1         | 2         |           | 2        | 6         | 1         | 1         |           |           |          | X          | 13         |
| Debris         | 20        | 2         | 6         | 4         |          | 5         | 3         | 5         | 6         | 2         | 2        | 2          | 57         |
| Uninspected    | 3         | 7         | 5         | 9         | 2        | 8         | 1         | 1         |           | 16        |          |            | 52         |
| <b>Total</b>   | <b>24</b> | <b>12</b> | <b>14</b> | <b>15</b> | <b>4</b> | <b>19</b> | <b>11</b> | <b>53</b> | <b>20</b> | <b>18</b> | <b>2</b> | <b>24+</b> | <b>214</b> |

Table 9-3: PWR failure root causes in US plants. The table shows the number of failed rods, Strasser, et al., 2003.

| Cause               | 1991       | 1992       | 1993       | 1994      | 1995       | 1996       | 1997       | 1998       | 1999       | 2000      | 2001       | 2002     | Total       |
|---------------------|------------|------------|------------|-----------|------------|------------|------------|------------|------------|-----------|------------|----------|-------------|
| Handling Damage     | 3          |            |            | 1         | 3          |            | 2          |            |            | 1         |            |          | 10          |
| Debris              | 110        | 29         | 16         | 7         | 13         | 1          | 14         | 4          | 3          |           | 12         |          | 209         |
| Grid Fretting       | 46         | 73         | 103        | 13        | 41         | 95         | 89         | 108        | 69         | 37        | 30         |          | 704         |
| Primary Hydriding   |            | 23         |            |           |            |            |            |            |            | 1         |            |          | 24          |
| Crud/Corrosion      |            |            |            |           | 9          |            | 5          |            |            | 11        |            |          | 25          |
| Clad Creep Collapse |            |            |            |           | 1          |            |            |            |            |           |            |          | 1           |
| Other Fabrication   | 1          | 7          | 3          | 3         | 21         | 5          |            |            | 1          | 1         | 1          |          | 43          |
| Other Hydraulic     |            |            |            |           |            | 21         |            |            |            |           |            |          | 21          |
| Inspected/Unknown   | 14         | 49         | 36         | 42        | 10         | 12         | 12         | 2          | 7          | 13        |            |          | 197         |
| Uninspected         | 25         | 18         | 17         | 3         | 13         | 3          | 8          |            | 29         | 11        |            |          | 127         |
| <b>Total</b>        | <b>199</b> | <b>199</b> | <b>175</b> | <b>69</b> | <b>111</b> | <b>137</b> | <b>130</b> | <b>114</b> | <b>109</b> | <b>75</b> | <b>43+</b> | <b>?</b> | <b>1318</b> |

Table 9-4: EDF Fuel failure causes, all in Framatome fuel in the year 2001, “Fretting” indicates “grid-rod fretting”, Strasser, et al., 2003.

| <b>48 Failed Fuel Assemblies</b> |    |
|----------------------------------|----|
| Grid-rod fretting                | 40 |
| Debris fretting                  | 1  |
| Manufacturing defect             | 1  |
| Uninspected/unknown              | 6  |

Table 9-5 to Table 9-6 provides key data of some recent fuel failures cases.

## 10 CLADDING PERFORMANCE UNDER ACCIDENT CONDITIONS (PETER RUDLING)

### 10.1 INTRODUCTION

Three different design basis accidents are treated in this section: (i) Loss Of Coolant Accident, *LOCA*, (ii) Anticipated Transient Without Scram, *ATWS*, and, (iii) Reactivity Initiated Accident, *RIA*.

#### 10.1.1 LOCA – Background Information

The objectives of the Emergency Core Cooling System, *ECCS*, *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 in most countries are:

- Peak Cladding Temperature, PCT, < 1204°C (or 2200°F)
- Equivalent Cladding Reacted, ECR<sup>28</sup>, < 17%
- Hydrogen gas produced < 1%<sup>29</sup>.
- Fuel must have coolable geometry<sup>30</sup>.

