Zr Alloy Manufacturing and Effects on In-reactor, Design Basis Accident and Interim Dry Storage Performance

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Nomenclature

Unit conversion

1 Introduction

To ensure safe operation, specific nuclear fuel performance criteria must be met during normal operation, anticipated operational events, postulated accidents, and back-end operations (storage, transportation, and final disposal). Zirconium alloy material properties affect some of the most important performance criteria related to:

- Pellet Cladding Interaction (PCI), Pellet Cladding Mechanical Interaction (PCMI), corrosion and hydriding properties, which, in turn, depend on:
 - Chemical composition.
 - Crystallographic texture and cladding microstructure.
 - Cladding strength/ductility/fracture toughness.
- Re-opening of the pellet-cladding gap, or lift-off, which is partly related to the fuel cladding creep properties.
- Limiting dimensional changes of fuel components (such as, for example, Fuel Assembly (FA) bowing), which are a function of creep (including oxide-induced and residual stress relaxation creep), irradiation growth, and hydrogen pickup in the components.
- Cladding performance during a Loss-of-Coolant-Accident (LOCA), which depends on hydrogen pickup during the base irradiation before the LOCA event as well as during the high temperature LOCA oxidation transient.
- Zr alloy performance during Reactivity Initiated Accident (RIA) and seismic events where hydrides reduce ductility.
- Maintaining high levels of cladding integrity and minimizing loss of ductility and fracture toughness, especially under normal conditions during interim storage and transportation, which depends on such factors as fuel rod design (initial He pre-pressurization level, presence of inner or outer liners, presence of pellets coated with ¹⁰B-containing burnable poisons), cladding's end-of-life hydrogen content (hydriding), and alloy microstructure including the amount of residual cold work from fabrication (cold-worked, stress-relieved vs. partially or fully recrystallized) and the effects induced by neutron irradiation.
- The pressure tubes in Pressurized Heavy Water Reactors (PHWRs) must resist dimensional changes that cause interference with neighbouring components and reduce efficiency of heat transfer. The concentration of hydrogen isotopes must be minimised to avoid cracking by hydrides and high toughness must be maintained during irradiation to avoid rupture.

The Zr-alloy properties are a function of the reactor environment (fast neutron flux, temperature, water chemistry, etc.) and the Zr-alloy microstructure. The microstructure is a function of material chemistry and the manufacturing process. A better knowledge of the consequences of manufacturing changes on the Zr alloy microstructures and in-pile material properties can lead to improved capabilities to respond to current fuel performance issues and to ensure that any changes in the current manufacturing processes of the Zr alloys will not impair the fuel safety performance criteria. The overall objective of this Special Topic Report (STR) is to provide this knowledge.

2 Reactor characteristics (Charles Patterson)

There are essentially five different types of commercial water-cooled reactors. By design, the watercooled reactors in operation can be separated under thermodynamic aspects into Closed Cycle Systems (PWR, VVER, and PHWR) and Open Cycle Systems (BWR and RBMK) [Riess & Millet, 1994].

The separation into "Closed" and "Open Systems" has consequences when it comes to the interaction between radiation and the coolant. In a "Closed" system, the net decomposition of the coolant can be suppressed by the addition of hydrogen or ammonia, whereas in "Open Systems" volatile radiolysis species are produced and removed from the coolant while non-volatile species remain in the coolant.

The primary coolant in Closed Cycle System reactors operates under alkaline and reducing conditions with LiOH or KOH as the agents of pH control. This basic principle has been used for almost 50 years and has only been modified within the established framework of specified values for pH control. Such modifications are called "coordinated", "modified", "elevated lithium", or "coordinated at elevated pH" Li/B-chemistries.

The primary coolant in Open Cycle System reactors has historically operated under neutral and oxygenated conditions while keeping the coolant water as clean as possible. However, based on material integrity concerns, the coolant chemistry conditions have been modified in most reactors to reducing conditions by injecting hydrogen in to feedwater (Hydrogen Water Chemistry (HWC), Noble Metal Chemical Addition (NMCA) and Online Noble Chemistry (OLNC). The latter two technologies involve injecting a solution containing noble metals (Pt +Rh) in the case of NMCA and Pt in the case of OLNC.

The fuel in water reactors operates in a wide range of environments. Typical operating conditions for varying reactor types are shown in Table 2-1. Additional information on the thermal-hydraulic conditions in BWR and PWR cores is given in Figure 2-1. For reference, the number and relative generating capacities of each type of operating plant in Figure 2-2. The figure shows similar data for reactors under construction, Figure 2-3.

3 Fuel assembly designs (Peter Rudling)

3.1 BWR fuel design

The fuel rod array for BWRs was initially 6×6 but the 7×7 lattice configuration was adopted relatively early and there has been a trend over the subsequent years to increase the number of rods so that now most fuel assemblies are either of 9×9 , 10×10 or 11×11 square configuration designs. An example of a BWR fuel assembly based on a 10×10 lattice is shown in Figure 3-1. The driving force in the trend toward larger number of fuel rods in each fuel assembly was the reduction of Linear Heat Generation Rate (LHGR). Such changes produced a number of fuel performance benefits, such as lower Fission Gas Release (FGR) and increased margin to the Pellet Cladding Interaction (PCI) failure threshold, which is discussed later. However, to decrease fuel cycle costs, the LHGRs of 9×9 , 10×10 and 11×11 fuel assemblies have successively been increased so that peak heat generation rates are today almost comparable to those of the older 7×7 and 8×8 designs.

In all BWRs, fuel bundles are enclosed in open-ended, square tubes or "channels". The combination of a fuel bundle (rods, spacers, tie plates and related hardware) and a channel is commonly identified as a fuel bundle or, more generically, as a fuel assembly. The channels are ducts for coolant flow that prevent steam induced lateral flow of coolant among the assemblies operating at different power levels. They also form the inter-assembly space into which control blades are inserted.

Fuel assemblies are positioned in a reactor core support structures at the bottom and top of each assembly. Lower tie or bottom support plates, Figure 3-1, are supported by a pedestal that carries the assembly weight and directs coolant into the channel. The upper ends of fuel assemblies are supported laterally by a rectangular lattice structure which bears against wear pads on the outer surface of the fuel channels. Cruciform-shaped control blades are located in the region between the channels of selected sets of four fuel assemblies. Part of the incoming coolant is diverted into the region among fuel assemblies to transfer heat from the control blades and prevent boiling.

Irrespective of the many possible different shapes, sizes and configurations, the common fuel assembly design requirements are:

- Maintain proper positioning of the fuel rods under normal operating conditions and in Design Basis Accidents (DBAs); i.e., maintain a controllable and coolable (safe) geometry during normal operation, anticipated operating occurrences and during postulated seismic events as well as LOCA, RIA and (more recently) beyond design-bases accidents.
- Contain or at least confine fuel materials and fission products.
- Facilitate handling before and after irradiation.



Figure 3-1: GE 14 Fuel Design, provided courtesy of GE.

3.2 PWR fuel design

For PWRs there has also been a trend to greater subdivision of fuel rods, e.g. from 15×15 to 17×17 fuel assemblies in PWRs of the Westinghouse design. However, since the control rods and control rod drives in PWRs are designed for a specific lattice configuration, they do not have the same flexibility with respect to changing fuel designs as BWRs. There is however, one exception namely DC Cook 1 which was switching to 17×17 by changing the reactor internals. Figure 3-2 shows the current PWR fuel rod arrays.



Figure 3-2: Layouts of different PWR fuel assembly arrays. Rods marked with yellow colour are guide tubes into which the control rod cluster is inserted. The position marked by a red filled circle is the instrument tube position.

In most PWRs, the assemblies are positioned in the core by bottom and top fittings, and the lateral clearances are restricted by the assembly-to-assembly contacts at the spacer-grid levels. PWR control rods consist of Rod Cluster Control Assemblies, RCCAs, the poison part of which moves into guide thimbles or tubes (GTs). These guide thimbles are an integral part of the assembly structure. Figure 3-3 shows an example of the Westinghouse PWR fuel design.

4 The materials used in fuel assemblies (Alfred Strasser)

4.1 Introduction

The materials used for the fuel assembly components are zirconium alloys, nickel-based materials such as alloy X-750, alloy 718, alloy 625 and low-carbon, low-cobalt austenitic stainless steels (SS) [Strasser & Rudling, 2004]. Materials are selected based on their nuclear, physical and mechanical properties along with their compatibility with conditions in the core of water reactors.

Zirconium alloys are now used exclusively for fuel cladding and are also widely used in the grids (spacers) of modern fuel because of their low capture cross-section for the thermal neutrons. Stainless steel was used in some early fuel cladding and grids (1960–1970 era), but was replaced with zirconium alloys as commercial capabilities and the related technology emerged.

