ZIRAT-10 Annual Report

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CONTENTS

1 INTRODUCTION

The objective of the Annual Review of Zirconium Alloy Technology (ZIRAT) is to review and evaluate the latest developments in zirconium alloy technology as they apply to nuclear fuel design and performance.

The 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-10 Program, the following technical meetings were covered:

- Review of ANL LOCA and SNF Programs Argonne National Laboratory, August 10-11, 2004.
- Nuclear Safety Conference, Washington DC, Oct. 2004.
- IAEA Technical Meeting on “Fuel Assembly Structural Behaviour” (FA bow, IRIs, SG-rod interaction, vibrations, etc), Cadarache, 22-26 November 2004.
- Jahrestagung Kerntechnik, Nuremberg, Germany May 10-12, 2005.
- Symposium N (Nuclear Material) of the EMRS spring meeting, Strasbourg, France, May 31 – June 3, 2005.
- 35th International Utility Nuclear Fuel Performance Conference, Las Vegas, Nevada, August 29 - September 01, 2005.
- Environmental Degradation Conference, Salt Lake City, Utah, USA, August 14-18, 2005.
- 18th International Conference on Structural Mechanics in Reactor Technology (SMiRT 18), Beijing, China, August 7-12, 2005.
- 6th International Conference on WWER Fuel Performance, Modelling and Experimental Support, 19–23 September 2005, Albena Congress Center, Bulgaria.
- Water Reactor Fuel Performance Meeting, sponsored by AESJ/ANS/ENS, October 3-6, 2005, Kyoto, Japan.

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-10 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 Emiritus, University of Toronto; Al Strasser, President of Aquarius and, Peter Rudling, President of ANT International, Mr. Friedrich Garzarolli, Dr. Rolf Riess and Professor Ali Massih.

The work reported herein will be presented in three Seminars: one in Marco Island, Florida, on January 30-February 1, 2006 one in Nice on February 8-10, 2006 and one in Japan in 2006.

The Term of ZIRAT-10 started on February 1, 2005 and ends on January 31, 2006.

2 BURNUP ACHIEVEMENTS AND FUEL PERFORMANCE ISSUES OF CONCERN TO UTILITIES (ALFRED STRASSER)

2.1 TRENDS IN FUEL OPERATING CONDITIONS

Economic incentives supported by advances in materials technology and improved computational modeling methods have increased the demands on fuel performance levels significantly from the early days of the industry, when burnups of 25 GWD/MT in conservative power density plants were considered successful achievements. The LWR plant operating method modifications that were made to achieve the improved economics were:

- Annual fuel cycles extended to 18 and 24 months,
- Discharge burnups increased from mid-30 to mid-50 GWD/MT batch average exposures by higher enrichments, in PWRs higher Li and B levels in the coolant and increased number of burnable absorbers in the core,
- Plant power uprates that ranged from 5 to 20%,
- More aggressive fuel management methods with increased enrichments and peaking factors,
- Reduced activity transport by Zn injection into the coolant,
- Component life extension with hydrogen water chemistry (HWC) and noble metal chemistry (NMC) in BWRs.

The trend for ***increasing fuel cycle lengths*** in the US is shown on Figure 2-1. The major economic gain of extended fuel cycles is due to the increased capacity factors, in turn, gained by reduced refueling times. Historically the US plants have had longer refueling shutdowns than European plants and had more to gain from longer cycles, while European plants maintained their annual cycles. Changes in economics, maintenance practices and licensing procedures have resulted in a trend to 18 month cycles in Europe as well and a trend to 24 month cycles in the US.

Nearly all the US BWRs are trending toward 24 month cycles. The older, lower power density PWRs have implemented the 24 month cycles, but fuel management limitations, specifically reload batch sizes required, have limited implementation of 24 month cycles in the high power density plants. The economics of 24 month cycles tend to become plant specific since they depend on the balance of a variety of plant specific parameters.

The economics combined with the fuel management demands of longer cycles are the prime incentives for ***extended burnups***. The average batch burnups in the US BWRs and PWRs are shown in Figure 2-2 and more detailed data are given subsequently. A complete, up-to-date economic analysis of the economics of extended burnup that take into account all of the cost factors, including back-end fuel disposal/storage costs has not been published. The latest published analyses are still those summarized by this author in the ZIRAT-8 Special Topics Report "High Burnup Fuel Issues", Adamson et al., 2003, based on a Westinghouse/EPRI study. That summary concluded that economic incentives for extending burnup levels beyond the 60 to 70 GWD/MT range will disappear and other incentives will be needed to go beyond this level.

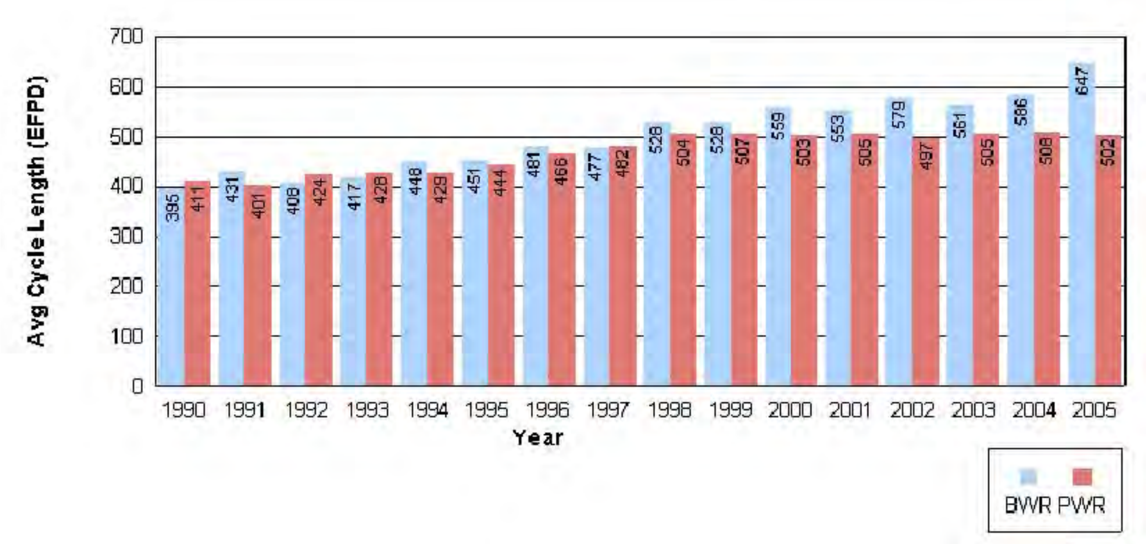

Figure 2-1: Average Fuel Cycle Length in the US, Yang et al., 2005.

Nevertheless the latest EPRI publication, Yang et al., 2005 states that "---studies indicate the fuel cycle economics can be improved significantly beyond 5% U-235 enrichment Burnup levels of 100 MWD/MTU or higher is economically desirable if performance issues can be resolved." Besides the poor grammar and presumably an error in the burnup units (undoubtedly meant to be GWD/MTU), *the statement is not backed up by the "studies" that are referred to, made by Westinghouse and sponsored by EPRI.* Those studies did not extend their economic analyses beyond the 70 GWD/MTU level and did not make economic analyses for enrichments >5%. The studies were summarized and evaluated in the above mentioned ZIRAT-8 STR and discussions between this author and the vendor agreed that there was no incentive to go beyond the 5% enrichment limit at that time.

In many cases the driving force to higher burnups are the economics of the fuel cycle length combined with the number of cycles. The choice of 18 or 24 month cycles are based on economic analyses that include fuel cycle as well as O&M costs and the multiple of the cycle chosen then determines the burnup. Perhaps that explains the lack of burnup level optimization as a function of fuel cycle costs.

The ***plant uprates, aggressive fuel management and extended burnup*** are based on higher enrichments and higher power operation. While all of the safety margins to normal operation, transients and accidents have to be maintained, the average fuel power will be increased at the cost of reduced margin in some areas. The fuel rod power census radially and axially across the core will increase with a plant power uprate. The fuel is designed to perform satisfactorily under these operating conditions; nevertheless, the reduced margin for a greater number of fuel rods increases the statistical probability of potential problems. In some PWR cases nucleate boiling may be initiated or increased with its attendant effects on crud deposition and possibly corrosion. The void fraction as a function of core height will increase in BWRs. To date no fuel failures have been related directly to power uprates, but peak power assemblies have been related to failures in BWRs.

The effect of ***water chemistry modifications*** are the most complex to understand since they involve the largest number of variables including the:

- global and micro-chemistry of the coolant at the cladding or other component surfaces,
- cladding temperature as a function of the power, heat flux and crud deposit characteristics,
- corrosion resistance of the cladding material,
- time of exposure.

Some of the chemistry additives can result in direct corrosion of the cladding, such as increased Li or direct contact of noble metals on fresh cladding. Other additives affect the crud morphology and can increase its thermal resistance, resulting in higher cladding temperatures and increased corrosion rates. These phenomena are discussed in detail in other sections of this report.

The effect of these operating parameter changes on zirconium alloy performance and the potential end-of-life (EOL) conditions that they may lead to are shown on Figure 2-3 for PWRs and Figure 2-4 for BWRs. While these figures have been shown since the ZIRAT-1 report, their updated versions are still the best overview of the zirconium alloy performance parameters most significant to their longevity. The effects of water chemistry should be considered separately from these charts, as each chemical additive or coolant impurity will need its own complex chart, best explained in subsequent sections.

One of the basic objectives of the ZIRAT Seminars is to identify and evaluate the latest data that become available in this chain of events and point out what effects they may have on the performance goals of zirconium alloys.

2.2 HIGH BURNUP FUEL PERFORMANCE SUMMARY

2.2.1 High Burnups Achieved in Utility Power Plants

The currently achieved burnup levels achieved in power plants are summarized in Table 2-1 for PWRs and Table 2-2 for BWRs based on publications during the past year and data provided by ZIRAT Members in response to our questionnaire on this topic. While the data are not complete, they represent a fair picture of the trends in the utilities of various countries that responded to the questionnaire or have published their data.

Table 2-1: Highest Burnup Achievements in PWR.

	GWD/MT Batch Ave.	GWD/MT Peak Assembly	GWD/MT Rod
USA (62 GWD/MT peak rod NRC limit)			
Wolf Creek, W RFA, ZIRLO	47	48.0	52.8
W RFA-2		48.0	
Indian Point 2 & 3	52	57 limit	60 limit
Catawba, McQuire, all ZIRLO			57
Oconee, M5 clad, GTs			57
North Anna			
3LTAs, M5 clad		52.0	
1LTA, M5 clad		67.6	
1LTA, ZIRLO clad			72
Other US PWRs Mk-BW, M5 clad	50.0	53.5	
W, RFA-2, ZIRLO clad	47.7	51.2	
Alliance LUA		48.9	
FRANCE (52 GWD/MT peak assy. regulatory limit)			
EdF plants, M5	47.0	51 UO_2	
		42 MOX	
LTAs in France and other countries with M5 clad		68 UO_2	80 UO_2
			60 MOX
GERMANY			
Unterweser, M5 clad, HTP spacer	47	49	53
Isar 2, Duplex ELS 0.8 clad HTP sp.	53	58	67
LTA all M5		17	
Brokdorc, Duplex ELS 0.8 clad, HTP sp.	49.1	58.9	62.9
Grafenrhein feld, Duplex ELS 0.8 clad, HTP sp.	52	61.8	67.1
Grohnde, Duplex ELS 0.8 clad, HTP sp.	49.1	58.9	62.9
JAPAN			
Kansai plants, Step 1 fuel		48	
Step 2 LTA: ZIRLO MDA, NDA clad		55	
4 LTAs (Ohi4) NDA clad		52	60
LT rods (McGuire+R2), NDA clad			84
			91 (pellet)
(Vandellos), MDA and ZIRLO clad		>55	75
Future planning		70	
SWEDEN			
Ringhals 2, W AEF+	45.3	47.1	
LTA Fragema AFA-3GAA		51.5	
Ringhals 3, Fragema AFA-3G	47.1	47.3	
LTA, Siemens HTP		57.2	
Ringhals 4, Fragema AFA-3G	46.2	47.6	
LTA, Fragema AFA-2G		50.3	

Table 2-2: Highest Burnup Achievements in BWRs.

