

# BWR Fuel Performance under Advanced Water Chemistry Conditions – a Delicate Journey Towards Zero Fuel Failures – a Review

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## Abstract

Boiling Water Reactors (BWRs) have undergone a variety of chemistry evolutions over the past few decades as a result of the need to control stress corrosion cracking of reactor internals, radiation fields and personnel exposure. Some of the advanced chemistry changes include hydrogen addition, zinc addition, iron reduction using better filtration technologies, and more recently noble metal chemical addition to many of the modern day operating BWRs. These water chemistry evolutions have resulted in changes in the crud distribution on fuel cladding material, Co-60 levels and the rod oxide thickness (ROXI) measurements using the conventional eddy current techniques. A limited number of Post-Irradiation Examinations (PIE) of fuel rods that exhibited elevated oxide thickness using eddy current techniques showed that the actual oxide thickness by metallography is much lower. The difference in these observations is attributed to the changing magnetic properties of the crud affecting the rod oxide thickness measurement by the eddy current technique.

This paper will review and summarize the BWR fuel cladding performance under these advanced and improved water chemistry conditions and how these changes have affected the goal to reach zero fuel failures. The paper will also provide a brief summary of some of the results of hot cell PIE, results of crud composition evaluation, crud spallation, oxide thickness measurements, hydrogen content in the cladding and some fuel failure observations.

**Key Words:** Boiling Water Reactor, Fuel Performance, Hydrogen Addition, Zinc Addition, Noble Metal Chemical Addition, Zero Leakers

## 1. Introduction

BWR fuel performance and reliability has improved over the years, and fuel failure incidents have decreased, as a result of continuing improvements in water quality. However, there is some variability in crud deposition and crud composition on fuel cladding materials possibly due to water chemistry changes adopted such as hydrogen addition, zinc addition, iron reduction and noble metal addition, as well as due to power uprates up to almost 20% in some plants. Increased deposition of corrosion products (“crud”) is undesirable because it can reduce heat transfer and increase fuel rod temperature, resulting in an increased corrosion of the fuel cladding, ultimately increasing the risk of fuel failures [1]. Some of the metallic impurities introduced into the reactor water through the feed-water system get incorporated in crud deposits. In the early years, the majority of crud deposits consisted of iron, chromium and nickel coming from the slow corrosion of stainless steel, and copper and zinc arising from the slow corrosion of brass condenser tubing. In the recent years, the introduction of intentional additives to control corrosion potentially may have increased the risk of crud

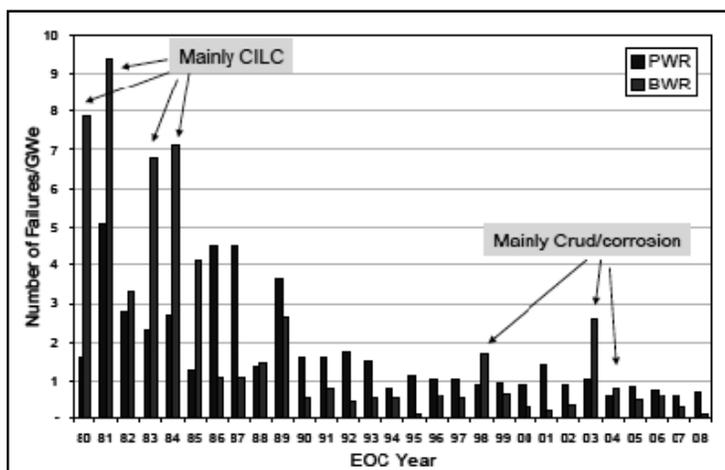


Figure 1. Number of Failed Fuel Assemblies in US Light Water Reactors [2].

buildup on fuel cladding. For example, high zinc usage in BWRs tends to increase the adherence of crud deposits on fuel cladding, which can result in undesirable oxide spalling in higher-rated cores. Crud/corrosion-related fuel failures have occurred at one BWR in 1998/99 and at four US BWR plants during the years 2002 to 2004 as shown in Figure 1 [2]. The Figure also shows the BWR fuel failures in the early years due to Crud Induced Localized Corrosion (CILC) and the successively decreasing trend in fuel failures except for the incidents in 1998/99 and 2002 to 2004 which were crud/corrosion related. With progressive uprating of fuel duty, the available margin to tolerate crud buildup has been reduced and additional care needs to be taken in specifying the water chemistry conditions to avoid undesirable fuel performance issues. Figure 2 shows the BWR fuel failure history from 2000 to 2007 along with the causes of failures identified [1]. A disappointing feature here is the presence of a significant number of failed fuel assemblies due to unknown reasons. When the reasons are unknown no corrective actions can be taken to guard against such failures.

