

Performance evaluation of new advanced Zr alloys for BWRs and PWRs/VVERs Vol II

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A handwritten signature in black ink, appearing to read 'Peter Rudling', with a stylized flourish at the end.

Mr Peter Rudling, President of ANT International

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1 Introduction

To meet the current situation with more aggressive reactor environments (higher burnups, changing water chemistries and loading patterns), and resolving fuel performance issues such as BWR channel bowing and PWR assembly bowing, a large number of zirconium alloys have been and are being developed. The main driver for the initial material development in Pressurized Water Reactors (PWRs) has been to reduce corrosion rates and Hydrogen Pick-Up Fractions (HPUFs), which have occasionally limited the maximum discharged burnup.

However, to ensure that the new Zirconium Alloys performs satisfactorily during normal operation, Anticipated Operational Occurrences (AOOs), postulated accidents and intermediate dry storage, it is crucial to assess the projected performance of components of the new zirconium alloy materials and relate the performance to the material characteristics. This assessment is the objective of this Special Topic Report (STR).

This ZIRAT22 STR is an update and expansion of the ZIRAT16 Report since the ZIRAT22 STR also includes BWR material development. However, since the amount of information is so large, this STR is divided into two separate reports, Vol. I providing the basic information to the detailed material performance in Vol. II (this report). The advantage of this structure is that staff familiar with the basics can only focus on reading Vol. II while staff with less experience can first start to digest the basics in Vol. I [Rudling, 2017] before reading Vol. II

This Report structure is as follows:

- Section 2 provides background information on Zr alloy developments for fuel rods and structural components in BWRs, PWRs/VVERs and CANDUs. The Section covers information related to normal operation, AOO, design basis accidents as well as interim dry storage.
- Section 3 tries to identify the best materials for fuel rods and for structural components in different reactors.

The following topics are covered in Vol. I:

- Section 2 provides background information on water reactors and their fuel,
- Section 3 discusses basics of irradiation and impact on water and materials,
- Section 4 gives a background to reactor safety and reviews design criteria related to the Zr alloy components (this means that e.g. fuel melting is not covered here),
- Section 5 presents in-reactor Zr alloy performance during normal operation and anticipated operational occurrences
- Section 6 and 7 contains information about design basis accidents and interim dry storage, respectively

More detailed information of the various topics covered in this report are listed below:

Corrosion and hydriding topics:

- Effects of Coolant Chemistry on Fuel Performance, LCC9 [Rudling et al., 2013]
- LCC7 and 8 STR on Introduction to Boiling Water Reactor Chemistry Volume I [Cowan et al., 2011] and Volume II [Cowan et al., 2012]
- LCC7 and 8 STR on PWR/VVER Primary Side Coolant Chemistry Volume I [Riess et al., 2011] and Volume II [Riess et al., 2012]
- LCC6 STR on Effect of Zink in BWR and PWR/VVER on Activity Build-up, IGSCC and Fuel Performance [Odar et al., 2010]
- ZIRAT12/IZNA7 STR on Corrosion Mechanisms in Zirconium Alloys [Adamson R. et al., 2007]
- LCC2 STR on CRUD in PWR/VVER and BWR Primary Circuits [Riess & Lundgren, 2006]

- ZIRAT9/IZNA4 STR on Corrosion of Zr-Nb Alloys in PWRs [Cox et al., 2004].
- ZIRAT8/IZNA3 STR on The Effects of Zn Injection (PWRs and BWRs) and Noble Metal Chemistry (BWRs) on Fuel Performance [Cox et al., 2003/2004]
- ZIRAT7/IZNA2 STR on Corrosion of Zirconium Alloys [Adamson et al., 2002/2003]
- The Effects of Zn Injection (PWRs and BWRs) and Noble Metal Chemistry (BWRs) on Fuel Performance, ZIRAT8/IZNA3 [Cox et al., 2003/2004].
- ZIRAT6/IZNA1 STR on CRUD ZIRAT6/IZNA1 STR on Water Chemistry and CRUD Influence on Cladding Corrosion [Wikmark & Cox, 2001/2002]
- Hydriding Mechanisms and Impact on Fuel Performance, ZIRAT5/IZNA1 [Cox & Rudling, 2000].
- Section 6 in ZIRAT/IZNA Annual Reports (ZIRAT5 and onward)
- PWR ZR Alloy Cladding Water Side Corrosion (PZAC) [Garzarolli & Garzarolli, 2012]

Thermal and mechanical property topics:

- Processes Going on in Nonfailed Rod During Normal Operation, ZIRAT15/IZNA10 [Patterson & Garzarolli, 2010].
- Processes Going on in Nonfailed Rod During Accident Conditions (LOCA and RIA), ZIRAT15/IZNA10 [Strasser et al., 2010b].
- Pellet Cladding Interaction and Pellet Cladding Mechanical Interaction, ZIRAT11/IZNA6 [Adamson et al., 2006/2007b].
- Mechanical Properties of Zirconium Alloys, ZIRAT6/IZNA1 [Adamson & Rudling, 2001/2002].
- Mechanical Testing of Zirconium Alloys – Vol. I And Vol. II [Adamson et al., 2013a and b]
- Mechanical Properties of Zirconium Alloys, ZIRAT6/IZNA1 [Adamson & Rudling, 2001/2002].

