

Key Emerging Issues and Recent Progress Related to Structural Material Degradation (BWRs and some PWRs)

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1 Introduction

The first “Fontevraud” conference was held in 1985. The main objective of this conference was to highlight the failure root cause analysis work performed by French laboratories, especially the EDF labs. A second objective was to allow Utilities to exchange information regarding field failures and plant maintenance.

The operating and maintenance experiences targeted by the “Fontevraud” conference are those resulting from materials failure or degradation.

From 1985 (“Fontevraud I”) to 2006 (“Fontevraud VI”), the conference took place in the Royal Abbey of Fontevraud, one of the largest monastic cities in the western Christendom, right in the heart of the Loire Valley. The conference then moved to Avignon in 2010 (“Fontevraud VII”) and was also held in the Popes’ Palace in 2014 (“Fontevraud VIII”), however, the original name of the symposium was maintained, due to its unique focus on materials failure and degradation issues.

Originally, “Fontevraud” focused only on PWRs but in 2006, the scope of the conference was expanded to include BWRs too. This same year, a track dedicated to civil engineering was also added to the program.

165 papers were presented at “Fontevraud VIII”, in September 2014, with the following breakdown:

- 108 papers dedicated to PWRs materials issues;
- 17 papers dedicated to BWRs materials issues;
- 17 papers applying to both PWRs and BWRs;
- 23 papers in the “civil engineering” track.

This report covers a total of 43 papers presented at the Conference that include 17 BWR papers, 3 BWR/PWR papers and 23 papers that comes under the civil engineering track.

The BWR/PWR papers are categorized under several headings such as 1) Irradiation effects on microstructure and segregation, 2) Irradiation assisted stress corrosion cracking (IASCC) initiation and susceptibility, 3) Crack growth in irradiated and unirradiated conditions, 4) Environmental fatigue, 5) Environmentally assisted cracking (EAC) of low alloy steel in BWR conditions, and 6) Miscellaneous topics that include bearing issues, colloidal effects and fuel performance.

The civil engineering papers that deal with concrete structures are categorized into several topics such as, 1) Structural issues, 2) Concrete containment buildings (CCBs), 3) Cooling Towers, 4) Corrosion and alkali-silica reaction, 5) Radiation effects on concrete, 6) Monitoring and 7) Miscellaneous topics.

2 Irradiation effects on microstructure and segregation of elements

Stainless steels exposed to neutron irradiation during service in light water reactors (LWR) can become susceptible to intergranular cracking, referred to as irradiation assisted stress corrosion cracking (IASCC). There are many different factors suspected of contributing to the cracking susceptibility, but with respect to the material itself they include material hardening, the localization of plasticity, and radiation induced segregation (RIS) at the grain boundaries.

The material phenomena cited above are a consequence of the cascade damage produced by the incursion of high-energy neutrons, which introduce interstitial, and vacancy-type point defects and clusters into the metal lattice. The presence, migration, and coalescence of these species introduce a variety of significant changes in the material microstructure, which in turn affect the material's performance. The hardening is generally attributed to the presence of small dislocation loops in the microstructure, which restrict dislocation movement, and therefore material flow, which is then manifested as an increase in the yield strength [Karlsen et al, 2014].

Radiation Induced Segregation (RIS) is a consequence of the irradiation-enhanced diffusion encouraging atoms like Ni and Si to enrich at the grain boundaries, while Cr becomes depleted at the grain boundaries [Karlsen et al, 2014].

Material hardening itself is not sufficient to produce intergranular stress corrosion cracking (IGSCC), which has been demonstrated by comparing the cracking susceptibility of materials hardened by cold-work with those hardened by irradiation, which found greater cracking susceptibility in the latter. When the hardness is accompanied by localization of plasticity, the consequent deposition of dislocations at the point of impingement on the grain boundary can result in local stresses and consequent sliding at the grain boundaries, which in turn can promote cracking. This has been shown to be true even of some cold-worked stainless steels, but is particularly important in irradiated materials [Karlsen et al, 2014].

On the other hand, if Cr is depleted in oxidizing LWR conditions, a stainless steel's passive oxide layer can be compromised, leading to susceptibility to cracking. Nonetheless, annealing experiments on proton-irradiated materials have suggested that Cr depletion alone cannot cause IASCC. However, it has also been speculated that Si is readily dissolved in hot-water conditions, so a grain boundary enriched in Si may promote corrosion processes at the grain boundary, which in turn can make them more susceptible to cracking [Karlsen et al, 2014].

The dislocation loop size distribution is plotted as a histogram for the material condition in Figure 2-1. The quantification shows that at the lowest annealing temperature and shortest time (500°C/6h) there was a reduction in the proportion of the smallest loops and a consequent increase in the proportion of the larger loops, which led to a broader distribution of loop sizes. Similarly, increasing the anneal time and temperature further reduced the density of loops and promoted a broader range of loop sizes.