In 2006, the fuel integrity improvement initiative established the challenging goal of eliminating fuel failures in US plants by 2010 that included water chemistry considerations on the formation of fuel deposits (crud) and potential effects on fuel cladding corrosion [3]. Although this goal has not been achieved, possibly due to continuing water chemistry changes, power uprates and core management changes, it is encouraging to note that there is a decreasing trend of fuel failures in the recent years, and no corrosion related fuel failures have occurred in the US over the past decade.

## BWR Chemistry Evolutions and Crud Spallation

### Fuel Surface Crud Spallation

The BWR has gone through many water chemistry evolutions since 1982 in attempts to mitigate intergranular stress corrosion cracking (IGSCC) and mitigate operating dose rates and radiation fields. Hydrogen water chemistry was implemented in BWRs to mitigate IGSCC of reactor internal components, Zinc addition and depleted zinc addition (DZO) was implemented to control and

manage radiation fields. The most recent water chemistry change introduced into BWRs is the noble metal chemical addition during hot standby conditions by a process called noble metal chemical application (NMCA) and on-line NMCA application (OLNC) during full power operation of the plant. Following the first NMCA application at Duane Arnold (DAEC) BWR in 1996, some crud spallation was observed followed by crud spallation at Peach Bottom 2 in 1998, the second NMCA plant. The location of crud spallation was different in the two cases where the crud spallation was confined more to the lower end of the fuel rod in the case of Peach Bottom 2 (PB2) while at DAEC crud spallation occurred in the higher duty region (~ 75 cm from the bottom end plug) [4,5]. Since the low elevation crud spallation at PB2, shown in Figure 3, is not a high duty region, the phenomenon is less well understood. This kind of low elevation crud spallation was never seen in this plant again, or at any other NMCA plant. However, as a precautionary measure, a limit on the noble metal input over the life time of a fuel bundle was imposed following the Peach Bottom 2 fuel crud spalling observation [6],

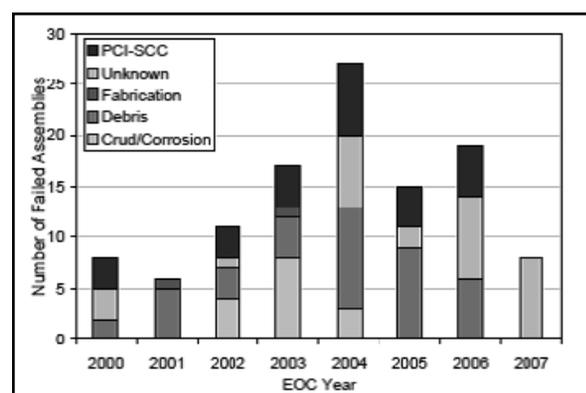


Figure 2. US BWR Fuel Failure History from 2000 to 2007 [1].

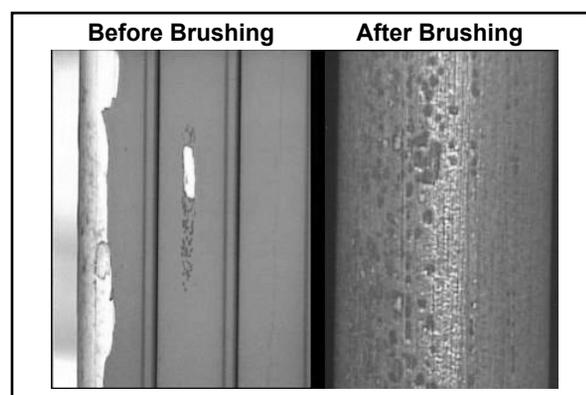


Figure 3. Tenacious Surface Crud Spallation at Peach Bottom-2 [4].

even though noble metal was not implicated in any of the corrosion related fuel failures that occurred during 2002 to 2004. It was later determined that the crud spallation is potentially more related to the higher levels of zinc usage than to the noble metal input, Figure 4 [2]. However, the allowable noble metal input over the life time of a fuel bundle still remains restricted to a level accepted by the industry despite the fact that noble metal has not been implicated in any of the fuel failures. Thus, guideline limits on both noble metal input and zinc input into BWRs have been established and there have been no fuel crud/cladding related fuel failures in US BWRs since 2004 [2].

### Eddy Current Liftoff Measurements

The eddy current liftoff measurements of crud deposited rods showed very large interference of the eddy current signal due to magnetic crud on the rod surface. Figure 5 shows the typical axial profile of the liftoff data from DAEC fuel rods as measured after correction to eliminate the interference. The corrected data (green trace) at each axial elevation represents the combined thickness of the zirconium oxide and the surface crud [4]. Such magnetic interference is common for fuel rods

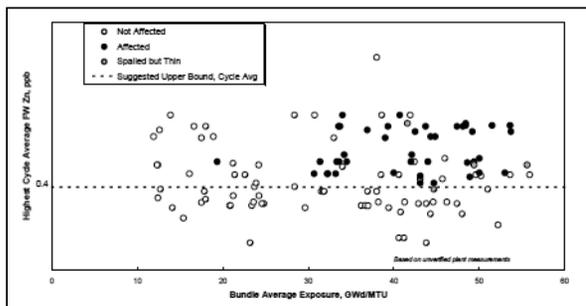


Figure 4. Feedwater Zinc vs. Bundle Exposure and Spalling Experience [1].