Core temperature maintained at low value for extended time<sup>31</sup>.

The first two criteria are addressing clad embrittlement. An embrittled fuel cladding could potentially result in loss of fuel coolable geometry due to fuel clad rupture during the post-*LOCA* oxidation phase.

The existing *LOCA* criteria were established in the 1973 ECSS Rule-Making Hearing and the development of the criteria were nicely reviewed by Hache & Chung, 2001 and summarized in the following.

At the 1973 ECSS Rule-Making Hearing, the Atomic Energy Commission, AEC, staff and commissioners and OECD-GSNI specialists were of the opinion that retention of clad ductility was the best guarantee against potential fragmentation of fuel cladding during post-*LOCA*. This potential fragmentation could occur due to not-so-well-quantified loading, such as thermal shock, hydraulic, and seismic forces, and the forces related with handling and transportation. *Later also the forces due to pellet-cladding bonding has been identified as a significant force during post-LOCA.*

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<sup>28</sup> The ECR is defined as the total thickness of cladding that would be converted to stoichiometric ZrO<sub>2</sub> from all the oxygen that are contained in the fuel cladding as ZrO<sub>2</sub>, and oxygen in solid solution in the remaining clad metal phase. Subsequently, in the *NRC* Information Notice 98-29 (August 3, 1998), the *NRC* stated that total oxidation, as mentioned in 10 *CFR* 50.46 (acceptance criteria for *LOCA* analysis), includes both preaccident oxidation and oxidation occurring during a *LOCA*.

<sup>29</sup> Total amount of hydrogen shall not exceed 1% of the hypothetical amount generated by the reaction of all the zirconium in the fuel cladding with the water/steam.

<sup>30</sup> The coolable geometry may be lost by either fuel clad ballooning causing coolant channel blockage or fuel cladding fragmentation due to clad embrittlement.

<sup>31</sup> After any operation of the *ECCS*, the core temperature shall be maintained at an acceptably low value and decay heat removed for the extended period of time required by long-lived radioactivity.

The Equivalent Cladding Reacted, ECR and Peak Clad Temperature, PCT, criteria were based on retention of clad ductility at 275°F (135°C, the saturation temperature during reflood) according to slow ring compression tests of double-sided steam oxidation non-ballooned unirradiated cladding Zircaloy-2 and -4 samples. The selection of the 17% ECR value was specific to the use of the conservative Baker-Just clad oxidation correlation. However, if a best-estimate correlation had used instead such as e.g. Cathcart-Pawel correlation, the threshold ECR would have been <17%. Most countries are using this criterion for ensuring adequate cladding ductility. In some countries it is assumed that the largest clad tensile stress<sup>32</sup>, during post-*LOCA*, is due to the thermal stresses during the quenching phase during the *LOCA*. In these countries the post-*LOCA* clad ductility criterion is specified such that the fuel cladding must be capable to withstand the quenching stresses without rupturing (which normally is a transversal break of the fuel cladding). Both in Russia and in Japan, the maximum allowable ECR during a *LOCA* transient is specified to ensure that the cladding can survive such a quenching without rupturing. In Russia a maximum value of 18% ECR is used for Zr1Nb claddings assessed in quench tests without any constraints of the clad during quenching. In Japan a corresponding value of 15% are used for Zircaloy claddings but assessed in quenching tests with significant constraints of the fuel clad.

The cold compression test is more conservative than the quench test, i.e., a fuel cladding that may pass the quench test may not pass the cold compression test. However, it may well be that the quench test is a more relevant test and that the cold compression test is overly conservative. The ongoing work in different countries may resolve this issue what type of test should be used to ensure that the fuel cladding will not fragment neither during the quench-phase of the *LOCA* not during post-*LOCA* events such as e.g. a seismic event.