Dimensional changes:

- BWR Fuel Channel Distortion [Garzarolli et al., 2011]
- In-Reactor Creep of Zirconium Alloys [Adamson et al., 2009a]
- Structural Behaviour of Fuel Components [Cox et al., 2005/2006]
- Dimensional Stability of Zirconium Alloys [Adamson & Rudling, 2002/2003]
- Irradiation growth (to be published within the ZIRAT22 Programme)

Fuel performance topics:

- Impact of Irradiation on Material Performance, ZIRAT10/IZNA5 [Adamson & Cox, 2005/2006].
- Structural Behaviour of Fuel Components, ZIRAT10/IZNA5 [Cox et al., 2005/2006].
- High Burnup Fuel Issues – Their Most Recent Status, ZIRAT8/IZNA3 [Adamson et al., 2003/2004].
- Dimensional Instability, ZIRAT7/IZNA2 [Adamson & Rudling, 2002/2003].
- High Burnup Fuel Design Issues and Consequences [Rudling P. et al., 2012b]

Other relevant topics:

- Welding of Zirconium Alloys, ZIRAT12/IZNA7 [Rudling et al., 2007/2008].
- Manufacturing of Zr-Nb alloys, ZIRAT11/IZNA6 [Nikulina et al., 2006/2007].

- Manufacturing of Zirconium Alloy Materials, ZIRAT5/IZNA1 [Rudling & Adamson, 2000].
- Performance Evaluation of New Advanced ZR Alloys For PWRS/VVERs [Garzarolli & Rudling, 2011]
- Processes Going on in Nonfailed Rod During Normal Operation [Patterson, 2010]

Performance during accident conditions and Interim Dry Storage:

- Dry Storage Handbook [Patterson & Garzarolli, 2015]
- Loss of Coolant Accidents, LOCA, And Reactivity Initiated Accidents, Ria, In BWRS and PWRS [Rudling et al., 2004/2005]
- Nuclear Fuel Behaviour under Ria Conditions [Rudling & Jernkvist, 2016]
- Processes Going on In Nonfailed Rod During Accident Conditions Vol II, (ZIRAT15/IZNA10 STR) [Strasser et al., 2010]

2 Material development and performance of Alloys

2.1 Introduction

2.1.1 Stainless Steel

The initial fuel rods of early BWRs and PWRs were very thin stainless steel (SS) clad (AISI 304, 316, 347 or 348) material, with a low wall to diameter ratio of 0.013 to 0.08, to keep thermal neutron absorption low. This material was selected due to its rather high strength and excellent corrosion resistance in high temperature water. The behaviour of these SS cladding was quite good in PWRs but manifested severe longitudinal intergranular cracking in BWRs after burnups in excess of 6 MWd/kgU, a mode of cracking that has not been observed out of pile in high purity water.

For the fuel element structural components originally mostly SS and Ni base alloys were used.

2.1.2 Zr alloys

2.1.2.1 Zr source materials

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 did 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”. And produces zirconium of high purity, (Table 2-1).

Table 2-1: Typical chemical composition of Zr produced by the Van Arkel Process.

Element	Zr-X-Bar* Impurities (ppm)
Fe	6
Cr	3
Al	2
Ti	<0.1
Si	0.2
Cu	0.1
W	<0.1
Hf	26
Mg	<0.1
O	70
C	25
N	<0.10
* Analysed by GDMS	
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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. The first step is to convert zircon into $ZrCl_4$, through a carbo-chlorination process performed in a fluidised bed at 1200 °C.

After the Zr/Hf separation process, Zr metal is obtained by Zr-reduction in which gaseous $ZrCl_4$ 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. This material is called “sponge Zr”.

Table 2-2: Typical chemical composition of Zr produced by the Kroll process.

Element	Impurity, ppm (Zr balance)
Fe	200-500
Cr	65
Al	25
Ti	25
Si	40
Cu	5
W	25
Hf	75
Mg	--
O	1100
C	110
N	45

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A third method was used in the former Soviet Union to electrolytically produce Zr metal. Before the electrolytic extraction of Zr, the concentration of Hf of the feed material is reduced by fractional distillation of zirconium silicate. The sand ($ZrSiO_4$) is first reacted with K_2SiF_6 to produce K_2ZrF_6 in a separate operation. In the extraction process, the electrolyte used is K_2ZrF_6 in KCl. The process operates in closed gas tight electrolytic cells at high temperature under an inert atmosphere. The current used is in the range 10 000 to 20 000 A, and the cell is water cooled externally so that the cell surfaces are covered with a layer of solidified electrolyte which avoids the pick-up of impurities. K_2ZrF_6 is consumed as Zr is deposited on the cathode. Zirconium is produced as a powder, and after pre-treatment it is mixed with pure scrap metal that has been purified by the Van Arkel process and the alloying addition, in most cases only Nb, either 1% or 2.5%. *The chemical content of Zr manufactured by the electrolytic process has lower impurity concentrations compared to that of Zr manufactured by the Kroll process but higher than that produced by the Van Arkel process.*

2.1.2.2 Development of Zry-1 to Zry-4

2.1.2.2.1 Zry-1

It was soon discovered independently by Battelle Memorial Institute and Iowa State College that the addition of tin to “impure” zirconium tended to overcome the deleterious effects of nitrogen and to lesser extent, that of carbon and aluminium. The amount of tin necessary to obtain optimum corrosion resistance to “impure” zirconium was found to be dependent upon the nitrogen content of the alloy. Estimations for the nitrogen tolerance limits were made by workers at Massachusetts Institute of Technology and it was recommended that an alloy, containing 2.5% tin that could counteract about 700 ppm nitrogen, be studied. This alloy—sponge zirconium plus 2.5% tin – was later known as Zircaloy-1.