Other materials such as stainless steel and nickel-based alloys are used in structural components, but have significantly larger cross-sections and can adversely affect reactivity and increase fuel-cycle costs depending on their mass and location relative to the active fuel. Upper and lower nozzles (tie plates) are usually constructed of cast, austenitic stainless steel. Springs are typically fabricated from materials with high strength and low rates of stress relaxation, such as alloy X-750 or alloy 718. These Ni-based alloys are solution heat treated and then precipitation hardened by means of thermal cycles that have been developed to achieve high strength while maximize their resistance to stress corrosion cracking. Low cobalt content is needed in Ni-based alloys and SS to minimize radiological exposure of plant personnel due to transmutation, corrosion and transport of activation products through the primary coolant system.

To lower the parasitic neutron absorption by grids, their structural components have historically been made of zirconium alloys such as Zry-2 or Zry-4. In most cases, zirconium alloy spacers utilize springs made of either alloy X-750 or alloy 718 to ensure adequate fuel rod support during irradiation. Some PWR designs utilize grids constructed entirely of a zirconium alloy. Alloy X-750 and alloy 718 is being used in an increasing number of fuel designs for the entire spacer; e.g., the top and bottom grids in some PWR designs and all spacers in some high-burnup BWR designs. In PWR fuel, Ni-base alloys can be used for the top and bottom grids because the neutron flux is much lower in these locations, resulting in a very small loss of thermal neutrons due to parasitic material absorption. In BWR fuel, Ni-based alloys are used in minimum-mass configurations (thin strips with all non-load bearing material removed) to reduce parasitic loss of neutrons. Such spacers are being used to minimize the pressure drop associated with thicker Zr-alloy spacers and to avoid hydrogen pickup and embrittlement of Zr-based spacers at fuel intended for high burnup, long residence applications.

It also should be noted that BWR zirconium alloys continue to be primarily Zry-2 or slight variants of Zry-2. PWR zirconium alloys no longer tend to be Zry-4, for reasons of insufficient corrosion resistance (and hydriding resistance) at high burnup, but have moved toward zirconium alloys with Nb additions.

4.2 Zirconium alloys

A number of different zirconium alloys have been developed for use in light and heavy water reactors. The principal alloys are described in Table 4-1. It is noteworthy that there are so many different Zralloys for PWR applications. Originally, Zr-4 was the standard material in PWRs. But increased corrosion rates and hydrogen pickup at extended burnups resulted in a need to develop more corrosion resistant alloys either as monotubes or as DUPLEX tubes (only used in Europe).

Both Zry-2 and Zry-4 have been used in BWRs; i.e., Zry-2 for fuel cladding and either Zry-2 or Zry-4 for fuel channels. With the composition and microstructural controls of current fuel, Zry-2 exhibits good corrosion resistance through high burnups as BWR fuel cladding. The pickup of hydrogen due to corrosion at the surface of fuel cladding is observed to increase with burnup. Such increases in hydrogen pickup have shown that alloys with better corrosion and hydriding resistance are needed. Improved versions of Zry-2 and alternate alloys are being developed. A similar condition exists for fuel channels. That is, Zry-2 is resistant to corrosion in BWR environments, but undergoes increases in

5 Performance of Zr Alloys

Table 5-1 lists the Zr alloy properties limiting the fuel performance during normal operation, AOO, DBA and Interim Dry Storage. The different "failures mechanisms" according to Table 5-1 are described in short in the following subsections.

Condition	Failure mechanism	Key Zr alloy material property	Design limit	How to increase margin to failure
Normal operation and AOO	Corrosion and Hydriding	Corrosion rate and HPUF	Maximum oxide thickness often limited to 100 μ m, to ensure that corrosion acceleration due to thermal feedback does not occur. The hydrogen concentration of zirconium alloys must be limited to prevent brittle mechanical failures due to the formation of hydrides during normal operation, handling during outage and seismic events.	Improve corrosion resistance by alloying and thermomechanical heat treatment during fabrication. Improving corrosion resistance also reduces the risk of hydride blister formation due to oxide spallation. Hydride blisters reduce PCMI margins during RIA. Decrease HPU to increase safety margins during LOCA, RIA, seismic events and handling accidents (including tip-over during interim dry storage).
	PCI during power ramping	SCC resistance, stress level	Margins assessed by test reactor ramping	Considering the fuel cladding, the best way to improve PCI performance is to add a Zr liner at the clad inside surface, as being used for BWR fuel cladding. PWR fuel cladding with Zr-liner has been ramp tested with excellent results. A thin graphite coating is added to the inside surface of CANDU fuel to minimise PCI. A thin graphite coating is added to the inside surface of CANDU fuel to minimise PCI.
	PCMI during power ramps NB: This failure mechanisms has not happened in commercial reactors but was observed for BWR fuel ramped in a test reactor.	HPU =Corrosion rate [*] HPUF	This failure mechanism may potentially happen for PWR fuel if there exist a solid hydride rim at the fuel clad outside surface and the ramp height is large enough following extended operation at low power	A prerequisite to form a hydride rim, a large enough heat flux and hydrogen content in the cladding are necessary. Reducing the hydrogen concentration to < 300 ppm may eliminate the formation of a hydride rim at the fuel clad outer surface. An even larger embrittlement effect may be obtained if a hydride blister with significant thickness is formed at the clad outer surface. In addition to a heat flux and large enough hydrogen content also spalled oxide is necessary for a hydride blister to form. Thus, developing cladding material that forms thin oxides that will not spall will eliminate this embrittlement effect.
	Lift-off	Creep resistance	Rod internal pressure must be limited to avert lift-off that potentially could lead to higher rod internal pressure and fuel failure due to an increase in fuel temperature as a result of degradation of heat transport between fuel and cladding inside surface.	Improved irradiation creep strength can be obtained by replacing SR with RX (or pRX) claddings or by alloying additions. The drawback with RX (or pRX) is that the PWR corrosion resistance in alloys containing significant amounts of Sn deteriorates compared with SR cladding. However, in BWRs, RX, pRX and SR tubes have similar corrosion properties.
	FA bowing (PWR/VVER)	Elongation rate of GTs	FA bowing (distortion) must be limited to ensure that control rods can be inserted anytime and that thermal margins (DNBR and LOCA) are maintained. Bowing may also cause grid damage during handling.	Tendency for FA bowing is mostly a design issue, i.e., with an appropriate FA design even a large GT elongation rate would not result in any significant FA bowing. However, lower GT elongation rate reduces the GT compressive stresses (as well as other fuel design features may do) and may reduce FA bowing. If GT have a weak point or low creep strength, FA bow can occur under the compressive stresses (depending on hold down springs) and cross flow etc. Key property of GT is to use a material with high creep strength under compression.
	Fuel Channel Bowing (BWR)	Irradiation growth, shadow corrosion, and HPU	Fuel channel bowing (distortion) must be limited to ensure that control rods movements are not hindered and that thermal margins (dry out and LOCA) are maintained.	 There are two main causes for fuel channel bowing today: Difference in irradiation growth rate of two opposing channel sides due to a fast fluence gradient (fluence gradient induced bow) – for most Zr alloys. Difference in the elongation of two opposing channel sides due to a hydrogen content difference (shadow induced bow) – only occurs for Zr alloys containing nickel-bearing SPPs such as e.g. Zry-2 Zr alloys containing Nb reduces 1) above and eliminate 2). Final beta-quenching of non-Ni containing Zircaloys minimizes both 1) and 2) above
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Table 5-1:	Zr alloy properties limiting performance during normal operation, AOO, DBA and interim dry storage.
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Table 5-1:	Zr alloy properties limiting performance during normal operation, AOO, DBA and interim dry storage.
	(Conťd)

Condition	Failure mechanism	Key Zr alloy material property	Design limit	How to increase margin to failure
LOCA	FR fracture and fuel dispersal	The embrittlement of the fuel cladding depends on the H content in the cladding which depends on: 1) the HPU during reactor operation prior to the LOCA and 2) the high temperature oxidation time during LOCA before break- away oxidation occurs (resulting in large HPU during the LOCA)	Retain some ductility of the fuel cladding (Post Quench Ductility, PQD) to ensure that fuel dispersal does not occur by making sure that the cladding does not fracture during the LOCA quenching phase or during post-LOCA events. The licensing criteria are currently being revised by USNRC Ensure that breakaway oxidation does not occur during HT oxidation and that PQD is met.	Reduced HPU during reactor operation before LOCA (through more corrosion resistant materials with less HPUF) will increase margins toward fuel dispersal (by ensuring some retained clad ductility). Ensure that breakaway oxidation (and thereby accelerated HPU) does not occur.
RIA	FR fracture (through PCMI) and fuel dispersal	The PCMI failure tendency increases with: 1) increased HPU during corrosion before the RIA 2) localised hydrides (blisters and rims) 3) increased volume fraction of radial hydrides	In most countries, FR failures are allowed above a certain enthalpy increase during the RIA. However, in Germany FR failures during RIA are not allowed. In all countries, fuel dispersal most not occur.	Reduced: 1) HPU (by Zr alloys with lower corrosion rate, or lower HPUF or both) and, 2) fraction of radial hydrides (by using SR instead of pRX or RX materials), formed during reactor operation before RIA (through more corrosion resistant materials with less HPUF). Elimination of any localised hydrides such as blister and rims will increase margins toward PCMI failures.
Dry Storage	Cladding rupture and fuel relocation during cask drop accident.	Creep strength Microstructural features affecting the orientation of hydride platelets upon initial formation and subsequent thermal cycling (heating followed by cooling).	In some countries, a maximum creep strain of 1% is established to ensure that creep rupture does not occur (Germany-based regulatory regime). In other countries, creep is considered to be a self-limiting deformation mechanism that is unlikely to result in cladding rupture (US-based regulatory regime). Peak cladding temperature, or cladding stress or both are limited to minimize the potential for formation of radial hydrides during the dry storage cooling phase. Although there is no requirement to maintain fuel integrity under cask drop accident conditions, the cask content must remain sub-critical under reflooding conditions. To satisfy this requirement, the applicant must be able to credibly assess the condition of the fuel, if it cannot be readily shown that the fuel remains intact.	Increased creep strength increases margins against creep rupture. Fuel design: By adding a liner/DUPLEX layer to the fuel cladding, little to no radial hydrides will form because the soluble hydrogen tends to diffuse to the liner material during cooling, thereby preventing the formation of radial hydrides in the non-liner part(s) of the cladding. In the absence of a liner, strong texture (basal pole preferentially aligned in the radial direction; no late beta quench) and minimization of grain boundaries aligned in the radial direction should be favoured.

