# Annual Report

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## Contents

1	Introduction (Peter Rudling)					
2	Burnup a	Burnup achievements and fuel performance issues (Alfred Strasser)				
	2.1	Trends in fuel operating conditions	2-1			
	2.1.1	General trends	2-1			
	2.1.2	Fuel cycle lengths	2-1			
	2.1.3	Burnup extension	2-2			
	2.1.4	Power uprates	2-4			
	2.1.5	Water chemistry	2-5			
	2.1.5.1	High lithium operation	2-5			
	2.1.5.2	Zinc injection	2-6			
	2.1.5.3	NMCA	2-8			
	2.2	High burnup fuel performance summary	2-8			
	2.2.1	High burnups achieved in utility power plants	2-8			
	2.2.2	High burnup fuel examination results	2-9			
	2.2.2.1	Actinide and fission product distribution in oxide fuels	2-9			
	2.2.2.2	HBS in UO <sub>2</sub>	2-14			
	2.2.2.3	Non-Destructive Testing (NDT) of FGR	2-15			
	2.2.2.4	FGR from MOX fuel	2-16			
	2.2.2.5	Additives to fuel pellets	2-16			
	2.2.2.6	Burnable absorbers	2-18			
	2.2.3	High burnup examination of zirconium alloys	2-19			
	2.2.3.1	Dimensional stability	2-19			
	2.2.3.2	Effect of hydrogen on alloy properties and performance	2-23			
	2.2.3.3	High burnup FR irradiations	2-29			
	2.2.4	Intermediate dry cask storage	2-32			
	2.3	Fuel reliability	2-32			
	2.3.1	Introduction	2-32			
	2.3.2	PWRs	2-37			
	2.3.3	BWRs	2-38			
	2.4	LOCA issues	2-40			
	2.4.1	Proposed United States Nuclear Regulatory Commission (USNRC) rule change	2-40			
	2.4.2	Pellet relocation during ballooning	2-47			
	2.4.3	Effect of LOCA on gas release from high burnup fuel	2-47			
	2.5	RIA tests and analyses	2-49			
	2.5.1	USNRC related activities	2-49			
	2.5.2	New NSRR test results	2-51			
	2.5.3	MOX RIA test analyses	2-55			
	2.5.4	Non-US regulatory positions	2-57			
	2.5.5	Cladding ductility at high burnup	2-59			
	2.6	Utility concerns regarding performance and regulations	2-60			
	2.7	Summary	2-60			
3	Zirconiun	n alloy manufacturing and alloy system	3-1			
4	Mechanic	al properties	4-1			
5	Dimension	nal stability (Ron Adamson)	5-1			
	5.1	Introduction	5-1			
	5.2	Summary given in ZIRAT12, 2007	5-11			

	5.3 5.3.1 5.3.2 5.3.3 5.3.3.1 5.3.3.2	New reports from the recent literature Creep and growth FR length Bow Channels FA bow	5-13 5-13 5-23 5-32 5-32 5-32 5-39
	5.4	Summary	5-40
6	Out-of-pile	and in-pile corrosion	6-1
	6.1 6.2 6.2.1 6.2.1.1 6.2.1.2 6.2.2 6.2.2.1 6.2.2.2 6.2.2.3 6.2.2.4 6.2.2.5 6.2.3	Out-of-pile corrosion In-pile corrosion & hydriding (Friedrich Garzarolli) Corrosion in PWRs and VVERs PWR FR corrosion process and its relation to the method of analysis New reported information Corrosion in BWRs Optimization history of Zry-2 State of knowledge on the acceleration of corrosion and hydrogen pick up of Zry-2 at high burnups Possible reasons of the acceleration of hydrogen pick up at high burnups and possibilities to optimize of Zry-2 for high burnup application Possibilities to optimize of Zry-2 for high burnups New reported information Summary	6-1 6-3 6-3 6-14 6-22 6-22 6-23 6-23 6-26 6-29 6-31 6-45
7	Primary fail 7.1 7.1.1 7.1.2 7.2 7.2.1 7.2.1.1 7.2.1.1.1 7.2.1.1.2 7.2.1.1.3 7.2.1.1.4 7.2.1.2 7.2.1.2.1 7.2.1.2.1 7.2.1.2.2 7.2.1.2.4 7.2.1.2.4.1 7.2.1.2.5 7.2.1.2.6 7.2.2 7.2.2.1 7.2.2.2 7.2.3 7.3	ure and secondary degradation – open literature data (Peter Rudling) Introduction Primary failures Secondary degradation Results presented in year 2008-2009 Primary fuel failures PWR and BWR general Westinghouse data EdF data (France) AREVA NP Data GNF Data Primary failure causes Manufacturing related failures GTRF Debris fretting PCI and PCMI Modelling FA bowing CRUD Degradation of failed fuel VVER fuel BWR Methods to assess fuel performance during operation Summary and highlights-year 2008-09	7-1 7-1 7-5 7-5 7-5 7-5 7-6 7-8 7-9 7-9 7-9 7-9 7-9 7-9 7-9 7-9 7-9 7-20 7-27 7-34 7-35 7-42 7-51 7-56 7-57
8	LOCA/RIA		8-1
9	-	nance during intermediate storage (Charles Patterson)	9-1
	9.1 9.2	Introduction Dry storage conditions relevant to fuel performance issues	9-1 9-5

	<b>9.3</b> 9.3.1	Status of fuel related regulatory requirements in the USA Introduction	<b>9-9</b> 9-9			
	9.3.2	General	9-9			
	9.3.3	Classification of fuel condition (ISG-1, Rev. 2)	9-13			
	9.3.4	Fuel retrievability and recoverability (ISG-2, ISG-3)	9-15			
	9.3.5	Heat transfer during and after transportation and storage accidents (ISG-7)	9-16			
	9.3.6	Burnup credit (ISG-8, Rev. 2)	9-16			
	9.3.7	Storage of components associated with FAs (ISG-9, Rev. 1)	9-18			
	9.3.8	Cladding considerations for the transportation and storage of spent fuel (ISG-11, Rev. 3)	9-18			
	9.3.9	Fuel integrity under cask drop and tipover conditions (ISG-12, rev. 1)	9-19			
	9.3.10	Subcriticality requirements (ISG-19, Rev.0)	9-19			
	9.3.11	Potential rod splitting due to exposure to an oxidizing atmosphere during short-term cask loading operations in LWR or other uranium oxide based fuel (ISG-22, Rev. 0)	9-19			
	9.4	Status of international fuel related regulatory requirements	9-19 9-20			
	9.4 9.4.1	Introduction	9-20 9-20			
	9.4.2	Comparison of international criteria	9-20			
	9.5	Thermal and mechanical issues affecting fuel in dry storage	9-20 9-23			
	9.5.1	Decay heat and cladding temperatures	9-23			
	9.5.2	Effects of stress levels and hydride re-orientation and cracking	9-32			
	9.5.3	DHC theories relevant to dry storage	9-43			
	9.5.4	Effect of creep	9-47			
	9.5.5	Mechanical modelling	9-49			
	9.5.6	Defective FR behaviour	9-54			
	9.6	Criticality considerations	9-59			
	9.6.1	Burnup credit	9-59			
	9.6.2	Source term definition	9-60			
	9.6.3	Effect of accidents on FRs	9-60			
	9.6.4	Effect of fire accidents	9-62			
	9.7	Cask design features	9-64			
	9.7.1	Introduction	9-64			
	9.7.2	Cask thermal capacities	9-65			
	9.7.3	New cask design features	9-67			
	9.7.3.1	TAD canister system	9-67			
	9.7.3.2	HOLTEC international	9-68			
	9.7.3.3	Transnuclear (AREVA)	9-70			
	9.7.3.4	NAC international	9-72			
	9.7.3.5	Mitsubishi Heavy Industries (MHI)	9-73			
	9.8	Conclusions	9-74			
10	Potential burnup limitations (Peter Rudling) 10					
	10.1	Introduction	10-1			
	10.2	Corrosion and mechanical properties related to oxide thickness and H pickup	10-1			
	10.3	Dimensional stability	10-3			
	10.4	PCI in BWRs and PWRs	10-4			
	10.5	LOCA	10-5			
	10.6	RIA	10-5			
	10.7	5% enrichment limits in fabrication plants, transport and reactor sites	10-5			
	10.8	Dry storage	10-6			
11	References		11-1			
Acron	yms and exp	ressions	11-1			
Unit c	conversion		11-5			

# 1 Introduction (Peter Rudling)

The objective of the Annual Review of ZIRconium Alloy Technology (ZIRAT) and Information on Zirconium Alloys (IZNA) is to review and evaluate the latest developments in ZIRAT 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 ZIRAT14/IZNA9 Program, the following technical meetings were covered:

- CQCNF-2009 conference at Hyderabad, India, Feb 18-20, 2009.
- International Atomic Energy Agency (IAEA) 13<sup>th</sup> International Topical Meeting on Research Reactor Fuel Management (RRFM), Vienna, Austria, 22 -25 March 2009.
- Reduction of Dose Rate in Pressurised Water Reactors (PWRs) Workshop, Würzburg, Germany, April 6-9, 2009.
- Jahrestagung Kerntechnik, Dresden, Germany, May 12-14, 2009.
- Nuclear Energy Institute (NEI) Dry Storage, FL, May 11-1.
- US Utility Conference, USA, summer 2009.
- SMIRT20, ESPOO, Finland, August 9-14, 2009.
- 14<sup>th</sup> International Conference on Environmental Degradation of Materials in Nuclear Power Systems-Water Reactors, Virginia Beach, Virginia, USA, August 23 27, 2009.
- Top Fuel, Paris, September 6-11, 2009.
- The 8<sup>th</sup> International Conference on Voda Voda Energo Reactor (Russian type PWR) (VVER) Fuel, September 26 to October 3, 2009, Bulgaria.
- VGB<sup>1</sup> Chemistry in Power Plants Conference, Dresden, Germany, October 28-29, 2009.

The extensive, continuous flow of journal publications is being monitored by several literature searches of worldwide 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.

<sup>&</sup>lt;sup>1</sup> See www.vgb.org.

The primary issues addressed in the review and this report is 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 (QC) 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 (Loss of Coolant Accident (LOCA), Reactivity Initiated Accident (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. The data published in this Report is only from non-proprietary sources; however, their compilation, evaluations, and conclusions in the report are proprietary to ANT International and ZIRAT/IZNA members as noted on the title page.