2.1.2.2.2 Zry-2, Zry-3 and Zry-4

Through an accidental contamination of a zirconium ingot by stainless steel it was realised that the impurities of Fe, Ni and Cr in zirconium drastically improved its corrosion performance, and the alloy development program was extended to evaluate the roles of these elements upon the corrosion characteristics of zirconium. Further investigations of the nitrogen tolerance of zirconium-tin ternary alloys with iron, nickel or chromium showed that the tolerance limits for the ternary alloys were higher than that for the binary zirconium-tin alloys. Concurrently, the Albany Station for the U.S. Bureau of Mines had made a number of refinements to the Kroll process for the preparation of sponge zirconium that yielded a product of lowered nitrogen content. The above information coupled with the knowledge that the corrosion rates of zirconium-tin alloys decrease with decreasing tin content lead to investigation into the properties of zirconium-1.8% tin alloys containing additions of iron, nickel or chromium that subsequently lead to Zry-2, -3 and -4.

2.1.2.2.2.1 Zry-2

The composition of early Zry-2 was fixed as follows:

Tin content – 1.5%

Since Zircaloy-1 proved to be unsatisfactory in regard to corrosion resistance, the tin content was dropped to 1.5% from 2.5%. The amount of tin in this early variant of Zircaloy-2 was sufficient to neutralise the effect of about 1000 ppm of nitrogen, which was considerably more than would be expected in zirconium sponge material at that time.

Iron content – 0.12%

Data from Westinghouse, Bureau of Mines and M.I.T. indicated that small quantities of iron increased the corrosion resistance in zirconium. An optimum value of 0.22% Fe was indicated from binary alloy studies, but since Zry-2 contained other beneficial elements and it was felt that the total alloy content should be kept to minimum to insure ease in fabrication, the iron content was fixed at 0.12%

Chromium content – 0.1%

The chromium content of Zry-2 was 0.1%. It had been found that chromium was beneficial in preventing corrosion of zirconium. Although data on binary zirconium-chromium alloys indicated that corrosion resistance of zirconium increased with increased chromium content, the amount of chromium was kept at 0.1% so that the hardness of the alloy would not be increased excessively.

Nickel content – 0.05%

Nickel was also shown to increase the corrosion resistance of zirconium, exhibiting its greatest influence of extending the life of zirconium alloys in high temperature steam. An optimum value was 0.25% Ni for binary zirconium-nickel alloys. Since Zry-2 contained other beneficial elements and since total alloy content had to be kept at a minimum to insure easy fabricability, the nickel content was fixed at 0.05%

The first important single property of Zry-2 was its excellent corrosion resistance in high temperature water, in which respect it was vastly superior to that of unalloyed zirconium.

2.1.2.2.2.2 Zry-3

In the spring of 1953 a new development program was undertaken to develop new zirconium alloys having corrosion resistance superior to that of Zry-2. This new program, which culminated in the alloy “Zircaloy-3”, was not dictated by any, at that time, known serious deficiency in the properties of Zry-2, but by the desire to improve the alloy. The alloy compositions and designations were as follows:

Zircaloy-3A 0.25% tin, 0.25% iron

Zircaloy-3B 0.50% tin, 0.40% iron

Zircaloy-3C 0.50% tin, 0.20% iron, 0.20% nickel

Concern over the corrosion resistance of Zircaloy-3 first occurred in the fall of 1956 when test results were obtained from the first lot of commercially produced ingots. The trouble with Zircaloy-3 manifested itself in the form of white corrosion product occurring in local areas strung out in the rolling direction (stringers). Subsequent examination of Zry-2 strip products fabricated in the same technique as the Zircaloy-3 strip products showed the presence of faint grey-white stringers. The principal difference was that the Zry-2 stringers were considerably smaller in size and more uniformly distributed. Further experimental work showed that the mechanism of stringer formation in the two alloys were different. For Zircaloy-3, the stringers were caused by segregated grain boundary precipitates while the stringers in Zry-2 were found to be related to gas voids formed in the metal during inert atmosphere melting. Subsequent experiments showed that vacuum arc-melting techniques greatly minimised the occurrence of stringers in Zry-2. Furthermore, beta quenching proved to be an effective remedial treatment for minimising the stringer effect in Zircaloy-3.

Since Zircaloys –2 and –3 exhibited similar water corrosion properties and the remedial measures for minimising the occurrence of stringers in Zry-2 were easier to apply (vacuum melting), the application of Zircaloy-3 was minimal.

Concurrently with the development of Zry-3, it was demonstrated in both out-of-pile and in-reactor experiments that the hydrogen absorption of zirconium alloys was enhanced when nickel was present in the alloy. In view of the above observations, it was considered prudent to initiate programs to re-evaluate the Zry-2 compositions, especially in respect to the nickel content and to determine the effect of coolant chemistry upon the hydriding of zirconium.