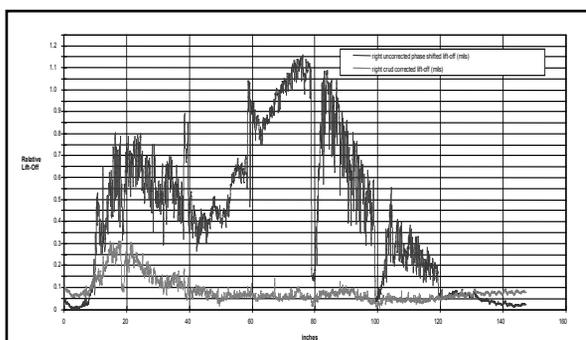


Figure 5. Eddy current liftoff measurement after one NMCA cycle showing large interference due to magnetic crud [4].

operated in plants with zinc injection, where the fuel crud typically contains a zinc ferrite, which is strongly magnetic [7]. It can be seen in Figure 5 that the zinc-enriched crud deposited along the active fuel length from 10-120 inch (25 to 305 cm) elevations. The magnetic effect persisted throughout 3 cycles of operation. Hot cell post irradiation examination (PIE) was performed to determine the actual thickness of the zirconium oxide and the crud. In general, the cladding oxide thickness measured by metallography was low at less than 27  $\mu\text{m}$ , this is lower than that suggested by the corrected eddy current trace (green) in Figure 5, and most of the eddy current liftoff was the result of the 10 to 56  $\mu\text{m}$  thick, tenacious crud layer as confirmed by hot cell PIE after 4.5 years of plant operation following NMCA [4,5]. A part of the crud layer appeared to be crystalline containing 30 to 35% zinc, consistent with the  $\text{ZnFe}_2\text{O}_4$  phase. No fuel failures were detected at DAEC during the monitoring period covering cycles 15 to 17.

### Crud Induced Corrosion Failures 1997-1999

Fuel failures in a BWR/6 occurred during the 1997-1999 period due to excessive crud deposition on fuel cladding material to an extent that in some cases the spacing between the rods were “almost” filled with crud that potentially would have increased the core resistance. The affected fuel rods were mostly first cycle peripheral rods and the crud loadings were 5 to 10 times higher than normal. The crud contained some Zn, Cu (from brass condenser tubing) and the rest being iron. The plant operated within the chemistry guidelines for feedwater iron and zinc at that time. The source of the iron crud that resulted in heavy crud deposits on fuel rods was not identified.

### Crud Induced Corrosion Failures 2002-2004

A BWR/6, experienced fuel failures during 2002-03 due to excessive crud deposition. The heaviest crud deposit occurred on the outer surface of peripheral rods at the second span (20 to 40 inches, 51 to 102 cm) from the lower end plug, Figure 6. Some peripheral rods failed due to local cladding penetration, while no interior rods exhibited heavy crud deposition or rod damage. Some peripheral

rods were found to bow away from the bundle, possibly due to higher cladding temperature and oxide growth on the peripheral side. Crud flake samples analyzed showed average metal contents in the crud in weight percent to be 71-84% Fe, 7-14% Zn, 2-8% Cu, 2-6% Si, and others <1%. Further analysis showed that the crud consisted of crystalline  $ZnFe_2O_4$  and an amorphous phase of zinc silicate  $Zn_2SiO_4$  filling the pores surrounding the zinc ferrite crystals. Analysis from two other BWRs confirmed the presence of zinc ferrite and zinc silicate in the crud deposits. Furthermore, when zinc silicate is present in the crud, the crud porosity can decrease from 50% to <10% due to pore filling by zinc silicate [2].

A BWR/4, Browns Ferry-2 experienced apparent corrosion-induced cladding failure of similar characteristics in 2002-04 and both had mid-cycle outages to remove failed assemblies and discharged all fuel in the reloads containing failed assemblies after 2 cycles [8]. Browns Ferry-3 also experienced fuel failures of similar characteristics in some 3rd cycle fuel rods as shown in Figure 7. The picture on the right was taken after brushing, exhibiting substantial surface spallation at the upper elevation of the failed rod [2].

An example of a maximum effective eddy current liftoff measurement that provides an estimate of the cladding corrosion made on a Browns Ferry-2 rod is shown in Figure 8, and the associated corrosion behavior is shown in Figure 9 [9]. Fuel rods in 63 fuel assemblies from reload 10 failed, while fuel rods from reload 9 and reload 11 that were also present in the core at the same time as reload 10 fuel showed normal liftoff behavior as illustrated in Figure 8 [9].