5.1 In-reactor performance

5.1.1 Introduction

The alloying elements in solid solution may affect the corrosion and hydriding properties as well as strength of the material, while the SPPs mainly affect corrosion and hydriding.

In the majority of cases, impurities will degrade the properties and characteristics of materials. Therefore, their concentrations are restricted by the standards and specifications for zirconium, its alloys and the resulting FA structural parts.

The basis for these restrictions is the requirements to preserve low neutron capture, high corrosion resistance, processability and crack resistance during manufacturing as well as reliable operation in reactors. Among the impurities that are most important in this respect are C, N, Al, H, Cl, P, Hf, F, as follows:

- N, Al and C impurities are most harmful for corrosion of zirconium.
- C, H, Si, Cl, P and F impurities decrease the ductility of zirconium and its alloys, thus, degrading processability and crack resistance.
- Cd, B, Hf and Co are elements that increase the neutron capture.
- C, N, Al, H, Cl, P, Hf, F increases strength and affects corrosion and ductility to some degree.
- Fe and Cr favourably influence the corrosion of zirconium, for this reason their allowable concentrations are much higher.

The typical requirements specified for impurity concentrations of zirconium and its alloys are summarized in Table 5-2.

Element	Pure Zr	Zry-2	Zry-4	Zr-Nb (2.4-2.8%Nb)	Zr-Nb (2.5-2.8%Nb)
Impurity, max	(ppm)				
AI	75	75	75	75	75
В	0.5	0.5	0.5	0.5	0.5
Cd	0.5	0.5	0.5	0.5	0.5
С	270	270	270	270	150
Cr	200	-	-	200	200
Со	20	20	20	20	20
Cu	50	50	50	50	50
Hf	100	100	100	100	100
Н	25	25	25	25	25
Fe	1500	-	-	1500	650
Mg	20	20	20	20	20
Mn	50	50	50	50	50
Мо	50	50	50	50	50
Ni	70	-	70	70	35
Ν	80	80	80	80	65
Pb	-	-	-	-	50
Si	120	120	120	120	120
Sn	50	-	-	50	100
Та	-	-	-	-	100
Ti	50	50	50	50	50
U	3.5	3.5	3.5	3.5	3.5
V	-	-	-	-	50
W	100	100	100	100	100

Table 5-2:	ASTM Standard B353 for Zr and its alloys used in nuclear industry (concentration of impurities).

The deduced FF⁵ for a burnup of <50 MWd/kg (the estimated post transition rate in comparison with the rate that would proceed on Zry-4 out reactor), deduced by Siemens calculations or own calculation on some published Westinghouse, Framatome, and Korean FR cladding corrosion data, are shown in Table 5-3 together with the average HPUF, and the creep characteristics for the interesting ZrSnFeCr-, ZrSnFeV-, ZrNb-, ZrNbCu-, and ZrSnNbTM-alloys.

⁵ Relates to the in-reactor to out-of-reactor corrosion rate for an alloy. A value of e.g. 4 means that the in-reactor corrosion rate is 4 times larger than that out-of-reactor at same tempeerature.

Table 5-3:Chemical composition, material condition and estimated corrosion and creep behaviour of alternate
ZrSnTM-, ZrNb-, ZrNbCu, alloys, being developed for PWR application.

Material	Sn (%)	Nb (%)	Fe (%)	Cr (%)	Others	(%)	Condition	∆D at 16 ¹ MWd/kg (%)	FF ¹	HPUF ² (%)
Best Zry-4	1.3		0.2	0.1			SR	0.48	2.5	~15/~25
Best Zry-4	1.3		0.2	0.1			RX	0.26	4	~15/~25
DX-D4	0.5						720°C/82%CW/SR	0.5	0.9	- /~25
DX-HPA-4	0.5		0.5		0.3 V		720°C/76%CW/SR	0.5	0.9	-/~10
M5		1	0.04				RX	0.35	0.8	<10
DX-Zr2.5Nb		2.5					720°C/>76%CW/SR	~0.5	0.4	<10
J3		2.5					RX	(0.35)	(1)	
HANA-6		1			0.05 Cu		PR		(1.3)	
AXIOM-X4		1		0.25	0.08 Cu		80%PR	~0.32	~1.3	
Quart ZrNbFe		1	0.1				RX	~0.35	~0.7	
ZIRLO	1	1	0.1				580°C/80%CW/SR	0.37	2.5	~15/~15
ZIRLO	1	1	0.1				RX	0.2		~15/~15
Low Sn-ZIRLO	0.67	1	0.1				580°C/80%CW/SR	~0.37	~1.5	- /~15
Optim. ZIRLO	0.67	1	0.1				580°C/80%CW/PR	~0.37	~1.9	- /~15
DX-Zr0.8Sn0.8NbFeV	0.8	0.8	0.2		0.1V		SR	~0.6	1.7	
Quart.ZrNbSnFe	0.5	1	0.1				RX	~0.23	~2	
Zr0.5Sn0.5Nb0.1Fe	0.45	0.55	0.1				SR		~1.1	~15/ -
AXIOM-X5A	0.45	0.3	0.35	0.25	0.05% Ni		50%PR	~0.3	~1.6	
HANA-4	0.4	1.5	0.2	0.1			PR		(1.6)	
Quart.ZrNbSnFe	0.3	1	0.2				RX	~0.27	~1.2	
AXIOM-X5	0.3	0.7	0.35	0.25			50%PR	~0.34	~1	
AXIOM-X1	0.3	0.8	0.15		0.12 Cu, 0).2V	80%PR	~0.25	~1.3	
Zr0.2Sn0.8Nb0.25Fe	0.25	0.8	0.25				SR	~0.76	~0.9	
^{1:} Diameter decrease i MWd/kgU () indicates a	^{1:} Diameter decrease in % of the cladding in the middle of the FR after the first cycle, normalized to a burnup of 16 MWd/kgU () indicates an uncertainty, ~ only few data,									

^{2:} HPUF in PWRs with high Ni/low Ni in the coolant. The HPUF is calculated from the average hydrogen content measured via the whole cladding subtracting the initial value of the cladding.

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The HPUF observed in Siemens PWRs is shown in Table 5-4. A particular aspect is the HPUF, which is in case of ZrSnFeCr alloys is different in Siemens PWRs and Westinghouse PWRs. Similar HPUF values were found in both reactor types for ZrSnNbFeCr alloys (with $\geq 0.5\%$ Nb) (~15%) and for binary ZrNb alloys an HPUF of $\leq 10\%$.

Material	FF	Mean pick-up fraction /%/	Number of measurements	Mean standard deviation /%/
1.5 Sn Zry-4	4	<10		
Low Sn Zry-4	2.5	>20		
DX ELS0.8b	1.1	23	32	7
DX D4	0.95	20	7	3
PCA-2b	1.3	12	2	4
DXHPA 4	1	10	6	3
Zr1Nb	0.8	9	12	6
Zr2.5Nb	0.4	9	10	5
	8	۶ <u>ــــــــــــــــــــــــــــــــــــ</u>	•	© ANT International 2017

Table 5-4: Mean pick-up fractions and statistical measurement data modified [Goll & Hoffmann, 2002].

5.1.2 Corrosion and HPU

5.1.2.1 Introduction

Corrosion of zirconium alloys is a thermodynamic and electrochemically based process affected by the following parameters, see Figure 5-1:

- The microstructure of the Zr alloy-metal surface.
- The water chemistry and the hydraulic conditions.
- The Zr alloy temperature (at the metal/oxide interface).



Figure 5-1: Parameters controlling corrosion performance of Zr Alloys.