	GWD/MT		
	Batch Ave.	**Peak Assembly**	**Rod**
USA			
Grand Gulf, GE-11, P 6/7 clad	46.8	51.1	56.4
going to ATRIUM 10 non-liner			67.9 (pellet)
River Bend, GE-11, P 6/7 clad	47.3	51.5	55.3 67.5 (pellet)
Fitzpatrick, Pilgrim	45-48		
Other US BWRs, GE-14	47.5	52.1 GE-11	
ATRIUM-10	43.1	53.0 GE-13	
FINLAND			
OL1, ATRIUM10B, LTP clad	38.7	43.6	
LTA, GE-14		14.5	
OL2, GE-12, P6 Triclad	38.1	41.3	
GERMANY			
KK Gundremmingen, MOX	nearly 50 ave.	58 ave.	
KK Isar 1	49.7 peak	53.3 peak	60 peak, LK3
JAPAN			
Fukushima Daini #1			
LTA 9x9 Step III, Zr2 liner clad		55	
LTA 9x9 Step III, HiFi clad		53	72 equiv. HiFi coupons
SPAIN			
Cofrentes, SVEA 96	44.6	49.7	60.3
			66.3 (pellet)
LTA, SVEA 96		53.0	63.4
			68.8 (pellet)
SWEDEN			
Forsmark 1, GE-12	41.2	44.7	
2 LTAs, GE-12	41.7	41.7	
Forsmark 2, SVEA-96S	41.8	45.3	
2 LTAs, SVEA-96S		39.7	
2 LTAs, GE-14		32.0	
6 LTAs, Optima 2		40.7	
Forsmark 3, SVEA 100	42.4	43.8	
2 LTAs, SVEA 100		41.3	
4 LTAs, GE-14		38.9	
8 LTAs, ATRIUM 10B		33.8	
Oskarshamn 1, Siemens 9x9	38		
SVEA 64, LK2 + clad		43.1	48.3
Oskarshamn 2, SVEA-64	42		
ATRIUM 10B		44.4	48.3
Oskarshamn 3, SVEA-Optima	46.6	48	53.8
SWITZERLAND			
2 LTAs, KKL SVEA96, LK3 clad		60+	68-73

The *highest burnup levels in PWRs* have been implemented in the US and Germany. The batch averages range between 47 and 53 GWD/MT, the peak assemblies between 48 and 62 GWD/MT and the peak rods between 53 and 67 GWD/MT. The burnup levels in the US plants have reached their maximum level permitted by the 62 GWD/MT max. rod exposure established by the NRC, until more data become available to justify increased regulatory burnup limits. The values above this level are in German plants. The current examinations of US rods from LTAs in the 65 – 75 GWD/MT range are planned to justify the increase of this limit to 70 or 75 GWD/MT.

While the French PWRs are limited to 52 GWD/MT assembly average burnup by their regulatory body, the current goal of the industry is to increase the limit to 70 GWD/MT. Similarly, the Japanese utilities, while they have relatively conservative current burnup levels, have an irradiation program to raise this to 90-100 GWD/MT rod burnup.

The lead test assemblies (LTAs) are usually exempt from regulatory limits and their highest peak rod burnups achieved have been in GWD/MT: 75 (US), 80 (France), 84 (Japan) 90 (previously reported for Switzerland).

The peak burnups achieved by the current cladding materials have been:

ZIRLO (GWD/MT):	48 batch,	53 assembly,	75 rod,
M5:	47 batch,	51 assembly,	80 rod,
Duplex ELS 0.8:	51 batch,	58 assembly,	64 rod.

The *highest burnups in BWRs* are in the US, Germany and Spain. The batch averages range between 43 and 50 GWD/MT, peak assemblies between 51 and 58 GWD/MT and the peak rods between 55 and 60 GWD/MT. The burnup levels in BWRs are catching up to those of the PWRs in the US as shown on Figure 2-2, probably because of the current NRC burnup limitation. Examination of fuel rods irradiated to 65 GWD/MT in Limerick are planned to justify a higher regulatory burnup limit. Irradiation results of a variety of BWR vendor fuel designs in KKGundremmingen and KKLeibstadt to extended burnups will be of significant interest in this regard when published.

The highest peak rod burnups achieved in LTAs have been in GWD/MT: 63 (Spain), 65 (US), 72 (Japan), 73 (Switzerland).

The peak burnups achieved by the current cladding materials have been:

Zircaloy 2 (GWD/MT):	49 batch,	53 assembly,	65 rod,
LK 3 (GWD/MT):	47 batch,	53 assembly,	60 rod.

The assembly and rod burnups listed above were achieved without failures; however, fuel assemblies and materials do not have a perfect performance record up to these burnups levels and the related problems are summarized in Section 2.3 and discussed in detail in subsequent sections.

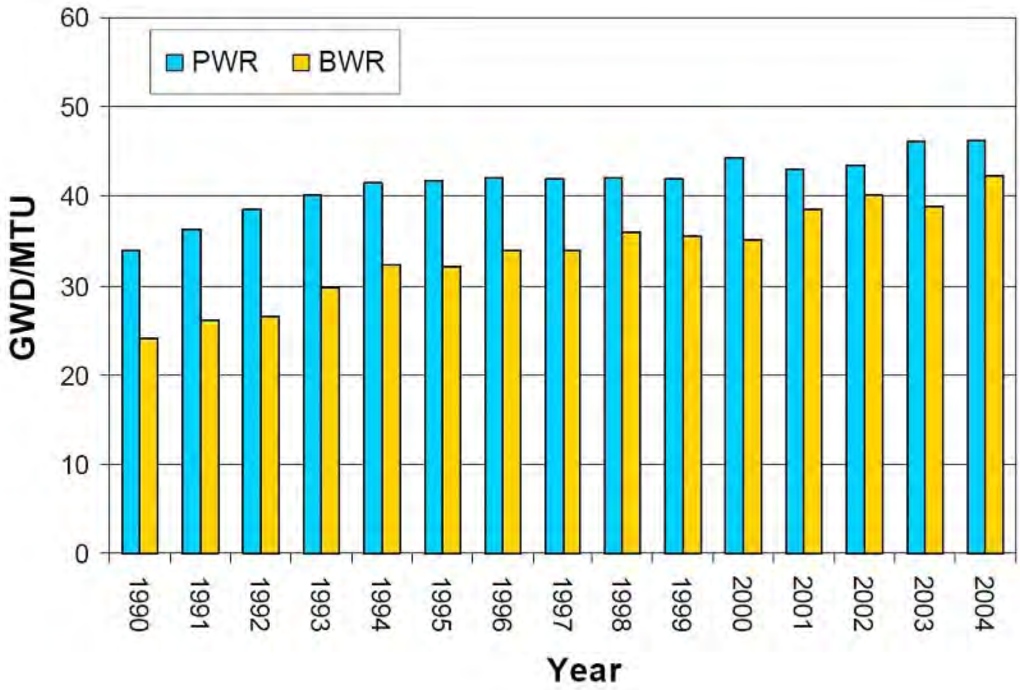

Figure 2-2: Average Batch Discharge Burn-up – U.S., Yang et al., 2005.

2.2.2 High Burnup Fuel Examination Results

The examination results of zirconium alloys after extended exposures are discussed in detail throughout this report under the appropriate headings. This section summarizes the most notable items of the highest burnup fuel examinations during the past year (>60 GWD/MT).

One of the most promising zirconium alloys for PWR applications has been Framatome's *M5 alloy (Zr-1%Nb)* a modified, patented version of the original Russian alloy of the same general composition. The alloy is the standard product for Framatome fuel cladding, guide tubes and spacers. The latest examination results reported peak fuel rod irradiations to 80 GWD/MT peak rod in France and 68 GWD/MT in the US, Mardon et al., 2005. Some significant results to date have been:

- Hydrogen pickup of 60 ppm in 80 GWD/MT cladding with "low" oxide thickness,
- Oxide thickness of <30µ on 68 GWD/MT cladding, (preliminary reports for 72 GWD/MT N. Anna rods estimated 30-33µ),
- Oxide thickness of <10µ on 57 GWD/MT guide tubes and 12µ at 80 GWD/MT on unfueled specimens,
- No accelerated cladding growth up to 71 GD/MT or guide tube growth to 58 GWD/MT,
- Slight spacer growth at 58 GWD/MT.

Irradiations of PWR fuel up to **peak rod burnups of 83 GWD/MT and pellet burnups of 90 GWD/MT** were reported for rods with the *Japanese NDA alloy cladding (Zr-1Sn, 0.27Fe, 0.16 Cr, 0.10 Nb, 0.01 Ni) with Al/Si doped large grain pellets* compared to standard fuel with Zircaloy-4, Ono et al., 2005. The fuel was irradiated in assemblies at McGuire, refabricated and exposed to the peak burnups in the R-2 test reactor. Some significant conclusions reached were:

- All rods were intact,
- At 83 GWD/MT rod burnup the NDA cladding had about 80µ oxide thickness compared to about 70µ for low Sn Zircaloy-4 and had consistently lower oxide thickness at the lower burnups as well,
- The large grain doped and undoped pellets did not have significant differences in fission gas release at 90 GWD/MT and were not significantly different from standard pellets at 30-40 GWD/MT.

Irradiation of PWR fuel up to **peak rod burnups of 67 to 75 GWD/MT** were reported for rods with *Japanese MDA alloy cladding (Zr-0.8 Sn,0.5 Nb, 0.2 Fe, 0.1 Cr) and ZIRLO (Zr- 1.0 Sn, 1.0 Nb, 0.1 Fe),* Watanabe et al., 2005. The fuel was irradiated in Vandellos 2 (Spain) for 4 cycles at average linear powers of 200/180/200/60 w/cm, subsequently 10 rods were removed and placed in the center of a fresh assembly and irradiated a 5th cycle at 200 w/cm. Some significant conclusions were:

- All rods were intact,
- Peak oxide thickness was 150-180µ for MDA at 72.1 GWD/MT and 110-150µ for ZIRLO at 69.4 GWD/MT. The high oxide thickness is believed to be due to the high power operation during the last cycle. (However, this may or may not be representative of extended burnup cycle operation).
- Hydrogen contents at pellet-pellet interfaces were 1300-1400 ppm compared to 900-1000 ppm opposite pellets; the pellet-pellet interfaces were the failure location of uniaxial tensile tests.
- The ring tensile tests for mechanical properties in the circumferential direction, more representative of fuel rod operation than the uniaxial tensile tests, resulted in uniform elongations <2% at 300-900 ppm H at 385°C for both alloys.
- (ZIRLO appears to be slightly better than MDA, although neither is very attractive for this power history and burnup).

Irradiation of BWR fuel to an **assembly average of 60 GWD/MT and peak rod burnups of 67-73 GWD/MT** were reported for rods irradiated in KKLeibstadt, Switzerland in a Westinghouse SVEA-96 assembly with LK3 cladding (ASTM Zircaloy-2 composition with alloy liner 0.1-0.4Sn, 0.02-0.045 Fe). Seven cycles of irradiation started with a linear power of 180-250 w/cm in the first two cycles declining to 80-100 w/cm in the seventh cycle. Some significant conclusions from the rods examined in the hot cell after exposures of 63-64 GWD/MT were:

- All rods were intact,
- Cladding oxide measurements in rod mid-sections (outside spacers) were an average of 18µ with peaks of 24µ,
- Total hydrogen content of the cladding was 180-208 ppm with 7-9% radial hydrides,
- 7µ thick crud layer contained hematite and zinc-iron spinel, the result of zinc injection.

A database as well as an experimental program for material properties of Zircaloy-4, ZIRLO and M5 has been established and a summary of some of the data has been published, Cazalis et al., 2005. Data include irradiation results on ZIRLO in Vandellos and M5 in Gravelines up to **75 GWD/MT rod burnup.**

A similar program has been established for fuel materials that includes UO_2, UO_2-Gd_2O_3, doped UO_2 pellets and MOX, Baron et al., 2005. Mechanical and thermal properties of the fuels are being assembled as well measured in a cooperative European program and used as input to modeling codes and to assist fuel design and performance evaluations.

2.3 FUEL RELIABILITY

Fuel performance reliability is of utmost importance to the economic and safe operation of the nuclear plants and it has improved significantly since the start of the industry. In the US reliability continued to improve from 1981 to 1991 and then decreased somewhat in the past years partly due to new plant operating parameters and partly to some new design features that were implemented. The number of defective assemblies per GWe as a function of calendar years are given on Figure 2-3. The failure cause statistics are summarized next, first for PWRs and then for BWRs.

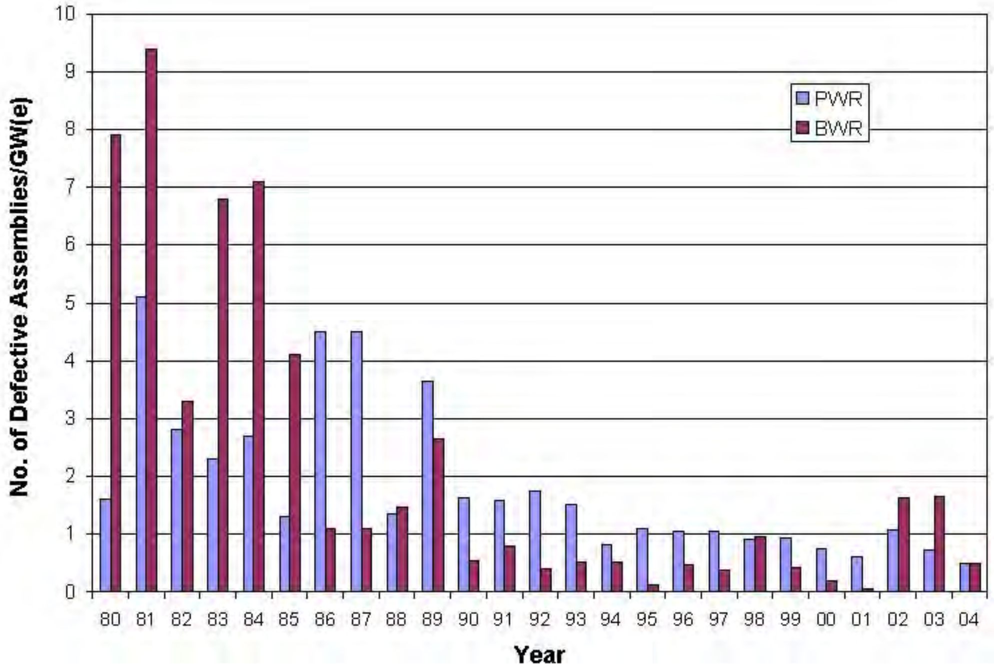

Figure 2-3: State of Fuel Performance in the U.S., Yang et al., 2005.

