ZIRAT-7 ANNUAL REPORT

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1 INTRODUCTION

The objective of the Annual Review of Zirconium Alloy Technology (ZIRAT) is to review and evaluate the latest developments in zirconium alloy technology as they apply to nuclear fuel design and performance. The large amount of data presented at technical meetings, published in the literature, and provided through discussion with the vendors in the year 2002 are evaluated and discussed to highlight the significance of the results and their potential effect on fuel performance and reliability.

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

The work reported herein will be presented in two Seminars: one in Budapest on January 14-16, 2003, and one in Clearwater Beach, Florida, on January 28-30, 2003.

The term of ZIRAT-7 is January 1 to December 31, 2002

2 UTILITY AND REGULATORY BODY PERSPECTIVES (PETER RUDLING)

2.1 INTRODUCTION

2.1.1 Regulatory perspective

Regulatory bodies in many countries are concerned about a series of fuel issues often related to normal operation (class I) and anticipated operational occurrences (class II) that are:

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

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

The different licensing burnup levels in various countries are provided in Table 2-1.

Table 2-1: Licensed maximum burnup in various countries. Information is also given whether the burnup limit is related to class I/II (normal operation/anticipated operational occurrences) and class IV events (LOCA/RIA).

Related to	no formal limit from regulatory body normal operation	normal operation	no formal limit from regulatory body	limited by RIA	normal operation	no formal limit from regulatory body Limits related to LOCA, RIA and normal operation	LOCA, RIA		
Batch Average Assembly Average Rod Average	49 rialitied Actual 49 62	43	ca	45	Germany. Limit roarby enovitin - most plante.	enrichment limit of 4.4 %-56 to 57 MWdrkg U (assembly average); one plant enrichment limit of 4.6 %-61 MWdrkg II (assembly average).	68 Cappelled a violation of the control of the cont	52 57 (2010)	50 55
Country	Czeck Spain	Slovakia	Sweden	BWR	Finland	Germany	Sn	France	Japan

2.1.2 Utility perspective

The utility environment is shown in Figure 2-1 and more described in the following.

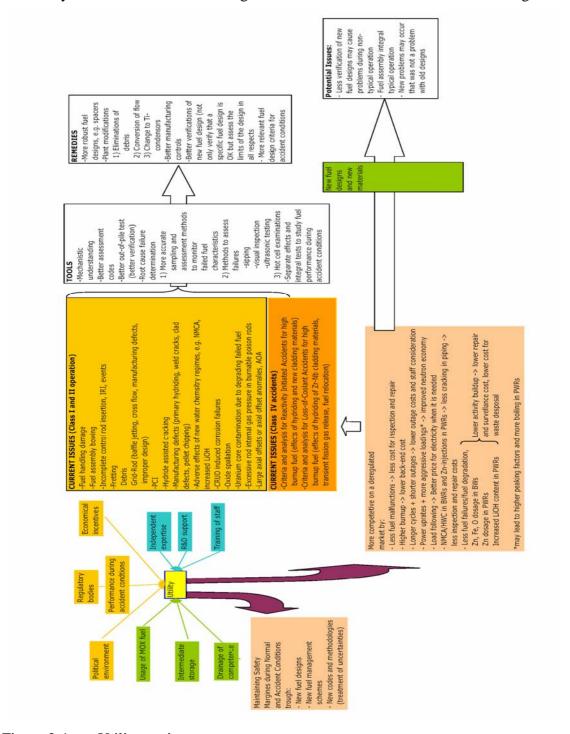


Figure 2-1: Utility environment.

One of the major present challenges to nuclear energy lies in its competitiveness. To stay competitive the industry needs to reduce maintenance and fuel cycle costs, while enhancing safety features. To meet these objectives, utilities are looking into the possibilities to:

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

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

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

The potential consequences of increased burnup on fuel are shown in Fig. 2-2 and 2-3.

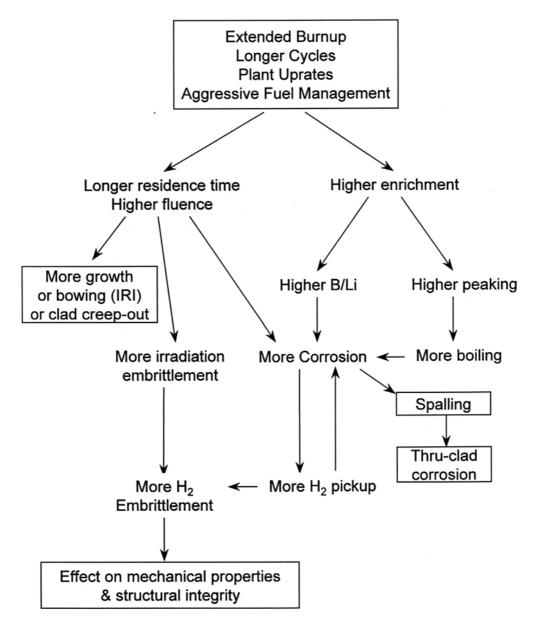


Figure 2-2: The effect of extended burnup on fuel performance – PWRs.

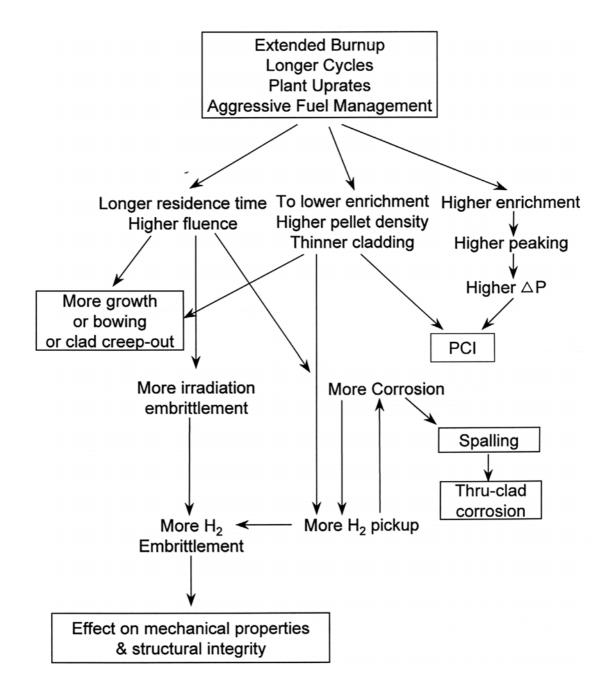


Figure 2-3: The effect of extended burnup on fuel performance – BWRs.

The planned and achieved burnups in BWRs and PWRs are provided in Table 2-2 and 2-3.

Table 2-2: Planned and achieved burnups in BWRs

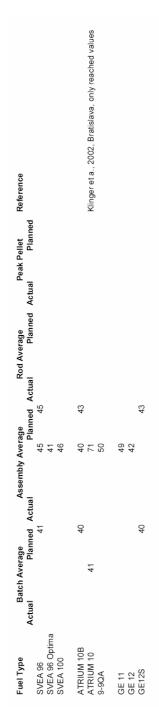


Table 2-3: Planned and achieved burnups in PWRs

3 2 6		4	2	₽ - E
Reference		*	4,7	m, Swe Nuclea tockholi
				ockholr ösgen Fuel, Si
Planned				uel, Ste n the G s, Top F
Licensed Bu Actual				c: ENS, Top F nriched Fuel ii ne", Proc: ENS Burnu
				ur, Prod High Ei gramm nce for
Planned 75		>70		ehaviou e with P V's Programar
Rod Average Actual				 A. Seibold, F. Garzarolli, R. Manzel, "Verifikation of High Burnup Materials Behaviour, Proc. ENS, Top Fuel, Stockholm, Swed Klinger et a., 2002, Bratislava W. Beck, R. Meinl Dr. G. Meier, "Fuel and Core Design for 5 Cycles Exposure with High Enriched Fuel in the Gösgen Nuclear J. M. Alonso, J. M. Alonso S. Azumi, S. Abeta, "Vandellos, 2 High Burnup LTA's Programme", Proc. ENS, Top Fuel, Stockholm S. Doi, S. Suzuki, M. Mori and T. Takahashi, "Advanced Fuel Design and Performance for Burnu
Planned 70	48 48 48			igh Burn gn for 5 C ellos, 2 H ced Fuel
0 0 7 0			46 46 46	ion of H e Desi "Vand
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Assembly Average Actual	44 47 58- Ref. 2 47			Manzel, "V /a eier, "Fuel a S. Azumi, S. nd T. Takal
Planned 65	44 74 74			zarolli, R. Pratislav Dr. G. Me Alonso S. M. Mori a
rage 50 50 50 .58nF				F. Gar. r., 2002 . Meinl o, J. M suzuki,
Batch Average Actual 5 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	33 - Ref. 2			1 A. Seibold, 2 Klinger et a 3 W. Beck, R 1 J. M. Alons 5 S. Doi, S. S
Fuel Type Batch Average Actual DX ELS0.8a clad. DX ELS0.8b clad. DX D4 HTP HTP S0 HTP X5 (5%enrichment) with Zr0.5SnF	W AEF+ AFA-3G AFA-3GA	17X17 W (ZIRLO) MHI (MDA) S2 cladding	MHI (Zry-4) 14x14 15x15 17x17	References

4.60% 4.40% 4verage Enrichment (%) 4.20% 4.00% BWR 3.80% ■ PWR 3.60% 3.40% 3.20% 3.00% 1991 1992 1993 1994 1995 1996 1997 1998 1999 2000 Year

The trends in the initial fuel enrichments are show in Figure 2-.