The authors of the report are Dr. Ron Adamson, Mr. Al Strasser, President of Aquarius Mr. Friedrich Garzarolli, Dr. Charles Patterson and, Mr. Peter Rudling, President of ANT International.

The work reported herein will be presented in three Seminars: one in Westin Hilton Head Island, SC, on February 1- February 3, 2010 one in Bilbao, Spain on March 16-18, 2010 and one in Japan in 2010.

The Term of ZIRAT14/IZNA9 started on February 1, 2009 and ends on March 31, 2010.

# 2 Burnup achievements and fuel performance issues (Alfred Strasser)

# 2.1 Trends in fuel operating conditions

# 2.1.1 General trends

The incentive for the continued improvements in operating economics and reliable fuel performance has resulted in significant advances in materials technology, software for modelling fuel performance and sophisticated instrumentation and methods for Post-Irradiation Examinations (PIE). These advances have been used to increase the demands on fuel performance levels and to put pressure on the regulatory bodies to license operations to increased burnup levels. The types of changes in Light Water Reactor (LWR) operating methods to achieve improved safety and economics have not changed in the past year and still include:

- Annual fuel cycles extended to 18 and 24 months.
- Increased discharge burnups from mid-30 to 58 GWD/MT batch average exposures by higher enrichments, increased number of burnable absorbers in the assemblies and in PWRs higher Li and B levels in the coolant.
- Plant power uprates that range from 5 to 20%.
- More aggressive fuel management methods with increased enrichment levels and peaking factors.
- Reduced activity transport by Zn injection into the coolant.
- Improved water chemistry controls and increased monitoring.
- Component life extension with Hydrogen Water Chemistry (HWC) and Noble Metal Chemistry (NMC) in Boiling Water Reactor (BWRs).

# 2.1.2 Fuel cycle lengths

The trend for increased fuel cycle lengths has come to a near "equilibrium" in the US with PWRs operating at an average of 500 Effective Full Power Days (EFFPD) per cycle and BWRs an average of 620 EFFPD per cycle, up to about 650 days for PWRs and 700 days for BWRs. 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 potential economic gains for cycle extension have decreased in the US since the downtimes for reloading and maintenance procedures have been significantly reduced.

Other countries that historically have had only one peak power demand per year in the winter compared to the two summer and winter power peaks in the US are also trending toward longer cycles as a result of changes in economics, maintenance practices and licensing procedures. PWRs are trending toward 18 month cycles in France, the UK (Sizewell B) and Germany. Japan is planning to increase cycle lengths in several steps, first to 15 months, which does not require relicensing, then to 18 and 24 months, which will require re-licensing.

### 2.1.3 Burnup extension

The major incentive for extended burnups is the potentially improved fuel cycle economy, although other incentives might exist. The improved economics depend in part on the decreased amount of spent Fuel Assemblies (FAs) to be purchased and handled. This is balanced by the increased amount of uranium and enrichment services required The economics of decreased assemblies could be impacted by the much longer cooling times required in spent fuel pools prior to on-site dry storage or transport to a storage facility as noted later.

The average batch burnups in US PWRs are generally in the range of 45-54 GWD/MT and in US BWRs in the range of 45-50 GWD/MT.

Some European plants operated in the 50-58 GWD/MT batch burnup range and have designed to go to 62 GWD/MT in their current cycles in both PWRs and BWRs. This is feasible, in part, due to their greater margin to regulatory burnup limits. The maximum burnup Lead Use Assemblies (LUAs) are in the range of 67 – 79 GWD/MT for both reactor types.

The trends in assembly average burnup extension in French plants are shown in (Figure 2-1). The low values for Mixed Oxide (MOX) fuel and the 1500MW<sub>e</sub> plant UO<sub>2</sub> fuels are expected to increase as the equilibrium of the new core management methods will be reached, [Thibault et al 2009].

The VVER-440 fuel is up to 57 GWD/MT assembly burnup with plans to go to 65 GWD/MT. The VVER-1000 fuel is up to 55 GWD/MT assembly burnup with plans to go to 68 GWD/MT, [Molchanov et al 2007]. Rod burnups of 73 GWD/MT burnup have been achieved in both VVER-440 and VVER-1000 fuel, [Markov et al 2008].

The Japanese are planning to raise current BWR 45 GWD/MT batch average burnup to 50 GWD/MT without exceeding the 55 GWD/MT peak assembly burnup regulatory limit for 9x9 "Step 2" fuel, [Itagaki & Murata 2007]. This will be done in conjunction with cycle extension from their current 13 month to future 16 or 19 month cycles. The PWR assembly burnups will also be raised to 48 and 55 GWD/MT for all rod arrays. The highest burnup reached by a PWR assembly has been 47 GWD/MT.

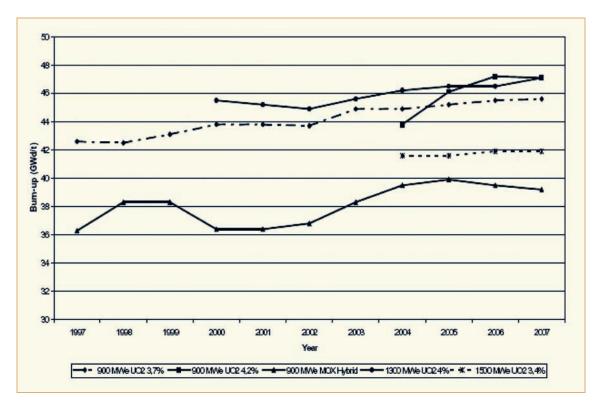


Figure 2-1: Assembly average discharge burnup trends in France, [Thibault et al 2009].

The burnups are currently limited by the regulatory agencies more than by technical limitations, except for LUAs and rods. Some of the current regulatory limits are summarized in (Table 2-1).

Table 2-1:	Dogulatory	ournun lim	ite in	
Table Z-T.	Regulatory b	Julliup IIIII	112 111	GVVD/IVIT.

USA	62.5 peak rod	Korea	60 peak rod
Belgium	55 UO <sub>2</sub> assembly	Netherlands	60 peak rod
	50 MOX assembly	Sweden	60 assembly
Finland	45 assembly		64 rod
France	52 assembly	Switzerland	75 peak pellet
Germany	65 assembly	Taiwan	60 peak rod (PWR)
Japan	55 UO <sub>2</sub> assembly		54 peak assembly (BWR)
	(66 peak rod)	UK	55 peak pellet
	45 MOX assembly		

Economic analyses reported in past ZIRAT reports indicated that economic incentives for extending burnups beyond the 60-70 GWD/MT batch average range will disappear and that other incentives will be needed to go beyond this level. The biggest block to increasing burnup is the essentially universal regulatory limit on 5.00% initial uranium enrichment. The cost of equipping and licensing the industry to handle enrichments greater than 5% would be very great and time consuming; and for that reason such a move is not being considered for the near future.

### 2.1.4 Power uprates

The power of the majority of the operating plants has been uprated, the maximum being 20%. While the fuel performance limits remain the same for the uprated conditions, the number of FAs operating at higher power change and the margin to the limits might be reduced. The effect of higher flow rates on hydraulic and water chemistry effects and their interaction with the fuel must also be considered. These changes have not affected the failure rate or apparently the fuel performance; however, it does increase the statistical probability of increasing power and burnup related factors on the fuel performance.

The power uprates approved in the US since 2007 are:

BWRs:		PWRs:			
Hope Creek	15.0%	Millstone 3	7.0%		
Susquehanna 1 and 2	13.0% each	Comanche Peak 1 and 2	4.5% each		
Browns Ferry 1	5.0%	Vogtle 1 and 2	1.7% each		
Cooper	1.6%	North Anna	1.7% each		
Crystal River	1.6%				
Davis Besse	1.6%				
Calvert Cliffs 1 and 2	1.4% each				

Power uprate applications under review are:

BWRs:		PWRs:	
Browns Ferry 1, 2 and 3	15.0%	Point Beach 1 and 2	17.0% each
Monticello	12.9%		

The total of about 165% amounts to approximately another high power unit since 2007. The effect of plant uprating and capacity factor improvements to an average of 91% since 1973 have increased the generation capacity by an equivalent of 27 plants as shown in (Figure 2-2), [Schutz 2009].

Power uprating is of course practiced outside the US as well. As an example, Electricité de France (EdF) is considering increasing the power of their 1300 MW<sub>e</sub> plants by 7.8% to be completed by 2017.

The effect of power uprating on fuel power distribution was discussed in ZIRAT12 and the effect on water chemistry was discussed in the ANT International Special Topical Report "Consequences of Power Uprating" to the LWR Coolant Chemistry (LCC3) Seminar.

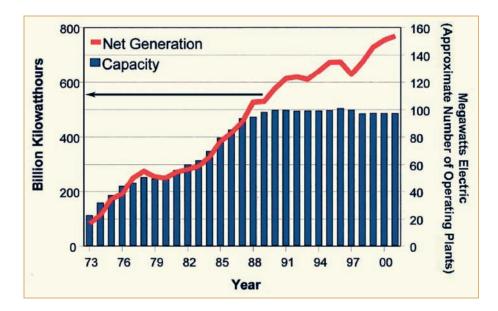


Figure 2-2: US nuclear power industry efficiency improvement, [Schutz 2009].

An example of an analysis of the effect of uprating on HWC and its potential effectiveness to prevent InterGranular Stress Corrosion Cracking (IGSCC) of core components was reported in ZIRAT13. The analysis made by the National Tsing Hua University, Taiwan [Wang et al 2007]) and published again recently [Yeh & Wang 2009] concluded that the increase in flow rate associated with a power uprate reduced hydrogen content and ECP in several core locations. This has been disputed by General Electric (GE) investigators by stating that they have not seen the drop in Electrochemical Corrosion Potential (ECP) at the hydrogen levels analyzed by Yeh in the reactors where they have applied HWC. While the actual measurements by Global Nuclear Fuel (GNF) are closer to reality than modelling, we would still recommend modelling as well as actual testing by the vendor in order to evaluate the effect of power uprating on the HWC effectiveness at various hydrogen levels and in various parts of the core.