Further analyses of BF 2 rods showed that the rod perforation was caused by OD-initiated cracks through a locally hydrided region as shown in Figure 10 [9]. Outside-in cracking of BWR fuel rods can occur under power ramp conditions that promote radial hydride formation on the outer surface with modest bulk cladding hydrogen levels [10]. There was no significant wall thinning of the rods. As is evident from the Figure, fuel cladding cracking exhibited extensive multi-branch type cracks within a zone of large hydride localization. Cracks appear to have originated both from the inner as well from the outer surface. The cracks in the zirconium oxide layer are an indication that the oxide layer is non-protective [9].

The examination of a sound partial length rod had surface characteristics similar to the failed rod with heavy spalling and corrosion with a ~ 90 micron eddy current liftoff peak near 95 inch (241 cm) elevation as shown in Figure 11 [9]. Despite the appearance and the increased eddy current liftoff, there was no visible perforation or through wall corrosion in this rod [9].

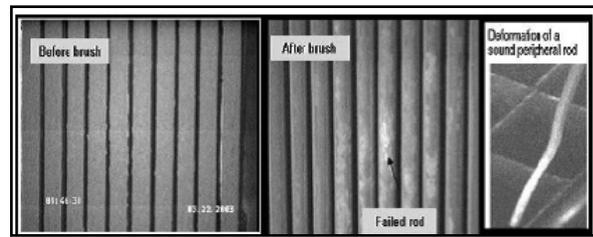


Figure 6. Tenacious Crud Build-up on Fuel Rods and Bow of a Peripheral Rod [2].

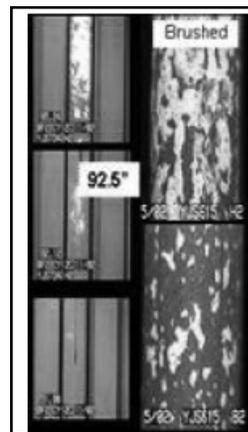


Figure 7. Browns Ferry-2 Cycle 12 "corrosion" Failed Rod before and after Brushing [2].

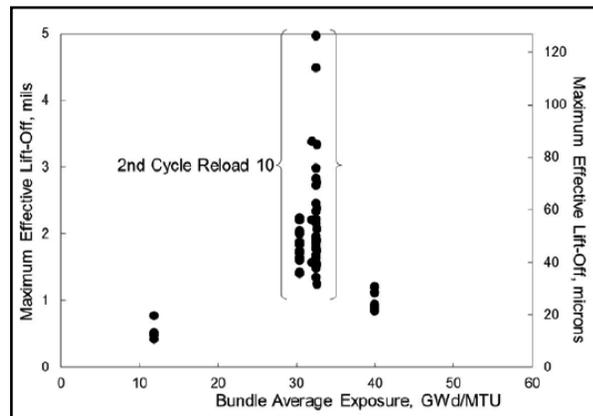
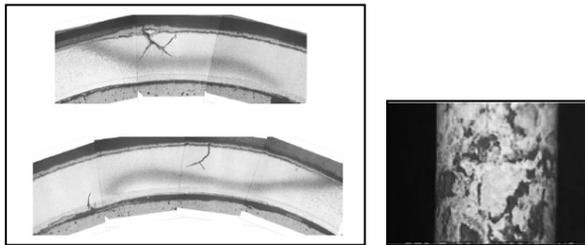
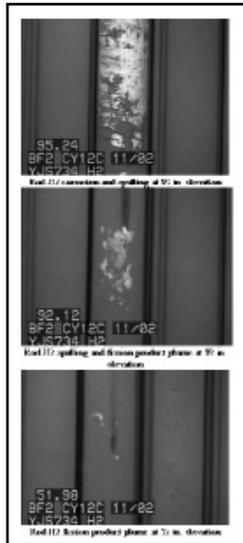
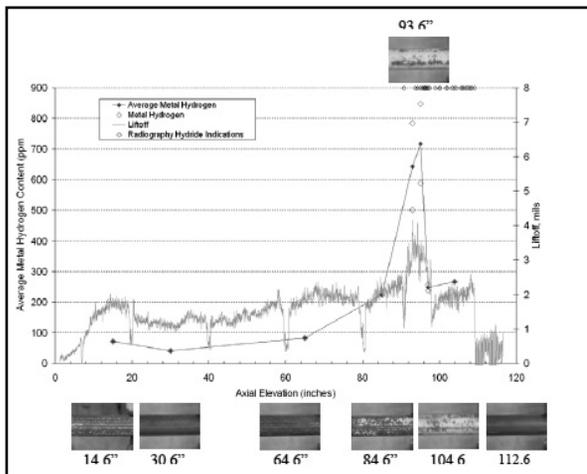


Figure 8. BF-2 Lift-Off vs. Exposure Showing Affected Reload 10 Fuel Rods Compared to Normal Performance of Earlier (Reload 9) and Later (Reload 11) Reloads [9].