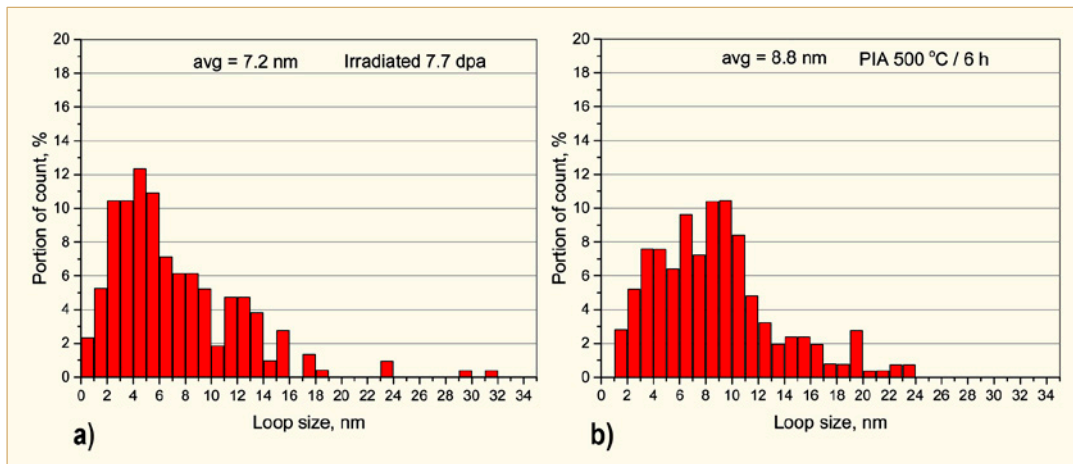


Figure 2-1: Distribution of dislocation loop sizes in 7.7 dpa 304 stainless steel in as-irradiated (a), and PIA heat-treated for 6 h at 500°C (b) [Karlson et al, 2014].

Marked radiation induced segregation was found in the as-irradiated state of the material, while marked reduction in the level of segregation was apparent after PIA at 500°C for 25 h, Figure 2-2.

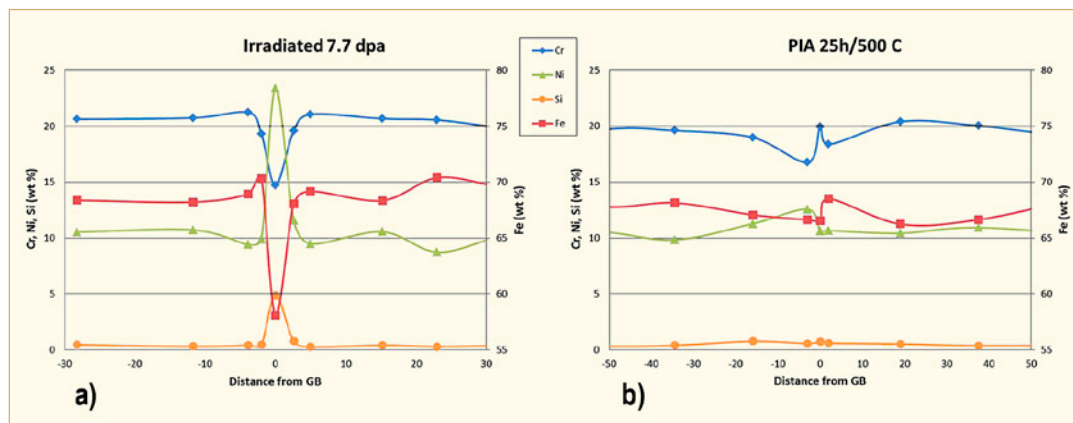


Figure 2-2: Example EDS profiles at a grain boundary before (a), and after PIA for 25 hours at 500°C (b) [Karlson et al, 2014].

Another paper investigated the microstructural changes in the RPV steels at low flux/low fluence conditions in BWR surveillance capsules [Nishida et al, 2014a]. Embrittlement of reactor pressure vessel (RPV) materials at low flux/low fluence conditions is of concern for BWRs. Therefore, in order to enhance the BWR surveillance database, Electric Power Research Institute (EPRI) conducted a Supplemental Surveillance Program (SSP) where a wide range of commercial RPV steels were irradiated in U.S. commercial reactors at various fluence and flux conditions. Seventeen materials of both base metals and weld materials containing different Cu and Ni contents, Cu: 0.22 wt.%, Ni: 0.51 wt.%; Cu: 0.16 wt.% Ni: 0.62-0.64 wt.%; Cu: 0.11 wt.%, Ni: 0.60-0.61 wt.%, irradiated in the supplemental surveillance program were examined in this study.

A relatively linear correlation was found between $\sqrt{V_f}$ (Microstructure) and ΔT_{41J} (Mechanical property) for all materials as shown in Figure 2-3.

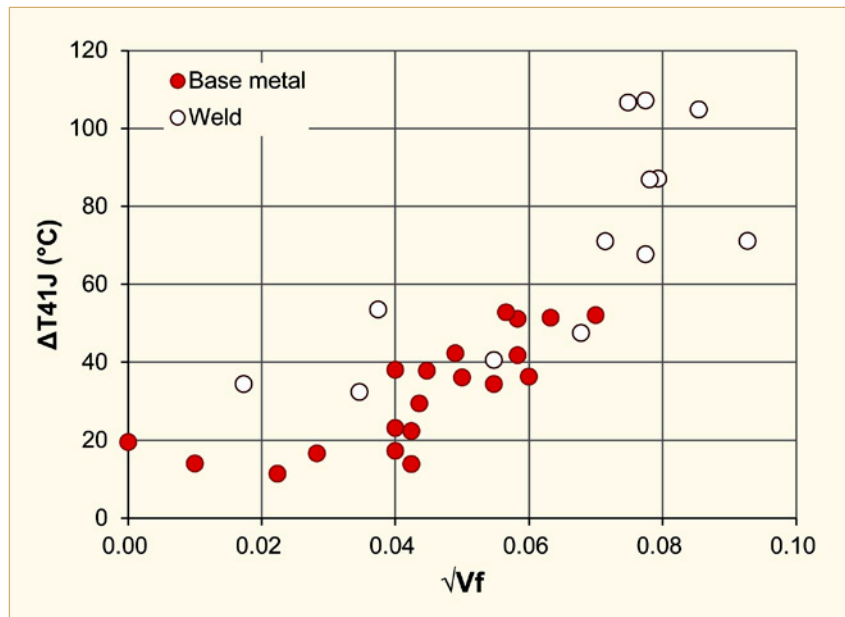


Figure 2-3: The linear relationship between microstructure $\sqrt{V_f}$ and mechanical property ΔT_{41J} [Nishida et al, 2014a].