### 2.1.5 Water chemistry

### 2.1.5.1 High lithium operation

Increasing burnup, cycle lengths and enrichments levels, combined with the recommended high, 7.4 Ph operation has increased the required lithium (Li) levels above the previously used maximum 3.5 ppm concentrations. The potentially increased zirconium alloy corrosion levels due to increased Li levels has created a cautious approach to increasing the Li levels; the concentration of Li in high Chalk River Unidentified Deposits (CRUD) levels and/or in combination with nucleate boiling is of particular concern. Operation with 5 to 6 ppm Li at the Beginning of Cycle (BOC) has become standard at Comanche Peak 1 and 2 as well as Ringhals (RH) 2 and 3 based on demonstration programs that indicated no measurable damage to the fuel. Details of these programs were described in previous ZIRAT reports.

More recently, EdF qualified an increase from their more conservative standard 2 ppm Li to the 3.5 ppm Li level by monitoring a plant at the higher level for several years, [Thibault et al 2009]. The 3.5 ppm Li at constant pH of 7.2 is being applied to all of their 1500 MW<sub>e</sub> units.

### 2.1.5.2 Zinc injection

One of the successful innovations in operating methods has been zinc (Zn) injection into the coolant to reduce activity transport within the primary coolant systems of both PWRs and BWRs. A total of 25 PWRs are injecting Zn in the US. EdF has tried out Zn injection successfully in Bugey 2 and 4 and is considering applying it to their high activity plants.

Activity transport has been reduced by the range of 20-35% without significantly affecting fuel cladding corrosion, be it Zircaloy-2, -4, ZIRconium Low Oxidation (ZIRLO) or M5. The presence of Zn, however, promotes the deposit of a Zn containing spinel on the cladding probably in the form of NixZnyFezO4, especially if sufficient Fe is present. While these CRUD deposits have not been observed to affect corrosion of the cladding, they have been suspected of promoting boron deposits that can result in Axial Offset Anomalies (AOAs) also called CRUD Induced Power Shifts (CIPSs) in plants with subcooled nucleate boiling. A key question has therefore been the effect of Zn injection on cores with nucleate boiling.

All of the US BWRs are operating with Zn injection as shown on (Figure 2-3), with a successful reduction in activity transport. Tenacious CRUD can form on BWR cladding as it has formed on PWR cladding; however, the BWR CRUD has a somewhat different composition, because the impurities in the coolant are different from those in the PWR. The tenacious CRUD layers have been thicker in BWRs and have also been the cause of failures. The CRUD analysis from the River Bend failure, a BWR6, was 71-84% Fe, 7-14% Zn, 2-8% Cu and 2-6% Si representing compounds of hematite (Fe<sub>2</sub>O<sub>3</sub>), zinc ferrite (ZnFe<sub>2</sub>O<sub>4</sub>), zinc silicate (Zn<sub>2</sub>SiO<sub>4</sub>) and traces of ZnO. While Fe was distributed uniformly in the CRUD layer, Zn and Cu were concentrated on the inner and outer surfaces of the CRUD layer that was 100µ thick. The brass condenser was the source of the Zn and Cu, [Cheng et al 2009].

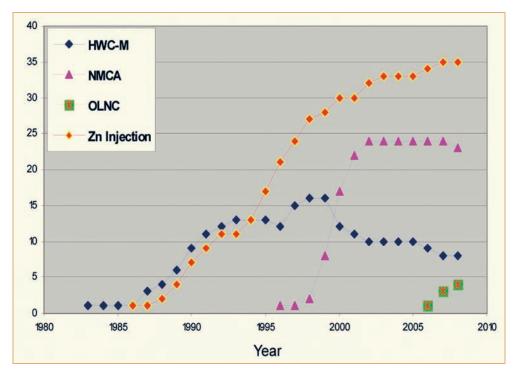


Figure 2-3: Trends of US BWRs operating under moderate HWC, Noble Metal Chemistry Addition (NMCA), On Line Noble Chem (OLNC), and zinc injection, [Cheng et al 2009].

The tenacious CRUD formed by Zn injection in a BWR 4 varied as a function of exposure in the range of 20-30  $\mu$  thickness. After 1 cycle, the CRUD consisted of about 2  $\mu$  zinc ferrite crystals and after two cycles the space between these crystals was infiltrated by zinc silicate. The porosity of such a layer can decrease to <10% by zinc silicate infiltration and increase its density (Figure 2-4). The Feed Water (FW) Zn content is recommended to be at a level <0.4 ppb to minimize such CRUD formation that could develop into a thermal barrier.

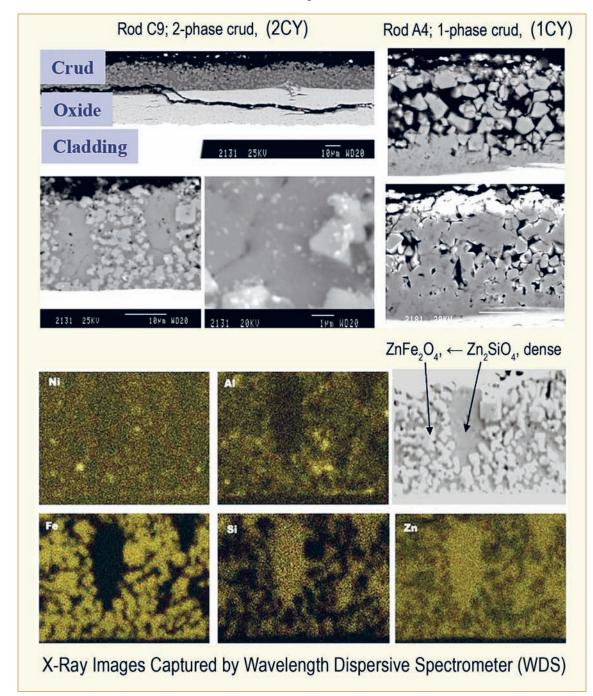


Figure 2-4: Tenacious CRUD taken on a Fuel Rod (FR) retrieved from a BWR5, [Cheng et al 2009].

# 2.1.5.3 NMCA

The reduction of the BWR Stainless Steel (SS) structural components' sensitivity to IGSCC has been accomplished by the application of HWC, the addition of up to 1.8 ppm hydrogen to the coolant, which in turn lowers the ECP by the recombination of the oxidants. Crack initiation and propagation will not occur at the target level of  $< 230V_{SHE}$ . The efficiency of the recombination process was improved by the addition of the noble metals platinum (Pt) and sometimes rhodium (Rh) that catalyze the recombination at significantly lower hydrogen levels (0.15-0.30 ppm), resulting in a potentially more effective function within the cracks and a reduction in the Main Steam Line Radiation Dose (MSLRD) due to <sup>16</sup> N promoted by the higher hydrogen contents. This NMCA is a process patented by GE.

Initially the Pt additions, in solution form, have been added batchwise at the End of Cycle (EOCs), and more recently an On-Line NMCA (OLNC) process has been initiated that injects the noble metal periodically during the operating cycle at full power. Usually the application is made on an annual basis. Several OLNC applications have been made at KernKraftwerk Mühleberg (KKM) in Switzerland starting in 2005 and the first application in the US was at a BWR2 in 2006 and at Peach Bottom 3 (BWR4) in 2007. Subsequently OLNC has been applied at Perry, Nine Mile Point 1 and 2. It is expected that an increasing number of US plants will adopt OLNC. In Europe OLNC has been applied at KernKraftwerk Leibstadt (KKL). All proceeded according to plan.

Post-OLNC fuel examinations have not been made in the US.

An alternative to OLNC, application of noble metals prior to fuel loading at 78° to 93 °C, has been tried by Exelon. This Low Temperature Noble Chemistry (LTNC) process has been tried successfully at La Salle-2, Quad Cities 1 and 2.

# 2.2 High burnup fuel performance summary

### 2.2.1 High burnups achieved in utility power plants

The *highest burnup levels in PWRs* have been implemented in the US, Germany, Russia and Switzerland. The batch averages range between 47 and 58 GWD/MT with plans to go to 60-62 GWD/MT in Germany. The peak assemblies range between 48 and 72 GWD/MT and the peak rods between 53 and 106 GWD/MT burnup.

The batch average burnup levels in the US plants have reached their maximum level permitted by the 62 GWD/MT max. rod exposure established by the Nuclear Regulatory Commission (NRC), until more data become available to justify increased regulatory burnup limits. Peak assembly average burnups in both the US PWRs and BWRs are in the range of 58 to 68 GWD/MT. The values above this level are in German, Russian and Swiss plants.

MOX assemblies have reached peak assembly average burnups of 62 GWD/MT in PWRs and 58 GWD/MT in BWRs in European plants.

The current examinations of US rods from LTAs above the 62 GWD/MT limit are intended to justify the increase of the current 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. This situation has not changed in the past years.

The Lead Test Assemblies (LTAs) are usually exempt from regulatory limits and their highest peak rod burnups achieved in the past years have been in GWD/MT: 72 (US, Russia), 80 (France), 84 (Japan), 90 (Switzerland).

The peak burnups in GWD/MT achieved by the current PWR cladding materials have been:

	Batch	Assembly	Rod
ZIRLO, standard	55	58	75
ZIRLO, optimized		70	>70
M5	54	68	80
E-110		62	73
Duplex (DX), various	58	62	68
PCA2b	49	55	58
M-MDA <sup>2</sup>		68	73

The highest burnups in BWRs are in the US, Germany, Spain and Switzerland. The batch averages range between 43 and 57 GWD/MT, peak assemblies between 51 and 68 GWD/MT and the peak rods between 55 and 73 GWD/MT. The burnup levels in BWRs are catching up to those of the PWRs in the US, probably because both reactor types are reaching the current NRC burnup limitation. Irradiation results of a variety of BWR vendor fuel designs in KKGundremmingen and KKL to extended burnups are 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), 75 (Germany).

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

Zircaloy 2 (GWD/MT)	:	49 batch, 71 assembly, 78 rod.
LK <sup>3</sup> 3 (GWD/MT)	:	47 batch, 53 assembly, 81 rod.

The assembly and rod burnups listed above were achieved without failures; however, FAs and materials do not have a perfect performance record up to these burnups levels and the related problems are summarized in Section 2.4 and discussed in detail in subsequent sections.

### 2.2.2 High burnup fuel examination results

### 2.2.2.1 Actinide and fission product distribution in oxide fuels

Knowledge of the quantification and distribution of the actinides and Fission Products (FP) at high burnup in the fuel oxides is important for understanding and modelling fuel performance during normal and accident conditions and storage of spent fuel. The data obtained on this topic affects the knowledge of the temperature distribution in the fuel, Fission Gas Release (FGR), swelling, stresses on the cladding during operation, criticality and burnup credit during fuel handling and storage, and decay heat during storage.