**Figure 9. Visual Appearance of Failed BF-2 Reload 10 H2 Rod; Leaking Locations Near 52 and 92 inches (132 cm and 234 cm) and Heavy Spalling Starting Near 95 inches (241 cm) [9].**



**Figure 10. Cladding Cross-section from a Failed BF-2 Reload 10 Rod at 95.9 inches (244 cm) Showing Cracks Within Massive Hydride Localization near Primary Failure Location along with a Photo Showing Excessive Crud/Oxide Spallation [9].**



**Figure 11. Visual Appearance, Lift-off, Cladding Metal Hydrogen Content, and Radiography Hydride Localization Indications in Sound BF-2 Reload 10 Rod [9].**

Since Browns Ferry-2 and the second BWR were both NMCA plants and the failures occurred after about 6 to 7 months of NMCA operation, it became necessary to determine whether NMCA had any role in these fuel failures. However, the failure of thrice burned rods at Browns Ferry-3 was able to suggest that the corrosion failures were not a result of NMCA, because sibling rods from the same lot that were discharged after one cycle of operation without being exposed to NMCA, exhibited accelerated corrosion at both the 50 to 100 cm and 254 to 305 cm elevations [2]. The conclusion was that corrosion of the rods initiated before NMCA application.

The investigation established that the BF-2 Reload 10 fuel failed by accelerated cladding nodular corrosion that resulted in the absorption of corrosion generated hydrogen. Localization of hydrides is postulated to have resulted from local thermal gradients that are caused by local differences in oxide growth and spalling of oxide. Massively hydrided regions then fractured under tensile loading that arose with accumulation of exposure during the course of normal operation. The specific manifestation of hydrogen and its localization in the BF-2 corrosion failures is a new, or previously unrecognized, BWR failure mechanism that has not been published in the industry [9].

### Counter Measures against Crud Induced Corrosion Failures

As a counter measure against CILC failures some material improvements were made that included employment of materials less susceptible to nodular corrosion. At the same time, some plants employed deep bed demineralizers in the condensate system for better removal of copper, while other plants replaced the admiralty brass condenser tubing with titanium to eliminate the copper source that was thought to be playing a major role in CILC related fuel failures. Currently, more than 20 plants have titanium condenser tubing, more than 10 plants have stainless steel condenser tubing, and the plants that still have brass condenser tubing have employed deep bed or deep bed plus filter demineralizer system combination to better remove the copper source.

Minimizing the formation of thick and tenacious crud is desirable due to the concern to fuel integrity because of the potential increase in cladding temperature and corrosion

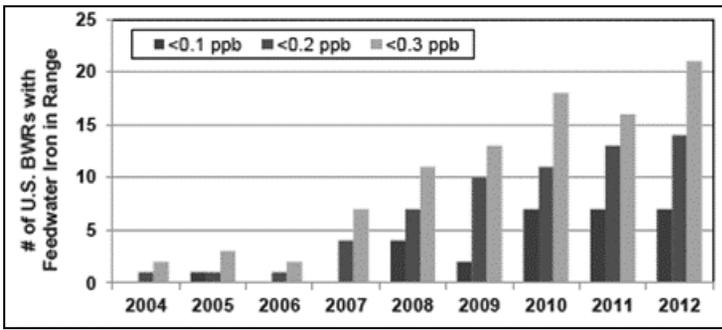


Figure 12. Evolution of Low Feedwater Iron Operation at US BWRs since 2004 [11].

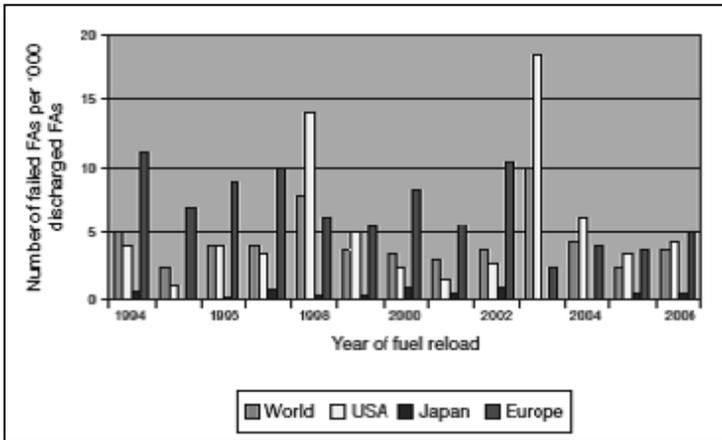


Figure 13. BWR Fuel Assembly Failure Rates Presented as Leaking Assemblies per 1000 Discharged Assemblies [14].