Figure 2-4: Trends in initial fuel enrichement, Cheng, 2002.

2.2 SUMMARY

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

- 1. Longer cycles
- 2. Higher discharge burnups
- 3. Modified water chemistries
- 4. Plant power uprates
- 5. Increased coolant temperatures
- 6. More aggressive fuel management methods

However there are a number of different issues that needs resolution to be able to successfully implement the above mentioned changes, Figure 2- and Figure 2-. The most important issues are:

- 1. Oxidation levels higher than predicted,
- 2. Excessive internal gas pressure in burnable poison rods,
- 3. Incomplete control rod insertion (IRI) events,

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- 4. Large axial offsets or axial offset anomalies (AOA),
- 5. Fuel failures due to high fuel duty,
- 6. Adverse effects of water chemistry,
- 7. High crud buildup, and
- 8. Accelerated growth of rods and assemblies
- 9. Fretting
- 10. Criteria and Analysis for Reactivity Accidents
- 11. Criteria and Analysis for Loss-of-Coolant Accidents
- 12. Criteria and Analysis for BWR Power Oscillations

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

3 ZIRCONIUM ALLOY SYSTEMS (PETER RUDLING)

3.1 INTRODUCTION

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

Table 3-1: Zirconium base material currently used, Garzarolli, 2002, (ZIRAT-7 Special Topical Report on Corrosion).

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

Table 3-1: Zirconium base material currently used, Garzarolli, 2002, (ZIRAT-7 Special Topical Report on Corrosion). Cont'd

Alloy	Sn %	Nb %	Fe %	Cr %	Ni %	O %	Others alloy elem.	%	Ref.	
2. Experimental alloys, Cont'd										
ZrSnFeO	0.5	-	0.4	-	-	0.22	-		13)	
Alloy-C	0.4	-	0.5	0.24	-	0.18	-		13)	
HPA-4	0.4-0.6	-	Fe	-	-		V		14)	
Valloy	-	-	0.15	1.2	-	-	-		7)	
Zr.7Fe0.7Ni	-	-	0.7	-	0.7	0.03	-		8)	
Zr0.25Fe0.2V	-	-	0.25	-	-		-		7)	
E635	1.1-1.4	0.9-1.1	0.3-0.5	-	-	0.05-0.07	-		3)	
Alloy-A	0.5	0.3	0.35	0.25	-	0.15	-		13)	
Alloy-E	0.7	0.4	0.45	0.24	-	0.13	-		13)	
NSF 0.2	1	1	0.2	-	-	0.1	-		7)	
T18/I18	1	0.6	0.2	-	0.05		-		15)	
NSF 0.5	1	1	0.5	-	-	0.1			15)	
Zr3Nb1Sn	1	2-3	-	-	-		-		9)	
Ozhenite 0.5	0.2	0.1	0.1	-	0.1		-		7)	
M3	0.5	0.5	0.25	-	-	0.12	-		10)	
0.2 Nb Zry-2	1.5	0.2	0.15	0.10	0.06		-		15)	
0.5 Nb Zry-2	1.5	0.5	0.15	01	0.05		-		12)	
D3	1.4	1	0.2	0.1	-		-		11)	
EXCEL	3.5	0.8	-	-	-		Mo	0.8	7)	
XXL	1.2	0.3	-	-	-		Mo	0.3	12)	
T12-15/I12-15	1	1-2	-	-	-		Mo	0.2-0.5	15)	
T19/I19	1.4	0.4	-	-	-		Te	0.2	15)	
T20/I20	1.2	-	-	-	-		Te	0.6	15)	
BAG	-	0.5	-	-	-		Bi	1	12)	
T68	0.8	-	0.3	0.1	0.1		Cu/Ta	0.2/0.2	16)	
T40	1	-	0.25	0.1	-		Cu/Ta	0.1/0.2	16)	

¹⁾ ASTM; ²⁾ Shebaldov, et al., 2000; ³⁾ Solonin, et al., 1999; ⁴⁾ Comstock, et al., 1996; ⁵⁾ Garzarolli, 2001; ⁶⁾ Mardon, et. al., 2000; ⁷⁾ Cox, et al., 1998; ⁸⁾ Amaev, 1971; ⁹⁾ Garzarolli, et al., 2001; ¹⁰⁾ Mardon, et. al., 1994; ¹¹⁾ Besch, et al., 1996; ¹²⁾ Ishimoto, et. al., 2000; ¹³⁾ Garde, et al., 2001; ¹⁴⁾ Seibold, 2001; ¹⁵⁾ Etoh, et al., 1996;

¹⁶⁾ Takeda and Anada, 2001

3.2 ZR-NB

The precipitation of the β Nb phase from α was studied in Zr-Nb alloys containing 0.55 to 2.5 wt% Nb at a temperature of 570C from 2 to 5000 hours by Toffolon et al., 2001. The $(\alpha+\beta_{Zr})$ and $(\alpha+\beta_{Nb})$ microstructures are shown in Figure 3-1. The precipitation process was studied by the calorimetric and the thermoelectric power methods, Figure 3-2 and 3-3. Based upon the results the authors suggest that the structure development is the following:

$$\alpha Zr + \beta Zr_{(\approx 20\% Nb)} \rightarrow \alpha Zr + \beta Zr_{(\approx 20\% Nb)} \downarrow + \beta Nb_{(\approx 80\% Nb)} \uparrow \rightarrow \alpha Zr + \beta Nb_{(\approx 80\% Nb)}$$

The results indicate that, the development of the stable $(\alpha + \beta_{Nb})$ structure in Zr-Nb alloys containing 1 % Nb may take a much longer time than that used during manufacturing of commercial alloys such as M5 and ZIRLO. It is well known that a $(\alpha + \beta_{Nb})$ structure is a prerequisite for good in-pile corrosion performance.

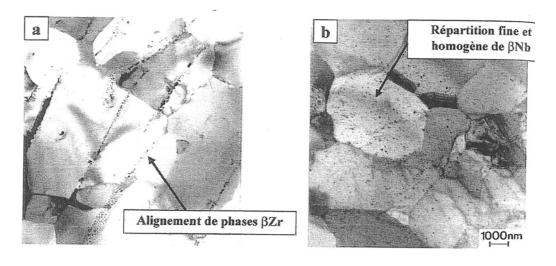


Figure 3-1: Micrographs showing (a) $(\alpha+\beta_{Zr})$ structure and, (b) $(\alpha+\beta_{Nb})$ structure, Toffolon et al., 2001.

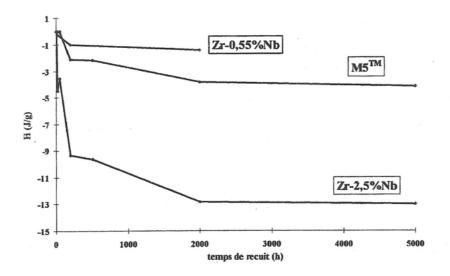


Figure 3-2: Precipitation of the β_{Nb} phase from a temperature of 570C studied by the calorimetric method, Toffolon et al., 2001.

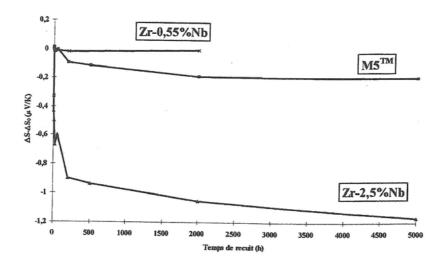


Figure 3-3: Precipitation of the β_{Nb} phase from a at a temperature of 570C studied by the Thermoelectric Power method, Toffolon et al., 2001.