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<sup>32</sup> During a *LOCA*, such as ballooning of the rod near the spacer grid, rod-grid spring chemical interaction and the friction between the fuel rod and spacer grids can restrict the axial movement of the cladding thus imposing clad tensile stresses during reflooding. In recognition of this, the AEC Staff wrote during the 1973 Rule-Making Hearing that “the loads due to assembly restraint and rod-to-rod interaction may not be small compared to the thermal shock load and cannot be neglected”. Subsequently, it was concluded that: “The staff believes that quench loads are likely the major loads, but the staff does not believe that the evidence is as yet conclusive enough to ignore all other loads”. *The NRC position is still that the best way to ensure that the fuel cladding will not fragment during post-LOCA event is to retain clad ductility since it may be difficult for codes to calculate exactly the post-LOCA stresses in the cladding*, Meyer, 2002.

During the late 70's – early 80's, slow ring-compression tests of ballooned and bursted samples showed that the 1973 criteria failed to ensure retention of ductility at 135°C in narrow local regions near the burst opening<sup>33</sup>, where H content exceeds about 700 ppm. This phenomenon was not known in 1973. However, the 1973 criteria still ensured resistance to 0.3 J impact tests, and survival after fully constrained quench tests for low-burnup Zircaloy<sup>34</sup>. The implications of the results are such that for fuel claddings with hydrogen content exceeding about 700 ppm near the burst opening<sup>35</sup>:

- The 17% ECR criterion may fail to ensure retention of ductility at 135°C
- The clad will not survive a fully constrained quench test without rupturing while it may survive an unconstrained quench test.

Also, the 1204°C peak cladding temperature (PCT) limit was selected on the basis of slow-ring compression tests that were performed at 25-150°C. However, samples oxidized at 1315°C are far more brittle than samples oxidized at 1204°C in spite of comparable level of total oxidation. This is because oxygen solid-solution hardening of the prior-beta phase is excessive at oxygen concentrations >0.7wt%. Consideration of potential for runaway oxidation (*due to that the oxidation process of the Zirconium material becomes to exothermic to be cooled by water*) was a secondary factor in selecting the 1204°C limit. **The 1204°C PCT and the 17% ECR limits are inseparable, and as such, constitute an integral criterion.** The post-quench ductility and toughness of the cladding material are determined primarily by the thickness and the mechanical properties of the transformed-beta layer.

The *LOCA* sequence can be divided into three phases, Figure 10-1:

- Ballooning and burst of the cladding occur since the rod internal pressure becomes much higher than the system pressure of the reactor pressure vessel and strength of the fuel cladding decreases as the temperature increases.
- The cladding is oxidized by steam and it becomes brittle when severely oxidized.
- The embrittled cladding may rupture by thermal shock caused by rapid cooling during the reflooding stage.

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<sup>33</sup> It appears that the stagnant conditions of water/steam in this location will significantly increase the hydrogen pickup during LOCA clad oxidation.

<sup>34</sup> Following these results, Japan modified the basis of its ECR criterion to ensure survival after fully constrained quench tests by specifying a maximum of ECR 15%.

<sup>35</sup> The hydrogen originates from the corrosion hydrogen pickup during: 1) base irradiation prior to the LOCA event and 2) the LOCA event

## **11 FUEL RELATED ISSUES DURING INTERMEDIATE STORAGE AND TRANSPORTATION (ALFRED STRASSER)**

### **11.1 INTRODUCTION**

The lack of a licensed, permanent spent fuel repository in any country has placed increased, if not total reliance on intermediate storage. As a result, the dry cask storage technology has expanded significantly and become a major activity and business component of today's fuel "cycle". A new word is clearly needed to describe the one-way route of the fuel from ore to permanent depository.

The regulations for storing fuel are "settling down" as more data and analyses are becoming available. The latest US Reg. Guide (*ISG 11*, Rev. 3, November 2003) shifts the regulatory criteria from burnup level, strain and oxidation limits to peak clad temperature, stress and temperature cycling limits. The modifications of the licensing criteria shift the burden to meeting the cladding temperature limit at the start of the dry storage cycle, a limit that has to be met by either longer cooling times in wet storage, or by modified higher heat capacity cask designs. Designs to meet higher thermal limits are becoming available.