2.1.2.2.2.3 Zry-4

The results of the program, re-evaluating the Zry-2 composition, resulted in two modifications of Zry-2 – nickel-free Zry-2 and Zry-4. The first alloy, nickel-free Zry-2 is essentially Zry-2 without the nickel addition. The second alloy, Zry-4, contained the same chromium and tin levels as Zry-2 but the iron content was increased to compensate for the removal of nickel.

It was, however, found that nickel-free Zry-2 exhibited poor corrosion resistance at elevated temperatures in steam. Thus it was anticipated that the removal of nickel would result in decreased high temperature steam corrosion resistance. This was verified in 800 and 850°F steam tests where nickel-free Zry-2 samples completely disintegrated within short periods of time, while Zircaloys –2 and –4 showed similar good corrosion properties.

Additional results showed that nickel-free Zry-2 consistently picked up less hydrogen in both water and steam than that of Zry-2, while Zry-4 showed lower hydrogen absorption only in water.

2.1.3 Zirconium alloys used outside Russia

The first information of the use of zirconium alloys in nuclear reactor environments appeared with the U.S. publication of some of their development work on Zr-Sn based alloys at the First U.N. Conference on the Peaceful Uses of Atomic Energy in Geneva during August 1955. Although little was said during the actual conference sessions, the simultaneous publication of “The Metallurgy of Zirconium”, [Lustman & Kerze, 1955], presented data on these alloys; and remained the “reference text” for many years. Although it took sometime to get details of the development of the Zircaloys, [Kass, 1962], Lustman and Kerze actually made no mention of the composition of the Zircaloys!

At the next U.N. Conference on the Peaceful Uses of Atomic Energy (also in Geneva in 1958) the Russians presented extensive results on a range of Zr-Nb alloys. These ranged from the all α ozhennites, through the Zr-1%Nb (E110) alloy, to higher alloys with up to 5%Nb, [Ambartsumyan et al., 1958; Ivanov & Grigorovich, 1958 and Korobkov, et al., 1958]. It was quite sometime after this before it was evident that only the Zr-1%Nb (E110) and the Zr-2.5%Nb (E125) alloys were being used commercially in Russian reactors, and that the ozhennites (despite their apparently better corrosion resistance) were not being used, Table 2-3, [IAEA-TECDOC-996, 1998].

Table 2-3: Compositions of Zr-Nb Alloys.

Composition	Ozhennite 0.5	Zr-1%Nb		Zr-2.5%Nb		Zr-1%Nb/1%Sn/Fe		Otto Hahn
		E110	M5	E125	CANDU	E635	ZIRLO	
Nb %	0.1	1.0	1.0	2.4-2.8	2.5-2.7	1.0	1.0	3.0
Sn %	0.2			0.005	0.01	1.2	0.7-0.9	1.0
Fe %	0.1	100	300-650	300-650	<650	0.4	0.1	0.06
O ppm		600-700	1400	900-1300	1200-1500			
C ppm			50-100	<270	<100		<270	
H ppm	25	25	25	25	<5	25	25	25
S ppm	-	-	25	-	-	-	-	-
Ni ppm	0.1%	<70	<70	<70	<70	<70	<70	<70
Hf ppm	<100	<100	<100	<100		<100	<100	<100

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The Russian revelations at the 1958 Geneva Conference started a “Bonanza” of studies of the corrosion of Zr-Nb alloys elsewhere, especially where reactor designs other than the U.S. PWRs and BWRs were under development.

In the U.K.A.E.A at Harwell a study of the corrosion of the whole range of Zr-Nb alloys from Zr-0.5%Nb to 100%Nb, was started, [Cox, 1960; Cox et al., 1960; Cox et al., 1961; Cox et al., 1962; Cox, 1963; Cox & Johnston 1963 and Cox & Read, 1963]. Meanwhile in the Industrial Group of the U.K.A.E.A at Culcheth work focused particularly on the Zr-2.5%Nb alloy as an alternative to Zry-2 for the pressure tubes of the proposed Steam Generating Heavy Water Reactor (*SGHWR*), [Perryman et al., 1963 and Allen et al., 1966]. Ultimately, although the Harwell studies, [Cox, 1960] suggested that an alloy with a composition close to Zr-2%Nb-0.5%Sn offered the best corrosion resistance among the higher strength Zr-Nb alloys, the U.K. programme focussed on the Zr-2.5%Nb alloy, once it became clear that this was the pressure tube alloy adopted by the Russians for their boiling water reactors (*RBMKs*). Some years later a joint Scandinavian/U.K programme (*SCANUK*) studied a number of ternary and quaternary alloys aimed at improved fuel cladding with the ability to withstand short, high-temperature excursions, [Tyzack et al., 1977].

None of these alloys were significantly better than Zry-2 either in- or out-reactor, however, under BWR (*SGHWR*) conditions.