4 MECHANICAL PROPERTIES (PETER RUDLING)

4.1 INTRODUCTION

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

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 $31\tilde{0}^{\circ}$ 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 $4\cdot10^{17}$ n·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 400°C for 24 hours, resulting in a structure consisting of elongated grains of hexagonal-close-packed (hcp) α -Zr, partially surrounded by a thin network of filaments of body-centred-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 α -phase particles that are low in Nb, surrounded by an Nb-enriched β -Zr matrix. The hcp α -Zr grains are oriented with their unique c-axes aligned in the radial-transverse plane, mostly tilted towards the transverse direction.

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

- the fuel assembly will not *fail* during normal operation (class I) and anticipated operational occurrences (class II). *Failing* in this sense has a broader meaning, namely that the fuel rod may not be breached and that the dimensional changes of the assembly during irradiation must be limited. The latter requirement is to ensure that control rods can be inserted and that the fuel can be handled during shutdown. Also the BWR fuel outer channel cross section must not have increased to such an extent that it is impossible to pass it through the upper core grid during reloading.
- the fuel remains coolable during an accident (class III and IV). Class IV design basis accident are *LOCA*, *RIA* and earthquake. During class III and IV situations limited fuel failures are however accepted. Another criterion that must be fulfilled in these situations is that it should be possible to insert the control rods.
- During class I and II operation, the following mechanical *failure* mechanisms and corresponding design criterion for the fuel assembly, including its components, are listed in *SRP* section 4.2:
- Plastic deformation the component is regarded as *failed* if it is plastically deformed and the appropriate criterion is that the stresses must be lower than the yield stress. *SRP* section 4.2 also state what type of methodology should be used when calculating these stresses. In these calculations the stress in the assembly location subjected to maximum stresses is calculated. In calculating this stress, all types of stresses are taking into account, such as welding residual stress, thermal stress, stress imposed by rod-system differential pressure, etc. It is interesting to note that the criterion on maximum allowable oxide thickness on fuel rods is related to this criterion. If the oxide thickness becomes too large in a *PWR*, the oxide thickness will increase the cladding temperature due to its lower thermal conductivity and would then increase corrosion rate. The oxide thickness would increase further, raising the clad temperature and corrosion rate, resulting in thermal feedback. Since increasing temperature decreases the yield strength of the material, the material would eventually mechanical fail, i.e. plastically deform, provided that the cladding stresses are large enough.
- Excessive creep deformation that could either result in creep fracture or too large plastic deformations that could e.g. lead to *dryout* due to excessive outward creep of the fuel cladding diameter that would limit coolant flow. Creep occurs at a stress level lower than the yield stress. The corresponding criterion is very general and just specify that the creep deformation must be limited.
- Fatigue failure Most fuel assembly components are subjected to fatigue stresses and *SRP* section 4.2 provides the maximum allowable fatigue stress level.

- *PCI* The criterion to eliminate this type of failure is by limiting the elastic and uniform plastic deformation in the cladding circumference during a class I and II transient to 1%. This value is of course not sufficiently to ensure that *PCI* failures do not occur. However, the fuel vendors are still designing their fuel so this 1 % limit is achieved in their design.
- Hydride embrittlement The criterion just mentions that the hydrogen content in the material must be limited so the fuel assembly component will not fail.

During accident conditions such as *LOCA* and *RIA*, the mechanical performance of the fuel cladding is crucial to meet the objective that the fuel must remain coolable during these types of accidents. In both situations, it is important that the fuel cladding may not fail in a brittle fashion during the *reflooding*¹ phase during *LOCA* and due to *PCMI*² during the *RIA* transient.

Fuel vendors have developed codes to model the fuel assembly mechanical performance during class I, II, III and IV situations. To be able to do the modeling correctly, data on mechanical performance of the fuel assembly must exist. The data are generated in two types of tests, either separate effect tests and integral tests. The former test studies only the impact of one parameter at a time on the mechanical performance, see e.g. Adamson and Rudling, Special Topical Report on Mechanical Properties, 2001. This could e.g., be the impact of hydrogen content on ductility. The data from these separate effect tests are then used by the fuel vendor to develop adequate models in their fuel performance codes. To then verify that the code comes up with the correct prediction on fuel assembly mechanical performance e.g. during a *LOCA*, the code predictions are benchmarked towards integral tests. In the integral test, the fuel assembly design and environment is as similar as possible as is existing in the situation that is simulated in the test, e.g. a *LOCA*.

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).

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4.2 NEW RESULTS

4.2.1 Cladding tubes

Bérat-Robert, et al., 2001, studied the impact of oxide thickness layers on mechanical properties of Zry-4 cladding tubes. *The results may increase the knowledge of what may happen with the fuel during intermediate storage*. The tubes were preoxidised at 450°C in CO₂ gas to eliminate hydrogen pickup that would otherwise occur during oxidation in water. Subsequently, the oxidized claddings were subjected to uni-axial tensile test at 400°C. Test specimen length was 85 mm and the strain rate was 2.5·10⁻⁴s⁻¹. The data in Figure 4-1 and Table 4-1 shows that increased oxide layer thickness increases material strength and reduces ductility. *No mechanistic explanation is given in the paper why the oxide layer increases the material strength*.

Table 4-1: Chemical composition of tested Zry-4 material, wt%, Bérat-Robert, et al., 2001.

Γ	Sn	Fe	Cr	0
Γ	1.30	0.21	0.12	0.128

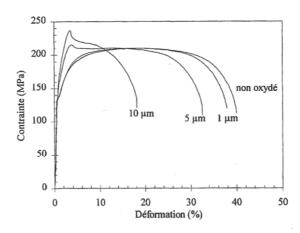


Figure 4-1: Tensile test results of RXA Zry-4 tubes, Bérat-Robert, et al., 2001.

Table 4-2: Tensile test data obtained in the 400°C tensile test, Bérat-Robert, et al., 2001.

Epaisseur d'oxyde par face (μm)	R _{p0,2} (MPa)	R _m (MPa)	Ar	A _t
0	134	. 210	0,176	0,393
1	134	210	0,163	0,378
5	145	214	0,027	0,321
10	160	241	0,023	0,190

To be able to model the fuel rod performance in-pile more accurately, mechanistic models are needed. In a study presented by Fandeur, et al., 2001, the impact of texture on the anisotropy of the mechanical properties was studied to develop a mechanistic model, Figure 4-2.

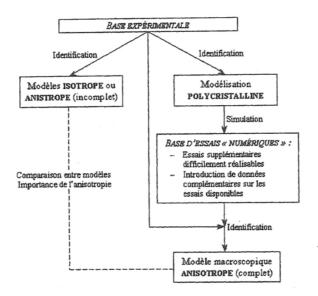


Figure 4-2: Procedure to completely characterize the mechanical behaviour of the Zry- 4 material, Fandeur, et al., 2001.

4.2.2 Pressure tubes

Investigations of fracture toughness and DHC velocity were carried out using unirradiated Zr-2.5Nb and Zr-1.2Sn-1Nb-0.35Fe pressure tubes of the CANDU and RBMK types that had different compositions, structure-phase states, textures and strengths, Table 4-3, Markelov, et al., 2002. The tests were perfromed at 250°C on precracked compact tensile specimens, Figure 4-3. Before testing for DHC the specimens were hydrided to the hydrogen contents of 60-100 ppm.

Table 4-3: Characteristics of Pressure Tube Materials Studied, Markelov, et al., 2002.