The flux effect on the material properties can be summarized as follows [Nishida et al, 2014a]:

- Flux effect is evident in higher Cu materials.
- ΔT_{41J} becomes larger in higher Cu and lower flux conditions.
- Cluster number density (Nd) becomes higher in higher Cu and lower flux conditions.
- Cluster diameter is similar in $\sim 10^9$ and $\sim 10^{10}$ n/cm²/s flux conditions, but is smaller at higher flux of $\sim 10^{12}$ n/cm²/s. The cluster size is larger at lower flux.
- Change in V_f (microstructure) is consistent with change in ΔT_{41J} (mechanical property). The volume fraction is larger at low flux conditions.
- Clusters are formed between Ni/Si/Mn with little copper atoms, and Ni enhances the formation of clusters.

In LWR operation, the fracture toughness of irradiated steels decreases with increasing fluence as shown in Figure 2-4. Although the microstructure of the irradiated material is expected to play a role in the deterioration in fracture toughness, the relationship is not clearly understood [Chou et al, 2014]. Figure 2-5 summarizes data collected from irradiated stainless steels retrieved from BWR components. The fracture toughness clearly decreases with increasing fluence, but the Frank loop sizes and densities are essentially unchanged across the fluence range. Therefore, Frank loops cannot be the only significant radiation-induced defects that degrade fracture toughness. However, prior studies have not identified other likely factors either [Chou et al, 2014]. The paper discusses irradiation-induced defects, detected by atom probe tomography (APT), as potential candidates.

3 Crack initiation and irradiation assisted stress corrosion cracking (IASCC) susceptibility

The objective of the crack initiation work was to follow slip band emergence as a precursor event for initiation of SCC on unirradiated type 304L SS in two different environmental conditions. The important findings of the study [Khan et al, 2014] are:

- 1) Slip band emergence was observed to act as a precursor event for SCC. Emergence of the slip bands in air and the preferential attack along the slip bands in chloride containing environment at strains as low as 0.35% were observed by three different microscopic techniques.
- 2) The SCC attack in chloride environment resulted in deeper inter-slip band (roughness) regions as compared to those in the specimens tested in air at room temperature (26°C). This further supports the hypothesis of slip bands being preferentially attacked and thereby acting as precursor for SCC.
- 3) Formation of locally strained regions inside the grains was a main feature observed during SSRT in air. The enhanced slip band heights at higher strains were also observed, causing an increase in surface roughness. In addition, Cracks due to SCC were observed to follow the path of slip bands for their growth.
- 4) SCC of type 304L SS in chloride environment (0.5M NaCl + 0.5M H₂SO₄) was evident at room temperature on straining to and beyond 3.5% strain, Figure 3-1. However, at very low strain levels, features of only localized attack were observed and can be attributed to preferential corrosion attack on adjacent slip bands within individual grains.
- 5) The SSR test carried out in high temperature and high pressure water (288°C, 10 MPa, simulating BWR conditions with around 1 ppm DO) up to a strain of 3.55% did result in initiation of fine stress corrosion cracks. The formation of slip bands disturbing the oxide layer caused preferential oxidation thus acted as a precursor to crack initiation. This confirms the effectiveness of slip step emergence as a precursor event for SCC initiation in BWR simulated environment. The oxide formed on type 304L SS under these conditions manifested as an inner adherent layer and an outer layer with big crystallites.

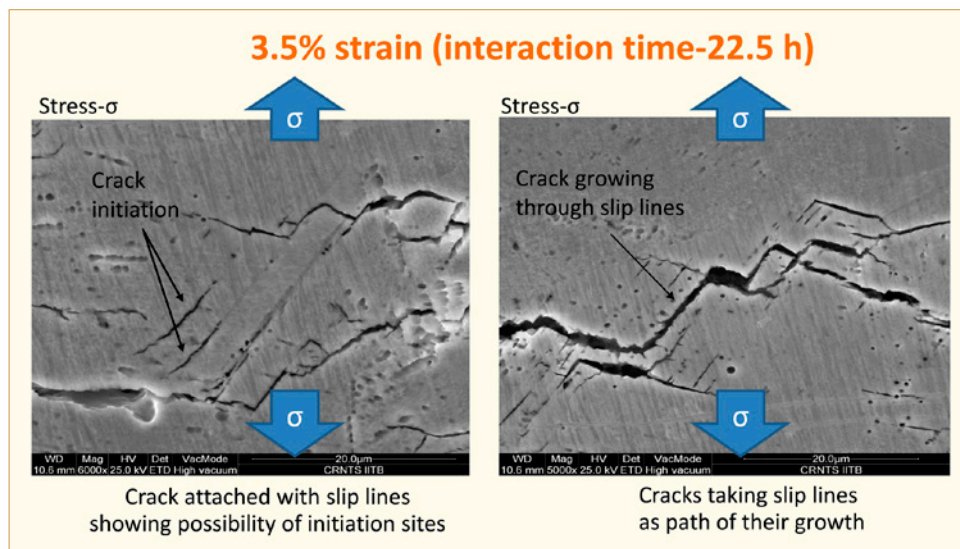


Figure 3-1: SEM images in gauge length of the specimen after SSRT in 0.5M NaCl + 0.5M H₂SO₄ at 26°C for 3.5% strain showing crack linked with slip lines [Khan et al, 2014].