A similar trend occurred in the Canadian programme, where, once again, an alloy for pressure tubes was required. There, however, the desire to maximise the mechanical properties of an alloy that was heat-treatable led to CO₂ extensive work in heat-treated Zr-2.5%Nb, after some studies of a range of Zr-Nb-Sn alloys, [Dalgaard, 1960 and Dalgaard, 1961]. Typically the alloy was first water-quenched from the ($\alpha+\beta$) phase field (~880°C) or the β -phase (~960°C), given a pre-determined amount of cold-work to accelerate the approach to an equilibrium structure, and then annealed at the temperature (480-500°C) below the monotectoid temperature (590-610°C depending on the oxygen content). In the β -quenched condition the corrosion resistance was very poor, and this improved with both the amount of cold-work and the annealing time, [LeSurf, 1969 and Cox, 1976a]. Again the knowledge that the Russians were already using the Zr-2.5%Nb alloy for pressure tubes in the extruded (in the

$\alpha+\beta$ phase field) and annealed condition, Table 2-4, led to a change to a similar fabrication route, but with 25-30% cold-work and only a 400°C/24 hr stress-relief. Subsequent comparisons have shown that, although the Russian and Canadian fabrication routes are similar, the Russian tubes have slightly better mechanical properties than the Canadian tubes. The Canadians also developed a higher strength Zr-2.5%Nb-0.5%Cu alloy, which behaved reasonably well in high temperature water, [Daugaard, 1960], but was mainly of interest for situations where strength was more important than corrosion resistance, such as for the “garter springs” that separate the pressure and calandria tubes in CANDU reactors. Concerns about the long-term effects of hydrogen embrittlement by the annulus gas (originally N₂ with some D₂ that diffuses through the stainless steel end-fittings-now changed to CO₂) have led to their progressive replacement with Incoloy 728 garter springs.

Table 2-4: Pressure Tubes – Material and Operational Conditions.

Reactor	Material	Finished ready condition	Configuration and moderator	Outlet temp., °C	Hoop stress, MPa	Fast neutron flux, 10 ¹⁷ n/cm ² s
CANDU	Zr-2.5%Nb	Cold worked	Horizontal, D ₂ O	308	135	3.7
FUGEN	Zr-2.5%Nb	Solution heat treated	Vertical, D ₂ O	280-286	96	2.7
RBMK	Zr-2.5%Nb	Annealed	Vertical, Graphite	288	86	1.7

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The German studies of Zr-Nb alloys started at Metallgesellschaft AG, where a higher strength alloy than the Zircalloys was sought for the fuel cladding in the “Otto Hahn” nuclear ship reactor, [Richter & Wilhelm, 1975 and Matucha et al., 1978]. Extensive early corrosion results were presented for a range of Zr-Nb alloys, [Rösler, 1961; Jangg et al., 1971; Anderko et al., 1962; Beyer et al., 1967 and Rückdeschel & Wincierz, 1967], and led to the selection of a Zr-3%Nb-1%Sn alloy for the Otto Hahn cladding. The corrosion resistance of the ZrNb₃Sn₁ alloy was good both in- and out-reactor, but heat treatment of the welds was a problem (the corrosion rates of β -quenched alloys with $\geq 2\%$ Nb, and sensitivity of these welds to delayed hydride cracking were known problems).

In Canada the same problem faced the proposed use of Zr-2.5%Nb cladding in the organic-cooled, D₂O-moderated WR-1 prototype reactor. Cracking of unannealed welds in stored fuel elements for this reactor provided the first direct observations of DHC (Delayed Hydride Cracking) in a commercial situation, [Simpson & Ells, 1974]. These results came just too late to allow remedial action before the first in service DHC cracking of pressure tubes in Pickering A#3 on August 10th 1974, [Cox, 1976b and Perryman, 1978].

In the U.S. investigations of Zr-Nb alloys by Naval Reactors Branch at the Knolls Atomic Power Laboratory centred on a Zr-2 at %Nb-2 at %Sn alloy, [Bibb et al., 1960 and Fisch, 1961]. The alloy generally oxidised somewhat faster than Zry-2 in both 360°C water and 400°C steam, but hydrogen uptakes were lower at ~10-15%. There is no evidence of whether this alloy was used by Naval Reactors Branch, however, it is suspected that a Zr-3%Nb alloy may have seen some service.

Zry-2¹ became the preferred cladding for Western Boiling Water Reactors (BWRs), while Zry-4², with its lower hydrogen uptake rate in hydrogenated water, was the preferred cladding in Western PWRs.

PWR Shipping port (1958) and the BWR Dresden I (1960) were the first commercial reactors that used Zry-2. The material condition selected in the early 1960s for fuel rod claddings of BWR and PWR was a cold worked and stress relieved condition to fulfil the ASME pressure vessel criteria, i.e. no plastic deformation (except creep deformation) of the fuel cladding is allowed even at the largest possible system to rod internal differential pressure.

¹ Zry-2: 1.2-1.7%Sn, 0.07-0.20%Fe, 0.05-0.15%Cr, 0.03-0.08%Ni

² Zry-4: 1.2-1.7%Sn, 0.18-0.24%Fe, 0.07-0.13%Cr, <0.007%Ni

3 Conclusions for the optimum PWR/VVER and BWR cladding and structure material

3.1 Introduction

New Zr alloy materials with increased margins towards the fuel design criteria during normal operation, AOO, DBA as well as dry storage condition are needed. The Zr alloy properties with the smallest margins towards the criteria will become limiting. Thus, it is crucial to develop new Zr alloy materials with properties that increase these margins (Table 3-1).