Tub a matarial	Characterine Diverse Charles	Texture parameters			
Tube material	Structure-Phase State	f_R	F_{T}	f_L	
CANDU Standard Zr-2.5Nb (Ø 103×4 mm)*	Plate – like α – Zr grains strongly extended along tube axis and separated by thin β – Zr interlayers.		0.56 ÷ 0.61	0.04 ÷ 0.09	
Experimental 1 Zr-2.5Nb (Ø 103×4 mm)*	Extended along tube axis α – Zr grains and β – Zr intelayers (~15% of volume).	0.46	0.52	0.02	
Experimental 2 Zr-2.5Nb (Ø 103×4 mm)*	Plate – like residual α – Zr grains extended along tube axis (~20% of volume) and equiaxed α – Zr grains with β – Nb precipitates.	0.54	0.42	0.04	
RBMK Standard Zr-2.5Nb (Ø 80×4 mm)*	Plate – like α – Zr grains extended along tube axis (~60-80% of volume) and recrystallized equiaxed α – Zr grains with β – Nb precipitates.		0.45 ÷ 0.46	0.05 ÷ 0.08	
RBMK TMT-2 Zr-2.5Nb (Ø 80×4 mm)*	Equiaxed residual α – Zr grains and transformed plate – like α – Zr grains with precipitates of β – Nb and residual β – Zr phase	0.37	0.56	0.07	
RBMK TMT-1 Zr-2.5Nb (Ø 80×4 mm)*	RBMK TMT-1 Zr-2.5Nb Equiaxed residual α – Zr grains and martenistic type transformed α – Zr grains with β – Nb precipitates		0.42	0.17	
RBMK Zr-1.2Sn-1Nb-0.35 Fe (Ø 80×4 mm)*	Plate like α – Zr grains and recrystallized equiaxed α – Zr grains (~55-70% of volume) with intermetallic Zr-Nb-Fe precipitates.		0.48	0.06	
Experimental 3 Zr-1.2Sn-1Nb-0.35 Fe (Ø 103×4 mm)*	Residual α –Zr grains extended along tube axis (~20% of volume) and finely dispersed transformed α – Zr grains with intermetallic Zr-Nb-Fe precipitates.		0.45	0.05	

*Inner diameter and wall thickness of tube

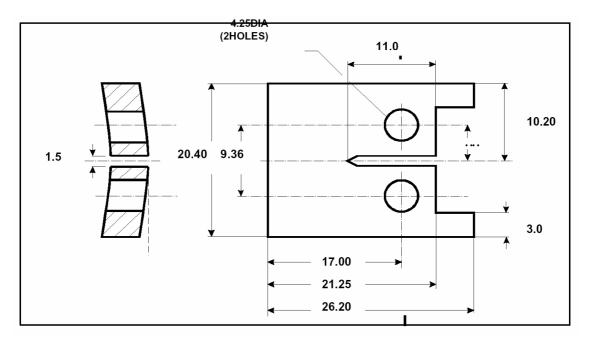


Figure 4-3: Compact tensile specimen for fracture toughness and DHC tests, Markelov, et al., 2002.

Markelov, et al., 2002 reported that the fracture toughness of CANDU pressure tubes of Zr-2.5Nb alloy produced on sponge Zr base is governed by the chlorine impurity content that in turn is related to the specific manufacturing process to produce CANDU pressure tubes based upon Zr sponge material. However, the content of chlorine in Zr-2.5Nb alloy in RBMK pressure tube is much lower since this material is produced from mixed iodide and electrolytic Zr. Thus, the fracture toughness of RBMK pressure tubes will be governed by other factors (fluorine as well as texture and structure-phases in the alloy) than the chlorine content.

Table 4-4 lists the results of fracture toughness testing pressure tube specimens of different types fabricated from Zr-2.5Nb produced in Russia of mixed electrolytic and iodide zirconium base. It is observed that the standard CANDU pressure tubes show lower fracture toughness than that of the experimental CANDU pressure tubes. The table also shows that fluorine content of < 5 ppm in Zr-2.5Nb alloy fabricated from electrolytic and iodide Zr mixture does not influence its fracture toughness. The f_R/f_T provides the ratio of grains with the basal pole in the pressure tube radial to the tangential direction. Both from Table 4-4 and Figure 4-4, it appears that increasing f_R/f_T ratio increases fracture toughness.

Table 4-4: Fracture Toughness of Electrolytic and Iodide Based Zr-2.5Nb Pressure Tubes at 250₆C (dJ/da), Mpa, Markelov, et al., 2002.

Tube type and number	f _R /f _T	Content of fluorine, ppm	dJ / da, ÌPà mean ± rms
CANDU standard 1	0.68	4.8	291.0 ± 2.5
CANDU standard 2 (front end)	0.57	2.3	220.6 ± 10.0
CANDU standard 2 (back end)	0.63	2.4	257.1 ± 5.6
Experimental 1	0.88	4.5	322.6 ± 7.2
Experimental 2	1.29	4.3	354.6 ± 2.4
RBMK standard 1	1.04	4.7	339.9 ± 8.8
RBMK standard 2	1.07	4.9	336.9 ± 16.3
RBMK TMT – 2	0.66	1.2	242.9 ± 1.7

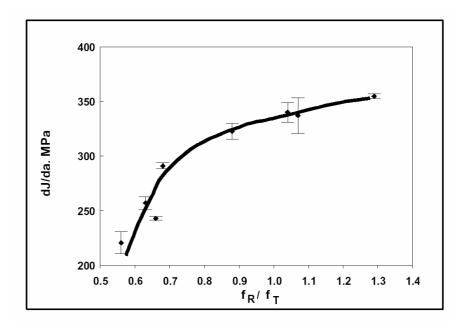


Figure 4-4: Fracture toughness (dJ/da) of Zr-2.5Nb pressure tubes vs texture parameter ratio f_R/f_T , Markelov, et al., 2002.

The DHC velocity is primarily defined by the pressure tube material strength and consequently, the highest DHC velocity is obtained by standard CANDU pressure tubes having the highest strength properties. However, the DHC velocity of RBMK pressure tubes subjected to standard processing may be more than an order lower.

The results of pressure tube specimens testing for DHC velocity are summarized in Table 4-5. It is noted that the highest DHC velocity is observed in standard CANDU pressure tube specimens.

Table 4-5: Results of pressure tube specimens testing for DHC velocity at 250°C, Markelov, et al., 2002.

		Ê₁		DHC	DHC
Tube material	YS [™] *	initial	Incubation time	crack length	velocity
	(ÌPà)	(ÌPà⋅m¹/²)	(min)	(mm)	(m/s)
CANDU		15.5	3.5	1.619	7.36×10 ⁻⁸
standard		16.0	4	2.276	11.56×10 ⁻⁸
Zr – 2.5Nb	596.7	15.1	14	1.829	8.59×10 ⁻⁸
21 - 2.0140	330.7	15.1	42	1.719	6.62×10 ⁻⁸
		15.5	6	1.542	5.83×10 ⁻⁸
		15.4	7	1.749	8.14×10 ⁻⁸
					0.1.1.10
RBMK		20.7	3240	0.977	1.71×10 ⁻⁹
standard 1	255.0	20.7	1244	1.257	3.22×10 ⁻⁹
Zr – 2.5Nb		20.8	1560	1.167	1.90×10 ⁻⁹
DDIMIC	0.40.0	47.0	400	0.400	
RBMK	342.0	17.9	160	2.488	6.04×10 ⁻⁹
TMT-2		19.1	140	2.025	6.14×10 ⁻⁹
Zr – 2.5Nb		18.8	197	1.695	6.62×10 ⁻⁹
RBMK		15.1	27.5	1.900	3.22×10 ⁻⁸
TMT-1	524.0	15.5	28.5	1.937	3.31×10 ⁻⁸
Zr – 2.5Nb	024.0	15.7	11	2.122	3.30×10 ⁻⁸
2.0145		16.0	11	1.896	3.20×10 ⁻⁸
RBMK	280.0	21.1	1104	1.718	4.89×10 ⁻⁹
Zr-1.2Sn-1Nb		21.0	3300	1.416	2.09×10 ⁻⁹
– 0.35Fe					
RBMK	365.0	18.6	82	1.579	1.00×10 ⁻⁸
standard 2		21.5	96	1.986	0.86×10 ⁻⁸
Zr – 2.5Nb		21.0	30	1.500	0.86×10
RBMK	455.0	17.0	29	1.975	1.32×10 ⁻⁸
standard 3		17.0	18	2.617	1.71×10 ⁻⁸
Zr – 2.5Nb					
Experimental	497.0	17.6	60	1,952	3.16×10 ⁻⁸
Zr-1.2Sn-1Nb-	497.0	17.5	42	2,060	3.16×10 ⁻⁸
0.35Fe		17.5	72	2,000	2.47×10
	at 250°C in tana	antial disantian of	t. do o		

^{* 0.2%}Yield Stress at 250°C in tangential direction of tube

It is known that β -Zr interlayers are present in Zr-2.5Nb CANDU pressure tubes and that these interlayers serve as fast diffusion paths for hydrogen diffusion to a crack tip. However, in the RBMK pressure tube material β -Zr interlayers does not exist independent of the RBMK pressure tube composition or thermomechanical treatment. This difference in microstructure may explain the lower DHC velocity in RBMK tube specimens. However, also the material strength impacts fracture toughness, Figure 4-5.