4 Crack growth in irradiated and un-irradiated conditions

The integrity of internal components of reactor pressure vessels of nuclear power plants is of significant importance regarding safe operation and shut-down. Any degradation by ageing, and in particular by cracking therefore is of highest interest and concern. Cracks may be initiated and grow due to mechanisms of environmentally assisted cracking (EAC). The most prominent modes of EAC concerning RPV-internals cracking are Inter-Granular Stress Corrosion Cracking (IGSCC) and Irradiation Assisted Stress Corrosion Cracking (IASCC). Susceptibility to IGSCC may be caused by either thermal sensitization of heat-affected zones at welds or by severe cold work. IASCC describes a particular mechanism of stress corrosion cracking, where susceptibility to intergranular environmentally assisted cracking is created by fast neutron irradiation. Fast neutron flux and fluence, as well as γ -radiation alter the parameters determining a corrosion system that consists of the given combination of material, environment and mechanical stress. This may lead to conditions with inherent susceptibility to environmentally assisted cracking. During the last decades numerous cracking incidents were observed in internals of both, BWRs and PWRs. This is a review paper that summarizes field failures in BWR RPV-internals due to intergranular cracking with a particular emphasis on BWR core shrouds [Roth, 2014].

The irradiated condition of a material shows a characteristic microstructure (interstitials, dislocations, voids, bubbles), a characteristic microchemistry (segregation of individual elements at grain boundaries) and a characteristic mechanical behaviour (increased strength and hardness, reduced macroscopic plasticity, localized deformation). Key phenomena of irradiation effects are well understood, and some remedial measures for field applications were successfully identified. In particular, it was revealed that the susceptibility to IASCC cannot be attributed to a single parameter or a combination of few parameters. Engineering assessment of RPV internals component behaviour is thus still a complex issue. Operational experience has not revealed complete failure or loss of integrity of materials in use [Roth, 2014].

Regarding the critical degree of irradiation, the accumulation of a threshold dose (or fluence) of fast neutrons has revealed to be a necessary, but still not a sufficient criterion. Critical fluence thresholds are different for BWR or PWR condition, based on existing observations from laboratory testing and field experiences (i.e. service failures). Typical values of critical fast neutron fluence thresholds are shown in Figure 4-1.

One of the most widespread degradation mechanisms that affected several BWR plants from different vendors worldwide is the intergranular cracking of core shrouds. IGSCC as well as IASCC were reported as the mechanisms of cracking. Thermal sensitization or neutron irradiation, surface cold work, weld residual stress, among others were identified as the root causes of stress corrosion cracking in such cases [Roth, 2014].

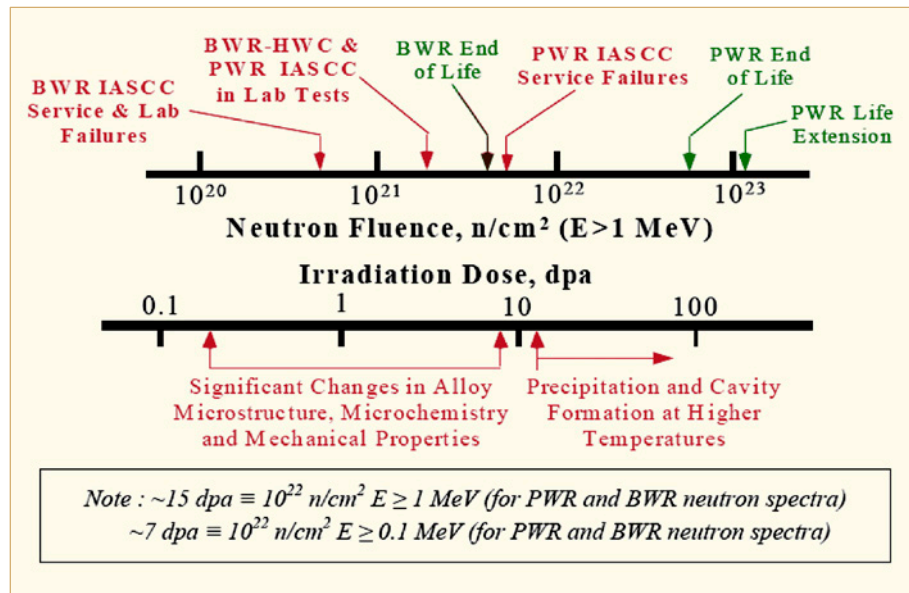


Figure 4-1: Threshold fluences for IASCC based on Bruemmer [Roth, 2014].

A variety of BWRs showed core shroud cracking since the 1990's. An example of cracking in a German BWR is shown in Figure 4-2. Based on the results from the failure analysis, the root cause of cracking was identified as IGSCC due to thermal sensitization, which occurred in this specific heat of concern despite stabilization with Niobium. It was revealed by the failure analysis that the 347 SS heat used for the upper and lower core support ring did not fulfil the specified requirements for the minimum stabilization ratio of Nb/C, as it was rather high in carbon and low in niobium. The Nb/C ratio (8 for the melt, 3.9 for the taken specimen) of this heat was much lower as the specified requirement of Nb/C ≥ 13 . However, core shrouds of other BWRs in Germany, were free of crack indications [Roth, 2014].