Attention has now turned to evaluating the effect of cask handling and transportation accidents on the fuel contained in the casks. The fuel must stay contained and remain subcritical during an accident, be retrievable after an accident and maintain regulatory dose limits as a result of an accident. Cask accident analyses and tests are underway.

Expansions of wet storage facilities are still an option, albeit seldom used, nevertheless also discussed in this report.

A total of about 440 nuclear plants in 31 countries supply slightly more than 16% of the global electricity supply and are the source of spent fuel at the rate of 10,500 THM/year (tons of heavy metal). The rate is expected to increase to 11,500 THM/year by the year 2010. Only about a quarter of this is reprocessed, a fraction that could decrease in the future, leaving about 8,000 THM/year for placement in interim storage facilities.

A total of about 170,000 THM of spent fuel were stored in a storage capacity of 243,000 THM in early 2003, most of it in spent fuel pools with a capacity of 163,000 THM and somewhat more than 12,000 THM stored in dry casks with an ever growing capacity. The geographic distribution of the spent fuel is shown in Table 11-1, Fukuda, et al., 2003.

Table 11-1: Global Status of Spent Fuel Storage, Fukuda, et al., 2003.

| t HM          |         |
|---------------|---------|
| Region        | Amount  |
| West Europe   | 36 100  |
| East Europe   | 27 700  |
| America       | 83 300  |
| Asia & Africa | 23 900  |
| World         | 171 000 |

Status January 2003

Assuming the current nuclear capacity and fuel cycles continue, the spent fuel discharged by regions is predicted as shown in Figure 10-1. North America will store all of its fuel and will require ever increasing storage capacity. The US is expected to run out of wet storage facility capacity in 2013, an unlikely availability date for a permanent disposal site. As a result, essentially all plants have implemented or are planning dry storage facilities. Currently about 40 Independent Spent Fuel Storage Installations (IFSI) are licensed in the US.

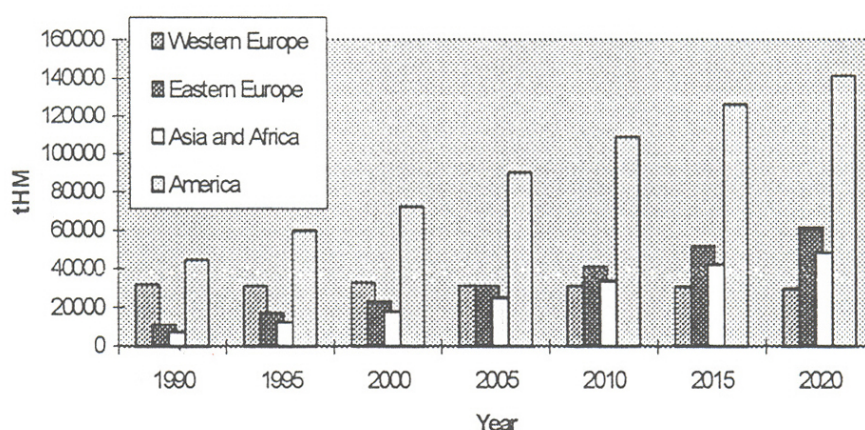


Figure 11-1: Global Spent Fuel Storage History and Future Requirements, Fukuda, et al., 2003.

Western Europe will have a slightly decreasing amount of fuel if reprocessing continues. However, Eastern Europe (former Communist Block countries) will double the amount of fuel discharged stored on site, since fuel contracts with Russia do not include the return of the fuel to Russia as they did in the time of the Soviet Union.

Asia, Africa and South America will also continue to discharge and store their fuel with the exception of Japan, which is planning a recycle economy.

Each country with nuclear plants has to plan its storage capacity needs individually since we have not managed to develop international sites yet, and some of these are discussed in the next Section 11.2.