During the initial oxidation/corrosion of zirconium alloys, a thin protective black oxide is formed. As the zirconium oxide grows in thickness the outer part of the oxide (facing the water/steam phase) is transformed into a greyish porous oxide. The oxide grows inwards into the zirconium alloy material.

In general, there exists common understanding that only atomic hydrogen that is generated by cladding corrosion is picked-up by Zr alloys and not the dissolved molecular hydrogen that is added into the reactor coolant. In high temperature water, Zr-based alloys build a thin, compact, black protective layer by early oxidation phase with water. Electrons released from Zr by oxidation are captured by hydrogen ions of the water that reduce them to hydrogen atoms. Some of these hydrogen atoms recombine to H₂ molecules and release from the Zr surface as hydrogen gas, whereas the other part can migrate into Zr matrix as soluble H atoms. Hydrides can form when the concentration of picked-up hydrogen exceeds the solubility limit for the Zr alloy, (often about 100 mg/kg at 330 °C) and any excess hydrogen formed by Zr oxidation (corrosion) is defined as Hydrogen Pick-Up Fraction (HPUF). This HPUF, depends on zirconium alloying content but also on temperature, water chemistry, and reactor start-up procedure. The total amount of hydrogen that is picked up by the Zr alloy is the product of the corrosion rate and the HPUF.

High Hydrogen Pickup (HPU) in the Zr alloy fuel cladding is one of the main concerns regarding fuel cladding performance. Increased HPU can lead to

- Formation of dense hydride rim, which results in early increase in cladding corrosion (hydride rim corrodes faster than the Zr alloys) during normal reactor operation.
- Increased dimensional changes of Zr alloy fuel assembly components (Hydride has a 15% higher specific volume relative to the metallic Zr).
- Develop embrittlement of the Zr alloy components during accident conditions (LOCA, RIA and handling and drop accidents (Zr-hydride is hard and brittle) [Garzarolli & Sabol, 2006].

The corrosion and hydriding process of Zirconium alloys is schematically shown in Figure 5-2.



Figure 5-2: Schematic diagram showing the corrosion and HPU process in zirconium alloys.

There are different types of corrosion modes:

• Nodular corrosion (almost only BWRs since it requires an oxidising environment) starting after 10 to 100 days of irradiation in material with large Zircaloy Second Phase Particles (SPPs) (Figure 5-3 and Figure 5-9). Nodular corrosion is characterized by much thicker local oxide patches that appear as white spots. The corrosion rate is initially very large but levels off at high burnups Due to the low oxidation potential in a PWR/VVER, only uniform corrosion normally exists (Figure 5-9). However, if for some reason there would be an increase in the coolant oxidation potential, nodular corrosion may also occur in PWRs/VVERs.

Oxide nodules have been observed on the outside surface of Zr-2.5Nb pressure tubes in two Indian PHWRs at one site. These nodules were attributed to traces of petrochemicals in the dry CO2 surrounding the pressure tube. Some of the nodules cracked and led to penetration of the tube walls. The tubes were replaced and the CO2 supply was changed to exclude petrochemicals

• Uniform corrosion (PWRs/VVERs/CANDUs/BWRs) that starts at the beginning of irradiation at a rapid rate, slows quickly (Figure 5-3 and Figure 5-9). With increasing oxidation and hydrogen absorption (pickup) at higher burnup, the concentration of hydrogen in Zr alloys can exceed their solubility limit and precipitate as hydrides. In components with heat transfer (e.g., fuel cladding), such hydrides tend to form in regions of lowest temperature. With the heat flux typical of LWR fuel rods, precipitation in regions of low temperature can lead to relatively high concentrations at the outer metal-oxide interface, which can, in turn, increase the oxidation rate. Hydrogen may also migrate to form hydrides at the interface of duplex and barrier cladding.

In PWRs, there is an increase in the oxide growth rate at a thickness of about 20-30 microns because hydrides may form in the Zr alloy at this point, thus increasing the oxide growth rate, Figure 5-8. At oxide thicknesses of about 100 microns and a large surface heat flux, there may also be a thermal feedback effect, which then leads to a second oxidation acceleration step.

In VVERs, there is normally no acceleration of the uniform corrosion process due to the very low hydrogen pickup. The reason for the low hydrogen pickup is due both to the normal use of the Zr1Nb alloy E110 and different coolant chemistry.

In BWRs, the SPPs (nickel-bearing and chromium-bearing SPPs) in Zry-2 starts to dissolve already at the start of irradiation due to the fast neutron flux. Dependent upon the initial size, type and chemistry of the SPP, the dissolution rate varies. At a certain fast fluence (corresponding to a certain burnup) the SPP has completely dissolved in the matrix, the fluence when this complete dissolution occurs increases with increased initial SPP size. When the SPPs have totally dissolved there is a dramatic increase in HPUF from 5-10% to over 100% (which means that part of the hydrogen must also come from the radiolytic hydrogen being produced) which eventually will lead to formation of a hydride rim at the metal/oxide interface. The hydride rim corrodes faster than the zirconium alloy which means that at this point there is a corrosion acceleration.



Figure 5-3: Corrosion morphology for Zircaloy [Adamson et al., 2007/2008a].

• Shadow corrosion (almost only in BWRs since it requires an oxidising environment) starting after a few days of irradiation (Figure 5-9), which may accelerate at high burnups. Although agreement does not exist regarding the underlying mechanism, shadow corrosion is frequently postulated to be a galvanic type of corrosion. Shadow corrosion has "always" been present in BWRs, but not in PWRs, primarily related to the high PWR hydrogen concentration which reduces or eliminates galvanic potentials between dissimilar alloy components. Shadow corrosion occurs in areas where a Zr alloy is in contact with or in close proximity to a dissimilar material such as nickel-based alloys (Inconel) or stainless steel, e.g., spacers in case of fuel rods (Figure 5-4), and control rods in case of BWR fuel outer channels (Figure 5-5 and Figure 5-6).



Figure 5-4: Oxide layer thickness profile of a BWR Zry-2 fuel rod opposite to Zry contact points (a) and Inconel contact point (b) after a burnup of 46 MWd/kgU, after [Garzarolli et al., 2001a].

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Figure 5-5: BWR channel control rod handle shadow corrosion.



Figure 5-6: Zirconium oxides near (a) and away from (b) a stainless steel control blade bundle [Adamson et al., 2000].

6 Zr component manufacturing

This section gives an overview on how the tubular manufacturing processes control the microstructure and in turn the fuel performance, (Figure 6-1).

							Strength							
							Ductility/Fracture Toughness							L
							Сгеер							
		11					Growth							
							Corrosion							
							Hydrogen Pickup Fraction							
							Stress Corr. Cracking							
Neutron-Flux	Neutron-Fluence	Power History	Burn up	Heat Transfer	Clad Temperature	Water Chemistry	Material Anameters Material Characteristics	Chem. Composition	Homogeneity	Precipitates	Grain Size	Dislocation Density	Texture	Curface Condition
							Melting							
							Hot Forming							
							Beta Quenching							
							Pilgering							
							Intermed. Annealing							
							Final Annealing							
							Surface Finish							



As an example of the effects of the parameters mentioned above on fuel performance we can consider PCI performance of a fuel rod that is related to:

- The stress corrosion cracking properties (material properties) of the fuel clad inner surface that in turn is a function of parameters such as:
 - Material characteristics (microstructure) that are a function of:
 - Chemical composition of the clad inner surface.
 - Homogeneity
 - Grain size of the fuel cladding
 - Texture
 - Clad inner surface condition

7 Metal production (Van Arkel, Electrolytic process, Kroll)

7.1 Introduction

The Swedish chemist Berzelius succeeded in 1824 to produce impure zirconium powder by reduction of potassium zirconium fluoride with sodium. In 1865 Troost was able to produce impure metal by reducing gaseous zirconium tetrachloride with magnesium. It was not until in 1925 that Van Arkel, DeBoar and Fast succeed in developing the first practical method for producing ductile zirconium. Their method was based upon an iodine process in which zirconium iodide decomposes on a hot filament. However, this method is very expensive and it would not be possible to produce large quantities of zirconium with this method. This material is called "crystal bar or iodide Zr".

In the second part of the 1940's, Dr. Kroll succeeded in developing a more economical process at Albany, Oregon, for production of ductile zirconium by reduction of zirconium tetrachloride with molten magnesium in an inert atmosphere. This material is called "sponge Zr".

A third method was used in the former Soviet Union to electrolytically produce Zr metal.

All fuel vendors are using Zr metal (sponge Zirconium) by the Kroll process except for the Russians that have been using Zr metal from a mixture of Zr produced through the Van Arkel and the electrolytic processes.

Nearly all the zirconium metal is extracted from zircon sand, Zr-Hf SiO₄, occurring in beach sand all over the world. The zirconium to hafnium ratio in zircon is about 50/1 but since Hf has a very large thermal neutron cross section, it is crucial that as much Hf as possible is separated from zirconium during the manufacturing process.