<sup>&</sup>lt;sup>2</sup> Mitsubishi Developed Alloy

<sup>&</sup>lt;sup>3</sup> Låg Korrosion (Low Corrosion in Swedish)

A detailed study of *actinide and fission product distribution* was made by the examination of a slice of UO<sub>2</sub> that reached 65 GWD/MT in a FR irradiated to an average 90 GWD/MT in a Siemens fabricated15x15 PWR assembly, [Walker et al 2009]. The burnup at the centre of the pellet was 57-62 GWD/MT and reached 140 GWD/MT at the pellet surface. The 3.5% enriched fuel operated at 500 to 600 °C except for up to 1000 °C in the first of 8 annual cycles at a power that decreased from 34 to 14 kW/m. The examination of actinide and FP was made by Secondary Ion Mass Spectroscopy (SIMS). SIMS has the advantage of a low detection limit of 0.001 ppm compared to 200 ppm for Electron Probe Micro Analysis (EPMA) and can detect fission gases in pores. It has the disadvantage that it cannot detect certain isotopes due to mass interference.

The distribution of the plutonium (Pu) isotopes is shown on (Figure 2-5) and shows the rapid rise toward the pellet surface, where the total Pu content reaches about 3.1%. The average abundance of the various Pu isotopes was:

239**D** 

1 ( 20/

					Radius, r/r <sub>o</sub>		
	0	)	0.2	0.4	0.6	0.8	1.0
	0.4x10 <sup>5</sup>	242Pu		 <del>\$\$ \$\$\$\$</del> MB MMM	<mark>∞∞∞∞∞∞∞∞</mark> <del>0999000000000000000000000000000000000</del>	NAN REPORT	
	Ion Intensity. c/s 0'8x10 <sub>2</sub>	<sup>239</sup> Pu	108-0 <mark>80</mark>	<del>co on</del> o¤	240		
	చ్ 0.8x10 <sup>5</sup>					<sup>241</sup> Pu + <sup>241</sup> Am	
	1.2x10 <sup>5</sup>		· · ·			<u> </u>	
Pu	15.7%						
Pu	8.7%						
Pu	29.4%						
Pu	46.2%						

Figure 2-5: Radial distribution of the major *Pu* isotopes in the fuel matrix, [Walker et al 2009].

It is important to remember that the odd isotopes are the fissionable ones and for handling, the even numbered isotopes are neutron emitters.

The other actinides analyzed americium and curium, <sup>243</sup>Am, <sup>244</sup>Cm, and <sup>245</sup>Cm had distribution shapes similar to the Pu curves. An exception was neptunium, <sup>237</sup>Np, which had a flat distribution due to the fact that it is produced from <sup>235</sup>U by capture of thermal neutrons and the other actinides are produced from <sup>238</sup>U by capture of epithermal neutrons concentrated in the outer fuel region.

Both the Pu redistribution curve and the flat Np curves were reproduced in MOX fuel by very short, High Temperature (HT) irradiations at about 1800 and 2000 °C by Japanese investigators using EPM, [Maeda et al 2009].

The stable element neodymium (Nd) is used for burnup determinations and the distribution of its isotopes is shown in (Figure 2-6). The isotope <sup>148</sup>Nd, which is generally used for determining burnup, is not shown because of interference from samarium isotopes; however, its shape will follow that of the other Nd isotopes.

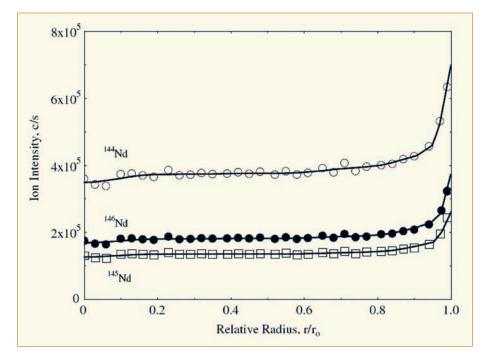


Figure 2-6: Radial distribution of the stable *Nd* isotopes <sup>144</sup>Nd, <sup>145</sup>Nd AND <sup>146</sup>Nd in the fuel matrix.

Studies of the fission product distribution concentrated on the fission gases and high vapour pressure FP. A SIMS scan of the High Burnup Structure (HBS) shows the concentration of the fission gases xenon and krypton, <sup>132</sup>Xe and <sup>84</sup>Kr, with the peaks representing their presence in gas bubbles (Figure 2-7). The calculated pressures indicated very few points above 25 MPa or maxima reaching 230 MPa, the maximum that the authors claim is sustainable by the fuel. The average pressure of 45 MPa was stated by the authors to be over-pressurized, but not excessively so. The mean pore size was 1.5 µ.

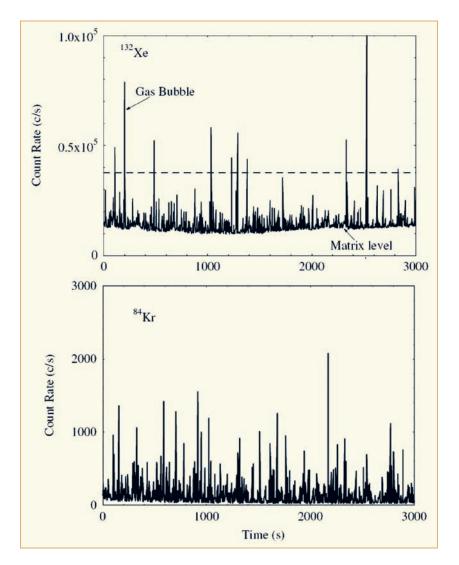


Figure 2-7: Depth profiles for <sup>132</sup>Xe and <sup>84</sup>Kr in the high burn-up structure. (The intensity spikes mark the location of gas pores in the microstructure. The dashed line on the Xe depth profile indicates the ion intensity that is equal to a gas pressure of 25 MPa in the pores. Time corresponds to erosion depth), [Walker et al 2009].

A comparison of the EPMA concentration profile that only measured the Xe retained in the matrix only to the SIMS measurements of the matrix, the pores and bubbles showed that almost all the gas missing from the fuel matrix is contained in the HBS structure (Figure 2-8), [Walker et al 2009]. The EPMA studies were published earlier and included the examination of samples from the peak burnup section of the rod at 95 GWD/MT that operated between 1650 and 1150 °C during the first two cycles and between 850 and 950 °C the remaining period, [Manzel & Walker 2002]. This HT portion of the rod showed the same pattern of the majority of gas release from the central portion of the rod, probably due to grain Recrystallisation, rather than from the HBS. They also made the important observation of about a 3X increase in FGR from the rods as they increased the rod average burnup from 50 to about 100 GWD/MT.

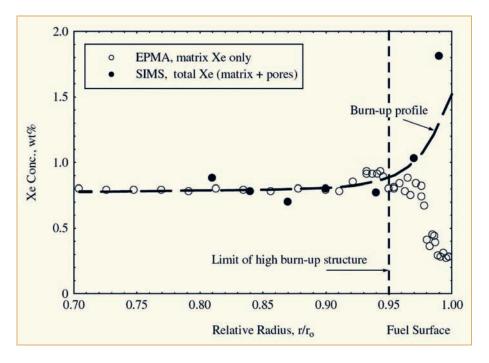


Figure 2-8: The local concentration of retained Xe in the outer region of the fuel as measured by SIMS and EPMA. The burnup profile determined from the local concentration of Nd measured by EPMA is included for the purpose of comparison, [Walker et al 2009].

These studies confirm the conclusion on the distribution of FGR made by the Paul Scherrer Institute (PSI) and reported in ZIRAT12.

The validation of the *computer models for calculating the isotopic inventory* of high burnup UO<sub>2</sub> and MOX fuel using the lattice code CASMO-4E depletion module were made against actual measurements made by the Paul Scherrer Institute (PSI). The burnup ranges were up to 121 GWD/MT for UO<sub>2</sub> and up to 71 GWD/MT for MOX, [Türk et al 2009]. The quantities and distributions of <sup>235</sup>U, <sup>239</sup>Pu, <sup>244</sup>Cm and <sup>148</sup>Nd were predicted well within ±5% with the exception of <sup>235</sup>U at very high burnups when the quantity of that isotope is very small. Special evaluations were made of the neutron producing isotopes since these are of significance for interim and permanent storage of spent fuel. The largest neutron sources due to spontaneous fission are <sup>242</sup>Cm, <sup>244</sup>Cm and <sup>252</sup>Cf and due to ( $\alpha$ ,n) reactions they are additionally <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, and <sup>241</sup> Am. The results for the high burnup samples are shown on (Figure 2-9). The largest neutron source for the first 100 years is <sup>244</sup>Cm. After about 3000 years the neutron sources have decreased significantly. The authors recommend more accurate calculations for the interim periods.

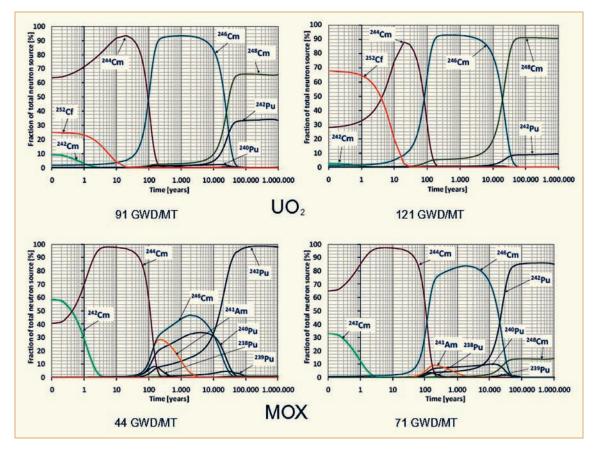


Figure 2-9: Neutron source intensity with time after discharge, [Türk et al 2009].

### $2.2.2.2 \qquad \text{HBS in UO}_2$

The HBS at the pellet periphery is of interest since it may affect fuel performance, particularly under accident conditions, by a rupture of the overpressurized fission gas pores, cracking of the fuel and sudden release of fission gases. An extensive evaluation of 210 HBS data points that have been published were made by Korean investigators to develop a correlation of the HBS width with burnups up to 100 GWD/MT, [Koo et al 2009]. The data were from a variety of PWRs, BWRs and Heavy Water Reactors (HWRs) on fuel of varying fabrication methods, enrichments and power histories. Width measurement methods included EPMA, Scanning Electron Microscope (SEM) and Optical microscopy (OP). The data were related to pellet average rather than rim local burnup.