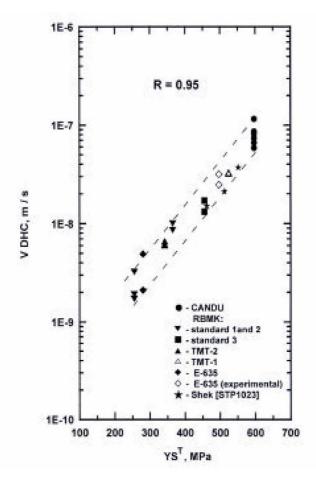


Figure 4-5: DHC velocity vs yield stress in the direction of normal to hydride cracking plane in Zr alloy at 250°C Markelov, et al., 2002.

The DHC process is known to proceed in a step-wise mode and the fracture surface is characterised by formation of striations. After DHC the 250° C the interstriation spacing in specimens of a standard Zr-2.5Nb CANDU pressure tube averages 20.6 μ m, Figure 4-6.

Figure 4-7 shows that the DHC velocity decreases with increased interstriation spacing in hydride crack fracture normalized to the strength of Zr-2.5Nb alloy.

5 DIMENSIONAL STABILITY (RON ADAMSON)

The broad topic of dimensional stability of reactor components is normally reviewed here. Topics include Component Growth, Fuel Rod Growth, Creep, Mechanisms, Experimental Facilities, and Summary Highlights. In the wake of the 13th ASTM Symposium on Zirconium in the Nuclear Industry in Annecy in June 2001, and since the ASTM book on the Symposium has not yet been issued, very few reports on these topics have appeared in the open literature in the past 12 months. Since an extensive report on dimensional stability, Adamson and Rudling, 2002, was issued in 2002, no literature review will occur in Section 5 this year.

In review, the primary sources of dimensional changes of reactor components (in addition to changes caused by conventional thermal expansion/contraction) are:

- irradiation growth
- irradiation creep
- thermal creep
- stress relaxation (which is a combination of irradiation and thermal creep)
- hydrogen and hydride formation

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

Hydrogen effects are arguably independent of irradiation, although that is yet to be conclusively shown. Factors which influence corrosion and hydrogen pickup by the zirconium alloy drive the substantial dimensional changes that can be caused by hydrogen or hydrides. It is noted that although it is known that many variables affect hydrogen absorption and distribution in Zirconium alloys, and much has been learned recently about the impact of hydrogen and hydrides on dimensional change, not much is known on the effects of those variables on the observed changes.

Many factors affect the size and magnitude of dimensional changes, as illustrated in Table 5-1.

Table 5-1: Parameters which affect dimensional changes caused by various mechanisms.

		Growth	Irradiation Creep	Thermal Creep	Hydride
•	stress		V	V	?
•	dislocation density, cold work	\checkmark	$\sqrt{}$	$\sqrt{}$?
•	fluence	$\sqrt{}$	$\sqrt{}$	$\sqrt{}$?
•	flux		$\sqrt{}$	$\sqrt{}$?
•	temperature	$\sqrt{}$	$\sqrt{}$	$\sqrt{}$?
•	texture	$\sqrt{}$	$\sqrt{}$	$\sqrt{}$?
•	metallurgical factors	$\sqrt{}$	$\sqrt{}$	$\sqrt{}$?
•	heat flux		?	?	?

Considerable effort in the past 30 years has resulted in a good empirical description of creep and growth; that is, predictions can be made of dimensional changes expected under various service conditions. Also good progress has been made, particularly in the last decade, in understanding the mechanisms involved. And in the past few years, an enlightened understanding of the effects of hydrides on dimensions has been attained. The past several ZIRAT reviews have covered all these topics in detail. There are, however, several areas for which more data and understanding is required to assure optimum component performance at the high fluences and burnups currently achieved and proposed for modern fuel designs. These areas include but are certainly not limited to

- factors affecting formation of <c> component dislocations
- effects of alloying elements such as Nb, Fe and Sn and impurities such as C and S
- temperature dependence of growth and creep over the full range of operating temperatures
- long term effects of small residual stresses
- lack of comparable high fluence data for the variety of new alloys being introduced for corrosion resistance
- effects due the influence of heat treatment on microchemistry and texture
- detailed influence of neutron flux and energy spectrum on creep (and perhaps growth, although the current wisdom indicates no effect)
- effects of hydride orientation and distribution on dimensional changes.

Active research and development programs continue in the important area of dimensional stability. Most technical conferences concerning nuclear materials have papers reporting new results. New developments will be followed by ZIRAT. It is anticipated that the current Special Topics Report will provide ample background to facilitate interpretation of future results.

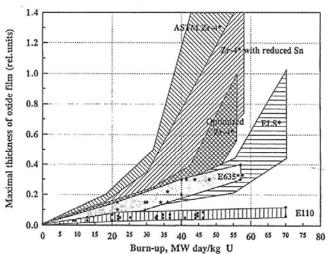
6 CORROSION AND HYDROGEN UPTAKE (BRIAN COX)

6.1 INTRODUCTION

The extensive Special Report on Corrosion covers the present state of knowledge on the in-reactor corrosion and hydrogen uptake mechanisms. Thus, there is no necessity to summarise this report here. However, a brief listing of the mechanistic conclusions could be useful.

- Although oxide films on zirconium alloys grow by the inward diffusion of oxygen ions through some thin residual barrier layer of oxide at the oxide/metal interface. The rate limiting process is actually the conduction of electrons in the opposite direction. Thus, the major increase in electronic conductivity in the zirconium oxide resulting from the in-reactor β/γ (fields can result in enhanced oxide growth under certain conditions (e.g. proximity of a dissimilar metal "shadow corrosion"). Significant potential differences between Zircaloys and steels or nickel alloys are present only in BWR water chemistry. Dissolved hydrogen eliminates these.
- There is no evidence of significant damage (e.g. point defect clusters or dislocation loops) from fast neutrons in zirconia films that could lead to enhanced oxygen diffusion and hence increased rates of growth of pre-transition oxide films.
- Irradiation enhancement of corrosion rates in-reactor commences at the rate transition to the linear post-transition corrosion period. The causes of the enhanced post-transition corrosion are primarily the degradation of the corrosion resistance of the Zircaloys by the knock-on displacement of Fe atoms from the unstable intermetallics particles (SPPs) into the Zr matrix. Alloys that contain more stable SPPs or no Fe SPP phases (e.g. Zr-Nb alloys) do not show this degradation of corrosion resistance in-reactor and in some instances (e.g. Zr-2.5%Nb) show improved corrosion resistance in-reactor.

4 type alloys in PWRs arises from several feed-back effects operating with increasing time and radiation dose. The degradation of the corrosion resistance gets progressively worse with increasing fast-neutron fluence; the thermal conductivity of the oxide film may also degrade with increasing oxide thickness (although this is far from being proved); and a further degradation in corrosion resistance can result from hydride accumulation at the oxide/metal interface if hydrogen absorption percentages are high. All these feed-back effects result in increasing temperatures at the oxide/metal interface, especially when heat ratings in the fuel are high. It is this temperature, combined with the degraded corrosion resistance of the metal that is oxidising at the oxide/metal interface that results in the characteristic shape of the in-reactor corrosion curves. Improvements have generally been achieved (Figure 6.1), either by reducing the amount of tin oxide in the oxide film (Low Sn alloys) or by eliminating degradable Fe intermetallics (Zr/Nb alloys).



- *) According to the data submitted by A. Seibold, E Ortied to the conference on structural material for PWR, Karlsrue, Germany.
- **) Corrosion of fuel cladding under VVER conditions (experimental reactor, surface boiling, oxygen concentration ≤ 0.02 ppm).

Figure 6.1 Comparison of in-reactor corrosion of Zr alloys in PWRs

• The hydrogen uptake rate is determined by a competition between the recombination and evolution of hydrogen atoms from the cathodic part of the corrosion reaction with the entry of hydrogen atoms at weak points in the oxide. These weak points are thought to be locations where flaws in the oxide approach the oxide/metal interface most closely (i.e. sites of easy electronic transport from the metal to the environment). In Zr-2.Nb alloys low hydrogen uptake has been correlated with more porous oxide films (i.e. enhanced ease of hydrogen escape into the environment).

During each yearly review the improved understanding of the oxide film structure in addition to reports of actual in-reactor behaviour are reported.