Table 3-1: Zr alloy properties limiting performance during normal operation, AOO, DBA and intermediate dry storage.

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 µm, 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 event and cask drop accident (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.
	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, e.g., by replacing SR with RX (or pRX) claddings or by alloying additions. The drawback with RX (or pRX) is that the corrosion resistance in PWRs deteriorates compared with SR cladding irrespective of alloying contents. However, in BWRs RXmpRX 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 (dryout and LOCA) are maintained.	There are two main causes for fuel channel bowing today: <ol style="list-style-type: none"> 1) Difference in irradiation growth rate of two opposing channel sides due to a fast fluence gradient (fluence gradient induced bow) – for most Zr alloys. 2) 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 Zircalloys minimizes both 1) and 2) above

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Table 3-1: Zr alloy properties limiting performance during normal operation, AOO, DBA and intermediate dry storage. (Cont'd)

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 impacting 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 and/or cladding stress are limited to minimise 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.

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In the following, the above limiting Zr alloy properties during different conditions are discussed.

3.2 Corrosion and HPU

3.2.1 Introduction

Future Zr alloy materials should have excellent corrosion resistance and low HPUF. Corrosion acceleration occurs due to:

- 1) SPP dissolution in Zircalloys (e.g. Zry-2 and Zry-4) both in BWRs and PWRs. The tendency for dissolution starts at the beginning of irradiation and at a critical fast neutron fluence (corresponding to a certain burnup) the SPPs have dissolved and at that time corrosion acceleration occurs. Since E110 (ZrNb) alloy is used in VVERs this is not an issue.
- 2) Hydride rim formation at the oxide/metal interface in BWRs and PWRs (this does not occur in VVWRs due to the extremely low oxide thicknesses and hydrogen contents – the reason for this is the excellent E110 material and that KOH instead of LiOH is used in the coolant to increase the PH).
- 3) At an oxide thickness of >100 microns corrosion acceleration may occur in PWRs (not in VVERs because the oxide thickness is so small) due to corrosion rate thermal feedback.

For any material the RXA condition results in higher corrosion rates than that of SRA material with the same chemical composition in PWRs but in BWRs the corrosion rate of SRA and RXA Zry-2 is the same. At a critical oxide thickness oxide spallation occurs. In fuel rods with a heat flux, oxide spallation may result in hydride blister formation, provided that:

- The difference in oxide thickness between spalled and non-spalled areas are large enough and,
- The surface heat flux is large enough

Hydride blister may reduce PCMI failure threshold during a RIA event. The HPU is a product of corrosion rate and HPUF. Low HPU results in larger margins during seismic event, LOCA, RIA and cask drop accident.

3.2.2 BWR

The initial SPP size in Zry-2 has a large impact on corrosion and HPU behaviour. Increasing SPPs will decrease the tendency for accelerated HPUF and corrosion acceleration.

As shown in Figure 3-1 in the case of Zircaloy, SPP size has a very pronounced effect on corrosion in 350°C water, 500°C steam, as well as in PWRs and BWRs. Uniform corrosion rate increases out of pile in 350°C water and in pile at a SPP size below 70-80 nm and nodular corrosion appears in-BWR and in 500°C steam in lots with a SPP size larger than 150 nm.