The alloy homogeneity and chemical impurity concentrations of the final products is mainly established in the very first steps, starting from the zirconium sand, i.e. hafnium separation, ZrO_2 reduction, scrap recycling and ingot melting. Only the gas contents can be influenced in later processes.

The three commercial processes that are used to produce Zr metal are described in the following subsections; i.e., the Kroll process (used by most Zr manufacturers), the Van Arkel process and the electrolytic process.

7.2 Process

7.2.1 Kroll process

The first step is to convert zircon into ZrCl₄, through a carbo-chlorination process performed in a fluidised bed at 1200 °C, (Figure 7-1).

After the Zr/Hf separation process, Zr metal is obtained by Zr-reduction in which gaseous ZrCl₄ is reduced by liquid magnesium at 850 °C in an oxygen-free environment (Kroll process). Any Mg remnants are subsequently removed from the sponge cake by distillation at 1000 °C. The sponge cake is then fractured and the pieces are carefully inspected and contaminated pieces showing up as discoloration are discarded. *The sponge impurity concentrations of Cl and Mg are determined by this process step*.



Figure 7-1: Zr sponge manufacturing.

The appropriate American Society for Testing and Materials (ASTM) specification for nuclear grade zirconium sponge is B349.

7.2.2 Van Arkel process

High purity Zr can be obtained by the Van Arkel process which consists of reaction of Zr with iodine at moderate temperature, gaseous phase transport as ZrI4 and decomposition of the iodide at high temperature on an electrically heated filament, Figure 7-2 and Figure 7-3. This process is considerably more expensive than either the electrolytic or the Kroll process but produces zirconium of high purity, as can be seen in Table 8-1.

8 Ingot melting process

8.1 Process

This is the first stage where zirconium appears as an alloy. The scrap has already been recycled. The chemical composition will not be changed significantly by further fabrication steps, except for a possible gas (H, O, N) uptake. An outline of the ingot manufacturing process is shown in Figure 8-1.



Figure 8-1: General process flow diagram for Ingot.

Briquettes of zirconium sponge recycle material from earlier manufacturing, and alloying elements blended with sponge are put together into an electrode in a specific sequence distributed along the length of the electrode. The alloying elements are added as compounds with a similar melting temperature as that of zirconium for homogeneity purposes. Normally, sponge-based briquettes containing the alloying compounds and solid recycle material are assembled and welded either by electron beam in vacuum or plasma arc welding in argon atmosphere into an ingot, Figure 8-2.



Figure 8-2: Consumable electrode showing sponge compacts and solid recycle sections [Schemel, 1989].

The subsequent melting is then normally done in vacuum to prevent oxygen and nitrogen pickup from the air. The consumable electrode is melted into a water-cooled copper mold slightly larger than the ingot diameter, Figure 8-3. As the arc melts the metal a fraction is solidified and forms a solid layer of the zirconium alloy in between the mold and the molten metal. Only a small fraction of the whole ingot is molten at any time during the continuous consumption of the electrode. Larger molten pool depth and volume increases homogeneity of the solidified ingot since diffusion and convection will take place during longer time before the melt has solidified. Larger amperage results in a large pool size. If the cooling of the mold is not done properly, copper may contaminate the ingot.

To increase the size and homogeneity of the ingot, the zirconium alloy electrode is melted three or four times. Ingots are melted four times to remove F in electrolytic material (to reduce high temperature oxidation) or Cl in Kroll material for CANDU pressure tubes (to increase fracture toughness.)

After each melting, the resulting ingot is cut up in cylindrical pieces and these pieces are put together and welded (EB welding in vacuum) in a different sequence compared to that of the previous electrode.



Figure 8-3: Schematics showing Consumable Vacuum Arc Melting.

8.2 Ingot quality

Process parameters that influence the characteristics and quality of the ingot are:

- 1) Quality of the raw material.
- 2) Molten pool diameter and depth which are functions of amperage and speed of melting.
- 3) The stirring of the melt by a magnetic flux during the melting process
- 4) Number or remelts of the ingot, an increased number of remelts tends to increase the homogeneity in the material and reduce impurities with low vapour pressures. Most manufacturers are using three remelts (triple melting).
- 5) Sequence of order of cut up cylindrical pieces from ingot from previous melting also contributes to ingot homogeneity. Ingot alloying element inhomogeneities within an ingot result in variation in properties of the final products.
- 6) Electrode welding and copper mould atmosphere composition.

The homogeneity of the alloying elements and impurities is permanently established by a good quality melting process. Changes in homogeneity cannot be made by subsequent processing except for the gaseous impurities. Inhomogeneities in the ingot will cause variability in properties among components fabricated from that ingot, and could result in some deviate compositions that would affect performance. Hence it is vital that the ingot melting proceed by a well-qualified procedure and that changes in the procedure be re-qualified.

8.3 Impact of ingot melting on in-pile performance

8.3.1 Microstructure

The alloying elements and impurities of modern zirconium alloys, such as Sn, Nb, Fe, Cr, Ni, O, Si, C are either fully dissolved in the alloy matrix (Sn, O) or dissolved to a large extent (Nb) or almost fully precipitated in small intermetallic second phase particles (Fe, Cr, Ni, Si, C), Second Phase Particle (SPPs).

In-reactor dissolution of SPPs - At normal Light Water Reactor (LWR) temperatures (270-370 °C (543-643 K)) the SPPs change under irradiation in a combination of two ways – amorphization and dissolution.

Amorphization means that the original SPP crystalline structure is converted to an amorphous structure. A critical temperature exists above which the annealing processes are fast enough to prevent the accumulation of defects needed to transform crystalline SPPs to amorphous structures. For low temperature irradiations, amorphization of both $Zr(Fe,Cr)_2$ and $Zr_2(Fe,Ni)$ in Zry-2 occurs readily at temperatures near 100 °C (373 K). For the temperatures (300 °C (573 K)) and neutron flux encountered in a LWR, $Zr(Fe,Cr)_2$ becomes amorphous but $Zr_2(Fe,Ni)$ does not. Above about 330 °C (603 K), neither type of SPP becomes amorphous.

The fluence required to produce complete amorphization depends on neutron flux, temperature and SPP size and chemistry. But, for typical $Zr(Fe,Cr)_2$ SPPs of initial size near 0.1 µm, the entire SPP is amorphous by end of bundle life burnups, <50 MWd/KgU ($1x10^{26}$ n/m², E>1 MeV). *The rate of dissolution depends on the SPP size, with higher rates for smaller sizes. The extent of dissolution depends on size and fluence.* It has been demonstrated in a BWR that small (<0.04 µm) SPPs in Zry-2 can completely dissolve at low to moderate burnups. Also in a PWR at temperatures near 290 °C, SPPs in Zry-4 with an average size of 0.2 µm were >80% dissolved at moderate burnup (1×10^{26} n/m², E > 1MeV). *The consequence of the complete dissolution of the SPPs in Zry-2 and Zry-4 is a dramatic increase in the Hydrogen Pick-Up Fraction (HPUF) and, after some time, also an increase of the corrosion rate in BWRs.* It must be noted, however, that nodular corrosion in BWRs can be adversely affected by the presence of large SPPs at the outer cladding surface and that cladding performance depends on the composition and size of the precipitates across the wall thickness.

For the Zr-Nb type alloys preferentially used in PWRs neither the β Nb nor Zr (Nb, Fe)₂ SPPs become amorphous (or dissolve) for irradiation temperature >330 °C (603 K).

SPP amorphization in itself does not appear to affect material behaviour; however, dissolution of both amorphous and crystalline SPPs does influence corrosion and growth.

8.3.2 Effects of Alloying on LOCA Breakaway Oxidation³⁴





Figure 8-4: Ring Compression Tests (20 °C) [Malgin et al., 2018].

³⁴ *Breakaway oxidation* means, for the purposes of this section, the fuel cladding oxidation phenomenon in which weight gain rate deviates from normal kinetics. This change occurs with a rapid increase of hydrogen pickup during prolonged exposure to a high temperature steam environment, which promotes loss of cladding ductility.

[Malgin et al., 2016] studied the effect of Nb, Sn and (Fe) on high temperature oxidation/oxidation breakaway and ductility of different Zr-Nb and Zr-Nb-Sn-Fe alloys with the following results, Figure 8-5 and Figure 8-6:

- For Zr-Nb alloys the increase of the Nb concentration has no significant effect on oxidation kinetics.
- Zr-1.0Nb alloy has the lowest high temperature oxidation rate in the temperature range 1000-1200 °C of all tested alloys.



Figure 8-5: High temperature oxidation at 1000 °C, Arrow indicate onset of breakaway oxidation for Zr1Nb1.08Sn0.33Fe while the star symbol indicates sample showing increased hydrogen pickup indicative of breakaway oxidation, see Figure 8-7 [Malgin et al., 2018].



Figure 8-6: Oxidation kinetics at 1100 °C and 1200 °C [Malgin et al., 2018].