In the US, an intermediate away-from-reactor (AFR) dry storage site is in the licensing stage, located in Skull Valley, Utah. The site is located on the reservation of the Skull Valley Band of Goshute Indians in a truly desolate location. The project is sponsored by a consortium of utilities that formed the Private Fuel Storage (PFS) Group. The *NRC* issued an SER in 2002 concluding that the facility is safe and meets the licensing requirements. The Atomic Safety and Licensing Board (ASLB) has held several meetings since then and cleared all issues except for the issue of an F-16 fighter aircraft from a nearby airbase crashing into the facility. The presumably final hearing on that topic was held in August, 2004 and the ASLB is now considering the case. HOLTEC and other organizations made analyses to show that the casks will protect its contents in case of a crash. The review of the data is in progress and the ASLB ruling is expected in January, 2005. If the ruling is positive, the Commissioners will instruct the *NRC* to issue a license. The expected appeals will proceed in parallel.

The State of Utah passed laws to stop the establishment of this site, all of which were ruled unconstitutional. All of the appeal courts upheld those decisions and the State is now taking the case to the Supreme Court.

## **11.2 STATUS OF FUEL RELATED REGULATORY REQUIREMENTS IN THE US**

### **11.2.1 Introduction**

The two basic regulations governing dry storage 10 *CFR* Part 71 (“Packaging and Transportation of Radioactive Material”) and 10 *CFR* Part 72 (“Licensing Requirements for the Independent Storage of Spent Nuclear Fuel and High Level Radioactive Waste”) are expanded and explained in NUREG-1536, the “Standard Review Plan (*SRP*) for Dry Cask Storage Systems”. In a further increased level of detail the *NRC* Spent Fuel Projects Office (*SFPO*) has issued a series of Interim Staff Guidance documents (*ISG*) some of which provide the fuel related detailed regulatory criteria.

The *CFRs* and *SRPs* were discussed in prior ZIRAT reports and have not changed, although the *SRP* will be modified to conform to the revised *ISGs*.

The *ISG* most relevant to cladding behavior during storage, Revision 2 of the Interim Staff Guidance (*ISG*) No. 11, issued in August, 2002 “Transportation and Storage of Spent Fuel Having Burnups in Excess of 45 GWD/MT” (reviewed in ZIRAT-7), has made some significant, reasonable changes to the regulatory criteria for storage of high burnup fuels by shifting from burnup level, strain and oxidation limits to peak temperature and temperature cycling limits. A further modification and clarification was issued as Revision 3 of *ISG* 11 under the title “Cladding Considerations for the Transportation and Storage of Spent Fuel” in November, 2003. It was too late for inclusion in the ZIRAT-8 report and is discussed in the next Section 11.2.2

## 12 POTENTIAL BURNUP LIMITATIONS

### 12.1 INTRODUCTION

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

- Corrosion limits of Zry-4 in high power *PWRs*, are extended by the alternate use of improved cladding alloys. Improved corrosion performance by the new alloys may allow the utilities to use the added margins, to modify plant operation e.g., to lower fuel cycle cost. However, this modified operation will in most cases result in higher corrosion duty of the zirconium materials. Thus, it is believed that the corrosion will always be limiting for plant operation even with the new type of alloys.
- Bowing of *PWR* fuel assemblies contributed in part by irradiation growth, creep and hydriding of Zry-4, has been reduced by improved guide tube materials (i.e., lower irradiation growth and hydriding rates), and reduced assembly holddown forces, but not yet finally eliminated.
- Bowing of *BWR* channels, extended by improved manufacturing processes, design changes such as variable wall channel thickness with relatively thicker corners, and in-core channel management programs.
- *RIA* and *LOCA* related burnup licensing limits are in the process of being assessed by additional experimental data and analyses. It would appear that the current *LOCA* limits are sufficiently conservative for high burnup fuel, although this may change in the future. The *RIA* limits are and will decrease as a function of burnup.
- The categories of event likely to eventually limit reliably and safely achievable burnup levels are outlined below. The zirconium alloy component most sensitive to the limits and potential methods for extending the limits are noted below.