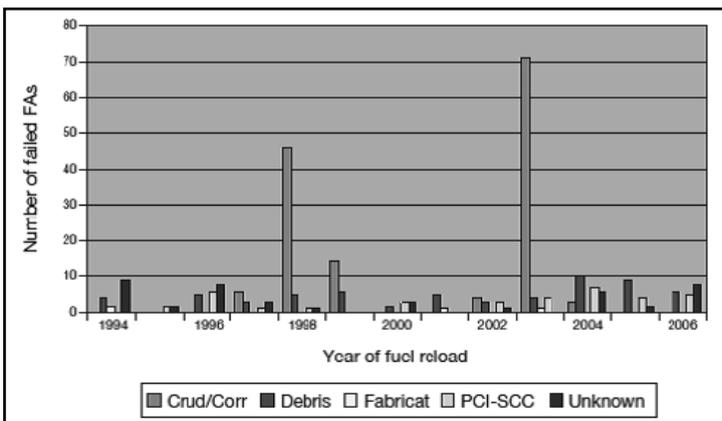


Figure 14. BWR Fuel Failure Causes in the United States [14].

suffered CILC failures due to deposition of copper and other soluble impurities in the reactor water into delaminating nodular oxide on Zircaloy-2 fuel cladding during the period from 1978 to mid-1980s [13]. The CILC fuel failure mechanism was substantially eliminated by process improvement of Zircaloy-2 cladding to minimize nodular corrosion susceptibility and, in most cases, replacement of brass condenser tubing with stainless steel or titanium alloy [2].

rates [4]. In order to further control fuel crud, US plants also adopted the use of low feedwater iron inputs in the range  $< 0.1$  to  $< 0.3$  ppb using better filtration technologies. The reduction of feedwater iron also has the added benefit of increasing the performance of zinc in reducing radiation fields. Figure 12 provides a representation of employing better filtration technologies that resulted in lower feedwater iron levels year after year [11]. BWR operators are also using  $< 0.4$  ppb zinc in the feedwater, per EPRI Water Chemistry Guidelines [12], under NMCA conditions in order to minimize the tenacious ferro-magnetic crud formation on fuel cladding material. However, there are a few NMCA plants that still use  $> 0.4$  ppb feedwater zinc because of the overwhelming benefits of zinc addition on radiation field reduction without any impact on long-term fuel performance.

Understanding the interaction of water chemistry parameters with fuel cladding material and the potential adverse impact of tenacious crud on heat transfer, and cladding corrosion and hydriding has become more important as local fuel rod power peaking tend to increase with higher duty, higher burnup and longer cycle fuel design and operation. Finally, fuel surveillance results indicate the need to further minimize Zircaloy-2 cladding corrosion to reduce the effect of tenacious crud on surface spallation [2].

### BWR Fuel Failure Rates

It was shown earlier in Figure 1 that the US industry annual average fuel failure rates in number of failed fuel assemblies per gigawatt electricity produced for both BWRs and PWRs. About eight BWR4 and BWR 5 units in the US

In the last decade, BWR fuel failure rates have been decreasing systematically, and Figure 13 shows the BWR fuel assembly leak rates from 1994 to 2006 for US, Japan and European region [14]. The Figure shows leaker rate maximums in 1998 and 2003 for US while the leaker rate in Japan was close to zero. The average fuel assembly (FA) failure rates per 1000 discharge FAs for the entire period 1994 to 2006 are : World average – 4.4, US – 5.4 (4.3 if massive BF2 failures were eliminated), Japan – 0.4 and Europe (Finland, Germany, Spain, Sweden, Switzerland) – 6.8.

The distribution of failure causes in the US BWRs is shown in Figure 14. Accelerated cladding corrosion and tenacious crud deposition were identified as being behind massive failures in 1998 (46 failures were due to crud induced localized corrosion-CILC [15] and in 2003 (63 crud/corrosion induced failures in the Browns Ferry Unit 2 during cycle 12 [8]. If BF-2 failures are excluded from the statistics, corrosion/crud and debris related fuel failures in US BWRs are at the same level.

Debris related failures dominate in European BWRs with a significant amount of PCI-SCC failure types before 2002 as shown in Figure 15, with a decreasing trend of debris failures after 2002 [14].

Figure 16 illustrates an estimate of BWR fuel failure causes worldwide. It is seen that crud/corrosion and debris related failures occur at the same frequency. PCI-SCC related failures are more or less uniformly distributed throughout the period 1994–2006, practically independent of the implementation of barrier cladding. Fabrication related failures were not observed during the last three years (2004–2006). However, the significant fraction of unknown failures is still a concern.

The fuel rod leaker rate comparison of all light water reactors over 4 year periods beginning from 1987 is presented in Figure 17. Overall, there is a decreasing trend of fuel rod leaker rates of LWRs since 1987.