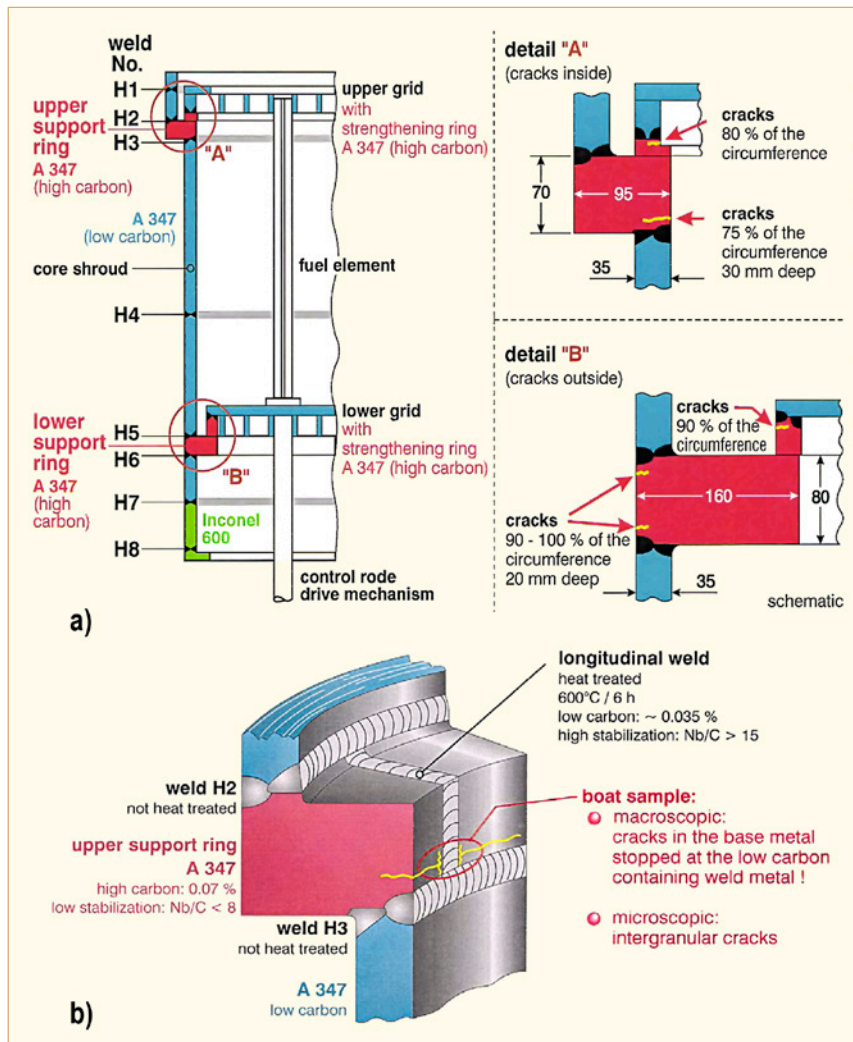


Figure 4-2: a) Location of cracks in the core shroud of Würgassen BWR (KWW), and b) location of cracks in the upper core support ring and material related information, [Roth, 2014].

Despite numerous similarities of the known core shroud cracking incidents, resulting actions worldwide have been significantly different as shown below.

- Case 1:

Cracks were considered as acceptable, provided the introduction of extended NDE and a variety of remedial measures, such as the application of tie rods and modifications in water chemistry. This was the most common reaction.

- Case 2:

Cracks in the core shroud were considered as not acceptable and core shrouds were thus replaced to allow further operation of these plants. This was applied in some cases, e.g. in some NPPs in Japan.

- Case 3:

Cracks were considered as not acceptable. However replacement or other repair was not performed due to economic reasons. The final solution was then a decommissioning of the plant. This was the case with KWW.

5 Environmental fatigue in stainless steel and environmentally assisted cracking of low alloy steel in LWR conditions

A paper from Switzerland addressed the environmental-assisted fatigue (EAF) initiation and subsequent short crack growth behaviour of different austenitic stainless steels that were characterized under simulated BWR/PWR conditions by cyclic fatigue tests with sharply notched fracture mechanics specimens. The paper discussed the effect of pH, dissolved hydrogen, load ratio/mean stress, long static load hold times and load sequences on EAF [Seifert et al, 2014]. This section summarizes only the data generated under BWR conditions.

At low ECP, the physical EAF initiation life moderately decreases with increasing dissolved hydrogen content and decreasing pH. Both parameters have little effect on the subsequent short EAF crack growth within the investigated range. Notch strain amplitude thresholds for environmental effects on physical EAF crack initiation decrease with increasing load ratio and mean stress. At small notch strain amplitudes, the effect of mean stress is more pronounced in BWR/HWC environment than in air and predicted by typical fatigue life mean stress corrections. Under certain loading conditions, long static load hold times result in an increase of the physical EAF initiation life, which saturates for very long hold times. On the other hand, little effect of hold times on subsequent stationary short EAF crack growth rates is observed. The physical EAF initiation life under load sequence loading in high-temperature water may be moderately shorter or significantly longer than predicted by a linear damage accumulation rule and corresponding constant load amplitude tests depending on the load history [Seifert et al, 2014].

At low ECPs under BWR/HWC conditions, a relevant environmental reduction of fatigue initiation life occurs in all investigated low-carbon and stabilized SSs for the combination of notch strain rates $\leq \sim 0.1\%/s$, temperatures $\geq \sim 100^\circ C$ notch strain amplitudes $\geq \sim 0.3\%$. The behaviour of stabilised and low-carbon SSs is very similar and sensitization affects the EAF behaviour under highly oxidizing BWR/ normal water chemistry (NWC) conditions and slow strain rates only. BWR/HWC environments usually result in acceleration of short fatigue crack growth by a factor of 3 to 20 with respect to air.

Figure 5-1 shows the effect of notch-root stress amplitude $S_{a,LEFM}$ and load ratio on EAF initiation in simulated HWC environment at $288^\circ C$ at a strain rate of $4 \times 10^{-2}\%/s$ for different load ratios. An increase of the load ratio from 0 to 0.7 and slight positive mean stress results in a strong reduction of the physical EAF crack initiation life in HWC environment. Furthermore, the stress thresholds for environmental effects seem to decrease with increasing load ratio.

A tremendous effect of load ratio is observed, if the notch stress amplitude at the low load ratio is below the stress threshold for environmental effects as shown in Figure 5-2. Here, EAF life is reduced by a factor of 60 to 100, when increasing the external load ratio from 0.05 to 0.5. In corresponding tests in air at room temperature, fatigue life is reduced by a factor of ~ 2 .