The conclusion of the study divided the HBS widths into three burnup zones (Figure 2-10). The onset of HBS formation was noted at 30 GWD/MT pellet average burnup while other references cite 40 GWD/MT. Local burnups, however, above which original as-fabricated grains begin to subdivided to form HBS, lie in a wide range of 50 to 80 GWD/MT. The differences in the local burnups for the onset of HBS can be due to differences in operating conditions such as power histories as well as methods of defining and measuring the HBS. The HBS width in the 1<sup>st</sup> region, 30 to 60 GWD/MT is determined by the accumulated, burnup related, fission damage in combination with a sufficiently low temperature to prevent thermal annealing. The HBS width is reduced slightly in the 2<sup>nd</sup> region, 60 to 75 GWD/MT local burnup, due to temperatures that result in partial annealing of microstructure damage. Above 75 GWD/MT, the 3<sup>rd</sup> region, the HBS width reaches a plateau of about 1.5 mm because temperatures are reached that induce extensive thermal annealing combined with high FGR from the central portion of the pellet, which limits the formation of the HBS region.

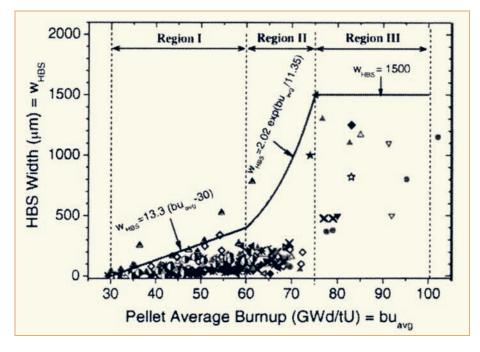


Figure 2-10: A conservative HBS width in LWR UO<sub>2</sub> fuel pellet as a function of pellet average burnup in the three regions, [Koo et al 2009].

An evaluation of the effect of enrichment and GS on the HBS width found them to be insignificant compared to the effects of burnup and temperature.

### 2.2.2.3 Non-Destructive Testing (NDT) of FGR

The NDT of FGR by the gamma scanning of the FR plenum for <sup>85</sup>Kr (10.75 year half life) has been frustrated for many years, because of the interference of the <sup>58</sup>Co peak (70.9 day half life) in the measurement which requires a long decay time before measurements can be made. The <sup>58</sup>Co comes from the <sup>58</sup>Ni source in the SS or nickel alloy plenum spring. (This author has recommended carbon steel or high chromium steel springs to avoid this problem).

An improvement in the separation of these peaks and that of ruthenium and rhodium (<sup>106</sup>Ru/Rh 372 day half life) was reported by the measurement of 22 SVEA96S Optima/L design rods. The rods had been irradiated in Teollisuuden Voima Oyj (TVO') Unit 2 in 2 assemblies to rod average burnups of 43 to 61 GWD/MT. The LADAKH and SEDAS software was used to calculate the total FGR based on these measurements and compared to the FGR calculated by the Westinghouse STAV code.

The results included some good correlations to the STAV code, some under and some overpredictions. Unfortunately there were no comparisons to actual FGR measurements made by puncturing and analyzing the gases. The NDT and STAV results were all in the FGR rate range of 1% to 8%, which is reasonable for the high duty fuel operating between 350 w/cm to150 w/cm to these high burnups. An outlier was analyzed to have about 10% gas release, but this was attributed to the effects of channel bowing up to 10 mm, which seems somewhat high for a SVEA channel.

The FR internal gas pressure was calculated to be within the Finnish regulatory limit of 7 MPa except for the rod affected by channel bowing.

### 2.2.2.4 FGR from MOX fuel

FGR from MOX fuel has been noted over the past years and generally attributed to high Pu content agglomerates in inhomogeneous fuel. A detailed examination of this has been made now by Commissariat à l'Energie Atomique (CEA), based on the fact that the fission gas isotopic composition from the high Pu agglomerates is different from the uranium matrix, [Lemoin & Guerin 2009]. The study was based in the heterogeneous MOX fuel made in the MELOX plant by the Micronized Master Blend route (MIMAS) process using the free flowing Ammonium Uranyl Carbonate (AUC) powder. The product has agglomerates that reportedly contain about 27% Pu while the U matrix contains only a small amount or any Pu.

The yield of xenon (Xe) is very similar for the U and Pu isotopes; however, the krypton (Kr) yield is a factor of 2 to 3 higher for U than Pu and varies with the fissile isotopes. The Xe/Kr ratio can then be used for tracing the source of the fission gas: high ratios show the dominance of the U matrix and low ratios the dominance and source from the agglomerates. The isotopic composition of the fissile species changes during the irradiation of the MOX fuel. In the agglomerates <sup>239</sup>Pu is dominant in the early part of the irradiation, then decreases and <sup>241</sup>Pu increases; this evolution is slower at higher Pu contents. In the U matrix Pu is created by neutron capture of <sup>238</sup>U and the major part of the fissions occur in <sup>239</sup>Pu as its content increases. The contribution of fissions in <sup>238</sup>U by fast neutrons in the early part of the irradiation, is significant, although much less than that in the Pu agglomerates. The origin of fission gases. The decrease in Pu fissile content in the agglomerates tends to be balanced by their increase in the U matrix decreasing the heterogeneity, the differences in integral composition, however, remain significant.

The data sources for these analyses were 80 PWR MOX FRs with 2.9% to 7.3% Pu, irradiated in the EdF 900 Mw<sub>e</sub> plant to burnups of up to 60 GWD/MT and subsequently punctured and sampled for FGR. The analyses were made by the ever increasingly improved CEA codes Margaret and METEOR. The conclusions reached by the study were:

- At low burnups the released gas is produced preferentially in the agglomerates.
- At high burnups the gas release is closer to a homogeneous release from the pellet, by probably a thermal mechanism.
- The high FGRs measure in MIMAS fuel (up to 10-13%) is mainly due to a high thermal mechanism and that the Pu rich agglomerates play a minor role at steady state conditions.
- A limited contribution of the agglomerates cannot be excluded by pellet cracks or other mechanisms that allow gas release from the pores of the restructured agglomerates.
- Under transient conditions, especially rapid transients, the contribution from the agglomerates can be significant.

The similarity in FGR between MOX and  $UO_2$  FRs at high burnup was re-enforced by the examination of a MOX rod with 5.8% Pu, irradiated at Halden in IFA-565 to 56 GWD/MT compared to BWR  $UO_2$  rods, [Ozawa 2009].

### 2.2.2.5 Additives to fuel pellets

Pellets with  $Cr_2O_3$  additives have reached commercial application by AREVA and Westinghouse. The advantages of this type of fuel are:

- Reduced FGR based on increased GS.
- Higher density and improved mechanical integrity due to improved sinterability.
- Improved Pellet Cladding Interaction (PCI) resistance due to more plastic mechanical properties.

The  $Cr_2O_3$  additive is designed to stay under the 0.16% solubility limit in  $UO_2$  in order to prevent second phase formation. Irradiations in power reactors reached 60 GWD/MT by both vendors' fuel as of last year and are being exposed up to 70 GWD/MT. The details were reported in ZIRAT12 and 13. This year AREVA reported the initiation of testing MOX fuels with the additive, [Arslan & Castelli 2009]. A total of 8 rods are being fabricated for insertion in 2 PWR assemblies. The microstructure of MOX is compared with and without additive in (Figure 2-11).

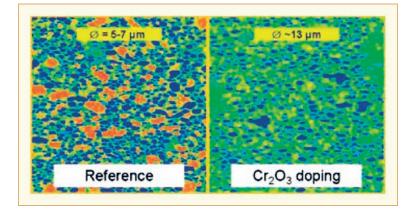


Figure 2-11: Effect of chromium additive on MOX microstructure, [Arslan & Castelli 2009].

GNF has been developing *pellets with*  $Al_2O_3$ - $SiO_2$  *additives* since 1980 with LUA irradiation tests in BWRs in the US, Italy, and more recently in Forsmark 1 (Sweden), Fukushima 1 (Japan) and Gundremmingen C (Germany), [Lingenfelter et al 2009]. Reloads have been initiated in Japan. The objective of the additives is to increase the creep properties, or soften the pellets at HTs and reduce release of certain FP to increase the margins to PCI. The mechanism, which accomplishes this performance improvement is by coating the UO<sub>2</sub> grain boundaries with about 2500 ppm of the additive as shown in (Figure 2-12). The GNF FGR model has overpredicted gas release by several percent (at an unstated burnup) and the authors report that no "adverse" swelling or rod growth behaviour has been observed.

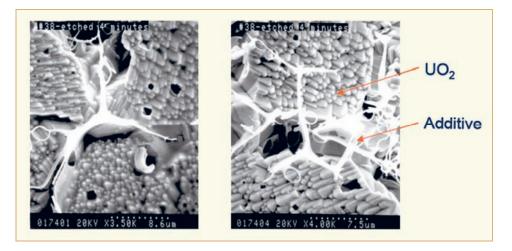


Figure 2-12: SEM images showing the morphology of grain boundary phases present in GNF additive fuel, [Lingenfelter et al 2009].

Segmented rods irradiated in 10x10 assemblies in Forsmark (burnup not stated) have been ramp tested at Halden to 17 kw/ft and some exceeding 20 kw/ft without failures. High burnup exposures in four Gundremmingen-C LUAs, currently at 80 GWD/MT, are expected to reach 98 GWD/MT in 2011. Meanwhile (U,Gd)O<sub>2</sub> fuel is being provided in reload quantities in Japan.

### 2.2.2.6 Burnable absorbers

Measurements of Westinghouse 17x17 PWR FRs in Korea indicated that the  $UO_2$ - $Gd_2O_3$  containing FRs grew more than the  $UO_2$  rods, [Yoo et al 2009]. After 3 cycles of irradiation (no burnups given) the Gd rods grew 0.57% compared to 0.42% for the  $UO_2$  rods. The actual increase of lengths of the Gd rods over the  $UO_2$  rods was 5 mm (0.19 in.) after the 1<sup>st</sup> cycle, 7 mm (0.26 in.) after the 2<sup>nd</sup> cycle and 8 mm (0.31 in.) after the 3<sup>rd</sup> cycle.