For Zr-1Nb- ySn-zFe alloys increasing tin concentration reduces the time to breakaway oxidation at 1000 °C onset leads to a decrease in duration of normal oxidation until the development of breakaway (see Figure 8-7).

9 Cladding tube (fuel rods, including liner) guide tube manufacturing

9.1 Tubing production (cladding, guide tubes, water rods)

9.1.1 Introduction

The tube shell is then subjected to a number of pilgering steps with intermediate anneals to restore the material ductility, see Figure 9-1.





After each cold working step an intermediate anneal must be performed to restore the material ductility.

Poor cleanliness of the tube hollow may result in surface contamination deteriorating its workability during subsequent pilgering steps and/or deteriorating the corrosion resistance of the final cladding tube. Insufficient removal of the oxide scale may result in generation of surface defects on the tube hollow that may reduce PCI performance of the final fuel cladding tube.

Flaws in tube hollow may reduce PCMI margins. Tube hollow dimensional deviations may result in tubes that will not meet the final tube dimensional requirements, e.g. ovality and wall thickness variation of final tube may degrade PCMI performance.

Temperature-time variability during all tube hollow heat treatments will result in similar variability of SPP microstructure (for Zircaloys and Zr-Nb alloys), resulting in variability of corrosion resistance of the final tubes produced from different tube hollows. Specifically for Zr-Nb alloys, if the temperature exceeds the monotectoid temperature, the corrosion resistance may be deteriorated due to β -Zr remnants in the final cladding tube.

Late beta-quench: overheating at OD may result in too large beta grains resulting in parallel plate structure with poor mechanical properties. Thus product may form cracks during subsequent manufacturing that may end up in the final product. OD overheating and too slow quenching rate may also result in too large SPPs resulting in poor in-pile corrosion properties. Underheating may cause insufficient homogenization; e.g., the formation of "stringers" that may result in significant variation in mechanical and corrosion properties of the final product. Significant variation of betaquenching process parameter values (within specification) may result in variation in SPP characteristics at the outer surface of different tube shells causing variation in corrosion properties between the tubes form the different tube shells.

Too large tendency for radial hydrides may reduce PCMI performance of fuel claddings.

9.1.2 Pilgering

9.1.2.1 General

The further processing of the extrusion to cladding proceeds in several steps by cold pilgering, also called rocking or tube reducing. Each pass through the pilger machine can accomplish up to 85% reduction in cross-sectional area. In most cases, three to four reduction passes are required to reduce the extrusion to the desired fuel cladding size. The degree of cold deformation of the final pass can be important for the texture, final grain size and, as a result, the in-reactor behaviour of the cladding.

After each reduction pass, the tubes must be cleaned and vacuum annealed, usually at 580-750 °C, to prepare it for the next cold reduction pass. The annealing treatments promote recrystallisation and soften the hollow. The time and temperatures of these anneals have been found to play an important role in developing optimum in-reactor behaviour. The ratio between wall reduction and diameter decrease during the pilger steps (especially the last one) affect the texture of the final tubing. Tube reduction by preferential wall reduction results in a sharp radial texture of basal poles, with rather high Kearns-factors⁴¹ in radial direction (0.6-0.8), whereas preferential reduction of the clad diameter results in a more tangential texture. At the usual degrees of cold deformation almost no basal poles are directed in the axial direction. The Kearns-factors in the axial direction is ≤ 0.1 .

In this cold reduction process, the tube elongates over a tapered stationary mandrel by means of two grooved rolls, or dies, which roll back and forth, (Figure 9-2) to (Figure 9-4). The roll groove is also tapered. The in-going tube is rotated and advanced by a small increment before each compressive pass of the rolls. The tube diameter and wall thickness are continuously reduced in a series of these small increments.

⁴¹ The Kearns-factor gives an indication of the fraction of grains with basal poles parallel to a specific direction.



Figure 9-2: Cold pilgering system with: the mandrel (tool inside the tube) and the dies (external tools with semicircular grooves), [Girard et al., 2001].



Figure 9-3: Illustration of tube movement during reduction stroke to determine initial and final tube dimensions at a location of interest. "F" designates the feed rate per strike and "CE" the cumulative elongation that has taken place in the stroke, [Schemel & McKenzie, 1974].

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The objective of the pilgering operation is reducing the diameter and wall thickness of the tubing while generating a texture that is favourable to in-core operation.

Process parameters that influence tube hollow quality include:

- *Quality of dies and mandrel.*
- *Type of lubricant and its cleanliness.*
- Degree of reduction as a function of tube wall thickness/diameter.

9.1.2.2 Texture

9.1.2.2.1 Microstructure

Plastic deformation in zirconium alloys occurs both by slip and twinning. Because Zr has a nonsymmetrical HCP structure, the resistance to deformation is different in different directions. [This is unlike alloys such as steel or Inconel that have symmetrical cubic crystal structures and deform equally in many directions.] Slip occurs in the $a < 11\overline{2}0 >$ direction on the prism and pyramidal planes as shown in Figure 9-5. The combination of twinning and slip leads to a strong preferred orientation of the crystals during cold working. Most of the deformation in cold pilgering is by slip, however twinning is the process that rotates the crystals from one orientation to another.



Figure 9-5: Crystallographic characterizations of the hcp elementary cell, after [Tenckhoff, 2005].

The texture that develops during rocking or drawing of Zr-alloy cladding tubes depends on the Q-value, i.e., the ratio of wall reduction to diameter reduction, which is given by:

Equation 9-1:

-		$Q = ln(W/W_0)/ln(D/D_0)$
where	W	is the final wall thickness,
	Wo	is the initial wall thickness,
	D	is the final diameter,
	$D_{ m o}$	is the initial diameter.

High *Q* values for a reduction pass tend to rotate the crystals so that their basal poles are closer to the radial direction. Reductions with corresponding values less than one tends to produce crystal orientations closer to the circumferential direction, (Figure 9-6).



Figure 9-6: Illustration of the effect of the Q ratio on texture development during pilgering, [Schemel, 1989].

The texture of cladding tubes that develops can be described by the texture factors f_r , f_t , and f_a , the resolved basal pole fractions in the radial, tangential, and axial directions, respectively, which sum to 1 ($f_r + f_t + f_a = 1$).

Usually f_a is ≤ 0.06 while f_r is $> f_t$ for cladding tubes. [Nagai et al., 1982a] found the following correlations between the texture factors and the *Q* value for stress relieved and RX Zry:

Equation 9-2:

$$f_t(as \ rolled \ or \ SR) = 0.078 \ ln(Q) + 0.5$$

Equation 9-3:

$$f_t(RX) = 0.05 \ln(Q) + 0.54$$

Recrystallisation annealing does not change the basal pole density distribution significantly while the prism pole distribution is changed as can be seen from Figure 9-7. Typical texture factors for different products are summarized in Table 9-1. It is somewhat surprising that E110 which is fabricated by a tube rolling procedure, characterised by a very high Q value does not have a more radial texture than that of pilgered recrystallised Zry-4.

Material	fr	ft	fa	Reference
CANDU Zr2.5Nb Pressure tubes	0.29-0.37	0.58-0.66	0.04-0.09	[Holt et al., 2000]
Zry cladding tubes	0.6-0.8	0.2-0.4	0.05	[Garzarolli et al., 1996c]
Zry-4 cladding tubes, SR	0.57	0.33	0.1	[Delobelle et al., 1996]
Zry-4 cladding tubes, RX	0.56	0.33	0.11	[Delobelle et al., 1996]
Zry-2, tubes, 80%CW, PR	0.64	0.31	0.08	[Perkins & Shann, 1994]
Zry-2, tubes, late β -quenched, 80%CW, PR	0.61	0.37	0.08	[Perkins & Shann, 1994]
E110 and E635 tubes, RX	0.57	0.33	0.10	[Shishov et al., 2005]
E110 and E635 sheets, RX	0.63	0.29	0.08	[Shishov et al., 2005]



Table 9-1:Typical texture parameters of fuel cladding tubes and strips.



10 Spacer strips, Channel sheet manufacturing

The first plastic deformation steps are accomplished by hot forging in the β phase, $\alpha + \beta$ phase and/or in the upper α phase. The ingots are heated to about 1100 °C and the forging is performed in a number of steps bringing down the diameter or thickness of the work-piece. Before each forging step, the work-piece needs to be reheated several times to increase its plasticity. It is generally desirable to minimize deformation in the $\alpha + \beta$ region. At high temperature, the oxide layer that develops during forging is not protective and consequently elements such as O, N and H from the air can be absorbed by the bulk of the ingot if no precautions, such as limiting the time at high temperature in unprotected atmosphere, are taken.

For sheet and strip the ingot is forged into a rectangular slab, and after it reaches a certain thickness it goes to hot rolling, beta-quenching and then cold rolling Figure 10-1. For spacer strip it is continuous cold rolling. The cold deformation of the individual cold rolling passes is lower than usually applied in pilgering. If continuous rolling and annealing is used the times at temperature after each cold deformation step are rather low (only a few minutes).