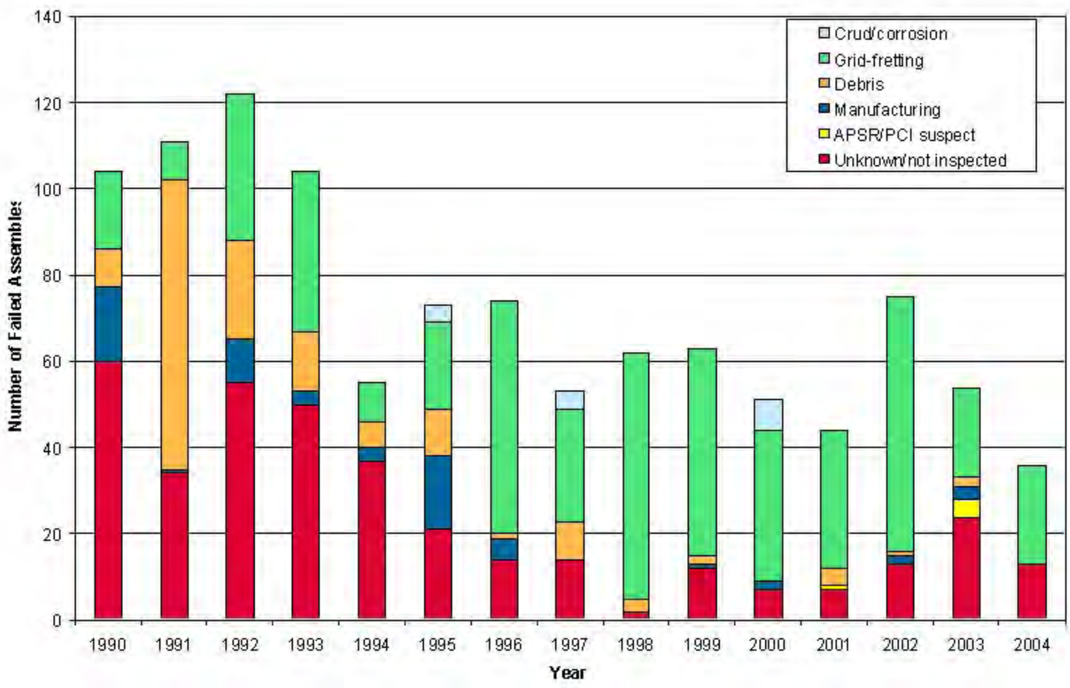

Figure 2-4: State of Fuel Performance in the US PWR. Yang et al., 2005.

PWRs

The major cause of cladding breaches in PWRs for the past years has been ***fretting between the cladding and the spacer grids***. In the US the fretting failures peaked in the early 90'ies, decreased in 2003, but still represent the highest % of failure causes as shown on Figure 2-4, Yang et al., 2005. The majority of the failures were in Westinghouse type fuel assemblies.

The French, EdF, experience, representing 58 operating PWRS, is similar, not surprisingly since Framatome is a former Westinghouse licensee. Their fretting failures peaked in 2001-2003 and decreased significantly in 2004, Provost & Debes, 2005 and Dangouleme, 2005. In 2003 all 4 of the identified failure causes in the 11 failed assemblies were due to fretting and in 2004 6 of the 10 identified failure causes in the 22 failed assemblies were due to fretting. The fretting failures represented 46% of all the failures in French PWRs since the start of their operation.

The Japanese, Kansai, experience representing 11 PWRs, currently by Westinghouse licensee Mitsubishi, experienced 69 fretting failures (31%) out of a total of 225 fuel failures as shown on Figure 2-5, Yamada, 2005. These peaked in the '78-'81 period and have been reduced to essentially zero, or 1 fretting failure out of a total of 2 failures in 2004.

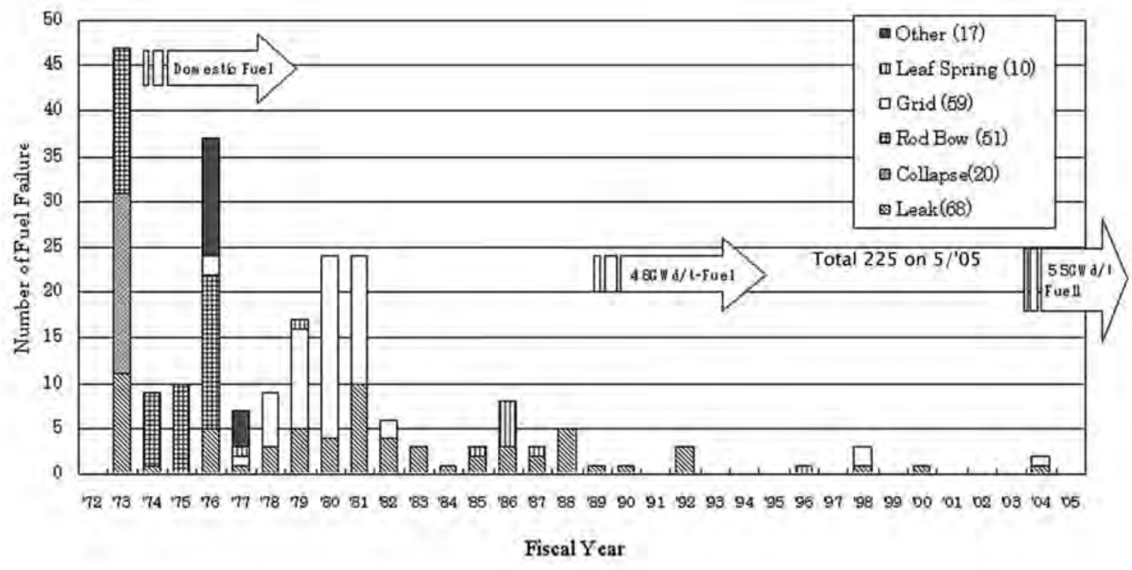

Figure 2-5: Experience of Kansai PWR Fuel in Japan, Yamada, 2005.

The cause of the fretting has been fuel rod and spacer vibration, in turn caused by some (not all!) inadequate spacer designs for holding the fuel rod, primarily an inadequate spring design, and/or hydraulic forces that were not anticipated or analyzed. The hydraulic forces range from local turbulence around the spacer to cross flow resulting from different design assemblies in the core that have different pressure drop characteristics. Extensive redesign of spacer grids and ex-reactor hydraulic testing to evaluate the spacers as well as hydraulic forces between different assembly designs have reduced the fretting events significantly, in addition to the increased use of spacers that performed adequately to start with. The discharge of assemblies that cause the fretting problem and replacement with improved designs takes time, so that this problem, while significantly reduced, has not been eliminated.

Failures due to ***fretting of the cladding by debris*** were a significant problem in the US in the early '90ies, but as the result of the implementation of debris resistant designs and better maintenance procedures have been reduced to only occasional failures. A similar trend in the reduction of debris failures was observed in France, where the overall debris related failures were 40% of all the failures experienced since the start of their plant operations. Of particular note is the total lack of debris failures in the Japanese, Kansai plants as noted on Figure 2-5. This can be attributed to their very meticulous operating and maintenance procedures, noted already in a review of their procedures some time ago, Strasser et al., 1989.

Two new failure types have been noted in US PWRS. Failures in B&W plants have been related to the pulling of axial power shaping rods (APSRs) and the suspected failure mechanism is PCI --- perhaps the first in the PWR industry with zirconium alloy fuel rods. A total of 3 plants had such failures. A hot cell examination is considered.

The second type of failure is related to the smaller diameter Westinghouse OFA rods. Over 15 such fuel rods have failed and a hot cell examination has been planned. The root cause is not known at this time.

Fuel rod and assembly bow due to differential growth of fuel rods and structural components and the interaction with hold-down forces by springs and hydraulic forces has come under reasonable control as the result of design changes. Increasing bow and control rod drag forces have been reported by EdF for 14 foot cores, but bow has been reduced by design changes implemented in the AFA-3 fuel assembly. Bowing has probably been a factor in spacer damage during handling reported for the 14 foot assemblies, Dangouleme, 2005.

Some bowing problems have occurred due to the *low* growth of M5 guide tubes that was not compatible with the M5 fuel rod growth and the spring hold-down forces.

Corrosion is a heavily monitored issue, discussed extensively in this report and at fuel performance meetings; however, it has rarely been a cause of fuel failure in PWRs, perhaps because it is so closely watched and its limits controlled.

The ***most important PWR fuel performance issues***, not necessarily related to failure causes, at this time are:

- Establishment of zirconium alloy mechanical properties as a function of high burnup, H content and distribution,
- Establishment of corrosion and H pickup limits for advanced Zr alloys,
- Effect of Zn injection on cladding at nucleate boiling conditions,
- Elimination of design specific assembly bowing,
- Reduction of internal pressure generated in B containing burnable absorber fuel rods.

The list assumes that clad fretting by spacers and debris are being resolved and will no longer be a problem.

BWRs

The major problem in the US BWRs in the past 3 years has been ***corrosion due to heavy crud deposits, combined with high duty and potentially corrosion sensitive cladding*** materials. While the failures were primarily in four plants, (Browns Ferry 2 and 3, Vermont Yankee, River Bend) the large number of failures determined these to be the highest percentage of all failures. The changes in water chemistry practices at the time of the failures and the differences in water quality between these plants have made the interpretation of the failure mechanisms more difficult. High fuel duty was a single consistent variable in the failures. Potential crud densification by Zn silicate formation in River Bend was also a potential cause. The failures are shown as a function of calendar years on Figure 2-6 along with other failure causes, Yang et al., 2005, and discussed in detail in the ZIRAT-8 and -9 Reports and this one as well. While ***manufacturing*** related factors are not mentioned in the reference, uncontrolled variables in the manufacturing process could potentially be contributors to decrease corrosion resistance of the cladding.

Debris fretting failures peaked in the early ‘90ies, similar to the PWRs; however, while the number of debris failures decreased, they were never eliminated in spite of ever improved debris filter designs on the fuel assemblies. The larger and more varied primary circuit of the BWRs may be a contributor to this.

The few recent failures with identified causes in the Swedish and Finnish BWRs appear to be primarily debris failures.

Pellet-clad interactions (PCI) have resurfaced since the ‘70ies and their subsequent suppression by operating restrictions and Zr liner fuel. The root cause has not been determined and hot cell examinations are in process in order to try to identify it.

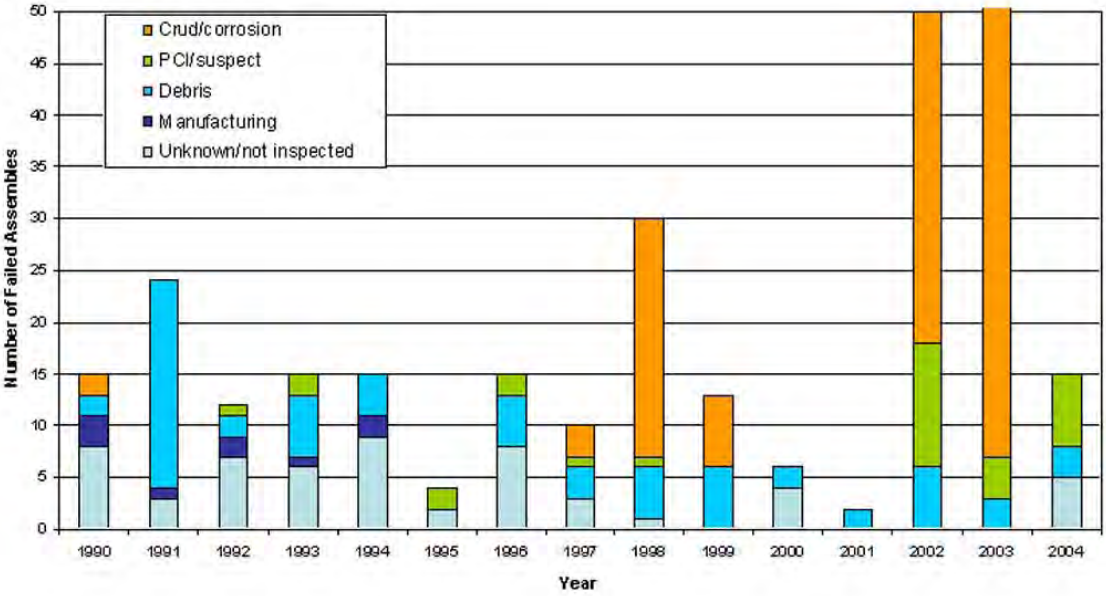

Figure 2-6: State of Fuel Performance BWR, Yang et al., 2005.

The ***most important BWR fuel performance issues,*** no necessarily related to fuel failures, at this time are:

- Establishment of zirconium alloy properties as a function of high burnup and H content and distribution,
- The effect of H pickup on assembly structural components at high burnup, its effect on properties and assembly integrity, (H pickup appears to increase >50 GWD/MT),
- Identification of the effect of water chemistry (Zn injection, HWC, NMC, and impurities), power and material variables on the crud deposition and corrosion of cladding,
- Channel bow in newer, C lattice, plants,
- Reliable local fuel duty analyses for new, complex design modifications,
- Development of zirconium alloys with better corrosion resistance and lower H pickup than Zircaloy-2.

A Fuel Reliability Data base (FRED) has been developed by EPRI and so far has collected data from 26 US utilities for 94 plants as of the beginning of 2005, Deshon, 2005. The type of data collected from utilities is summarized in Figure 2-7 and includes fuel cycle descriptions and fuel failure reports. Additional contributors and users internationally are invited by EPRI.