Hot cell measurements also indicated that the cladding creepdown (cladding type not identified) was greater over the  $UO_2$  enriched axial blanket section than the Gd section, and also that the Gd containing fuel column grew significantly more than the axial blanket column. The densification of the Gd pellets was measured to be less than those of the  $UO_2$  pellets and the subsequent swelling and contact with the clad assumed to occur sooner. The authors concluded that the differential growth was due to the large cladding creepdown over the axial blanket combined with the "large" or at least earlier swelling and extension of the Gd fuel stack that resulted in axial creep in the early cycles of irradiation. The differential decreases in the last cycle as the axial blanket swelling takes over the effects of densification.

Just when we thought we know everything there is to know about *gadolinium (Gd) cross sections*, some new data emerge. Recent calculations of UO<sub>2</sub>-Gd<sub>2</sub>O<sub>3</sub> absorber fuel pins in a SVEA-96, Optima 2 assembly did not match measured values, [Perret et al 2009]. The calculations of fission rates using MCNPX with the JEFF-3.1 cross sections library underestimated and calculation of the ratio of  $^{238}$ U capture (C<sub>8</sub>) to total axial and radial fission (F<sub>tot</sub>) overestimated the measured values in the Gd pins. After significant efforts to explain the discrepancies, they were resolved by the use of recently measured Gd cross sections at the Rensselear Polytechnic Institute (RPI), which differed significantly from current library values, [Leinweber et al 2006]. A new set of resonance parameters were analyzed up to 300 eV and based on these the new parameters and cross sections were merged into the ENDF/B-VII.0 DATA set; values above this energy level remained unchanged. The resonance ranges of seven Gd isotopes, numbers 152, 154, 155, 156, 157, 158 and 160 were modifies to account for the new measurements. The new capture cross section of <sup>157</sup>Gd is compared to the older one over the applicable energy range in (Figure 2-13).

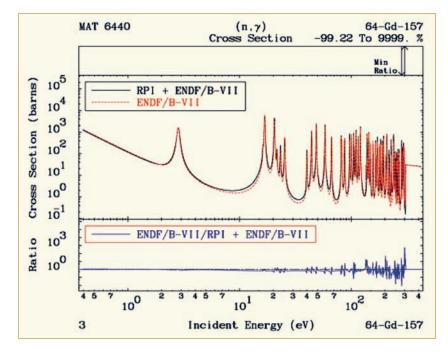


Figure 2-13: Gadolinium-157 neutron capture cross section from 0.4 to 400 eV derived from ENDF/B-VII.0 or JEFF-3.1 (dashed) and from ENDF/B-VII.0 with the resonances measured by RPI (solid), [Perret et al 2009].

# 4 Mechanical properties

There were limited number of results related to mechanical properties published in the past twelve months. This topic will be covered in greater depth in the ZIRAT15/IZNA10 AR.

# 5 Dimensional stability (Ron Adamson)

# 5.1 Introduction

One of the most unique aspects of material behaviour in a nuclear power plant is the effect of radiation (mainly neutrons) on the dimensional stability (i.e., a change in dimensions during service) of the reactor components. Three phenomena may cause zirconium alloy fuel components to change their dimensions:

- Stress-free axial growth due to fast neutron irradiation.
- Hydriding of the Zr-alloy component.
- Creep in Zr-alloy components that are stressed.

Creep is defined as a time dependent change in dimension of a reactor component (or any material) under a stress, even if that stress is below the yield stress. The most important applications of creep are in in-reactor performance of fuel bundle components in BWRs/PWRs, Vodo-Vodyanoi Energetichesky Reactor (in English Water-Water Energetic Reactor) (VVER) and, CANDU and Reaktor Bolshoi Mozhnosti Kanalov (in English Large Boiling Water Channel type reactor) (RBMK) pressure tubes.

Outward creep of the pressure tubes may limit the lifetime of the CANDU reactor.

Inward creep of cladding early in life and outward creep later in life are very important to FR performance. A burnup limiting, fuel design, safety criterion in many countries requires the fuel clad outward creep rate to be no larger than the fuel pellet swelling rate. This requirement addresses the condition in which the rod internal pressure exceeds the primary system pressure due to a large release of fission gas and thereby induces outward cladding creep. If the rate of cladding creep exceeds the rate of pellet swelling, the pellet/clad gap will re-open; i.e., the cladding will *liftoff* from the fuel pellets, (Figure 5-1). Due to the low thermal conductivity of larger gaps, such liftoff can produce a thermal feedback effect that is defined as an "unanalyzed condition" in licensing space and could potentially lead to fuel failures. Thus, a fuel cladding with higher creep strength will result in larger margins towards *liftoff*.

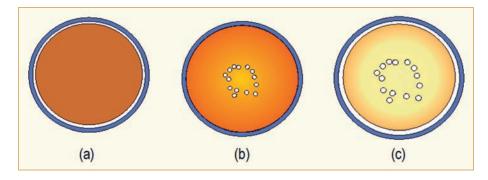


Figure 5-1: Schematics showing how the pellet-cladding gap may change over burnup, (a) low burnup-a significant pelletcladding gap exist, (b) intermediate burnup – no pellet-cladding gap and (c) high burnup in a high power rod with significant FGR – reopening of the pellet-cladding gap.

Also FRs, PWR/VVER Guide Tubes (GT), GT assemblies, and BWR channels bow by a combination of creep and growth. Specifically, creep strength of the PWR GTs is to a large extent involved in the FA bowing mechanisms, (Figure 5-2). If we compare two similar hypothetical FA designs (thus, the FA stiffness is the same) with the same GT compressive forces but with GTs with different creep strength both designs would bow to the same degree in the reactor. However, in the case of the low creep strength GT material the FA distortion would remain after the FA is removed from the core. On the other hand, the FA with GTs with high creep strength would become straight again after off –loading the assembly from the core (since the elastic distortion of the assembly in-core would fully relax after off-loading) BWR channels also creep (bulge) outward due to a pressure differential between the core flow region inside the channel and the core by-pass region outside the channel, (Figure 5-1). These bowing and bulging processes can limit bundle burnup by inhibiting control rod movement or by decreasing thermal margins in LOCA or dryout/DNB situations. Excessive FA bowing in PWRs has also resulted in grid damage during shuffling of FAs in refuelling outages. Also unloading of channels through the core grid can be complicated due to excessive bulging of BWR fuel channels.

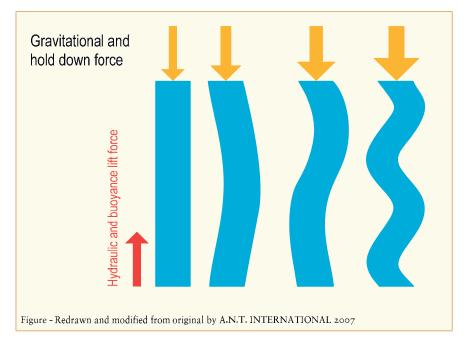


Figure 5-2: Schematics showing FA bowing. Increased GT growth my result in larger holding down forces (figures going from left to right). The first mode of bowing is C-shape (the second drawing from the left), while the second and third mode of bowing is S-shape and W-shape, respectively.

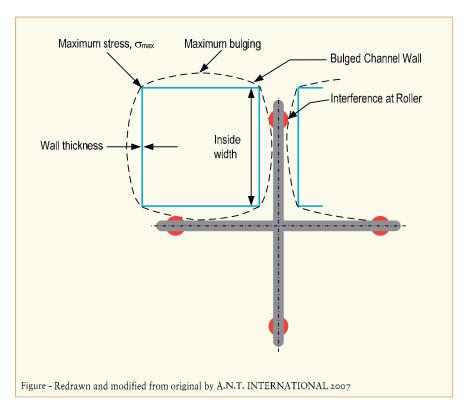


Figure 5-3: Channel CB interference, modified figure according to [Gorman & Lipsey 1982].

The above discussion leads to the concept that understanding the mechanisms of creep 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, which is one of the objectives of this STR. Another objective is to get a better understanding of how creep impacts dimensional instability of FA components and thereby how larger margins towards liftoff, FA bowing and fuel outer channel bulging may be obtained by fuel design changes.

An idealized creep curve (strain versus time) is given in (Figure 5-4). High burnup implies long times, but for in-reactor service the third stage (tertiary or unstable) creep is rarely reached. Generally the secondary (steady state) creep stage is approached. An exception is for the case of missing fuel pellets in the rod, which promotes local creep down that can get into the tertiary creep stage, [Franklin & Adamson 1988].

# 6 Out-of-pile and in-pile corrosion

# 6.1 Out-of-pile corrosion

There were limited number of results related to out-of-pile corrosion published in the past twelve months. This topic will be covered in greater depth in the ZIRAT15/IZNA10 AR.

# 6.2 In-pile corrosion & hydriding (Friedrich Garzarolli)

The entire area of corrosion (and the accompanying absorption of hydrogen in the zirconium metal matrix) is of prime interest when considering performance of the core components and therefore the performance of the entire reactor. For instance, for PWRs a practical corrosion limit exists (about 100 µm oxide thickness, which is associated with a critical amount of hydrogen absorption). The growth of oxide films on Zr-alloy FR Claddings and Structural components has been treated extensively in the literature.

The most important parameters affecting zirconium alloy cladding corrosion are:

- Temperature.
- Exposure time.
- Water chemistry, including impurities and CRUD deposition.
- Neutron flux.
- Zr-alloy chemistry and condition.
- Formation of a dense hydride layer.

Due to the different FR surface temperatures and water chemistry in the different types of power reactors, such as PWR, VVER, BWR, RBMK, CANDU, corrosion of the Zr-alloy claddings and structural components proceeds quite differently in the different reactor systems.

Out of reactor in oxygen free water the corrosion forms at first a dense protective oxide film. The rate of corrosion decreases with increasing oxide layer thickness, S ( $S^3$ =k't, where t is time and k a constant). At a certain thickness ( $S_{Trans}$ =1.5-3 µm) the oxide layer becomes porous causing a rate transition and the further corrosion exhibits a more or less constant rate governed by the thickness and quality of the innermost part of the oxide layer, often called *barrier layer*. The buildup rate of the uniform oxide layer (corrosion rate, *ds/dt*) before and after the rate transition shows an exponential temperature dependency:

(Eq. 6-1) 
$$ds/dt = C \exp(-Q/RT),$$

where

C is the corrosion constant that depends on Zr-alloy composition and condition.