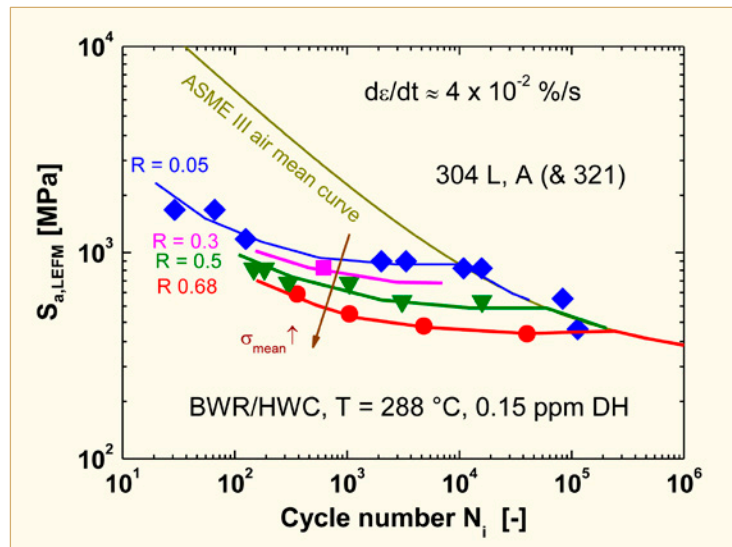


Figure 5-1: The effect of notch stress amplitude and load ratio (mean stress) on EAF initiation life [Seifert et al, 2014].

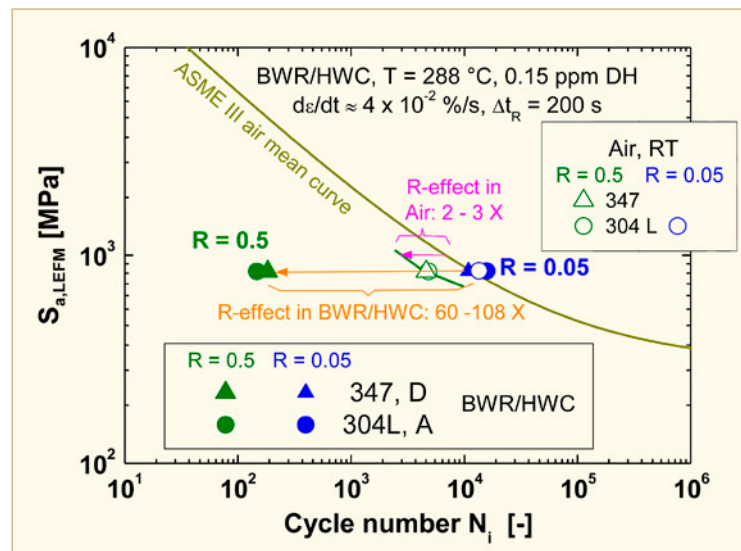


Figure 5-2: The effect of load ratio R on EAF initiation at a notch stress amplitude of 840 MPa [Seifert et al, 2014].

The study also included evaluation of EAF initiation life under different static load holding times, and authors concluded that these require further studies.

A German study undertook the evaluation of chloride ions on the EAC of low alloy steels (LAS) under BWR conditions. The pressure boundary components manufactured from LAS, e.g. the reactor pressure vessel, are usually not directly in contact with the primary coolant since they have an austenitic stainless steel cladding overlay. However, depending on the plant design, unclad pipes made of LAS may be in direct contact with the primary coolant, e.g. in feed water piping of some BWR designs. Furthermore, cladding discontinuities of the reactor pressure vessel surface may cause a direct contact between LAS and the primary coolant. This situation is postulated for conservative safety analyses. Therefore, for pro-active safety assessments it is assumed that cooling water can be in direct contact with the LAS materials of the pressure boundary [Herbst et al, 2014].

6 Miscellaneous topics

The papers below were selected to be covered under miscellaneous topics since they did not fall into the specific categories listed in the previous chapters. They are, 1) SCC mitigation in BWRs by platinum addition, 2) Colloid sampling, 3) Operational experience with 900mm R1T pocket-type bearings at Oskarshamn unit 3 nuclear steam turbine generator, and 4) Fuel related papers. Since these papers are on unrelated topics, they are discussed under different sub headings in this chapter.

6.1 SCC mitigation in BWRs by platinum addition: effect of environment and injection rate

On-line noble metal chemical addition has been developed to mitigate stress corrosion cracking (SCC) in reactor internals and recirculation pipes of BWRs avoiding the negative side-effects of the conventional hydrogen water chemistry. For a more efficient reduction of the electrochemical corrosion potential (ECP) Pt is injected into the feed water during power operation. Pt deposits as very fine metallic particles on all water-wetted surfaces of reactor internals and appear to remain electro catalytic over long periods of time.

The gradual development of various BWR SCC mitigation technologies from hydrogen water chemistry (early 80's), to noble metal chemical addition – NMCA (early 90's), to On-line NMCA (early 2000's) is shown in Figure 6-1.

To assess the SCC mitigation by Pt addition technology, the deposition and distribution of Pt on stainless steel coupon specimens, exposed to simulated BWR water conditions, using a sophisticated high-temperature water loop, has been investigated in detail at Paul Scherrer Institute (PSI) in Switzerland. During the tests Pt solution was injected into the high temperature loop and Pt was deposited on the specimens. Scanning and Transmission Electron Microscopy techniques and Laser Ablation – Inductively Coupled Plasma – Mass Spectrometry were used to characterise the Pt distribution and concentration on the specimens [Ritter et al, 2014].