### The Delicate Journey Towards Zero Fuel Leakers

Figure 18 shows the percentage of leaker free BWR units

worldwide and in Japan for the period 1994–2006, and in the United States for the 2000–2006 time frame. The average value for Japan throughout 1994–2006 was 96.0%. The United States and worldwide averages for 2000–2006 were, 63.3% and 77.6% respectively.

The delicate journey of trying to achieve zero fuel leakers by 2010 in the US was hampered by the constantly changing reactor operating conditions, including core management changes and power uprates, and the ever evolving water chemistry conditions. Thus, the journey towards zero fuel leakers has not been smooth because of these changes. However, it must be noted that in the US, no corrosion related fuel failures have occurred over the past decade, which is extremely encouraging.

In March 2013, US BWR customers operated over 1.4 million fuel rods with no leakers, a milestone that opened the door for a new era of fuel reliability [16]. Implementation of lessons learned from the past failure events has played the most important role in the systematic identification and elimination of failure mechanisms. As an example,

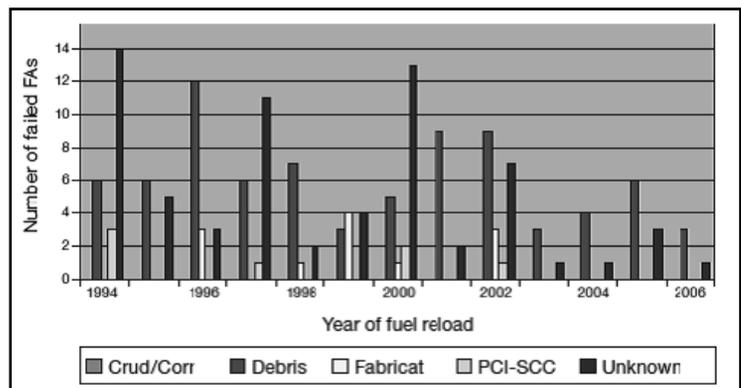


Figure 15. BWR Fuel Failure Causes in Europe [14].

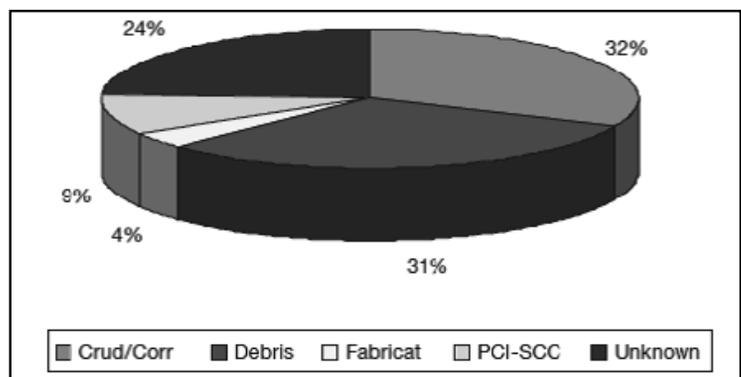


Figure 16. Estimated World Distribution of BWR Fuel Failure Causes [14].

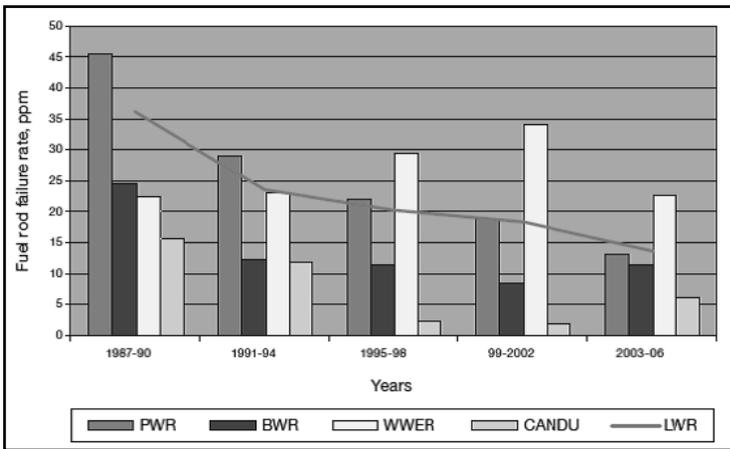


Figure 17. Fuel rod leakage rate calculated for the period 1987–2006 [14].