6.2 PROGRESS IN 2002

This has been a year without a major conference on zirconium alloys or on fuel cladding behaviour. The proceedings of the 2001 ASTM "Zirconium in the Nuclear Industry" meeting have yet to appear, although the corrosion papers were discussed in ZIRAT – 6 (based on authors' drafts). We have yet to see whether any significant points arose in written discussions, or as a result of authors modifying their papers. The proceedings of the 2001 TMS Symposium on "Defect Properties and Mechanical Behavior of HCP Metal and Alloys" held in New Orleans, LA, Feb. 11-15th [TMS (2002)] have appeared. Although much basic physical metallurgy relevant to mechanical properties and delayed hydride cracking was presented nothing of direct relevance to the corrosion process (e.g. defect mechanisms for iron redistribution) was reported.

A noticeable new trend in the general literature relevant to the properties of zirconia has been observed. In addition to numerous studies of bulk (usually stabilised) zirconia, a big upsurge in interest in the electrical properties of very thin oxide films is being evinced in the semiconductor field, with the aim of using very thin zirconia films in MOSFETs. Although a whole range of fabrication techniques (other than oxidation) is being reported, some information relevant to (e.g.) "Shadow Corrosion" may be found.

This year's report will cover the results from the general literature first, and attempt to relate them to in-reactor behaviour.

7 WATER CHEMISTRY IMPACT (GUNNAR WIKMARK)

7.1 INTRODUCTION

Last year we reported on the Axial Offset Anomaly, AOA, problem in PWRs and could conclude that a large number of PWR cores in the US, approximately 30 cores, had been affected, and almost none outside of the USA, Despite this, the details of the basic phenomena are not known, such as the composition and amount of crud formed in the AOA plants, the chemical conditions establishing the crud formation, the amount of subcooled boiling required, etc. Several directions for trying out remedies were, however suggested, and a number of them had also being started to be applied last year.

The impact of water chemistry on accelerated corrosion was described for three cases last year. On case was the PWR Palo Verde-2, which had recorded AOA and obviously crud induced local corrosion, leading to several fuel failures. Several cases were reported were apparent oxide flaking and elevated Eddy Current lift-offs were recorded in plants that had fuel in an NMCA environment for at least one cycle. At that stage, it was stated that all known cases where any fuel having operated three 18 months cycles and the last had been after NMCA, such enhanced corrosion had been noted. In addition, another BWR had experienced enhanced corrosion, but no fuel failures, apparently due to a CILC (crud induced localised corrosion) phenomenon, assumedly driven by the copper input.

7.2 *PWRS*

7.2.1 General Water Chemistry Trends

The options considered for EDF regarding their PWR operation was recently reviewed by Nordmann, 2002. The main point in the paper was that all changes, such as increased pH, modified hydrogen level, increased lithium, etc., are regarded as more and more interconnected. It is hence not any more possible to optimise the water chemistry with only one goal in mind, e.g. lowering the PWSCC (primary water stress corrosion cracking), without also considering the impact on activity levels, cladding corrosion, waste treatment, etc.

There is an obvious trend that water chemistry modifications in the European PWRs are becoming more and more common (Staudt, 2002) as shown in Figure 7-1, just as has been the case for BWRs for a longer time.

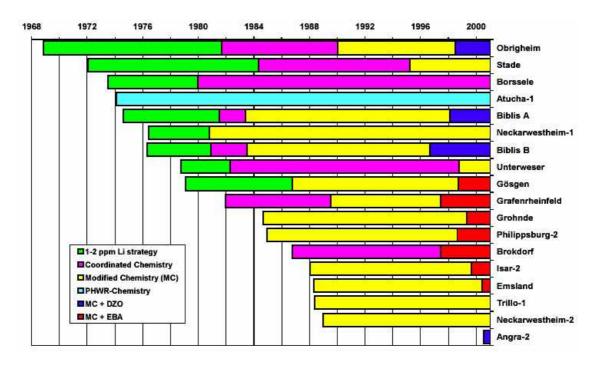


Figure 7-1. Water chemistry regimes in KWU plants. MC – modified chemistry (non-constant pH during cycle); DZ – depleted zinc addition; EBA – enriched boron addition. Staudt, 2002.

It is interesting to note that the introduction of EBA has not been only to cope with the inherently higher reactivity of the MOX fuel, as has been reported previously (ZIRAT-5, Section 7), but also for economic reasons: the very high burn-ups targeted in the KWU plants implies that larger volumes of the borating system would else be required. Despite the fact that EBA is not at all inexpensive, it has been found that re-building the borating system to allow a larger volume would be even more costly.

7.2.2 AOA and Fuel Crud Removal

As discussed in a previous report (ZIRAT-5, Section 7, 2000), axial offset anomaly, AOA (ZIRAT-6, Section 5, 2001; Wikmark and Cox, 2001)), has been postulated to be mitigated by removing most of the crud on fuel that is reinserted in the core again.

7.2.2.1 Fuel Cleaning

The efficacy of the ultrasonic (US) fuel cleaning method has recently been shown to be very good, with typically more than 85% removal of all deposited crud (Blok, 2002), as shown in the example in Figure 7-2.

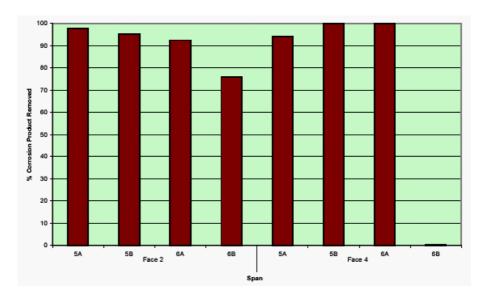


Figure 7-2. The relative amount of corrosion products removed by fuel cleaning with a prototype cleaner for a discharged assembly cleaned in Callaway October 2001. Blok, 2002

The main purpose for the AOA mitigation is of course the removal of the mass of crud, but a benefit in reducing the activity has also been observed in the Callaway fuel cleaning campain, Figure 7-3.

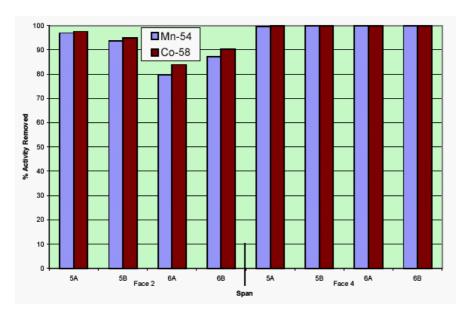


Figure 7-3. The fuel-cleaning efficacy for activity with a prototype cleaner for a discharged assembly cleaned in Callaway October 2001. Blok, 2002

8 ZIRCONIUM ALLOY MANUFACTURING (PETER RUDLING)

8.1 INTRODUCTION

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

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

8.2 NEW RESULTS

8.2.1 Zircaloy

During manufacturing, cold pilgering is a process that may result in cladding defects. Generally, larger degree of cold reduction in the clad radial versus the tangential direction will result in a more radial texture that is beneficial in PCI and hydride orientation respect. More cold reduction in each pilgering step may reduce the number of necessary pilgering steps to bring the clad to its final dimensions. This may save omney for the fuel clad manufacturer. However, to much cold reduction may result in cladding defects. Thus, a knowledge of the deformation processes during the pilgering process may be used to optimise the microstructure in the cladding without introducing defects.

Experimental evaluation of plastic shearing due to cold rolling Zircaloy-4 (Zy4) cladding tubes was performed, Girard, et al., 2001. An experiment design was used to evaluate the influence of three parameters on the shear stress: the feed rate, the frequency of rolling steps, and the type of interal lubricant. Calculation and experimental analysis have shown a good correlation between a cumulative damage factor (CDF) and depth of defects in cold-pilgered tubes. The CDF depends strongly on the shear strain, ε_{rr}^{5} .

⁵ This can be approximated by $\gamma/2$ and is the shear strain in the plane normal to the *r*-axis (radial direction) in the *z*-direction (axial direction), see Figure 8-3.

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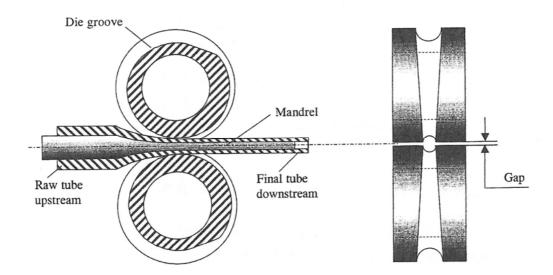


Figure 8-1: Cold pilgering system with: the mandrel (tool inside the tube) and the dies (external tools with semicircular grooves), Girard, et al., 2001.