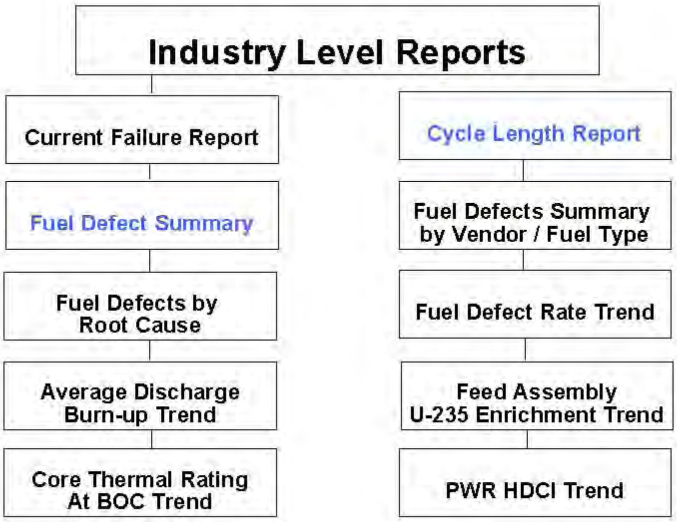

Figure 2-7: Industry Fuel Reliability Database FRED Rev. 1, Deshon, 2005.

2.4 *FUEL PERFORMANCE RELATED UTILITY CONCERNS*

A brief survey of ZIRAT member utilities discussed their primary concerns related to fuel performance issues and these are listed here in no particular order of importance.

PWRs

- The grid fretting problem has not been resolved in CE plants and is still present to a lesser extent in B&W plants with BZ spacers. The HTP spacer may solve the problem in the latter, but experience is not yet available. The problem seems to be solved in Westinghouse plants with their RFA2 spacer, although there is a record of one failure.
- An additional problem in the B&W plants is the physical interaction between the baffle and spacers of the assemblies on the edge of the core resulting in spacer wear. At this time it is not clear whether the assembly and/or the baffle are bowing.
- Assembly bow and twist is still a problem; in one case it started with implementation of M5 guide tubes.
- Caution is the word for water chemistry changes that involve higher Li and/or Zn injection.
- Power reductions and monitoring core designs to avoid AOA are costing money and should be avoided with improved water chemistry control. An AOA is suspected in a plant without nucleate boiling or Zn injection.
- The 24 month cycles are shutdown margin limited and as a result one utility had to stay with 18 month cycles as a result.
- Maintaining quality in fuel fabrication by utility and vendor oversight is necessary, particularly related to pellet fabrication to avoid chipping and subsequent performance problems, particularly in high duty fuel rods. (Applies to PWRs as well).

BWRs

- Channel bow in the more recent C lattice plants.

- Water chemistry related cladding corrosion issues.

- Fuel rod degradation is still an issue and in one case (not yet examined) it is either a non-liner fuel or alloy liner fuel, neither of which should degrade.

- What is the cause of the upsurge in failures (in PWRs as well)? and why is it so difficult to get at the root cause of failures?

- Zircaloy-2 may not be adequate to attain extended burnup goals: water rods have had 2,000 ppm H at 65 GWD/T, structural components had X00 ppm H at 52 GWD/MT and fuel cladding is lower, but still 300 ppm with a dominance of radial hydrrides.

- The cause of duty related failures after sequence exchanges and other control rod movements, presumably PCI for which the liners in the cladding should provide protection.

- Debris failures are not a past issue --- they keep happening; the debris sources are not clear.

2.5 FUEL RELATED REGULATORY ISSUES

PWRs

- Extended burnup batches are limited due to the 62 GWD/MT peak rod burnup and the limit should be extended (applies to BWRs as well),

- The alternate source term in the core established by the NRC places limits on the fuel rod linear heat generation rate (LHGR in kw/ft) vs. burnup, this defines the fraction of radio-nuclides in the pellet-clad gap that would be released during a transient and has been limiting in the related analyses.

- RIA and LOCA limits proposed are troubling and their status is summarized at the end of this section. RIA is particularly limiting for 2 loop Westinghouse plants, because they have particularly high rod worths and less margin during a simulated rod ejection accident.

- New NRC limits on corrosion for LOCA analyses may prove a problem for plants still using Zircaloy-4. One plant planning to switch to M5 is delayed in its licensing because of the time it takes to do the required applicable LOCA analyses.

- New spent fuel pool regulations require dispersion of high burnup or recently discharged fuel for security reasons and require an unobstructed flow path for water to the bottom of the pool to mitigate a pool LOCA.(Applies to BWRs as well).

BWRs

- Channel bow is a regulatory issue as well as a technical one.
- Power plant uprates are limited by some regulations.
- Unexplained PCI failures during “normal” operation are likely to influence regulatory actions on transient and accident criteria and limits.
- GNF’s Maximum Extended Load Line Analysis (MELLLA) provides a very small window in the power-flow map, limits flow control capability at high power and requires rods to be pulled. Since this is undesirable at full power, utilities lower power for the rod maneuvers that result in power losses. The improved MELLA+ model would alleviate this and is being reviewed by the NRC; however, the NRC is concerned that this modification could affect stability and action has been delayed because there is a need to show that stability is not affected.
- NRC reviews of modeling methods, as the above, are more extensive and take more time recently because of changing standards and newer personnel unfamiliar with the history of prior, related applications and licensing actions.

LOCA Status

The intent of the NRC is to change current LOCA embrittlement criteria defined by 10CFR50.46 to generic criteria that will require a specific cladding ductility to be retained for various combinations of time at temperature. The criteria will be independent of alloy identification, but each licensee will have to show how his alloy meets the criteria. This approach will cover M5, as an example, without calling it out specifically. Details of the criteria are still in the thinking/planning stage. It is believed that the 2200°F max. cladding temperature will be retained, but the 17% max. Equivalent Cladding Reacted (ECR) will probably not be retained.

Additional test data are needed to develop the criteria and these are expected in late 2005. The NRC believes the data should be from tests of irradiated cladding and Industry (EPRI, Utility Robust Fuel Group Members) is pressing to use unirradiated and pre-hydrided specimens as adequate. The irradiated, high burnup fuel planned for testing is:

- Zircaloy-4: Robinson PWR cladding currently at ANL-East, the initial test described above.
- ZIRLO: N. Anna PWR cladding being shipped from Studsvik to INEL (formerly ANL-West) for sectioning and characterization, then to be shipped to ANL-East for LOCA and mechanical property testing, (ANL-East can not handle full length assemblies and rods).
- M5: French PWR cladding being shipped from Studsvik with ZIRLO, as noted above, for the same testing sequence.

The above views are those of the NRC and its contractors. EPRI generally agrees to the approach for establishing new LOCA criteria, but disagrees that sufficient data could be made available and evaluated in time for the September 2006, or end of 2006 rulemaking. There are numerous technical issues that have not been resolved and the time is probably too short to do so in time for the current rulemaking agenda as pointed out by EPRI and in agreement with this author as well:

- The ECR is heat-up rate sensitive and the “design base” rate has not been defined. The relatively slow rates used by ANL are more benign than CEA’s fast rates as an example. The M5 alloy behaves in a brittle manner during the rapid rates used by the French compared to the more ductile results at ANL.

- The embrittling element in the former β layer is identified as the oxygen by ANL. But the hydrogen probably plays a yet unidentified role beyond increasing the oxygen solubility in the Zircaloy.

- Different ECR results are developed by one sided and two sided LOCA oxidation methods. The most applicable method (or both?) to LOCA has not yet been determined.

- Measured test results for ECR of unirradiated Zr alloys correlate well with the Cathcart-Pawel (C-P) correlation predictions; however, the tests with the irradiated Robinson rod samples had lower, measured oxidation thickness than the C-P predictions for reasons that are not entirely clear.

Rulemaking

In order to provide adequate criteria that will accommodate the potential embrittlement of the cladding, represented by the “former β layer” after a LOCA, the NRC is planning to change 10CFR50.46 accordingly. The rulemaking process to make changes in the Federal Register requires public hearings before the changes can be finalized. The scheduled goal for that is the latter part of 2006. The process to accomplish this has started with a LOCA Meeting at the NRC on February 10, 2005, and the NRC announcements at the OECD/SEGSFM meeting in Paris, April, 2005. The schedule of events planned for the rule making and related information meetings is as follows:

- September 30, 2005: NRC RES to issue a *draft* Research Information Letter (RIL) with the current licensing criteria and the data base for the proposed rulemaking. This is normally an internal document not released to the public.

- End of November, 2005: the NRC RES RIL will be made available to the public.

- Approximately February, 2006: Place the proposed rule changes in the Federal Register, allowing 6 months for public comment. This could include another ACRS meeting, perhaps a public meeting with the Commission.

- End of 2006: the CFR will be modified and issued in final form.

The strong point made by Industry was that sufficient data will not be available in time to meet this overly optimistic short schedule. There was no resolution of this disagreement.

RIA Status

The extensive differences in interpretation of the RIA test data and their application to the RIA limit curve continue to exist between the NRC and Industry (EPRI, ANATECH, Utility Robust Fuel Group).

Of the many points on which the NRC-Research Branch disagrees with the ANATECH/EPRI report the following claims by the NRC are the most significant:

- The limit curve should include the spalled rods that fail at significantly lower energy inputs.
- The fact that no fuel dispersion has been observed >10 ms pulses is not considered by the NRC and they believe that any failure may disperse fuel and potentially block flow --- therefore the coolability limit should be identical to the failure limit curve.
- They disagree that the higher clad strains on MOX tests are due to greater fission gas related swelling.
- As a result the NRC limit curve between about 10 and 70 GWD/MT burnup is at about 80 cal/g, significantly lower than the ANATECH curve at 170 cal/g for UO_2.

The procedures by which these disagreements are to be resolved were not clear as of the meetings held in the summer of 2005 and neither was the attitude of the NRC Regulatory Branch toward these issues.

Spent Fuel Transport

Insufficient data on high burnup cladding properties and their potential performance during hypothetical accidents have kept the NRC from issuing detailed criteria and guides related to the transport of high burnup fuel in dry storage casks. Each application for transport is taken on a case by case basis; however, since there are few if any such applications currently, this is not a high priority item.

3 ALLOY SYSTEMS (BRIAN COX)

There has been little work published recently on phase-diagrams and phase transformations in Zr-alloy systems. However, there are always a small number of papers worth reporting on. The most prolific group working in this field is probably the CNEA group in Argentina. This year Gonzalez & Gribaudo, 2005 have reported on an "Analysis of Controversial Zones of the Zr- Cr Equilibrium Diagram". This work was driven by some differences that can be observed in the zirconium rich zone of the proposed phase diagrams where invariant eutectoid and eutectic equilibria are proposed. In the zirconium-rich region of the phase diagram only limited results are available on the impact of oxygen concentration (an α-Zr stabiliser) on the eutectoid and eutectic reactions of Cr (a β-Zr stabiliser at high temperature). Using Zr base metal with either 0.24 at.% O, or 0.62 at.% O, and Cr of at least 99.85 wt.% purity (50 ppmw Fe main impurity), they fabricated 7 alloys within the range 0.3 to 26.5 at.% Cr. They do not quote the impurity contents of their base Zr alloys, which were 99.85 and 99.8 wt% pure Zr). The authors reported little observable difference in the $\alpha \rightarrow \beta$ transformation temperature for their two oxygen levels, and recorded partial isotherms at 4 temperatures for the Zr-Cr-O system, Figure 3-1, Gonzalez & Gribaudo, 2005. An isopleth section was also constructed at a level of 0.79 at.% O, Figure 3-2 and compared with earlier results of Rumball & Elder, 1969. The differences are quite significant above 1.0 at.% Cr. Below this Cr concentration only the $\alpha + \beta \rightarrow \beta$-Zr transformation temperatures differ significantly.