Q the activation temperature.

R the gas constant.

T the absolute temperature.

The temperature effect on corrosion rate is related to the increased transport rate of the corrosion species responsible for the oxide growth (electrons and/or oxygen ions) with increased temperature.

Q/R is reported to be between 11000 and 16000 K and appears to quite independent on Zr-alloy composition and condition. Probably the best Q/R value for Zry-4 corrosion in water is 12880 K, as deduced from long-time experiments at 270-360 °C, [Hillner et al 2000].

The general model for the out-of-reactor corrosion in oxygen free water is widely accepted, although it was pointed out, e.g. [Bryner 1979], that the quasi-linear corrosion is in reality a periodic corrosion behaviour, repeating the first stage of oxide growth and transition (see Figure 6-1) at least at short to intermediate exposure times. The repeated transitions can be seen in metallographic examinations by lateral cracks in the oxide layer. Considering the repeated transition occurrences, [Pêcheur et al 2004] and [Bonieau et al 2007] proposed to described the post transition model by a cyclic corrosion with repeated transitions.

Certainly, for the description of the out of reactor corrosion mechanism major discrepancies exist on used activation temperatures (Q/R) and corrosion constants. Some of the discrepancies are related:

- To the data base (the activation temperatures and corrosion constants obtained from a e.g. 360 °C water test and a 400 °C steam test are different since the material response in the two different environments is different and others.
- To the environmental condition (oxygen and hydrogen content) or,
- to the tested material (the alloying and impurity content as well as the material condition affect the corrosion resistance).

Furthermore, sometimes a late acceleration in corrosion rate can be seen, e.g. [Peters 1984]: A consideration of the late acceleration makes the (Eq. 6-1) more complex. However, most Zr alloy cladding materials do not show such a late acceleration. [Garzarolli et al 1989b] have reported such a late acceleration of the post transition corrosion rate only for Zry-type alloys with a rather small Second Phase Particle (SPP) size.

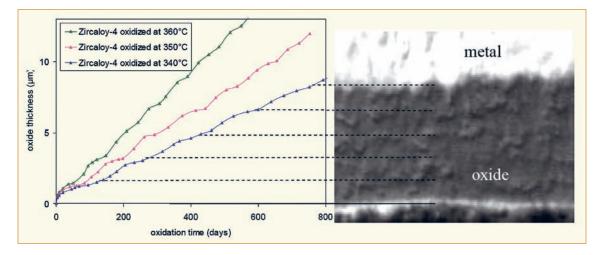


Figure 6-1: Corrosion of Zircaloy-4 in autoclave at 340-360 °C, [Bonieau et al 2007].

# 6.2.1 Corrosion in PWRs and VVERs

# 6.2.1.1 PWR FR corrosion process and its relation to the method of analysis

In this section it will be pointed out that any detailed description of the PWR corrosion process is very much correlated to the model applied to describe the important in-PWR/VVER effects, which are:

- a) The effect of heat flux, which depends largely on the thermal conductivity of the oxide layer and on the thickness and quality of the CRUD layer.
- b) The effect of neutron irradiation.
- c) The potential effect of the Li and B additions to the coolant.
- d) The effect of a potentially formed hydride rim at the outer colder surface.

There are several models for all these parameters that differ not only in the constants but also in the basic concept. In the following the major differences in considering the effect of heat flux and of neutron irradiation and the consequence on the in-PWR corrosion process are reviewed.

#### The effect of heat flux

Cladding tubes are subjected to a heat flux under service conditions. This <u>causes a temperature</u> <u>increase over the oxide layer</u> from the outside surface to the metal-oxide interface. Out-of-reactor experiments with electrically heated samples confirmed that the <u>corrosion rate under heat flux</u> <u>conditions is controlled by the temperature of the protective barrier layer adjacent to the metal</u> <u>surface</u>, [Stehle et al 1975]. This result also demonstrates the rate-controlling function of the inner part of the oxide layer.

The temperature drop through the oxide layer ( $\Delta T$ ) is:

(Eq. 6-2) 
$$\Delta T = S_{ox} \cdot q'' / \lambda,$$

where

 $S_{ox}$  = the oxide layer thickness (cm),

q'' = the heat flux (W/cm<sup>2</sup>) and,

 $\lambda$  = the thermal conductivity (W/cmK).

The increasing oxide/metal interface temperature with increasing oxide thickness (exposure time) and heat flux causes an acceleration of the oxide layer growth rate as shown in (Figure 6-2). This acceleration is often called *thermal feedback* effect.

# 7 Primary failure and secondary degradation – open literature data (Peter Rudling)

The open literature data are provided in the following sections.

# 7.1 Introduction

### 7.1.1 Primary failures

During reactor operation, the FR may fail due to a primary cause such as fretting, PCI manufacturing defects, corrosion, etc., Table 7-1.

Table 7-1:Primary failure causes for LWR fuel during normal operation and Anticipated Operational<br/>Occurrences (AOO).

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 <sup>14</sup> ), Enhanced Spacer Shadow Corrosion, (ESSC), <sup>15</sup> (in BWRs), dry-out due to excessive FR bowing.
Manufacturing defects	<ul> <li>Non-through-wall cracks in the fuel cladding developed during the cladding manufacturing process.</li> <li>Defects in bottom and/or top end plug welds.</li> <li>Primary hydriding due to moisture in fuel pellets and or contamination of clad inner surface by moister or organics.</li> <li>Too large a gap between the FR and the spacer grid supports (poor spacer grid manufacturing process) leading to excessive vibrations in PWR fuel causing fretting failures.</li> <li>Chipped pellets may result in PCI failures both in liner and non-liner fuel.</li> </ul>
PCI	PCI—an iodine assisted SCC phenomenon that may result in fuel failures during rapid power increases in a FR. 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 FR causing fuel failures. This situation may occur for example due to different pressure drops in adjacent FAs causing cross-flow. Baffle jetting failures - Related to unexpectedly high coolant cross-flows close to baffle joints.

<sup>&</sup>lt;sup>14</sup> CILC – an accelerated form of corrosion that has historically resulted in a large number of failures in BWRs. Three parameters are involved in this corrosion phenomenon, namely: 1) Large Cu coolant concentrations as a result of e.g., aluminium brass condenser tubes, 2) Low initial fuel rod surface heat flux – occurs in Gd rods and 3) Fuel cladding that shows large initial corrosion rates- occurs in cladding with low resistance towards nodular corrosion.

<sup>&</sup>lt;sup>15</sup> This corrosion phenomenon resulted recently in a few failed rods. The mechanism is not clear but seems to be related to galvanic corrosion. This corrosion type may occur on the fuel cladding in contact or adjacent to a dissimilar material such as Inconel. Thus, this accelerated type of corrosion occurred on the fuel cladding material at spacer locations (the spacer springs in alloy BWR fuel vendors fuel are made of Inconel). Water chemistry seems also 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*, even in aggressive water chemistry.

The failure statistics during 1980-2007 in US PWRs and BWRs are shown in Figure 7-1 and the percentage of failures mechanism during 2000-2007 is provided in Figure 7-2.

Most of the US BWR failure cases are related to CRUD-accelerated corrosion failures. Other US 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 GTRF; however, experience with new grid designs appears to be promising. During the last years 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.

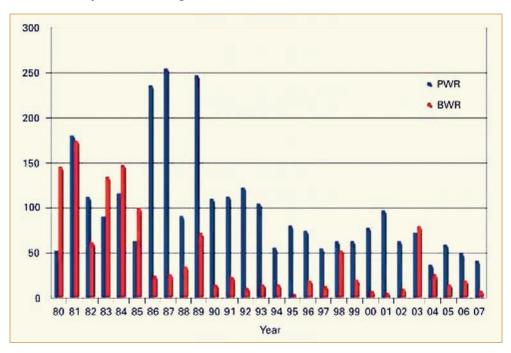


Figure 7-1: Fuel failures in US PWRs and BWRs, 1980-2007, [Tompkins 2008].

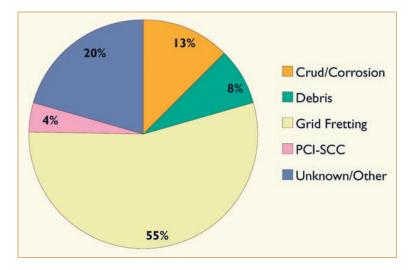


Figure 7-2: Percentage of fuel failures by mechanism for US PWRs and BWRs, 2000-2007, [Tompkins 2008].

#### Annual Report - 2009

In the European BWRs debris fretting is one of the major failure causes. Debris filters in the BWR fuels do not eliminate the debris fretting failures while the debris filters seems to be efficient in the PWRs.

(Table 7-2) and (Table 7-3) provide key data for some of the most recent fuel-failure cases.

Table 7-2: Summary of previous PWR failure key events, see previous ZIRAT/IZNA-reports for details.

Nuclear unit	Type of primary failure	Comment
TMI-1, Cy 10, 1995	Nine high peaking FRs, Zr-4 Cladding, failed after 122 days of operation. CRUD/corrosion related failures.	<ul> <li>All failed and degraded pins reportedly had Distinctive CRUD Pattern (DCP)<sup>16</sup>.</li> <li>High peaking factors, thermal-hydraulic conditions. Calculations indicated that no boiling should have occurred on the pins with DCP, although the pins with DCP were calculated to have a slightly higher temperature.</li> <li>Water chemistry (low pH at BOC, pH &lt; 6.9, max LiOH 2.2 ppm).</li> <li>Some, AOA effect was found reaching a maximum in the middle of cycle 10.</li> <li>The source of the CRUD could not be determined. The CRUD sampling showed that the nickel-to iron ratio was in the range 1.25 to 16.7, which was reportedly somewhat lower than in previous investigations.</li> </ul>
Seabrook, Cy 5, 1997	Five one-cycle ZIRLO rods failed. CRUD/corrosion related failures.	<ul><li>Longer cycle in transition to 24-month cycle.</li><li>Possibly CRUD-induced overheating resulting in substantial nucleate boiling.</li></ul>

<sup>&</sup>lt;sup>16</sup> This acronym implies that the fuel inspection revealed CRUD deposits on the fuel rod and that the deposits were uneven in the rod circumference.