Throughout the manufacturing of the different products, the material is kept in lots. A lot is defined in all specifications as those final products made by the same process from a single ingot and given the final heat treatment in a single furnace load.

Most sheet materials are in *RXA* condition to get optimum ductility to allow plastic deformation of the sheet into the final product configuration such as fuel channels and spacer grids without any risk of cracking during subsequent forming operations.



Figure 10-1: General process flow diagram for Strip/Sheet fabrication (References are provided to relevant section in the tube manufacturing section).

11 Effects of LWR fuel design and fabrication on fuel assembly component performance during dry storage and transportation

Figure 6-1 lists important cladding material properties and attributes (such as creep, oxidation susceptibility, etc.), which depend on one hand on unirradiated material characteristics (such as chemical composition, texture, etc.) and alloy fabrication process parameters (such as melting, pilgering, etc.) and on the other hand on in-reactor operational parameters (such as neutron fluence, cladding temperature, etc.).

Acceptable values of these cladding material properties and attributes are eventually pre-determined in order to meet specific performance criteria resulting from mandated safety, investment-protection, and economic considerations during reactor operation. The overarching goal in fuel design is the optimum performance of the nuclear fuel during reactor operations. It is up to spent fuel management to develop means to safely handle the discharged fuel, typically (1) by preventing or containing potential releases of radioactive material from fuel rods, and (2) by preserving or minimizing changes in the mechanical configuration of the fuel assembly. For example, the recommended maximum thickness of the ZrO₂ layer on the outside of a fuel rod cladding and the maximum hydrogen concentration in the cladding can be specified as 100 μ m and 600 wppm, respectively, for PWR rods. These limits were derived from considerations about the in-reactor performance of the fuel rods, independently of their expected performance during back-end operations. The latter will have to properly take into account a conservative range of actual oxide layers and hydrogen concentrations measured on spent SNF rods; actual values to take into consideration may turn out to be similar, lower, or higher than the recommended limits for in-reactor duty.

As a result, a discussion of the effects of fuel design and fabrication steps on fuel assembly component performance is mostly limited to:

- Fuel rod design, such as the presence of an inner liner in BWR fuel
- Specific microstructural features resulting from cold-worked, stress-relieved (CWSR) vs. recrystallized (RX) annealing.

A parameter, such as texture, for example, will not be discussed in great detail in the context of backend operations because its range of desirable values is dictated by in-reactor fuel performance. Given that the chosen process for fabricating PWR cladding tubes result in a fairly uniform cladding texture in essentially all commercial PWR rods, texture is a given characteristic of discharged PWR fuel, which is not susceptible to be even considered in fuel assembly design optimization for some purpose related to back-end operations.

The dependency of only two of the potential cladding degradation phenomena (see Section 5.3 of this report) on cladding composition and condition (microstructure, hydrogen concentration, neutron fluence) will be discussed in some detail; they are thermal creep and hydride reorientation. The impact of fuel rod designs containing (1) an inner liner in most BWR designs or (2) an outer layer in some PWR designs will also be discussed given their ability to suppress hydride reorientation as a potential cladding degradation mechanism. Three degradation phenomena (SCC, DHC, and hydrogen migration) will not be addressed for the reasons stated in Section 5.3.

11.1 Cladding temperature during dry storage

The temperature of fuel rods is not uniform in a storage or transportation system, as illustrated in selected axial (along the length of the rod) and radial (across circular sections of the cask) temperature profiles shown in Figure 11-1. When heat transfer inside the cask occurs mostly by radiation and conduction by the helium gas, the highest temperature is usually experienced in cladding sections located toward the center and mid-section of the cask; this is visible in the profiles measured in the cask in the horizontal position (Figure 11-1), for which heat transport is limited to conduction and radiation. When convection of He is also present, the highest temperatures are displaced toward the higher sections in the cask, as shown by the profiles measured in the cask in the vertical position

ZR ALLOY MANUFACTURING AND EFFECTS ON IN-REACTOR, DESIGN BASIS ACCIDENT AND INTERIM DRY STORAGE PERFORMANCE

(Figure 11-1), in which some amount of convective heat transfer is usually present. In large casks, cladding temperatures vary locally (axially and radially) and the differences in temperature can be very significant. The highest temperature reached by cladding is referred to as the peak cladding temperature, or PCT. Although only a small percentage of the cladding, a few percent at most, reaches or gets close to the PCT, it is an important parameter for assessing the mechanisms that may affect the performance of spent fuel assemblies in dry storage. High enough temperatures are conducive to activate several potential cladding degradation mechanisms such as thermal creep and hydride reorientation. *In fact, given a class of SNF with known properties and attributes, specifying the PCT is the only environmental parameter available to a dry storage system designer*. As a result, the performance of cladding heated to the PCT is one of the main technical regulatory focus, even though the cladding that experiences a temperature within 10 °C of the PCT represents only a small fraction of the total cladding surface area.



Figure 11-1: Measured axial (top) and radial (bottom) temperature profiles in a He-filled CASTOR V/21 storage cask standing in the vertical and horizontal positions [EPRI report NP-4887].

In summary, cladding temperatures vary significantly in a cask. The peak cladding temperature affects only a very small cladding fraction, but is important in assessing the most deleterious effects of the degradation mechanisms that are very sensitive to temperature. It should be noted that high temperatures can also have beneficial effects, for examples, by preventing brittle fracture in irradiated claddings and preventing hydride precipitation at a crack tip (and thus preventing DHC by suppressing crack propagation).

11.2 Thermal creep

The specific cladding variable that determines the possibility of rupture by thermal creep is the extent of deformation, or creep strain, of the cladding material, which depends on time (Figure 5-21), alloy composition, alloy condition (microstructure), hydrogen content (due to zirconium oxidation and hydrogen pickup), and neutron fluence.

11.2.1 Impact of alloy composition and microstructure

From the estimated relative creep behaviour (creep strain), given in Table 11-1 and Table 11-2, it can be concluded that most commercially used advanced claddings have less strain during dry storage than standard CWSR Zircaloy-4 claddings.

Improvements in creep resistance are especially noticeable for RX Zircaloy-2 and RX M5. The creep strain of the new alternate alloys is also expected to be significantly less than CWSR Zircaloy-4 due to the final fully (RX) or near-fully recrystallization (pRX) condition for these alloys.

The impact of alloy additives, tin (Sn), niobium (Nb), and oxygen (O), can be assessed using the SNO parameter discussed in Section 5.3.2.2. Finally, the impact of some impurities, such as sulphur, can be seen in Figure 8-51 (Section 8.3.3.10.1).

Alloy	Sn (%)	Nb (%)	Fe (%)	Cr (%)	Others	Condition	SNO Parameter*	Relative Creep Strain ^{**}
Best Zry-4	1.3		0.22	0.1	1200 ppm O	CWSR	2.08	1
Best Zry-2	1.3		0.22	0.1	1200 ppm O	RX	2.08	<0.54
E110		1	0.01		600 ppm O ₂ , F- impurity	RX	1.36	0.94
ZIRLO	1	1	0.1		1200 ppm O ₂	CWSR	2.72	<0.77
DX-3b	0.8		0.5		1200 ppm O ₂	CWSR	2.1	1
DX-D4	0.5		0.5	0.2	1200 ppm O ₂	CWSR	2.1	1
M5		1	0.04		1200 ppm O, 20 ppm S	RX	1.72	<0.71
MDA	0.8	0.5	0.2	0.1	1200 ppm O ₂	CWSR	2.52	>0.94
NDA	1	0.1	0.3	0.2	1200 ppm O ₂	SCWR	1.92	>1.1
Optim. ZIRLO	0.67	1	0.1		1200 ppm O ₂	pRX	2.39	<0.8
E110-Sponge		1	0.04		800 ppm O ₂ ,	pRX	1.48	<0.86
* Refer to Section 5.3.2.2 for a definition of the SNO parameter ** Estimates based on [Adamson et al. 2009a]								

 Table 11-1:
 Relative creep behaviour of commercially used advanced Zr Alloys.

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Alloy	Sn (%)	Nb (%)	Fe (%)	Cr (%)	Others	Condition	SNO Parameter*	Relative Creep strain**
Best Zry-4	1.3		0.22	0.1	1200 ppm O	CWSR	2.08	1
Best Zry-2	1.3		0.22	0.1	1200 ppm O	RX	2.08	<0.54
M-MDA	0.5	0.5	0.4	0.3		CWSR	2.22	>1.18
S2	0.8	0.1	0.3	0.1		RX	1.72	<0.71
J1		1.8				RX	1.72	<0.71
J2		1.6		0.1		RX	1.72	<0.71
J3		2.5				RX	1.72	<0.71
Quart.ZrNbSnFe	0.5	1	0.1			RX	2.22	<0.49
Quart.ZrNbSnFe	0.3	1	0.1			RX	2.02	<0.57
Quart.ZrNbSnFe	0.3	1	0.2			RX	2.02	<0.57
Quart.ZrNbFe		1	0.1			RX	1.72	<0.71
AXIOM-X1	0.3	0.8	0.05		0.12% Cu, 0.2%V	p(80%)PR	2.02	<0.6
AXIOM-X2		1	0.06			RX	1.72	<0.71
AXIOM-X4		1	0.06	0.25	0.08% Cu	p(80%)PR	1.72	<0.75
AXIOM-X5	0.3	0.7	0.35	0.25		P(50%)PR	2.02	<0.6
AXIOM-X5A	0.4 5	0.3	0.35	0.25	0.05% Ni	P(50%)PR	1.77	<0.64
HANA-4	0.4	1.5	0.2	0.1		pRX	2.12	
HANA-6		1			0.05%Cu	pRX	1.72	
* Refer to Section 5.3.2.2 for a definition of the SNO parameter ** Estimates based on [Adamson et al., 2009a]								

 Table 11-2:
 Relative creep behaviour of new alternate Zr Alloys.