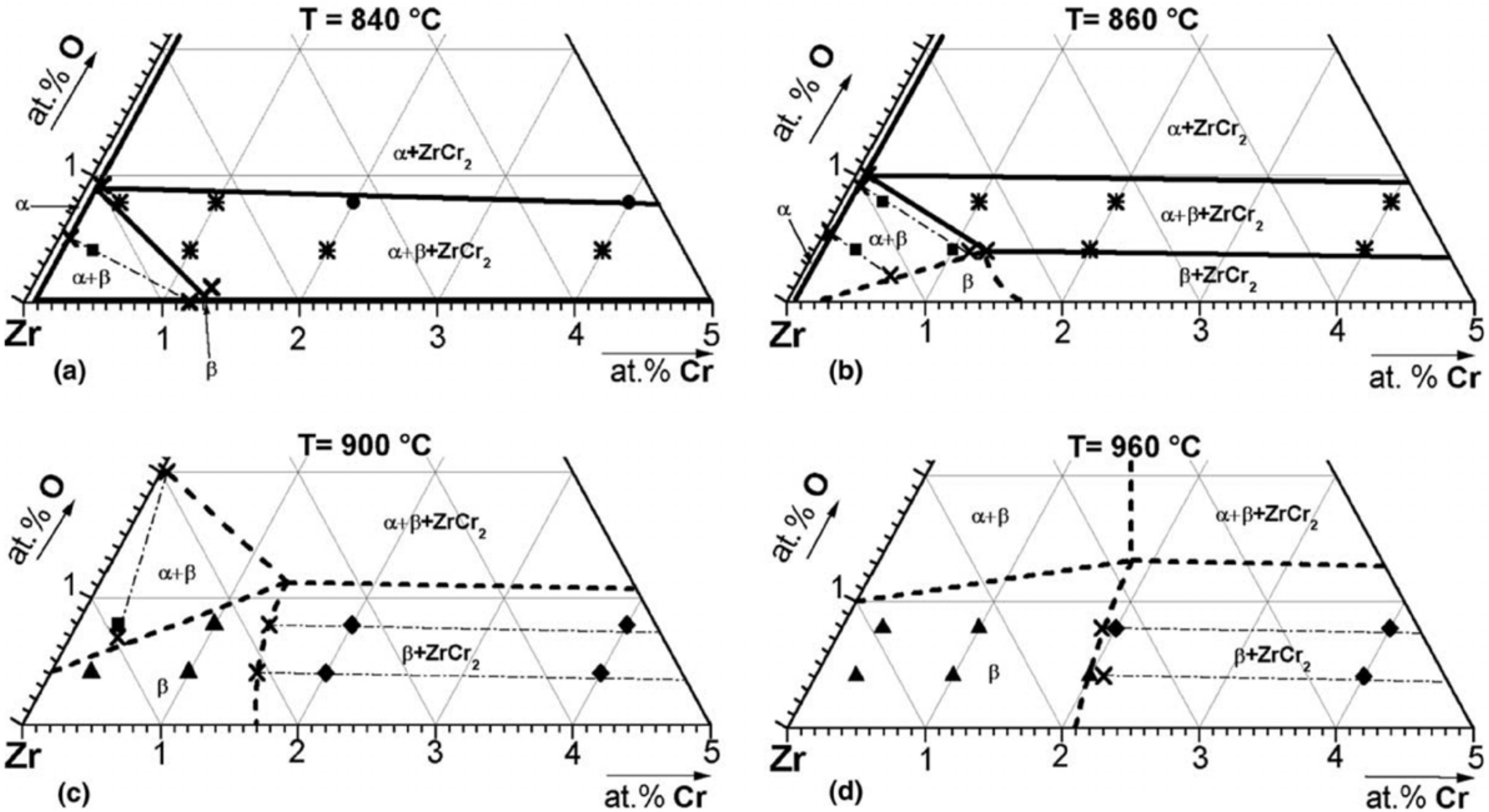

Figure 3-1: Partial isotherms at 840, 860, 900 and 960°C of the Zr-Cr-O system. (■) $\alpha + \beta$, (✱)$\alpha + \beta + ZrCr_2$, (▲)β,(◆) $\beta + ZrCr_2$, (●) $\alpha + ZrCr_2$, (×−−−×) experimental tie-line.

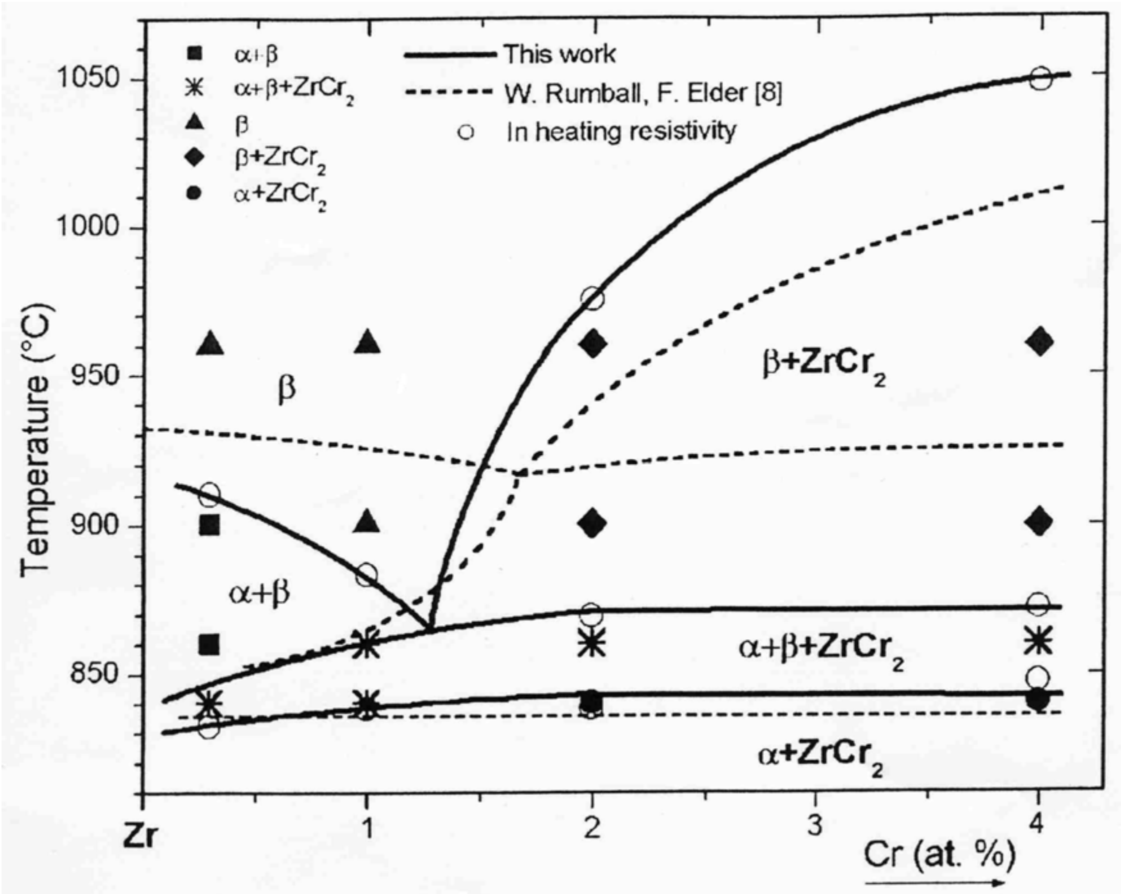

Figure 3-2: Isopleth section (0.79 at.% O) of Zr-Cr-O system as compared with similar results in Rumball & Elder, 1969.

The metastable Ω phase in Zr-Nb alloys continues to attract attention, Aurelio et al., 2005. In this instance the interest is in the Ω phase generated by the athermal $\beta \rightarrow \Omega$ transformation that occurs following β-quenching of a Zr-Nb alloy. The authors present the relationship between β-Zr (bcc) structure presented in its hexagonal analogue, and the hexagonal Ω phase, Figure 3-3. The difference consists of minor displacements of the central atoms in the unit cell, which allow the $\beta \rightarrow \Omega$ transformation to be athermal. This is a situation that is not met in nuclear reactor components since Zr-1%Nb alloy fuel cladding is used in the fully recrystallised condition, and Zr-2.5%Nb pressure tubes are extruded in the ($\alpha + \beta$) phase field, which allows segregation of the Nb into the β-Zr phase so that the initial β-Zr phase at the α-Zr grain boundaries contains about the monotectoid Nb composition of ~19%Nb. This does not spontaneously transform to Ω on cooling, but does during thermal decomposition of the β-Zr thermally, Hehemann, 1972, Figure 3-4. The authors do not consider this mode of formation and transformation – they do not even refer to the work of Hehemann, 1972. The information they present on variations in the “a” lattice parameters of the bcc, Figure 3-5 and Ω^q, Figure 3-6 phases are, therefore, of largely academic interest, as are the variation of the “c” lattice parameter of Ω, Figure 3-7.

5 MECHANICAL PROPERTIES (RON ADAMSON)

5.1 INTRODUCTION (PETER RUDLING)

The mechanical properties of essentially two different components are normally 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 $4 \cdot 10^{17}$ $n \cdot m^{-2} s^{-1}$.

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 400C for 24 hours, resulting in a structure consisting of elongated grains of hexagonal-close-packed (HCP) ά-Zr, partially surrounded by a thin network of filaments of body-centered-cubic *β*-Zr. These *β*-Zr filaments are metastable and initially contain about 20% Nb. The stress-relief treatment results in partial decomposition of the *β*-Zr filaments with the formation of hexagonal-close-packed ω-phase particles that are low in Nb, surrounded by an Nb-enriched *β*-Zr matrix. The hcp ά-Zr grains are oriented with their unique c-axes aligned in the radial-transverse plane, mostly tilted towards the transverse direction.

In 2005 there was an insufficient number of papers in the literature to allow a proper review of the above. Therefore that review will be postponed until ZIRAT 11.

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. This 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 criterions 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 specifies 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[1] phase during *LOCA* and due to *PCMI*[2] during a *RIA* transient.

To ensure that the various fuel design mechanical design criteria are met, different mechanical tests are performed. The data are generated in two types of tests, either separate effect tests or integral tests. The former test studies only the impact of one parameter at a time on the mechanical performance, see e.g. Adamson & Rudling, 2001. This could e.g. be the impact of hydrogen content on ductility. Table 5-1 gives a summary of some of the separate mechanical tests being used.

[1] This is the last phase during a *LOCA* situation when the core is reflooded with water that cools the fuel cladding surface imposing very large thermal stresses that may fracture the fuel cladding.
[2] Pellet Cladding Mechanical Interaction, i.e., interaction without the influence of iodine (that would instead result in *PCI* (Pellet Cladding Interaction).

6 DIMENSIONAL STABILITY (RON ADAMSON)

6.1 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 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 rare that a technical conference in the reactor performance field 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 & Rudling, 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, relaxation of differential residual stresses, or 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.

Review of the mid-2003 through late 2004 literature on dimensional stability was reported in the ZIRAT 9 Annual report, Adamson et al., 2004. Reported highlights include:

- "*BWR* channel bow is a significant issue. The observed bow is reported to be caused by the effects of differential hydride and fluence distributions. Shadow corrosion has been suggested to play a role.

- Final stage beta-quenching of *BWR* channel strip reduces irradiation growth (at least to moderate fluences) and reduces channel bow.

- *PWR* fuel assembly bow has been reduced by mechanical design modifications and by using low growth material such as M5 or ZIRLO.

- Collective bow (all in one direction across the entire core) has been observed at Ringhalls plants. The phenomena are not well understood, but bow has been reduced by mechanical design modifications and by use of low growth M5 guide thimbles and grids.

- Realization that corrosion, hydriding and irradiation growth can cause unacceptable grid envelope changes have led to mechanical and material fixes.

- Fuel rod and assembly growth has been reduced by informed material changes involving reduced hydrogen and corrosion levels and use of low growth materials like M5, PCA and HPA-4.

- There is hard data indicating that M5 has low and non-accelerating irradiation growth to about 1×10^{26} n/m^2 (E > 1 MeV) (50 MWd/kgU). Its Russian mother alloy, E110, has high and accelerating growth at lower fluences. An explanation for the differences is still lacking.

- Mildly cold worked E-635 alloy has low, non-accelerating growth out to high fluence, and is similar in growth to *RXA* E-635. This is very different from comparisons between cold worked and *RXA* Zircaloy, but may be similar to the behavior of cold worked or *RXA* ZIRLO. The low growth materials form only few <c>-component dislocations during irradiation.

- A special high-heating rate process has been developed for Zr2.5Nb alloys which results in small effective grain size and random texture. Irradiation growth and creep rates are sharply reduced by this process.

- The flux dependency of irradiation creep has been shown to be non-linear at low flux and tends toward being linear at typical *LWR* fluxes.

- New data has confirmed that for very high in-reactor strain or stressing rates, such as during a *PCI* event, the creep behavior more closely conforms to high stress, post-irradiation creep than lower stress steady state in-reactor creep.

7 CORROSION AND HYDRIDING

This year has been without a major Zr alloy Corrosion Conference so far. The IAEA Technical Committee Meeting on "Behaviour of High Corrosion Resistance Zr-Based Alloys" will not occur until 24-28th October, and a summary of it will be appended later. Meanwhile the final versions of papers presented at the June 2004 ASTM Zr Conference in Stockholm have been appearing in the on-line Journal of ASTM International. It does not appear that any other version of the proceedings will appear. The corrosion papers from this conference were reviewed in ZIRAT-9, and no major changes seem to have been made in the final versions that would necessitate a second review of the papers. The references for the final versions are given here (in order of discussion in the ZIRAT-9 Annual Report) so that readers have access to the final versions. Abolhassani et al., 2005, Barberis et al., 2005, Kakiuchi et al., 2005, Kapoor et al., 2005, Bojinov et al., 2005, Yueh et al., 2005, Motta et al., 2005, Elmoselhi & Donner, 2005, Lysell et al., 2005 and Takagawa et al., 2005. George Sabol's Kroll Medal Paper at the 2004 ASTM Zr Conference has now been published, Sabol, 2005. It provides an excellent review of the work involved in the development of ZIRLO.

New publications this year that have some relevance to the Zr alloy corrosion process will be discussed starting with work on bulk ZrO_2 studies.

7.1 ZRO$_2$ STUDIES (BRIAN COX)

It was pointed out last year that studies of tetragonal ZrO_2 in zirconium oxide films have, in the past, ignored the observation that there are three slightly different crystallographic forms for *t*-ZrO_2. At present no study of oxide films on Zr alloys has identified which of the three isomorphs has been present in the films that were studied. A recent study of the three tetragonal forms, designated *t*;*t*' and *t*'', Caracoche et al., 2005 notes that *t*' -ZrO_2. is the stable form with regular eight co-ordinated oxygen atoms surrounding each Zr site. *t*'–ZrO_2 is made up of defective Zr surroundings and is resistant to transformation to the monoclinic phase. The *t*''–ZrO_2 phase has only been reported recently for many of the ZrO_2–M_2O_3 systems (M = Y, Er, Nd, Sm, Yb). In this variant of the *t*–ZrO_2 system, displacements of the oxygen ions from the fluorite locations are smaller than in the t' phase, and the t'' phase is often referred to as pseudo-cubic because of this. The authors concentrate on the properties of the t'' phase and its transformation to cubic-ZrO_2. However, from the view point of Zr alloy corrosion, and the association of the corrosion rate transition with the change from *t*-ZrO_2 to m-ZrO_2 it becomes important to know whether it is the *t*-ZrO_2 or the *t*'–ZrO_2 that is formed in thin oxide films. If the *t*'–ZrO_2 phase is more resistant than the *t*–ZrO_2 phase to transformation to *m*-ZrO_2 then can we ensure that only *t*'–ZrO_2 is formed, and will this result in delayed post-transition corrosion.