## 12.2 CORROSION AND MECHANICAL PROPERTIES RELATED TO OXIDE THICKNESS AND H PICKUP

- *BWRs*: increased uniform and shadow corrosion, oxide thickness spalling.--- due to longer residence time, higher power and water chemistry changes. Current crucial issues are: shadow corrosion mechanisms, CRUD-chemistry-corrosion interaction and specific effects of NMCA with or without Zn-injection.
- *PWRs*: increased uniform corrosion, oxide thickness, spalling --- due to longer residence time and higher Li, higher power, more boiling. The introduction of Zr-Nb alloys may also result in accelerated corrosion at the welds, e.g., between the end-plugs and cladding tubes. Also, welding between dissimilar metals such as e.g. ZIRLO and Zry-4 may result in chemical compositions of the welds that show inferior corrosion resistance. Luckily, the corrosion temperatures at these elevations in the core is significantly lower than the peak temperatures and this may be the reason that no corrosion issues have been reported so far in the welds.
- Increased H pickup tendency<sup>44</sup>:
  - decreased ductility and fracture toughness during any situation (e.g., *RIA*, *PCMI*, *LOCA* and post-*LOCA* events, seismic event, transport container drop-accident conditions),
  - increases growth due to higher hydride volume
  - increased corrosion due to impact of hydrides at the cladding outer surface,
  - may impact creep behaviour of fuel claddings during class 1-IV events and during intermediate storage.
  - increased knowledge of the effects of irradiation and hydrides on the fracture toughness of thin-walled zirconium alloy components needed.

### Most sensitive component

Spacer and fuel claddings.

### Increase margin for *PWR*

- Improved knowledge of corrosion and hydrogen pickup mechanisms.
- Improved alloys with appropriate fabrication processes: ZIRLO/E635 (Anikuly), M5/Zr1Nb. Duplex is another alternative that may be necessary to achieve satisfactory mechanical properties.
- Change to enriched B soluble shim to reduce Li. There is however a fear that enriched B would increase AOA potential, i.e., more absorption per g. B, even though there may be less B.
- Improved water chemistry and CRUD control.

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<sup>44</sup> Due to the introduction of more corrosion resistant materials. For some types of Zr alloys, the hydrogen pickup fraction increases with decreasing corrosion rate.

- Increase corrosion resistance of steam generator materials.

Increase margin for *BWR*

- Improved knowledge of corrosion and hydrogen pickup mechanisms.
- Improved alloys under development.
- Modification of manufacturing processes (to get optimum sized, more stable second phase particles).
- Improved water chemistry and CRUD control.

*12.3 DIMENSIONAL STABILITY*

- Increased growth of components and differential growth between them resulting in reduced fuel rod spacing or even rod contact, guide tube bowing, fuel assembly bowing, spacer cell and envelope dimensions, *BWR* fuel channel and *PWR* fuel assembly bow may result in:
  - decreased thermal margins (*LOCA* and dry-out)
  - control rod insertion difficulties (safety issue)

Most sensitive component

Potentially all zirconium alloy components, but currently *PWR* guide tubes and *BWR* channels. Also *BWR* spacers have occasionally increased so much in dimensions that unloading of the assembly from the outer channel was very difficult.

Increase margin for *PWR*

- Alloys with lower growth and hydriding rates for guide tubes – ZIRLO, M5, M4, E635 (Anikuloy).
- Lower hold-down forces.
- Beta-quenched material after the last plastic deformation step during manufacturing. Beta-quenched materials do normally, however, show higher corrosion rate and lower ductility. These properties may be improved by an appropriate final heat-treatment in the alpha-phase.

Increase margin for *BWR*

- Uniform microstructure and texture throughout the fuel outer channel.
- Beta-quenched material in as-fabricated step.
- Channel management programs.
- More corrosion resistant material in channels and spacers is allow irradiation for longer cycles.
- Increased understanding of basic phenomena driving the channel bow process.

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