To observe the strain path in the last rolling pass of Zy4 cladding tubes, Girard, et al., 2001, used a technique pieces of rods were screwed in the thickness of the tube. The holes receiving the inserts were obtained by drilling, Figure 8-2. A Au-Ag-Cu alloy was chosen as the insert material since it has similar plastic deformation behaviour to that of Zry-4 and its contrast is suitable for subsequent analysis. Three Au-Ag-Cu alloy inserts, separated by 10 mm, were placed at 300 mm from the tip of a raw tube. After rolling, the angular postion of two successive inserts varies from a few degrees, Figure 8-3.

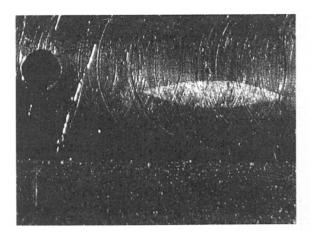


Figure 8-2: Insert with hole drilling photograph, Girard, et al., 2001.

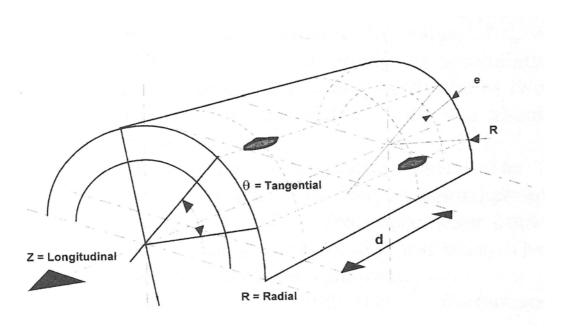


Figure 8-3: Insert position with rolling helix, Girard, et al., 2001.

9 PRIMARY FAILURE AND SECONDARY DEGRADATION (PETER RUDLING)

9.1 INTRODUCTION

9.1.1 Primary Failures

During reactor operation, the fuel rod may fail due to a primary cause such as fretting, PCI, manufacturing defects, corrosion, etc. A summary of failures statistics are shown in Table 9-1 to Table 9-3 and Figure 9-6.

The data indicate that historically the debris fretting failures is a major failure cause both in BWRs and PWRs. Also, corrosion related failures specifically in US has resulted in a large amount of failures in BWRs. In PWRs, grid-rod fretting seems to cause large number of failures.

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 ⁶), Figure 9-1, enhanced spacer shadow corrosion, ESSC, (in BWRs), Figure 9-2, dry-out due to excessive fuel rod bowing. Figure 9-3 shows failure due to excessive PWR corrosion.
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 gap between the fuel rod and the spacer grid supports (poor spacer grid manufacturing process) leading to excessive vibrations in the PWR fuel leading to 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 unirradiated state, Figure 9-4
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, Figure 9-5. This situation may e.g. occur 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

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⁶ Crud Induced Localised Corrosion – an accelerated form of corrosion that have historically resulted in large number of failures in BWRs. Three parameters are involved in this corrosion phenomenon, namely: 1) Large Cu coolant concentrations- as a result of e.g. aluminium brass condenser tubes, 2) Low initial fuel rod surface heat flux – occurs in Gd rods and 3) Fuel cladding that shows large initial corrosion rates- occurs in cladding with low resistance towards nodular corrosion.

⁷ This corrosion phenomenon resulted recently in a few failed rods. The mechanism is not clear but seems to be related to galvanic corrosion. This corrosion type may occur on the fuel cladding in contact or adjacent to a dissimilar material such as Inconel. Thus, this accelerated type of corrosion occurred on the fuel cladding material at spacer locations (the spacer springs in alloy BWR fuel vendors fuel are made of Inconel). Water chemistry seems also play a role if the fuel cladding material microstructure is such that the corrosion performance is poor. Specifically coolant chemistry with low Fe/(Ni-Zn) ratio seems to be aggressive (provided that the cladding material shows poor corrosion performance. A fuel cladding material with good corrosion resistance does not result in ESSC, enhanced spacer shadow corrosion, even in aggressive water chemistry.

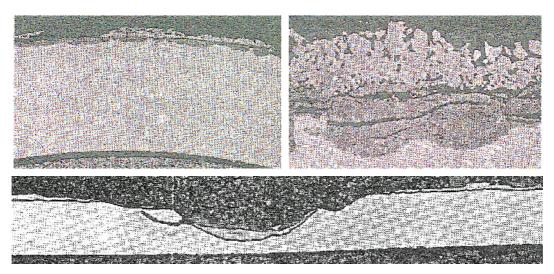


Figure 9-1: CILC corrosion, Marlowe, et al., 1985.

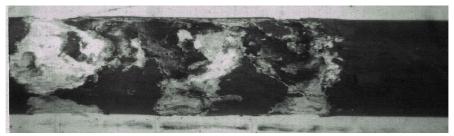


Figure 9-2: 5 cycle fuel in NPP Leibstadt (KKL) showing ESSC, Zwicky, et al., 2000.



Figure 9-3: PWR corrosion failure due to excessive corrosion, Baily, et al., 1991.

10 CLADDING PERFORMANCE UNDER ACCIDENT CONDITIONS (PETER RUDLING)

10.1 INTRODUCTION

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

10.1.1 LOCA

The objectives of the Emergency Core Cooling System, ECCS, LOCA criteria are to maintain core coolability and preserve heat transfer area and coolant flow geometry during the quench phase and post-quench phase of a LOCA. The criteria in most countries are:

- Peak Cladding Temperature, PCT, < 1204°C (or 2200°F)
- Equivalent Cladding Reacted, ECR³⁰, < 17%
- Hydrogen gas produced < 1%³¹.
- Fuel must have coolable geometry³².
- Core temperature maintained at low value for extended time³³.

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 fragmentation during post-LOCA.

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

³⁰ The ECR is defined as the total thickness of cladding that would be converted to stoichiometric ZrO₂ from all the oxygen that are contained in the fuel cladding as ZrO₂, 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), indudes both preaccident oxidation and oxidation occurring during a LOCA.

³¹ Total amount of hydrogen shall not exceed 1 % of the hypothetical amount generated by the reaction of all the zirconium in the fuel cladding with the water/steam.

³² The coolable geometry may be lost by either fuel clad ballooning causing coolant channel blockage or fuel cladding fragmentation due to clad embrittlement.

³³ 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.

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

The Equivalent Cladding Reacted, ECR and Peak Clad Temperature, PCT, criteria were based on retention of clad ductility at 275°F (135°C, the saturation temperature during reflood) according to slow ring compression tests of non-ballooned unirradiated cladding Zircaloy-2 and -4 samples. The selection of the 17% ECR value was specific to the use of the conservative Baker-Just clad oxidation correlation. However, if a bestestimate correlation would have been used instead such as e.g. Cathcart-Pawel correlation, the threshold ECR would have been <17%. Most countries are using this criterion for ensuring adequate cladding ductility. In some countries it is assumed that the largest clad tensile stress³⁴, 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.

³⁴ 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 (Ralph Meyer, Personal communication, 2002.*

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During the late 70's – early 80's, slow ring-compression tests of ballooned and bursted samples showed that the 1973 criteria failed to ensure retention of ductility at 135°C in narrow local regions near the burst opening³⁵, 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 Zircaloy³⁶. The implications of the results, are such that for high-burnup fuel cladding tubes with a H uptake, prior to the LOCA event, exceeding about 700 ppm:

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

Also, the 1204°C peak cladding temperature (PCT) limit was selected on the basis of slow-ring compression tests that were performed at 25-150°C. However, samples oxidized at 1315°C are far more brittle than samples oxidized at 1204°C in spite of comparable level of total oxidation. This is because oxygen solid-solution hardening of the prior-beta phase is excessive at oxygen concentrations >0.7wt%. Consideration of potential for runaway oxidation (*due to that the oxidation process of the Zirconium material becomes to exothermic to be cooled by water*) was a secondary factor in selecting the 1204°C limit. The 1204°C limit was subsequently justified by the observations from impact tests and handling failure of fuel rods exposed to high temperatures in the Power Burst Facility. 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 primarely by the thickness and the mechanical properties of the transformed-beta layer.

During the LOCA event, the cladding is primarily embrittled by the formation of ZrO_2 and oxygen stabilized α Zr(O) as well as by hydrogen pickup during irradiation and high temperature steam oxidation during LOCA. Figure 10-1 is a schematic representation of the oxygen distribution across the cladding, and Figure 10-2 shows the results of an oxidation test along with a metallographic view of the phases. It is the region of the cladding wall that becomes the β -phase and is later transformed to α during the quench that is the principal load-bearing material. The prior β -phase layer passes on the fracture resistance to the cladding and can prevent fragmentation during reflood. Increased thickness and ductility of this phase will consequently reduce the risk of clad fragmentation during lost-LOCA.