The tetragonal – monoclinic phase change in ZrO_2 is martensitic (i.e. diffusionless) and involves a significant volume change. This transformation can be achieved by exposure of partially stabilised ZrO_2 to water vapour at low temperatures. Deville et al., 2005 examined the surface topographical changes resulting from the *t–m* ZrO_2 transformation in 3Y-TZP and Ce-TZP induced by exposure to water vapour at 140°C and 2 bar, using an AFM. The observed topographical changes are shown in Figure 7-1a-d, and a model for how one of these topographical features might form is shown in Figure 7-2. As might be expected the stresses resulting from the volume change during the *t-m* ZrO_2 transformation induced cracks in the samples, Figure 7-3. The transformation always initiated at a grain boundary. Such a model might be applied to the generation of cracks in Zr corrosion films leading to a transition in the corrosion kinetics.

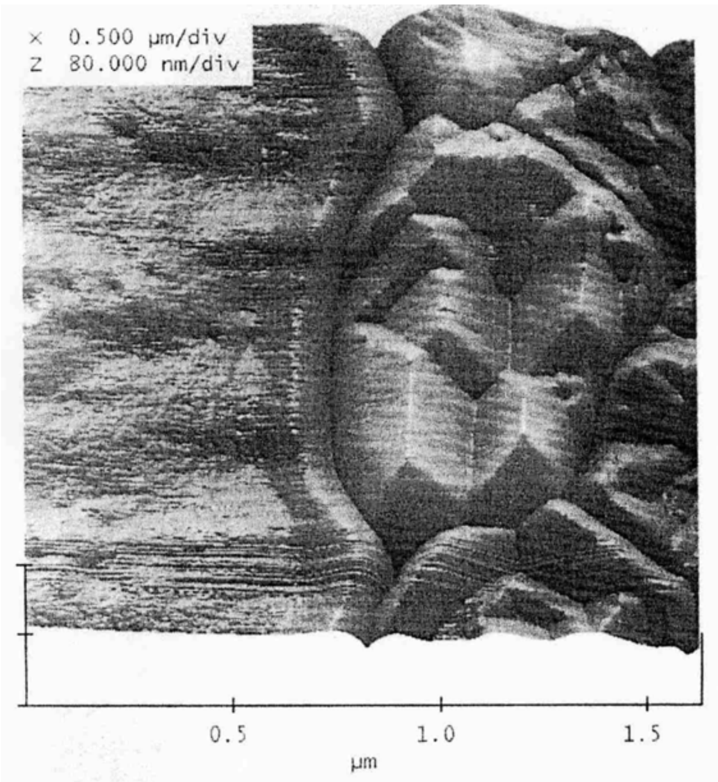

Figure 7-1a: Self-accommodating martensitic variant pairs with a more complex spatial arrangement in 3Y-TZP. The untransformed grain on the left is a stable cubic phase grain. Grain boundary thermal grooves are clearly visible.

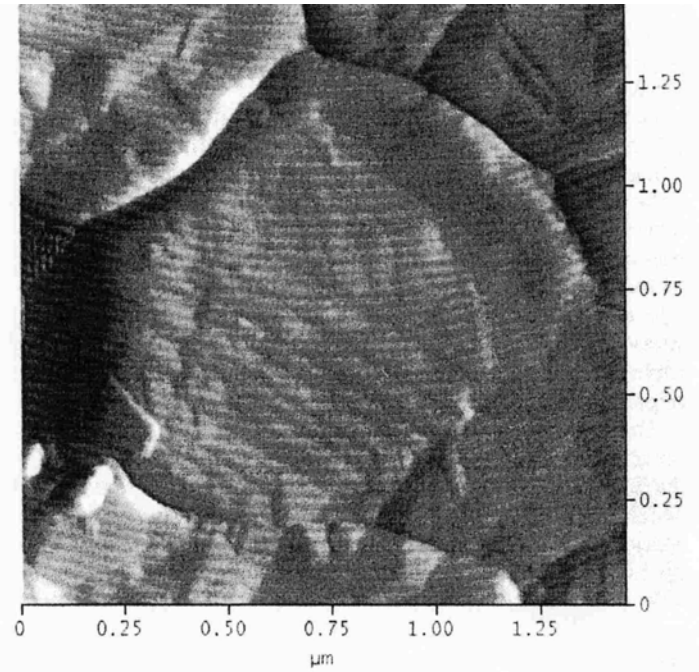

Figure 7-1b: Self-accommodating martensitic variant pairs in 3Y-TZP with either a junction plane almost parallel to the surface, leading to a rippled surface, or with the deformation strain being accommodated by slipping rather than twinning.

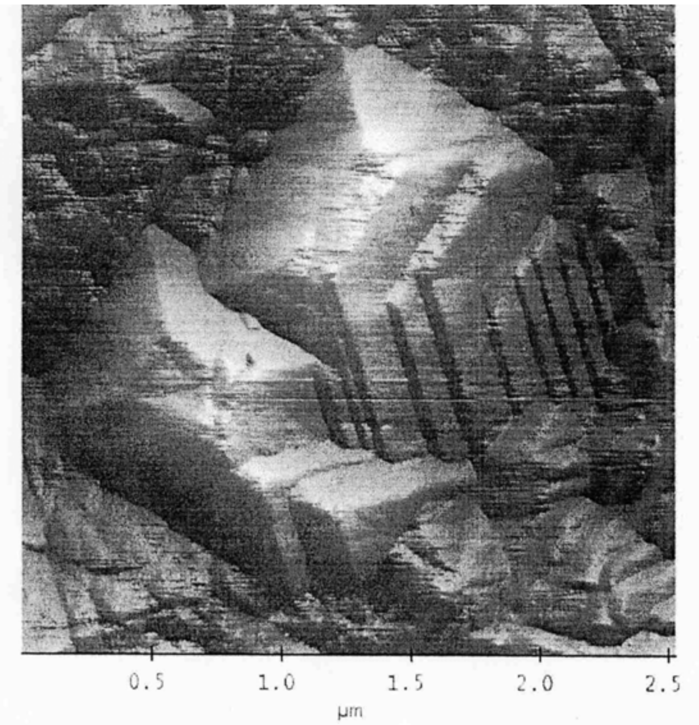

Figure 7-1c: Self-accommodating martensitic variant pairs in arrangement in Ce-TZP. Untransformed parts can be seen in between pairs.

8 EFFECTS OF WATER CHEMISTRY

8.1 PWR WATER CHEMISTRY (ROLF RIESS)

The primary coolant serves as a moderator and is the medium for transporting heat from the core to the steam generators. Hence, it must not endanger plant operation by the corrosion of materials and consequences thereof. The role of water chemistry can be divided into the following main points.

1) Metal release rates from the structural materials should be minimal.

2) The occurrence of localized forms of corrosion should be counteracted.

3) The transport and deposition of corrosion products should be controlled in such a manner, that contamination of the primary coolant system is kept low.

4) The deposition of corrosion products on heat transfer surfaces, particularly on fuel assemblies, should be prevented as far as possible.

5) The radiolytic formation of oxygen should be suppressed.

In certain instances, situations may be encountered where chemistry conditions that are optimum for achieving one goal can lead to a decreased level of achievement relative to other goals. As a result of such considerations the water chemistry specification must define parameters to achieve a balance among the five goals, recognizing that highest priority is assigned to materials and fuel integrity goals. Although the other goals are given second priority, they cannot be ignored. The materials which are in contact with the primary coolant are:

a) Austenitic stainless steels for components and piping of the primary system.

b) Zirconium alloys for cladding of fuel assemblies.

c) Incoloy 800, Inconel 690 TT (Thermally Treated) or, Inconel 600 MA (Mill Annealed) or TT for steam generator tubes. Stainless steel tubing is used in VVER SGs (Steam Generators).

d) High alloy materials (ferritic stainless steels) of low surface area for internals of the primary system.

The water chemistry conditions applied to these materials must fulfil the above mentioned requirements. Thus the primary coolant of PWRs, which contains boric acid (900-1800 ppm B at BOC) as a neutron absorber, is chemically conditioned by the addition of isotopically pure lithium (Li-7) hydroxide (2-5 ppm Li at BOC) as a non volatile alkalizing agent, and of hydrogen.

Recently, an increasing number of PWRs are adding zinc (5-40 ppb) in order to:
(1) reduce plant activation by reducing the metal release and by replacing cobalt isotopes in the oxide layer; and, (2) minimize stress corrosion cracking of Inconel 600 material.

In VVER plants NH_3 is added, which decays to H_2 by radiolysis. KOH is added instead of LiOH, so that the pH-control is accomplished by K + Li + Na (Li-7 is formed by the B-10 (n, α) Li -7) and NH_3. None of the VVER plants are adding zinc like the PWR plants.

Concerns Regarding Fuel Elements

From today's perspective it is most important to evaluate the factors that are of greatest concern for fuel element corrosion, and what have been driving forces (problems) for water chemistry in the last 10-to 15 years.

These driving forces are moves to improve Plant Availability and Fuel Economics which can be characterized by:

- Changing to 18 and 24 month cycle
- Core up-rating
- Higher enrichment fuel, increased burn-up
- Low leakage cores combined with increased sub-cooled nucleate boiling

These moves, based on operational experience, caused concerns over coolant additives and impurities because the fuel elements in the operating plants (especially in the US) experienced heavy crud deposition at positions where sub-cooled boiling created two negative effects, namely (1) accelerated corrosion effects and (2) Axial Offset Anomaly (AOA).

The corrective actions believed to be effective are:

- Higher pH primary water chemistry
- Zinc additions

For the pH strategy it is believed to avoid in any case pH_T-values of < 6.9 by increasing the LiOH concentration above a long time valid value of 2-2.2 ppm Li. However, such increases in lithium concentration may be a risk regarding the corrosion resistance of the zirconium alloys.

Specifically one environmental factor may be emphasized which is the corrosion products deposition on fuel surfaces, which can lead to increased cladding temperatures and increased corrosion rates. Such deposits have been identified as non-stoichiometric Ni-ferrites($Ni_xFe_{3-x}O_4$), Ni oxide or metallic Nickel. Such crud deposition occurs specifically at positions with sub-cooled boiling and may cause accelerated corrosion defects locally and axial power shifts by boron precipitation (AOA).

Zinc addition may also lead to a more degrading crud at positions with high steaming rates. Thus, surveillance programs after introduction of zinc are highly recommended, especially for PWRs with high duty cores. On the other hand, zinc reduces corrosion product release from system surfaces.

8.1.1 Higher pH Primary Water Chemistry; Lithium/B-Strategy

Historically, the starting point for all discussions about the correct pH for the PWR primary coolant can be found in P. Cohen's book "Water Coolant Technology of Power Reactors", 1969, and especially in the Chapter "The Physical Chemistry of Water and Aqueous Solutions". The central point in this document is the work of Sweeton et al., 1968, who reported measurements of the solubility of Fe from Fe_3O_4. They wrote the general dissolution reaction as

$$1/3\ Fe_3O_4\ (s) + (2-b)H^+ + 1/3\ H_2(g) = Fe(OH)_b^{(2-b)+} + (4/3-b)H_2O$$

This equation is valid for the temperature range of interest and applicable to dilute acidic and basic solutions. It also became clear that isotopically pure Lithium-7-hydroxide is the most suitable pH control agent to be used in the PWR Primary Coolant.

The result of Sweeton's work can be seen in Figure 8-1.