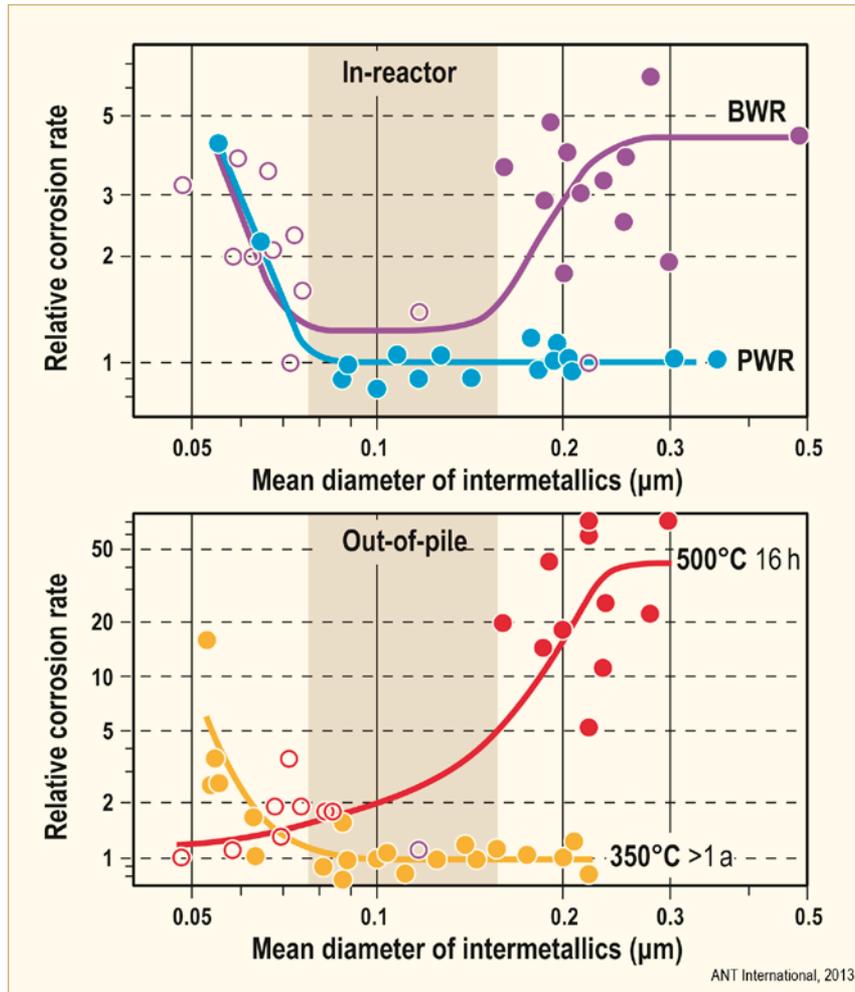


Figure 3-1: Corrosion of Zircaloy versus size of intermetallic precipitates, after [Garzarolli et al., 1996a].

Increased HPU at high burnups reduces margins during DBA and cask drop accident during interim dry storage. Figure 3-2 shows how the HPU increases with burnup for various Zry-2 BWR fuel rod designs.

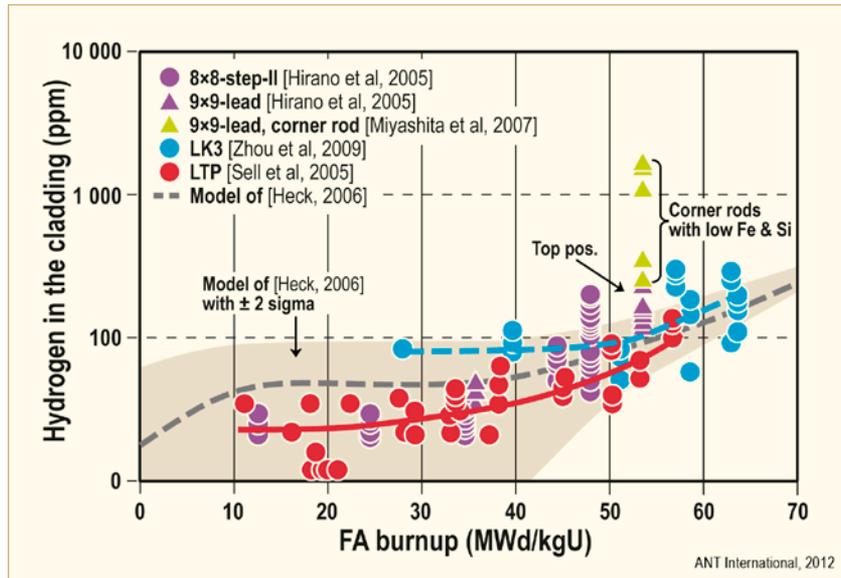


Figure 3-2: Hydrogen pickup of Zry-2 fuel rod claddings in BWR at high burnups, after [Hayashi et al., 2003], [Ledergerber et al., 2005], [Hirano et al., 2005], [Miyashita et al., 2007], [Zhou et al., 2009], [Sell et al., 2005], and [Heck, 2006].

The reason for the dramatic increase in HPUF for the Zry-2 materials is the result of the irradiation induced dissolution of the nickel-bearing second phase particles (SPPs) which does not exist in Zry-4. In 1995 [Huang et al., 1996] reported that after irradiation of Zry-2 coupons with fine SPP in a commercial BWR HPUF as well as the corrosion rate increased at high BUs due to the irradiation induced SPP dissolution. The authors showed that when SPPs virtually “disappeared”, within the resolution of Scanning Transmission Electron Microscopy (STEM) at that time, HPU and corrosion increased. Figure 3-3, illustrate that trend, especially for specimen 1R, whose SPPs disappeared between fluences of 2.5 and $8.5 \times 10^{25} \text{ n/m}^2$ ($E > 1 \text{ MeV}$).

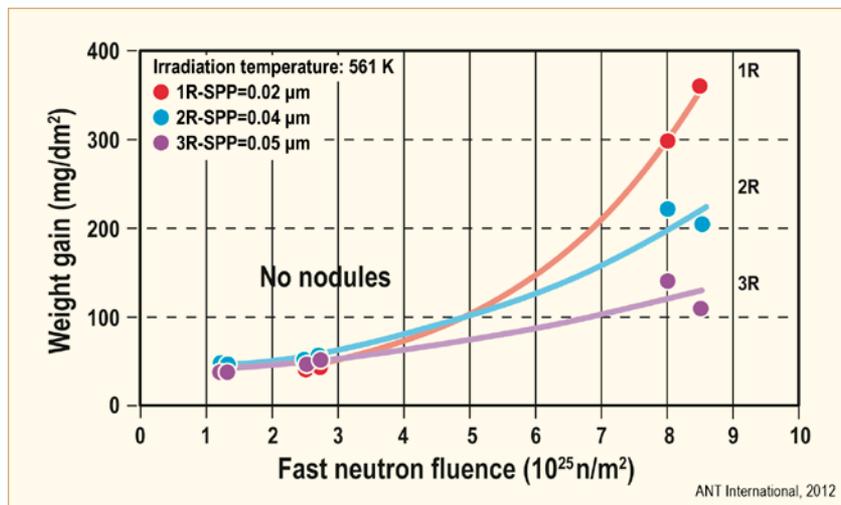


Figure 3-3: Corrosion as a function of fluence for Zry-2 for non-heat transfer surface specimens having different SPP sizes. An oxide weight gain of 15 mg/dm^2 corresponds to 1 micron of oxide, after [Huang et al., 1996].