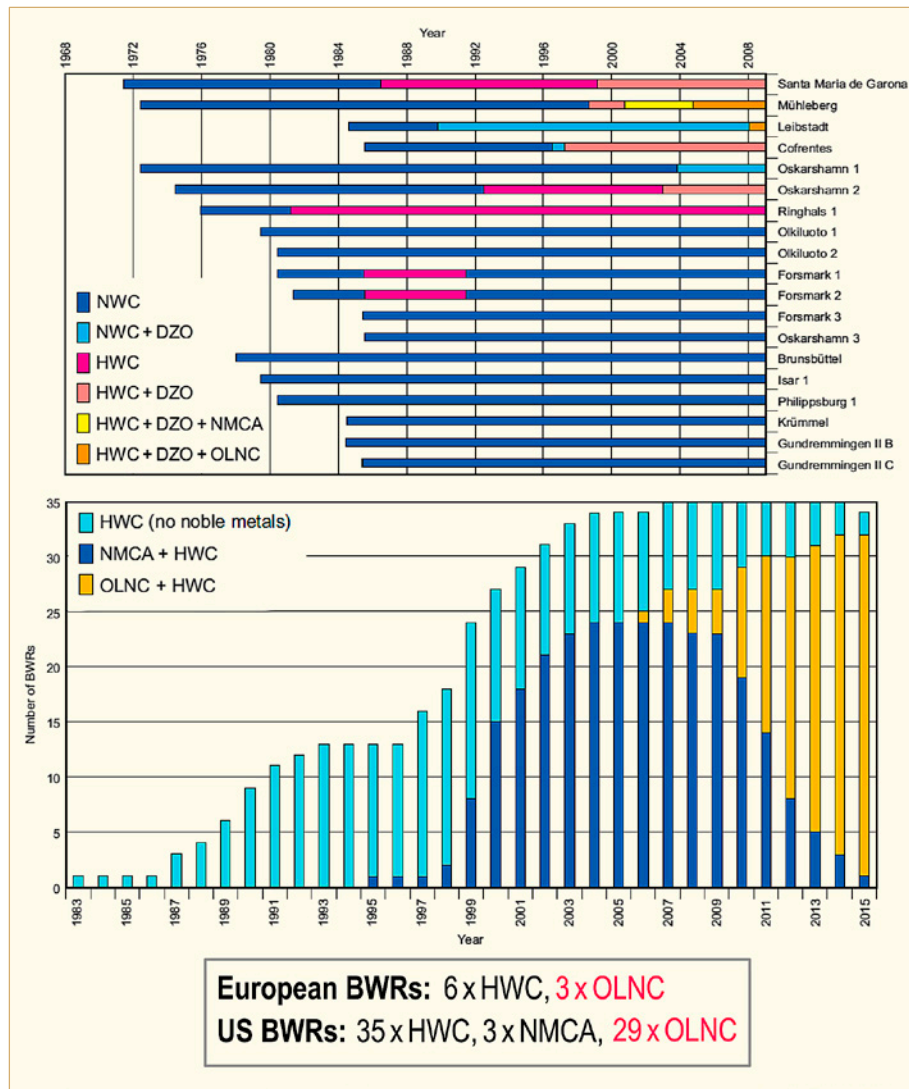


Figure 6-1: Application of SCC Mitigation Technologies in Europe and USA [Ritter et al, 2014].

This paper presents results from a set of experiments with varying concentrations of dissolved H_2 and O_2 in the high-temperature water, as well as results from tests with different Pt injection rates are presented and discussed. These experiments revealed a clear difference in the Pt deposition behaviour in terms of size and homogeneity in distribution depending on the water chemistry and Pt injection rate. A reducing environment and slow Pt injection rates resulted in the most effective Pt deposition, thus potentially in a better mitigation against SCC [Ritter et al, 2014].

As expected, it was found that the ECP of SS specimens responded more effectively to higher Pt addition rate as well as the total amount of Pt injected into the high temperature flow loop. Clearly, Figure 6-2a and Figure 6-2b show the SS potential response under reducing conditions to lower and higher Pt injection rates, respectively. Higher Pt injection rate provided a much lower ECP, as anticipated.

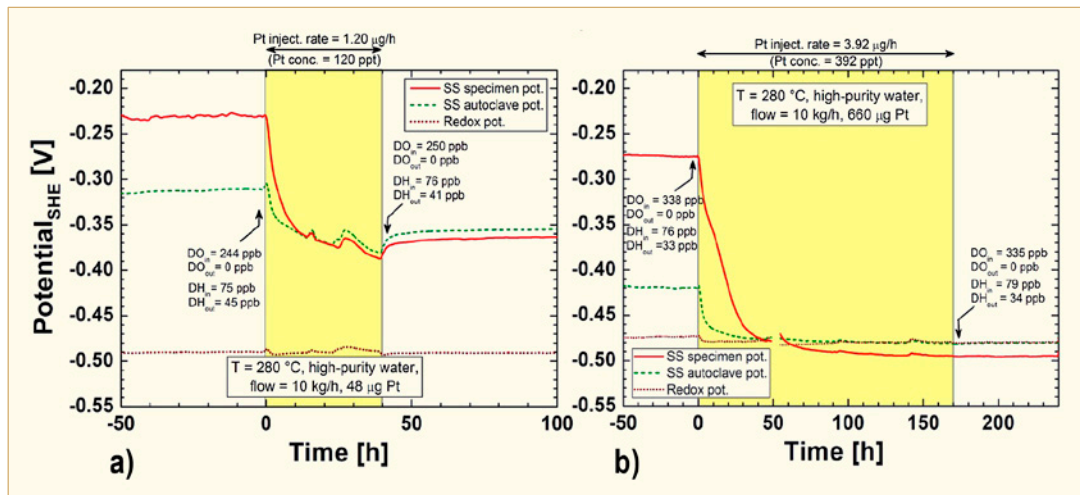


Figure 6-2: Redox potential response during different Pt injection rates [Ritter et al, 2014].