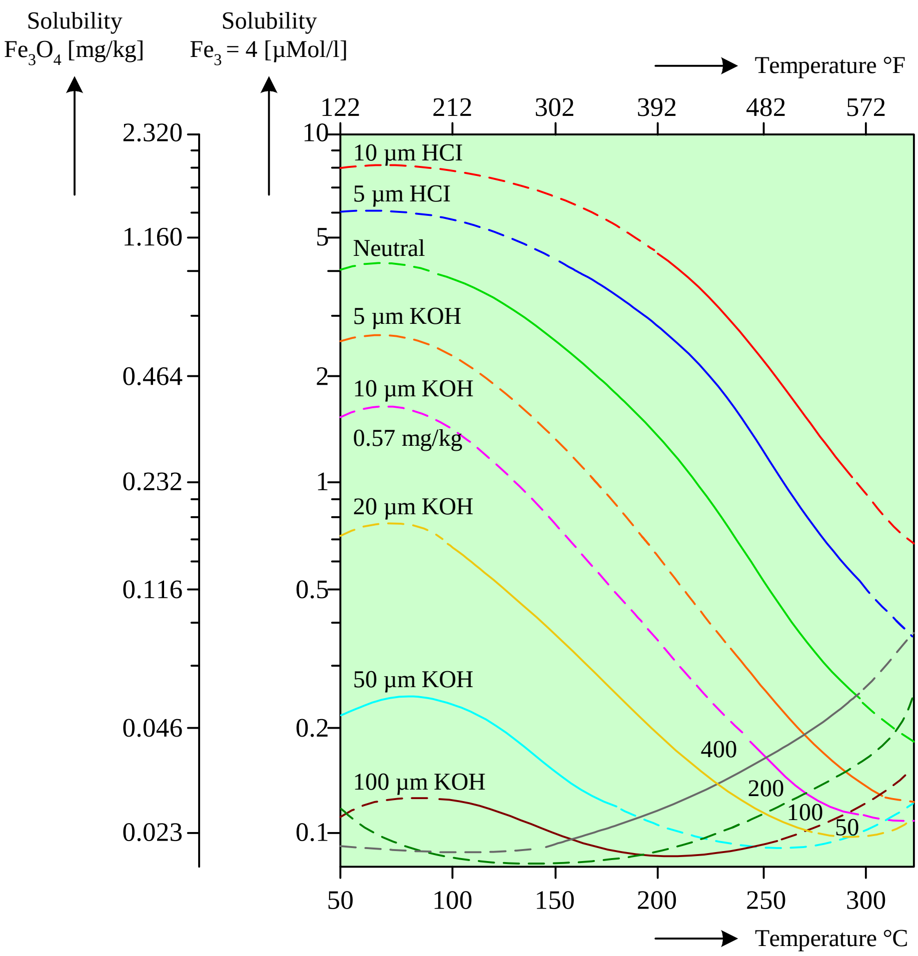

Figure 8-1: Magnetite Solubility by Sweeton et al., 1968.

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, pellet-cladding interaction (*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. This corrosion acceleration can be generated by e.g., CRUD deposition (CILC[11]), enhanced spacer shadow corrosion, ESSC,[12] (in *BWR*s), dry-out due to excessive fuel rod bowing.
Manufacturing defects	Non-through-wall cracks in the fuel cladding developed during the cladding manufacturing process. Defects in bottom and/or top end plug welds. Primary hydriding due to moisture in fuel pellets and or contamination of clad inner surface by moister or organics. Too large a gap between the fuel rod and the spacer grid supports (poor spacer grid manufacturing process) leading to excessive vibrations in *PWR* fuel causing fretting failures. Chipped pellets may result in *PCI* failures both in liner and non-liner fuel
PCI	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 *PCI* 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 an 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: Debris fretting in *BWR* and *PWR* Grid-rod fretting - Excessive vibrations in the *PWR* fuel rod causing fuel failures. This situation may occur for example due to different pressure drops in adjacent fuel assemblies causing cross-flow. Baffle jetting failures - Related to unexpectedly high coolant cross-flows close to baffle joints.

[11] Crud Induced Localised Corrosion – an accelerated form of corrosion that has historically resulted in a large number of failures in *BWR*s. 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.

[12] 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 to 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 2001-2004 were presented in the ZIRAT-9 Annual Report, Figure 9-1 to Figure 9-2. The failure statistics for the year 2004 in these figures are preliminary.

Most of the BWR failure cases are related to crud-accelerated corrosion failures. Other BWR failure cases involved six plants, which experienced PCI-like failures following control rod moves. Debris fretting also remains a problem even after the introduction of debris filters.

In PWRs the primary contributor to failure rates remains grid-to-rod fretting; however, experience with new grid designs appears to be promising. Last year it was noteworthy that some PCI-suspect failures were also experienced at three B&W-designed PWR plants following the movement of axial power shaping rods (APSRs) even though their calculated stress levels remained within the permissible range.

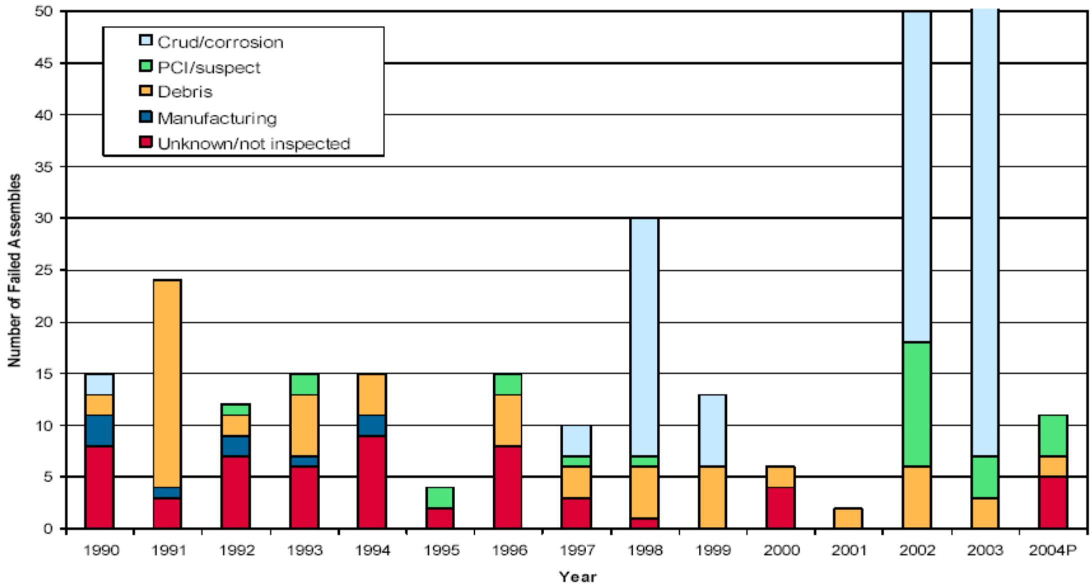

Figure 9-1: Trend in US failure root causes (2004 results are incomplete), Yang, et al., 2004.

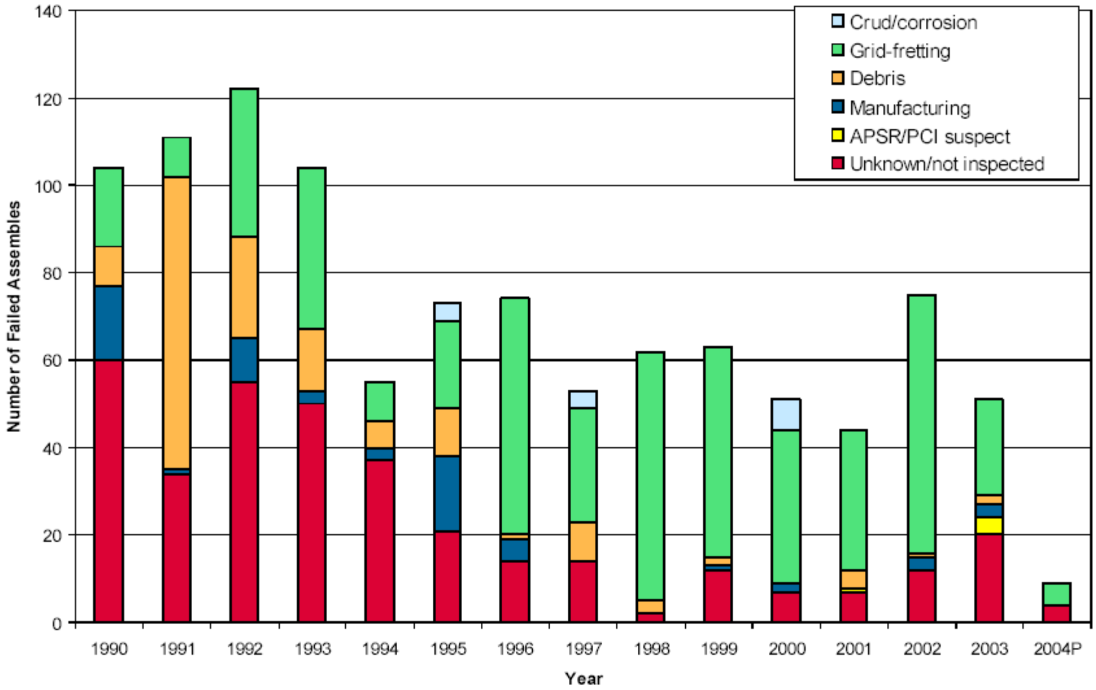

Figure 9-2: Trend in US *PWR* failure root causes (2004 results are incomplete), Yang, et al., 2004.

Fuel Reliability in Japanese PWRs and BWRs is very good. The fuel failure rate is less than 2.0E-6 or even lower in recent years, Ishiguma, 2004, Figure 9-3. *Also, for the Japanese BWRs the fuel failure rate has been very low with approximately 10E-6 failed rods per total operated rods over the last couple decades, Otsuka & Kitamura, 2004. The most probable root cause of most of these failures in ABWR plants is debris fretting.*

However, in comparing Japanese fuel performance with other countries one has to take into account that the outage duration is much longer in Japanese plants compared to plants outside Japan.

10 CLADDING PERFORMANCE UNDER ACCIDENT CONDITIONS

10.1 INTRODUCTION (PETER RUDLING)

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 utilised criteria in most countries are:

- Peak Cladding Temperature, PCT, < 1204°C (or 2200°F)
- Equivalent Cladding Reacted, ECR[37], < 17%
- Hydrogen gas produced < 1%[38].
- Fuel must have coolable geometry[39].

Core temperature maintained at low value for extended time[40].

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.

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[37] The ECR is defined as the total thickness of cladding that would be converted to stoichiometric ZrO_2 from all the oxygen that are contained in the fuel cladding as ZrO_2, 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 pre-accident oxidation and oxidation occurring during a LOCA.

[38] Total amount of hydrogen shall not exceed l% of the hypothetical amount generated by the reaction of all the zirconium in the fuel cladding with the water/steam.

[39] The coolable geometry may be lost by either fuel clad ballooning causing coolant channel blockage or fuel cladding fragmentation due to clad embrittlement.

[40] 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.

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. Moreover, *the forces due to pellet-cladding bonding have been identified as a significant force during post-LOCA.*

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 the reflood phase) 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.[41] However, if a best-estimate correlation had used instead such as, e.g., the Cathcart-Pawel correlation,[42] 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[43], 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, that is to say, what type of test should be used to ensure that the fuel cladding will not fragment neither under the quench-phase of the LOCA nor during the post-LOCA events such as e.g. a seismic event.

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[41] L. Baker and L. C. Just, 1962. Studies of metal-water reaction at high temperatures. III Experimental and theoretical studies of zirconium-water reaction, ANL-6548.

[42] J. V. Cathcart, 1976. Zirconium metal-water oxidation kinetics, IV. Reaction rate studies, ORNL/NUREG/TM-41.

[43] 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 1970's – early 1980's, slow ring-compression tests of ballooned and burst samples showed that the 1973 criteria failed to ensure retention of ductility at 135°C in narrow local regions near the burst opening[44], 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[45]. The implications of the results are such that for fuel claddings with hydrogen content exceeding about 700 wppm near the burst opening[46]:

- 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 oxidised at 1315°C are far more brittle than samples oxidised 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 greater than 0.7 wt%. Consideration of potential for runaway oxidation (*because the oxidation process of the zirconium metal becomes highly 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 oxidised by steam and it becomes brittle when severely oxidised.
- The embrittled cladding may rupture by thermal shock caused by rapid cooling (quenching) during the reflooding stage.

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[44] It appears that the stagnant conditions of water/steam in this location will significantly increase the hydrogen pickup during LOCA clad oxidation.

[45] 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%.

[46] 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 total reliance on intermediate storage. As a result the dry cask storage technology has become a major activity and business component of today's back-end fuel strategies.

The regulations for storing fuel are more or less settled as more data and analyses have become available. The US Reg. Guide (ISG 11, Rev. 3, November 2003) shifted 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 for the past 1-2 years has been on 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 in progress to provide regulations when such transports to the final (or other intermediate) storage site will occur. A major activity concerns the evaluation the properties of the fuel cladding and components, especially as related to the hydride distribution and orientation in the zirconium alloys, and their degree of resistance to fracture during hypothetical handling and transport accidents.

Expansion of wet storage facilities are still an option, albeit seldom used. In fact the threat of terrorism has spawned regulations that require rearrangement of fuel assemblies that can decrease currently available pool storage space.

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.

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.

Western Europe will have a slightly decreasing amount of fuel for storage if reprocessing continues. However, Eastern Europe (former Communist Block countries) have doubled the amount of fuel discharged and 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.

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. After 8 years of effort the Atomic Safety and Licensing Board (ASLB) gave a recommendation in February, 2005 to the NRC to issue a license for the site. The final ASLB hearing in August, 2004 was related to the risk of an F-16 aircraft crash into the site from a nearby Air Force base, a case that was presented successfully as an insignificant risk. The NRC Commissioners in turn requested the NRC staff in September, 2005 to issue a license --- an action that is expected before the end of 2005. Prior to construction there are still three items that need completion:

- The utilities that wish to use the site need to sign their contracts, (they are waiting for the license to be issued),

- The Bureau of Indian Affairs needs to give final approval to the lease of the land, (conditional approval has already been given pending issuance of a license),

- The Bureau of Land Management has to approve the rail line, based on part of a military spending bill that requires the Air Force to do a study, (other agencies have already approved the rail line).

The State of Utah will probably go to the Court of Appeals in Washington, DC or the 10th Circuit in Denver to appeal the license. The appeal might be able to proceed in parallel with construction. PFS hope construction could start in 2006, take about 2 years for *earliest receipt of shipments in 2008*, Martin, 2005.

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.