³⁶ 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 %..

³⁵ It appers that the stagnant conditions of water/steam in this location will significantly increase the hydrogen pickup during LOCA clad oxidation.

11 FUEL PERFORMANCE CRITERIA DURING INTERMEDIATE STORAGE (AL STRASSER)

11.1 INTRODUCTION

This year's update on intermediate storage of spent fuel will concentrate on dry storage. The issues related to wet storage have not changed since last year's review (ZIRAT-6) and will not be repeated here. The report will concentrate on the fuel related licensing issues, relevant data and analyses and fuel performance in dry casks. A brief update on storage sites and cask designs is given initially.

The wet storage pool capacities at the plant sites continue to fill up and the total filled will approach 50% of all the US pools in 2003, Figure 11-1. The lack of a licensed permanent government storage site keeps slipping into the future in spite of the congressional and senate approvals of Yucca Mountain. The European and Asiatic countries are no better off as reprocessing becomes more expensive than storage and licensed permanent repository sites are lacking universally. As a result there is a worldwide, strong, increasing trend toward dry storage of the discharged fuel.

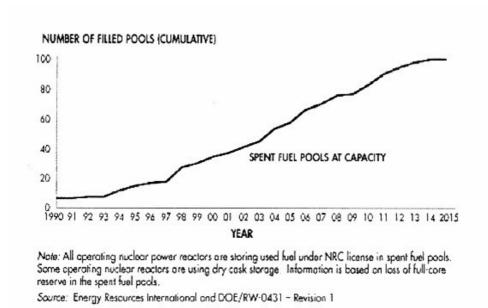


Figure 11-1: Spent fuel storage pool capacity in the U.S., Gruss, 2002.

A total of about 17,000 spent fuel assemblies (42,540 tons of fuel) are stored in the US of which 97% are At Reactor (AR) sites and 3% are Away From Reactor (AFR) sites, such as General Electric, Morris, IL.

Less than 10% of the total inventory are stored in dry casks and the remainder in spent fuel pools (SFP).

The currently licensed US dry storage facilities, also called Independent Spent Fuel Storage Installations (ISFSI), are up to a total of 22 and are shown on Figure 11-2. Additional 21 sites that are likely to be licensed in the near term are shown on Figure 11-3.

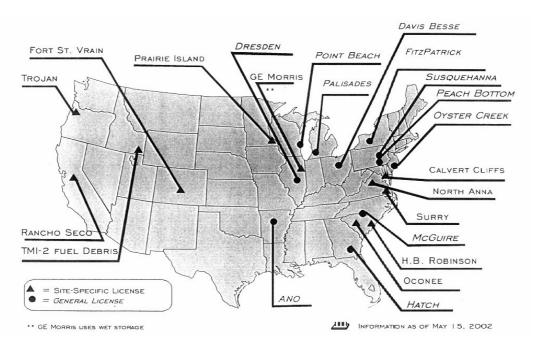


Figure 11-2: Operating ISFSIs in the U.S., Gruss, 2002.

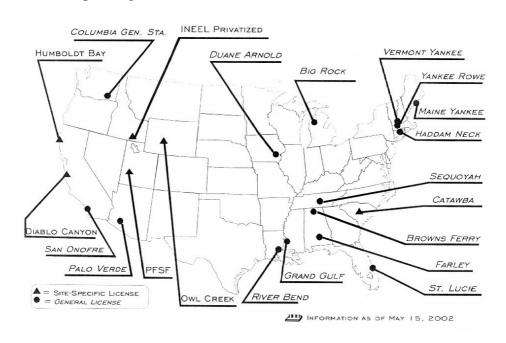


Figure 11-3: Potential near-term, new US ISFSIs, Gruss, 2002.

The most advanced intermediate AFR dry storage site in the US is at Skull Valley, Utah on the reservation of the Skull Valley Band of Goshute Indians. The site is sponsored by a consortium of utility companies that has applied for a license to build the facility. The licensing process is about 80% complete. The NRC released its SER in September, 2000 with the conclusion that the facility would be safe and meet regulatory requirements. Supplements on aircraft/cruise missile crashes were issued late in 2001, after Sept. 11, and deemed "not credible" based on a one in a million probability of occurring. A second set of hearing s by the Atomic Safety and Licensing Board (ASLB) was completed in July, 2002 and is (at this writing) evaluating the testimony. If their decision is positive, the NRC could issue a license in December 2002. The next step would be initiation of the design phase and a marketing campaign to obtain commitments from the utilities.

Assuming all goes well and work is initiated at the beginning of 2003, the site could receive fuel at the earliest in 2005. The license would permit a total of 4,000 casks and construction would be phased 1,000 at a time.

The outrageous, prohibitive laws passed by the State of Utah to prevent development of the Skull Valley site were declared unconstitutional by the District Court of Utah and the case is now going to the Federal Court of Appeals at the request of the State.

Numerous licensed, active dry storage sites exist in Europe and Asia and include Belgium, Bulgaria, Czech Republic, Germany, Hungary, Italy, Netherlands Rumania, Russia, Slovakia, Spain, Switzerland, Ukraine and Japan. The location of both dry and wet storage sites as well as the sites for reprocessing in Europe are shown on Figure 11-4.

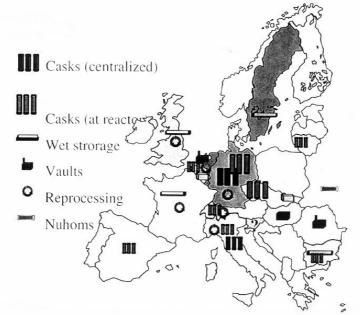


Figure 11-4: Interim storage sites in Europe, Hunter, 2002.

12 POTENTIAL BURNUP LIMITATIONS

12.1 INTRODUCTION

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

- Corrosion of Zry-4 in high power PWRs, extended by improved cladding alloys.
 However, 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. This modified operation will in most cases result in heavier corrosion
 duty of the zirconium materials. Thus, it is believed that the corrosion will always
 be limiting for plant operation even with the new type of alloys.
- Bowing of PWR fuel assemblies contributed in part by irradiation growth and hydriding of Zry-4, extended by improved guide tube materials (i.e., lower irradiation growth and hydriding rates), and reduced assembly holddown forces, but not yet finally eliminated. It is questionable, whether design changes to make the fuel assembly stiffer will resolve the issue. It may be that, these design changes will only increase the incubation time before bowing occurs and at that point it will be even more difficult to e.g. insert control rods.
- 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, in the process of being assessed by
 additional experimental data and analyses. Based upon the on-going test program,
 it is not clear today if the licensing limits will be extended or not. The limits may
 also actually be reduced.

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

BWRs: increased uniform and shadow corrosion, oxide thickness spalling.--- due
to longer residence time, higher power and water chemistry changes. Current
crucial issues are: shadow corrosion mechanisms, CRUD-chemistry-corrosion
interaction and specific effects of NMCA.

- PWRs: increased uniform corrosion, oxide thickness, spalling --- due to longer residence time and higher Li, higher power, more boiling. The introduction of Zr-Nb alloys may also result in accelerated corrosion at the welds, e.g., between the end-plugs and cladding tubes. Also, welding between dissimilar metals such as e.g. ZIRLO and Zry-4 may result in chemical compositions of the welds that show inferior corrosion resistance. Luckily, the corrosion temperatures at these elevations in the core is significantly lower than the peak temperatures and this may be the reason that no corrosion issues have been reported so far in the welds.
- Increased H pickup tendency⁵⁷:
 - decreased ductility and fracture toughness during any situation (e.g., RIA, PCMI,..)
 - increased corrosion due to impact of hydrides at the cladding outer surface

Most sensitive component

Spacer and fuel claddings

Increase margin for PWR

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

⁵⁷ due to the introduction of more corrosion resistant materials. For some types of Zr alloys, the hydrogen pickup fraction increases with decreasing corrosion rate.

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12.3 DIMENSIONAL STABILITY

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

Most sensitive component

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

Increase margin for PWR

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

Increase margin for BWR

- Uniform microstructure and texture throughout the whole fuel outer channel
- Beta-quenched material in as-fabricated step
- Channel management programs
- More corrosion resistant material in channels and spacers is needed to allow irradiation for longer cycles

12.1 PCI IN BWRS

• Increased sensitivity for PCI by alloy diffusion from Zry-2 base material into the liner. However, this may not be a concern with the current Fe-alloyed liner.

Most sensitive component

Fuel cladding

<u>Increase margin for BWR</u>

• Ramp testing of high burnup liner fuel

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