# 8 LOCA/RIA

There were limited number of results related to LOCA/RIA published in the past twelve months. This topic will be covered in greater depth in the ZIRAT15/IZNA10 AR.

# 9 Fuel performance during intermediate storage (Charles Patterson)<sup>26</sup>

# 9.1 Introduction

Although worldwide policy decisions and statements of intent with regard to new, nuclear power plants continue to the show the importance of nuclear power as an energy source, political decisions combined with financial conditions continue to reinforce the need for "interim" storage of spent fuel as part of the overall fuel cycle. Most countries that generate nuclear power are in the process of developing criteria, designs and sites for the permanent disposal of Spent Nuclear Fuel (SNF). As indicated in (Table 9-1), however, operating repositories have yet to become licensed realities. Meanwhile the pools at the nuclear plant sites are filling up with spent fuel and the utilities are transferring the spent fuel from the pools to dry cask storage sites that are located, mostly, at the plant sites but also at remote storage facilities. Exceptions are the central, large intermediate pool facilities that serve all the plants in Sweden (CLAB facility) and all the plants in Finland (KPA-STORE). The lack of a licensed permanent fuel repository in any country has placed total reliance on intermediate storage. As a result dry storage has become a major activity and business component of today's back-end fuel strategies.

The importance of dry storage in back-end fuel strategies is due to a combination of factors. As noted above, the absence of permanent, geologic repositories combined with limited, in-pool storage capacity at reactor sites has forced the use of dry storage technology in countries that utilize a once-through, direct-disposal fuel cycle, which is shown schematically in (Figure 9-1) for reference.

Country	National decision	Status	Operational target date
Finland	Geological Repository	Site selected	2020
Sweden	Geological Repository	Site selected	2020
USA	Geological Repository	Yucca Mt. suspended in 2009 To be re-reviewed	Unknown
France	Geological Repository		2025
Germany	Geological Repository		2030
Japan	Geological Repository		2030
Switzerland	Geological Repository		2040
UK	Geological Repository	Siting initiated Three communities interested	
Canada	Geological Repository	Siting expected to start in 2010	≥2035
Belgium	No decision	R&D	R&D
Spain	No decision	R&D	R&D

Table 9-1: Status of geologic repositories [after Moran 2009].

<sup>&</sup>lt;sup>26</sup> The chapter on interim storage by A. Strasser in the ZIRAT13/IZNA8 reports has been updated by

C. Patterson to reflect developments through late 2009.

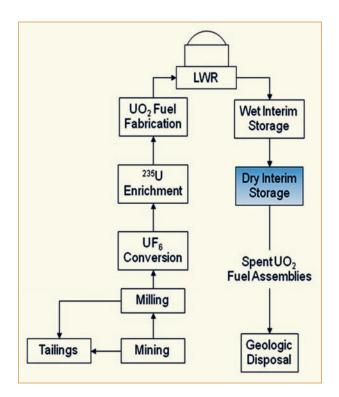
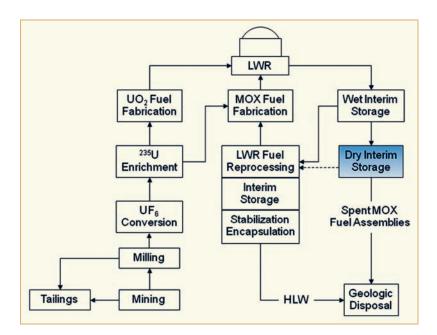
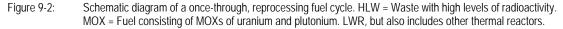


Figure 9-1: Schematic diagram of a once-through fuel cycle. LWR, but also includes other thermal reactors.

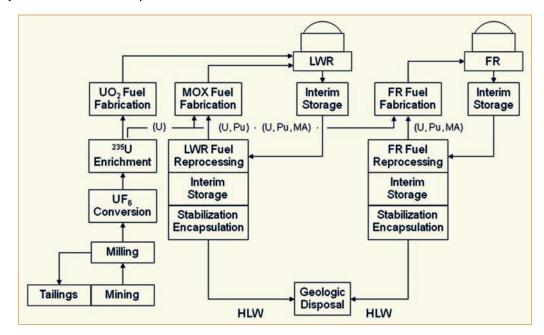
Economic considerations and capacity imbalances also contribute to the storage of spent  $UO_2$  fuel in countries that have commercial recycling capabilities. That is, dry storage offers a means of delaying reprocessing and recycling operations until the cost of fuel fabricated from virgin uranium or from highly-enriched, weapons-grade uranium justifies the recycling process. Dry storage also offers a means for balancing differences among the inventory of SNF, the capacity to reprocess irradiated fuel and the capacity to recycle the resulting plutonium as MOX fuel in the existing fleet of thermal-spectrum reactors.

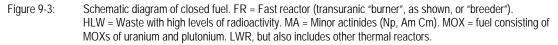
It should be noted that, in the absence of permanent geologic repositories, dry storage is also an inherent element of a reprocessing fuel cycle. The build up of actinides such as neptunium, americium and curium typically limit recycling to a single pass through a LWR. As shown in (Figure 9-2), this leads to the storage and disposal of spent, MOX assemblies in addition to the High Level Waste (HLW) that comes from the initial reprocessing operation. So, dry storage is common to both once-through and single-recycle fuel cycles.





In principle, a closed fuel cycle such as that shown in (Figure 9-3) could eliminate the need for dry storage. Reprocessing of spent fuel and the partitioning of transuranic nuclides (e.g., Pu, Np, Am, Cm), short half-life FP (e.g., Cs, Sr) and long-lived FP (e.g., Tc) are expected to reduce the volume of HLW by a factor of ten relative to the direct disposal of SNF, [Pereira et al 2006]. The transmutation of the transuranics and selected FP by means of (fast) reactors with high-energy neutrons or accelerator-based devices is intended to convert these high activity products to stable or short-lived nuclides. The overall effect of reducing the decay heat should allow for geologic disposal within a relatively short interval of time.





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# 10 Potential burnup limitations (Peter Rudling)

# 10.1 Introduction

The potential FA 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 limits of Zry-4 in high power PWRs, are extended by the alternate use of improved cladding alloys. Improved corrosion performance by the new alloys may allow the utilities to use the added margins, to modify plant operation e.g., to lower fuel cycle cost. However, this modified operation will in most cases result in higher corrosion duty of the zirconium materials. Thus, it is believed that the corrosion 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 FAs contributed in part by irradiation growth, creep and hydriding of Zry-4, has been reduced by improved GT materials (i.e., lower irradiation growth and hydriding rates), reduced assembly hold-down 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.

# 10.2 Corrosion and mechanical properties related to oxide thickness and H pickup

- BWRs: increased uniform and shadow corrosion, oxide thickness spalling, increased HPU, and formation of radial hydrides due to longer residence time, higher power, modern power histories, and water chemistry changes. Current crucial issues are: shadow corrosion mechanisms its effect on channel bow, late increased corrosion and HPU of Zry-2 at high burnups, and formation of radial hydrides, CRUD-chemistry-corrosion interaction, effect of water chemistry impurities, as well as specific effects of NMCA with or without Zn-injection.
- PWRs: increased uniform corrosion, oxide thickness, spalling, and new Zr alloys due to longer residence time and higher Li, higher power, more boiling. The development of new Zr-alloys is still ongoing. The database for several of the new alloys is still limited. Zr-Nb alloys are occasionally affected by accelerated corrosion due to surface contaminations and/or boiling. Welding of the new alloys may need improved processes (Zr-Nb alloys) and chemical compositions between dissimilar metals such as e.g. ZIRLO and Zry-4 may result in inferior corrosion resistance. Luckily, the corrosion temperatures at these elevations in the core are significantly lower than the peak temperatures.

- Decreased ductility and fracture toughness as consequence of the increased HPU during any situation (e.g., RIA, PCMI, LOCA and post-LOCA events, seismic event, transport container drop-accident conditions).
- Increased growth due to higher hydride volume and thick oxide layers.
- Increased corrosion due to impact of hydrides at the cladding outer surface.
- Impact of corrosion and HPU on creep behaviour of fuel claddings during class 1-IV events and during intermediate storage.
- Increased effects of irradiation and hydrides on the fracture toughness of thin-walled zirconium alloy components.

#### Most sensitive component

Fuel claddings and structural components such as spacers, PWR GTs, BWR channels.

#### Increase margin for PWR

- Improved knowledge of corrosion and HPU mechanisms.
- Improved alloys with appropriate fabrication processes: ZIRLO, MDA, New Developed Alloy (NDA), and M5/Zr1Nb. DX is another alternative that may be necessary to achieve satisfactory mechanical properties.
- Zr-alloys such as Optimised ZIRLO, Modified MDA, S2, Modified E635 with reduced Sn content in comparison to the original composition of ZIRLO, MDA, NDA and E635 are being explored.
- 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 HPU mechanisms at high burnups.
- Modification of manufacturing processes (to get optimum sized, more stable SPPs).
- Zry-4 fuel channels for controlled positions.
- Improved alloys under development.

Improved water chemistry and CRUD control.

# 10.3 Dimensional stability

- Increased dimensional changes of components and differential dimensional changes between them resulting in reduced FR spacing or even rod contact, GT bowing, FA bowing, spacer cell and envelope dimensions, BWR fuel channel and PWR FA 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 Zircaloy PWR GTs 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 GTs ZIRLO, M5, E635 (Anikuloy).
- Modified mechanical design to provide lower hold-down forces, stiffer assemblies, etc.
- Refinement of the understanding of the effects of corrosion and hydriding at moderate and high burnups.
- 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 might be improved by an appropriate final heat-treatment in the alpha-phase. Also applies to BWR materials control of texture (close to ideal isotropy) is critical for good performance at high burnups).

#### Increase margin for BWR

- Uniform microstructure and texture throughout the flow channel.
- Use of lower growth material, such as beta-quenched material in as-fabricated step, NSF or other Nb-modified zirconium alloys.
- Channel management programs, including assessment of degree of control over specific reactor periods.
- More corrosion and shadow-corrosion resistant material in channels and spacers.
- Increased understanding of basic phenomena driving the channel bow process, including flux and hydrogen-driven processes.
- Lowered hydride pickup and increased uniformity of hydride distribution in channels and spacers through heat treatment and alloy choice.

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