11.2.2 Impact of hydrogen concentration

In unirradiated RX Zircaloy-2 cladding tubes specimens, hydrogen results in an increase in creep rate as long as the hydrogen concentration remains below the hydrogen solubility limit (Figure 11-2) and in a decrease in creep rate when the hydrogen concentration becomes larger than the solubility limit, i.e., when zirconium hydrides are present (Figure 11-3). For unirradiated CWSR Zircaloy-4, increasing hydrogen concentration always results in a decrease in creep rates (Figure 11-4).

11.2.3 Impact of neutron irradiation

As previously noted in Section 5.3.2.2, creep rates and creep strains of irradiated cladding are, for all tested temperature conditions and stress regions, smaller than those of unirradiated cladding. Figure 11-5 and Figure 11-6 illustrate the impact of neutron fluence on creep rates of irradiated CWSR Zircaloy-4 and RX Zircaloy-2 compared to the same materials in the unirradiated condition.

12 CANDU pressure tube manufacturing and effects on in-pile performance

12.1 Fabrication of Zr-2.5Nb pressure tubes (Christopher Coleman)

The fabrication of Zr-2.5Nb pressure tubes (PTs) involves the following basic processing steps:

- 1) melt ingot,
- 2) hot work to bar,
- 3) machine or pierce hollow billets,
- 4) pre-heat billets and extrude into tubes,
- 5) cold-work to size, and
- 6) stress relief.

Several variations are used for both pressurized heavy water reactors (PHWR, (CANDU-type)) and graphite-moderated light water reactors (mostly RBMK).

In Canadian PTs the ingot is melted four times to attain a small concentration of Cl, which is responsible for lowering crack growth resistance. Press and rotary forging follows, to break down the as-cast microstructure and prepare the bar to fit the extrusion press. A hollow is machined from the bar that is quenched into cold water from the β -phase. The hollow is then clad in Cu, heated into the $(\alpha+\beta)$ -region, usually about 815 °C for about 15 minutes, then extruded with a ratio of 10:1. The extrusion is cold-worked, either drawing or pilgering, in two steps for a total strain of 27%, followed by stress-relieving at 400 °C for 24 h; to be indicated as CW-SR. The resulting microstructure consists of elongated hcp α -Zr grains containing a high dislocation density, and a thin layer of bcc β -phase at the α -grain boundaries (Figure 12-1). The crystallographic texture consists of basal plane normals parallel with the transverse direction, fewer in the radial direction and rarely in the longitudinal direction. This microstructure is developed during extrusion and the texture is slightly sharpened and the dislocation density increased with cold work.



Figure 12-1: Elongated grain structure in Zr-2.5Nb pressure tube material [Cheadle, 2010].



Figure 12-5: Microstructure of RBMK Zr-2.5Nb pressure tube in TMT-2 condition [Coleman et al., 2008].

	CANDU		RBMK	
	CWSR	CW-A	TMT-1	TMT-2
Oxygen concentration (ppm)	1370	550	550	550
fτ	0.57	0.42	0.39	0.52
Dislocation density (m ⁻² × 10 ¹⁴)	3.1	1.0	1.7	1.0
Grain size (μm)	0.5	5.0	3.0	3.0
Yield Strength (MPa)	807	483	570	464
	•	•	© A	NT International 2015

 Table 12-1:
 Four attributes that can affect tensile strength of Zr-2.5Nb pressure tubes.

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Nomenclature

ABB	ASEA Brown Boveri
ABSS	AB Sandvik Steel
AEC	Atomic Energy Commission
AECL	Atomic Energy of Canada, Ltd.
ANL	Argonne National Laboratory
ANT	Advanced Nuclear Technology
AOA	Axial Offset Anomaly
AOO	Anticipated Operational Occurrences
AR	Annual Report
AREVA	French nuclear reactor and fuel vendor
ASME	American Society of Mechanical Engineers
ASTM	American Society for Testing and Materials
ATR	Advanced Test Reactor
ATUCHA	Nuclear reactor in Argentina
AXIOM	Westinghouse alloy
BCC	Body Centered Cubic
Bct	body centered tetragonal
B-J	Baker-Just
BNFL	British Nuclear Fuel Limited
BOR	Research Fast Reactor
BQ	Beta-Quenched
BW	Basket-Weave
B&W	Babcock and Wilcox
BWR	Boiling Water Reactor
CABRI REP	RIA test performed in CABRI reactor
CANDU	Canadian Deuterium Uranium
CE	Combustion Engineering
CEZUS	Companie Europen Ziconium Ugine Sandvik
CILC	CRUD Induced Localized Corrosion
CR	Control Rod
CRUD	Chalk River Unidentified Deposit
CW	Cold Worked
CWSR	Cold-Worked Stress-Relieved
CWSRA	Cold Work and Stress Relieved Annealed
DAE	Department of Atomic Energy (India)
DBA	Design Basis Accidents
DBT	Ductile-Brittle Transition
DFM	Diffusion First Model

DHC	Delayed Hydride Cracking
DNB	Departure from Nucleate Boiling
DUPLEX	A fuel cladding with an outer thin corrosion resistant layer and a core with another creep resistant alloy
DX	Short for DUPLEX
EB	Electron Beam
ECBE	Effective Control Blade Exposure parameter
ECCS	Emergency Core Cooling Systems
ECR	Equivalent Clad Reacted
EFPD	Effective Full Power Days
ELS	Extra-Low Sn
ESSC	Enhanced Spacer Shadow Corrosion
FA	Fuel Assembly
FBR	Fast Breeder Reactor
FCC	Face Centered Cubic
FDBQ	Final Dimension &-Quenched
FDRH	Fuel Design Review Handbook
FFPH	Fuel Fabrication Process Handbook
FGR	Fission Gas Release
FMTR	Fuel Material Technology Report
GC	Guide Channels
GCR	Gas Cooled Reactor
GE	General Electric
GNF	Global Nuclear Fuel
GT	Guide Thimbles or Tubes
HCP	Hexagonal Close-Packed
HiFi	Zry-2 type of alloy with high Fe content
HPA	High Performance Alloy
HPU	Hydrogen Pickup
HPUF	Hydrogen Pick-Up Fraction
HT	High Temperature
HTGR	High-Temperature Gas-Cooled Reactor
HTP	High Temperature
HWC	Hydrogen Water Chemistry
IAEA	International Atomic Energy Agency
IASCC	Irradiation Assisted Stress Corrosion Cracking
ID	Inner Diameter
IGSCC	Intergranular Stress Corrosion Cracking
IPHT	In Process Heat Treated
IRI	Incomplete Rod Insertion

Unit conversion

TEMPERATURE			
°C + 273.15 =	K °C ×	1.8 + 32 = °F	
T(K)	T(°C)	T(°F)	
273	0	32	
289	16	61	
298	25	77	
373	100	212	
473	200	392	
573	300	572	
633	360	680	
673	400	752	
773	500	932	
783	510	950	
793	520	968	
823	550	1022	
833	560	1040	
873	600	1112	
878	605	1121	
893	620	1148	
923	650	1202	
973	700	1292	
1023	750	1382	
1053	780	1436	
1073	800	1472	
1136	863	1585	
1143	870	1598	
1173	900	1652	
1273	1000	1832	
1343	1070	1958	
1478	1204	2200	

	Radioactivity
1 Sv	= 100 Rem
1 Ci	= 3.7 × 10 ¹⁰ Bq = 37 GBq
1 Bq	= 1 s ⁻¹

N	IASS
kg	lbs
0.454	1
1	2.20

DIST	ANCE
x (µm)	x (mils)
0.6	0.02
1	0.04
5	0.20
10	0.39
20	0.79
25	0.98
25.4	1.00
100	3.94

PRESSURE				
bar	MPa	psi		
1	0.1	14		
10	1	142		
70	7	995		
70.4	7.04	1000		
100	10	1421		
130	13	1847		
155	15.5	2203		
704	70.4	10000		
1000	100	14211		

STRESS INTENSITY FACTOR		
MPa√m	ksi√inch	
0.91	1	
1	1.10	