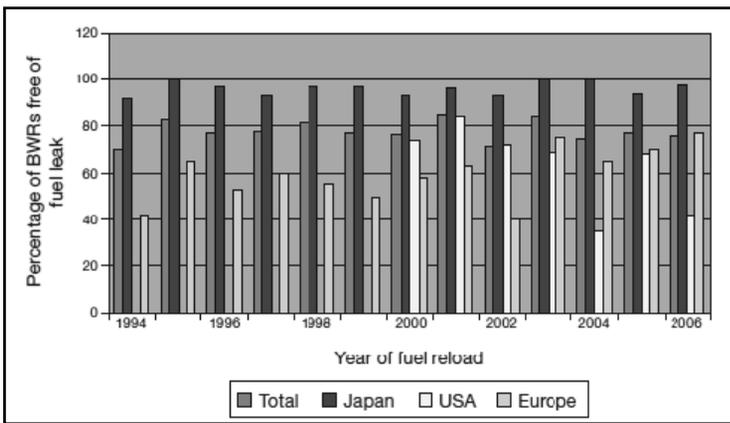


Figure 18. Percentage of BWRs with Zero Fuel Leakers up to 2006 [14].

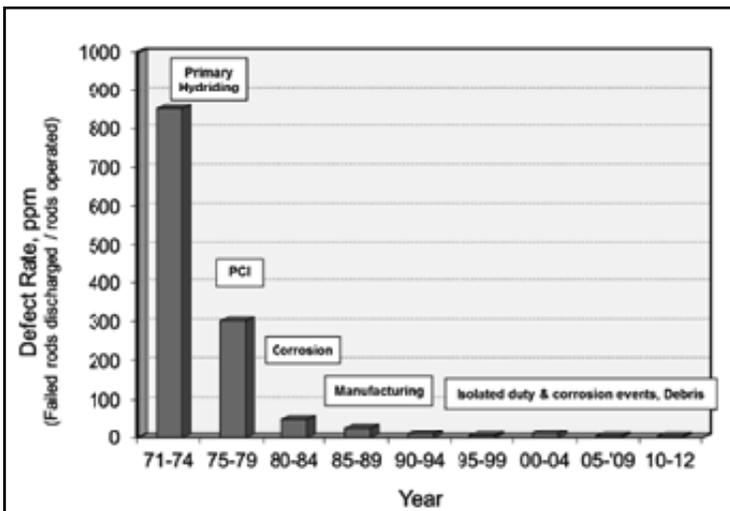


Figure 19. The Path to Zero Fuel Failures Over the 40 Plus Years [16].

debris filters have been installed in plants because of the results of higher debris inventories experienced in a smaller number of plants, but the entire BWR fleet benefitted from this experience.

On the journey towards reaching zero fuel failures, there are four broad failure mechanisms that have affected BWR fuel over the past twenty or so years: debris fretting; duty-related or PCI (pellet-clad interaction) type; manufacturing defects; and crud or corrosion. Three of these challenges have been largely resolved in the recent years. Crud/corrosion has not affected a US plant in approximately 10 years, manufacturing related failures have been eliminated for the most part, and PCI-type failures are rare now, largely due to the widespread implementation of operating practices to reduce the duty applied to the fuel. Recently, debris fretting has been the failure mechanism that has affected the most plants, caused the most fuel failures, and has been the most difficult to eliminate. Three factors are most important in the U.S. BWR fleet debris failure rate improvement seen since 2006 through today [16]. They are:

Reloads with lower-tie plate debris filters began operating in 2006 and are now near 100% of most Global Nuclear Fuel supplied cores [16].

Plants have been informed of the increased susceptibility to debris failures in BWRs with pumped forward feedwater heater drains. Most plants with this configuration, including a BWR/5, a BWR/6 and two BWR/4 units in the US, installed strainers in the heater drain lines to help protect this otherwise unfiltered stream that is approximately 35% of feedwater flow. All have seen their debris fretting failure rate decline dramatically. A BWR in Sweden opted to switch to cascade drains during

a 2011 mid-cycle outage and has operated since that time without a failure (after experiencing 19 debris failures in 5 annual cycles just before this change) [16].

Many plants where repeat debris failures had occurred significantly strengthened their Foreign Material Exclusion (FME) programs and practices [16].

The results of these efforts are reflected in the successful march towards zero leakers in the US as shown in Figure 19 [16]. However, sustaining this success is not an easy task since a single fuel failure event can affect the success achieved over many years concerted effort. Thus, it is critical to pay detailed attention to every activity that can potentially affect fuel failures in the BWR industry.

## 2. Conclusions

Due to sustained efforts, the journey towards zero fuel leakers in BWRs has been successful despite many challenges faced under demanding plant operating and advanced water chemistry conditions. CILC type corrosion has been eliminated by improving the cladding material and eliminating the copper source. Crud induced corrosion has been eliminated by controlling feedwater zinc and iron levels. Manufacturing related failures have been eliminated by paying more attention to material specification and PCI type failures have been circumvented by improved operating practices to reduce duty on fuel materials. Debris fretting related failures has been eliminated by implementing debris filters, modifications to forward pumped heater drain systems, and paying critical attention to FME programs. The challenge is to maintain the successes achieved with intense attention and scrutiny.

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