In the late 1990s, [Sihver & Hallstadius, 1999], [Limbäck et al., 2001], [Tägtström et al., 1999] reported that uniform oxide formation and Hydrogen Pickup, HPU of LK2 and LK2+ fuel cladding of ABB (today Westinghouse) accelerated at higher burnups while the LK3 material was much less affected (Figure 3-4). The reason for the different behaviour of three different materials was proposed to be the initial SPP size, which is very small for LK2 (low A-parameter) and largest for LK3. The

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List of common abbreviations

AECL	Atomic Energy of Canada Limited	IAEA	International Atomic Energy Agency
AISI	American Iron and Steel Institute	IASCC	Irradiation Assisted Stress Corrosion Cracking
ANSI	American National Standards Institute	ID	Inner Diameter
ANT	Advanced Nuclear Technology	IGSCC	Intergranular Stress Corrosion Cracking
AOA	Axial Offset Anomaly	INPO	Institute of Nuclear Power Operations
AOO	Anticipated Operating Occurrence	IRI	Incomplete Rod Insertion
AREVA	French Equipment Manufacturer	KKL	KernKraftwerk Leibstadt
ASEA	Allmänna Svenska Elektriska Aktiebolaget (General Swedish Electrical Limited Company)	KWU	KraftWerkUnion
ASME	American Society of Mechanical Engineers	LCC	LWR Coolant Chemistry
B&W	Babcock & Willcox	LHGR	Linear Heat Generation Rate
BCC	Body Centred Cubic	LME	Liquid Metal Embrittlement
BNFL	British Nuclear Fuels Limited	LOCA	Loss of Coolant Accident
BOP	Balance of Plant	LWR	Light Water Reactor
BWR	Boiling Water Reactor	MCP	Main Circulating Pump
CANDU	Canadian Deuterium Uranium	MPS	Missing Pellet Surface
CASL	Consortium for Advanced Simulation of LWRs	NDA	New Developed Alloy
CE	Combustion Engineering	NEI	Nuclear Energy Institute
CILC	CRUD Induced Localized Corrosion	NG	Nuclear Grade
CIPS	CRUD Induced Power Shift	NMCA	Noble Metal Chemical Addition
CP	Corrosion Product	NPD	Nuclear Power Demonstration
CR	Control Rod	NPP	Nuclear Power Plant
CRUD	Chalk River Unidentified Deposits	NRC	Nuclear Regulatory Commission
CWSR	Cold Work and Stress Relieved	NSSS	Nuclear Steam Supply System
DNB	Departure from Nuclear Boiling	NWR	Normal Water Chemistry
ECBE	Effective Control Blade Exposure	OD	Outer Diameter
EFID	Effective Full Insertion Days	OLNC	On-Line Noble Chemistry
ELS	Extra-Low Sn	OPG	Ontario Power Generation company
EOC	End Of Cycle	OTSG	Once-Through Steam Generators
EPRI	Electric Power Research Institute	PCI	Pellet Cladding Interaction
ESSC	Enhanced Spacer Shadow Corrosion	PCMI	Pellet Cladding Mechanical Interaction
FA	Fuel Assembly	PGS	Pickering Generation Station
FGR	Fission Gas Release	PSD	Power Spectral Density
FP	Fission Product	PWR	Pressurised Water Reactor
FRED	Fuel Reliability Data Base	PWSCC	Pressurised Water Stress Corrosion Cracking
FRI	Fuel Reliability Indicators	QA	Quality Assurance
GC	Guide Channels	QC	Quality Control
GE	General Electric	RBMK	Reaktor Bolshoi Mozhnosti Kanalov (in English Large Boiling Water Channel type reactor)
GNF	Global Nuclear Fuel	RCCA	Rod Cluster Control Assembly
GT	Guide Tubes	RCP	Reactor Coolant Pump
GTRF	Grid-To-Rod Fretting	RIA	Reactivity Initiated Accident
HAZ	Heat Affected Zone	RXA	Recrystallised Annealed
HBS	High Burnup Structure	SCC	Stress Corrosion Cracking
HCP	Hexagonal Close-Packed	SCI	Shadow Corrosion Induced
HFE	Healthy Fuel Examinations	SEM	Scanning Electron Microscopy
HPA	High Performance Alloy	SG	Steam Generator
HPU	Hydrogen Pick-Up	SNB	Subcooled Nucleate Boiling
HPUF	Hydrogen Pick-Up Fraction	SP	Spacer Position
HTP	High Thermal Performance	SPP	Second Phase Particles
HWC	Hydrogen Water Chemistry		

Unit conversion

TEMPERATURE		
$^{\circ}\text{C} + 273.15 = \text{K}$	$^{\circ}\text{C} \times 1.8 + 32 = ^{\circ}\text{F}$	
T(K)	T($^{\circ}\text{C}$)	T($^{\circ}\text{F}$)
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}

MASS	
kg	lbs
0.454	1
1	2.20

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

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 $\sqrt{\text{m}}$	ksi $\sqrt{\text{inch}}$
0.91	1
1	1.10