A brief summary of the status of the fuel related ISG’s is as follows:

- ISG 1, Rev. 1, Damaged Fuel, has not changed over the past year; however, a Rev. 2 will be coming out for public comment by early ’06 that will essentially endorse, with minor changes, the ANSI Std. 14.33-2005, “Characterization of Damaged Spent Nuclear Fuel for the Purpose of Storage and Transportation”.

- ISG 8, Rev. 2, Burnup Credit, no changes in the past year; data collection in process by DOE, EPRI, others,

- ISG 11, Rev. 3, Cladding Considerations for Transport and Storage, modifications are being discussed for a possible Rev. 4 and these are summarized in the next Section,

- ISG 19, Subcriticality Requirements During Accidents, no changes in the past year,

- ISG 22 is a new Guide that is in preparation to cover the issues raised by air blowdown of the moisture in the cask after loading and before seal welding. The cladding temperature could reach 350° – 400°C and the concerns are regarding potential problems with breached fuels.

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 *PWR*s, 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 may always be limiting for plant operation even with the new type of alloys. Furthermore, the influence of CRUD on corrosion may increase with increasing duty.

- 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), reduced assembly holddown forces, and other mechanical/themomechanical design changes, 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 fuel burnups up to 75 MWd/kgU. The *RIA* limits (threshold enthalpies) may continue to decrease as a function of burnup due to the increase in clad corrosion and hydrogen uptake.

- 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 MECHANCIAL PROPERTIES RELATED TO OXIDE THICKNESS AND H PICKUP*

- *BWR*s: 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, effect of water chemistry impurities, and specific effects of NMCA with or without Zn-injection, late increased corrosion at high burnups.

- *PWR*s: 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, due to surface contaminations and/or boiling. 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[54]:

- 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.

<u>Most sensitive component</u>

Spacer and fuel claddings.

<u>Increase margin for *PWR*</u>

- Improved knowledge of corrosion and hydrogen pickup mechanisms.

- Improved alloys with appropriate fabrication processes: ZIRLO/E635 (Anikuloy), M5/Zr1Nb. Duplex is another alternative that may be necessary to achieve satisfactory mechanical properties.

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[54] Due to the introduction of more corrosion resistant materials. For some types of Zr alloys, the hydrogen pickup fraction increases with decreasing corrosion rate.

- 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.

- 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 dimensional changes of components and differential dimensional changes 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, 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.

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APPENDIX A

Proceedings of the 2004 International Meeting on LWR Fuel Performance
Orlando, Florida, September 19-22, 2004
Paper 1002

Assessment Of Fuel Washout In LWRs – New Methodologies

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Abstract *– New approach for analysing fuel defects in BWRs and in PWRs have been developed. The main idea behind the models is to compare the activity release from one nuclide with short or very short half-life with that of one with long half-life. During operation of a non-defected core both the short and long-lived nuclides indicate the same amount of Fissile Materials (FM) on the core surfaces. During operation with a defected core the short-lived nuclide still reflects the amount of FM on the core surface while the source for the long-lived nuclide, Xe-133, is both from the defect itself as well as from the FM on the core surfaces. Generally, the dominating source for Xe-133 in the latter case is from the defect itself. Thus, when a defect occurs this can easily be seen since the models calculate a larger amount of FM based upon the measured Xe-133 activity compared to that based upon the short-lived nuclide.*

The occurrence of fuel washout can be seen with these models from the increasing amount of calculated FM based upon the short-lived nuclide. Analyses of routinely measured activity data from a large number of plants have shown that the defect itself has not given any significant contribution to the short-lived nuclide. This is a very important point that makes the models very useful. The new models can be used for the following:

1. *To detect a defect at start-up of the reactor after a shut-down. It is important to achieve the zero-failure goal at start-up to reduce the risk of fuel washout from failed rods.*

2. *To detect when a small defect occurs during operation with high sensitivity, even if the activity background level in the core is high.*

3. *To detect when fuel washout starts with very high sensitivity irrespective of applied water chemistry.*

4. *To quantitatively assess the increased amount of FM on the reactor surfaces when fuel washout occurs with high precision,*

For both BWR and PWR models, Xe-133 has been selected as the nuclide with long half-life, while, Sr-92 and Rb-89, respectively, has been selected to monitor the amount of FM on the core surfaces.

The developed methods and models can replace all currently used methods for analyses of fuel defects. In general, the new models are more sensitive and more precise than currently used methods and models.

I. INTRODUCTION

Activity release in relation to failed fuel has been a large issue for more than a decade and many different methodologies to assess the fuel rod defect status in the core have been developed over the years, e.g., by fuel vendors, EPRI, various research laboratories and universities. During certain conditions, the defected fuel rod may degrade by forming secondary defects transversal breaks and/or long axial cracks, axial splits. The activity release from a rod with secondary defects is normally larger than that of a rod with only primary defects. If the defect is large enough to allow water to get in contact with the fuel, fuel washout may occur. Fuel washout is a larger utility concern compared to a defected rod resulting in just high fission product release. The reason being that when the defected rod is extracted from the core, the activity in the coolant will decrease to the value prior to the failure. However, if fuel washout has occurred, significant amounts of Tramp Uranium, TU, will result in increasing the background activity level for many years. The TU levels will decrease over time due to that part of the TU is depositing on the fuel surfaces and during each outage a part of the core is replaced with fresh assemblies. To eliminate the risk of getting TU core contamination from a failed rod, the utility may choose to perform a forced outage to remove the failed pin (-s) before formation of secondary defects develop with potential TU core contamination as a result. However, the industry does not have today a reliable methodology to assess if emerging fuel washout occurs or not. Thus, large degree of fuel washout may already have occurred before the utility management realises that TU core contamination has taken place.

In PWRs, the current practice is to use I-134 as an indicator for fuel washout, but its half-life, 52.6 min., is far too long. An increasing I-134 activity may only be a consequence of a primary defect opening up, allowing water to get in contact with some of the fuel leaching out I-134 without getting fuel particles into the coolant. Thus, in this case fuel washout does not occur.

For BWRs, radio chemists are using Np-239 as a fuel washout indicator and this radionuclide works fine during Normal Water Chemistry, NWC. However, Np-239 cannot be used reliably during Hydrogen Water Chemistry, HWC, with/without Nobel Metal Chemical Additions, NMCA. This, since uranium and transuranium nuclides will form complexes that will deposit onto the system surfaces at reducing conditions. As a result the Np-239 concentration in the coolant at the sampling point can decrease with a factor of 100 or more when hydrogen is injected into the feedwater. Consequently, by measuring the Np-239 coolant concentration under such conditions, the fuel washout from the degraded rods would be underestimated by far.

The objective of this paper is to present a new approach to analyse radiochemistry data that reliably will assess if emerging fuel washout occurs both in PWRs and BWRs (both during NWC and HWC with/without NMCA). The BWR and PWR models are named ***BwrFuelRelease*** and ***PwrFuelRelease***, respectively.

II. SECONDARY DEGRADATION MECHANISMS

Degradation has historically been more of an issue in BWRs than in PWRs. During the period 1992-1993 six BWR plants in US and in Europe were forced into unscheduled outages because of concerns about failed *Zr-sponge liner fuel*[1], [1]. In all these cases, the very high off-gas activities resulted from only one or two failed rods. Both long axial cracks and significant loss of fuel pellet material were observed.

Failed rods in PWRs may degrade, but the amount of dispersed fuel as a consequence of this degradation is lower than that in a BWR. The rationale for the less severe behaviour in PWRs may be due to the fact that the coolant chemistry in a PWR is more reducing than in BWRs.

II.A. PWRs

In PWRs, the most severe type of degradation is normally the development of transversal breaks. The processes involved in developing a transversal break in a PWR rod is schematically shown in Figure 1 and Figure 2. Secondary hydride defects, that are a prerequisite for transversal breaks, tend to form at the position of the maximum clad surface temperature, which is in the upper part of a PWR rod. If the primary defect location is far enough from this part of the rod with highest temperature, the steam penetrating the primary defect will become "dry" enough to cause massive secondary hydriding in this location. If the hydride will penetrate the whole cladding thickness around the whole clad circumference, transversal break of the rod may occur, leading to fuel washout.

[1] *Zr sponge liner fuel* consists of a thin liner at the clad inner surface produced from Zr sponge material to improve PCI performance. No alloying elements have been added to this material and its major impurities are oxygen (about 600-900 wtppm) and iron (about 150-500 wtppm).

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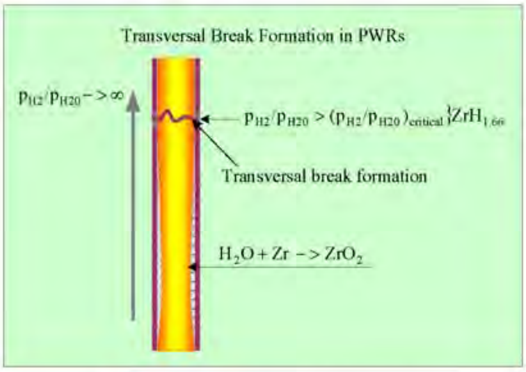

Figure 1: Schematics showing the events resulting in PWR transversal break formation.

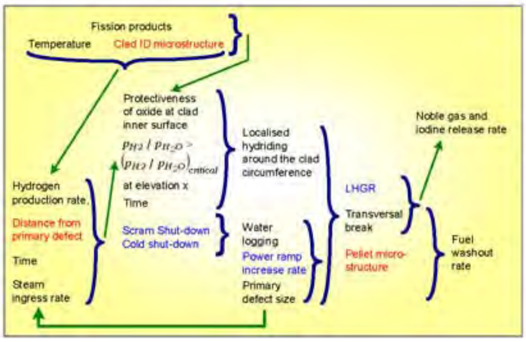

Figure 2: Schematics showing the parameters that may impact the PWR and BWR transversal break tendency. The key parameter that is related to operation is in blue colour while the corresponding parameters related to fuel design is in red colour.

II.B. BWRs

In BWRs, degradation of a failed rod may lead to either transversal breaks or axial splits, for more details see [2]. In some cases both of these types of secondary degradation defects are formed.

II.B.a. Transversal Break Formation

The transversal break formation is not correlated to power ramping (as formation of axial splits) but can result during operation of a failed rod during constant power, e.g., [3]. Sometimes, it seems that lowering of the reactor power to such an extent that the lower part of the rod may be filled with water, *waterlogging*, such as e.g. during a cold shut-down, may enhance the risk of getting a transversal break upon return to full power. The transversal breaks predominantly form in the lower part of failed rods with low burnup, Figure 3 and the parameters impacting the transversal break tendency are the same as those for PWRs, Figure 2.

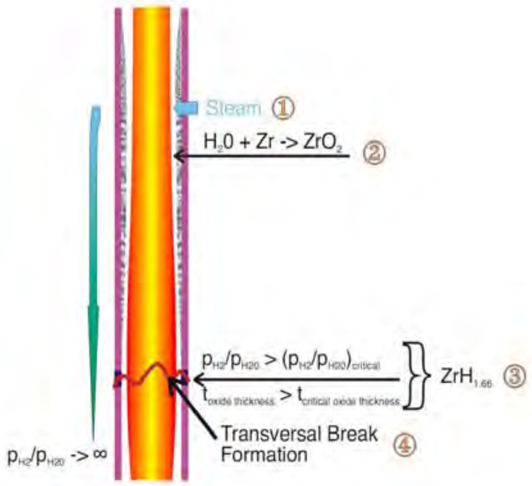

Figure 3: Schematics showing the events resulting in BWR transversal break formation. The numbers in the figure relate to the sequence of the different events that may lead to a transversal break.

II.B.b. Axial Split Formation

Literature data, e.g., [4] and [5] shows that long axial cracks, axial splits, only occur in conjunction with a power ramp of preferentially intermediate to high burnup rods. Thus, if a failed rod is not subjected to a power ramp, no axial split will form. It should be pointed out however, that power ramps must be performed in the reactor for other reasons and consequently, it will be impossible to run a plant without any power ramps. Also, most power ramps do not lead to axial split formation of failed rods.

The axial split formation is schematically shown in Figure 4. Figure 5 shows the different processes governing the fuel washout rate for a failed rod with an axial split.

APPENDIX B – UNIT CONVERSION

TEMPERATURE

°C + 273,15 = K

°C*1,8 +32 = °F

T(K)	**T (°C)**	**T(°F)**
273	**0**	32
289	16	61
298	25	77
373	**100**	212
473	**200**	392
573	**300**	572
633	360	680
673	**400**	752
773	**500**	932
783	510	950
793	520	968
823	550	1022
833	560	1040
873	**600**	1112
878	605	1121
893	620	1148
923	650	1202
973	**700**	1292
1023	750	1382
1053	780	1436
1073	**800**	1472
1136	863	1585
1143	870	1598
1173	**900**	1652
1273	**1000**	1832
1343	1070	1958
1478	1204	**2200**

DISTANCE

x (µm)	**x (mils)**
0,6	0,02
1	0,04
5	0,20
10	0,39
20	0,79
25	0,98
25,4	**1,00**
100	3,94

PRESSSURE

bar	**MPa**	**psi**
1	0,1	14
10	**1**	142
70	7	995
70,4	7,04	**1000**
100	10	1421
130	13	1847
155	15,5	2203
704	70,4	**10000**
1000	100	14211

MASS

kg	**lbs**
0,454	**1**
1	2,20

STRESS INTENSITY FACTOR

MPa√m	**ksi√inch**
0,91	**1**
1	1,10

CONVERSION OF DIMENSIONS	
1 Sv	= 100 Rem
1 Ci	= 3.7 x 10^{10} Bq = 37 GBq
1 Bq	= 1 s^{-1}