Figure 6-3 shows that the dissolved oxygen and hydrogen content in the feed water have a strong influence on the Pt particle size and its distribution. The fully reducing environment leads to finer Pt particles and a more homogenous distribution. The oxidising environment leads to much larger Pt particles and inhomogeneous distribution. Finer Pt particles, evenly distributed across the specimen surface, are regarded as advantageous for the mitigation of SCC compared to larger, inhomogeneously distributed, and therefore, fewer Pt particles [Ritter et al, 2014].

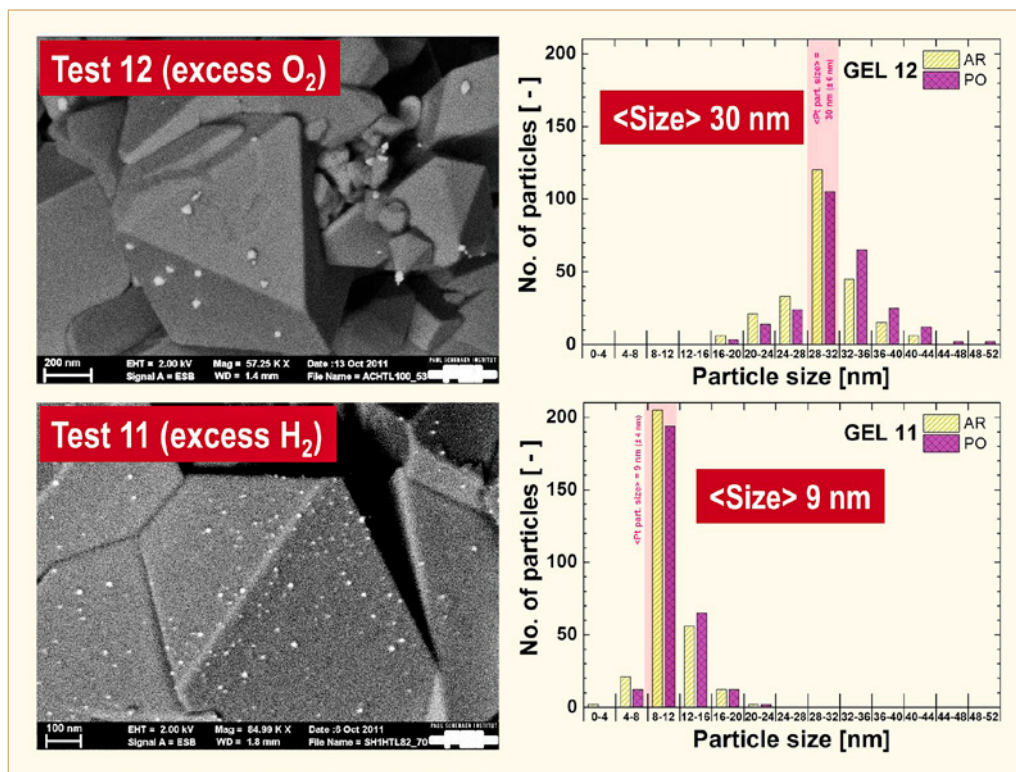


Figure 6-3: Effect of oxidising and reducing environment on Pt particle size [Ritter et al, 2014].

7 Integrity of concrete structures used in nuclear power plants

Some of the conference papers addressed civil engineering topics associated with nuclear power plant operation. This chapter of the report summarizes papers dealing with the integrity of concrete structures, which is becoming an important topic as more and more plants are seeking license renewal. The topics covered in this chapter include, aging and degradation of concrete structures, concrete containment buildings (CCBs), cooling towers (CTs), corrosion and alkali silica reaction (ASR)/ alkali aggregate reaction (AAR), radiation effects on concrete, monitoring and other miscellaneous topics.

7.1 Assessment of degradation and aging of nuclear power plants concrete structures

A paper from the US summarized the results of an expert-panel assessment of ageing degradation modes and mechanisms of concrete structures in NPPs, where, based on specific operating environments, degradation is likely to occur, or may have occurred; to define relevant aging and degradation modes and mechanisms; and to perform systematic assessment of the effects of these age-related degradation mechanisms on the future life of those materials and structures [Busby et al, 2014].

The following degradation modes and mechanisms have been identified as having the greatest potential impact on the ability of concrete structures to fulfil their safety related functions during long-term NPP operation [Busby et al, 2014]:

- 1) Corrosion of conventional reinforcement is difficult to assess because of inaccessibility to inspection.
- 2) Creep of pre-stressed concrete containments continuously affects the internal stress state and adds to tendon relaxation and gradual loss of pre-stress.
- 3) Irradiation of concrete lacks sufficient data for a clear evaluation of its effects on long-term operations.
- 4) Alkali-silica reaction potential consequences on the structural integrity of the containment.
- 5) Fracture/cracking, which is a well understood behaviour characteristic of concrete structures and is accounted for in structural design, plays a unique role in post-tensioned containments during de-tensioning and re-tensioning operations which may be undertaken as part of life extension retrofit work, resulting in delamination, and may evolve with time as a creep-cracking interaction mechanism.
- 6) Boric acid attack of concrete in the spent fuel pool involves knowledge gaps related to the kinetics and the extent of the attack (role of the concrete mix design).
- 7) Corrosion of the inaccessible side of the spent fuel pool and containment liners and the stress corrosion cracking of the tendons are important degradation modes due to the absence of in-service inspection.

In general, the performance of reinforced concrete structures in nuclear power plants has been very good. Incidents of degradation initially reported generally occurred early in the life of the structures and primarily have been attributed to construction/design deficiencies or improper material selection. This paper focuses on the expanded material degradation analysis (EMDA) that addresses aging and degradation of concrete materials in nuclear power plants (NPPs), with special emphasis on safety-related structures [Busby et al, 2014].

The NPP structures and their potential degradation modes are illustrated in Figure 7-1, Figure 7-2, Figure 7-3 and Figure 7-4.

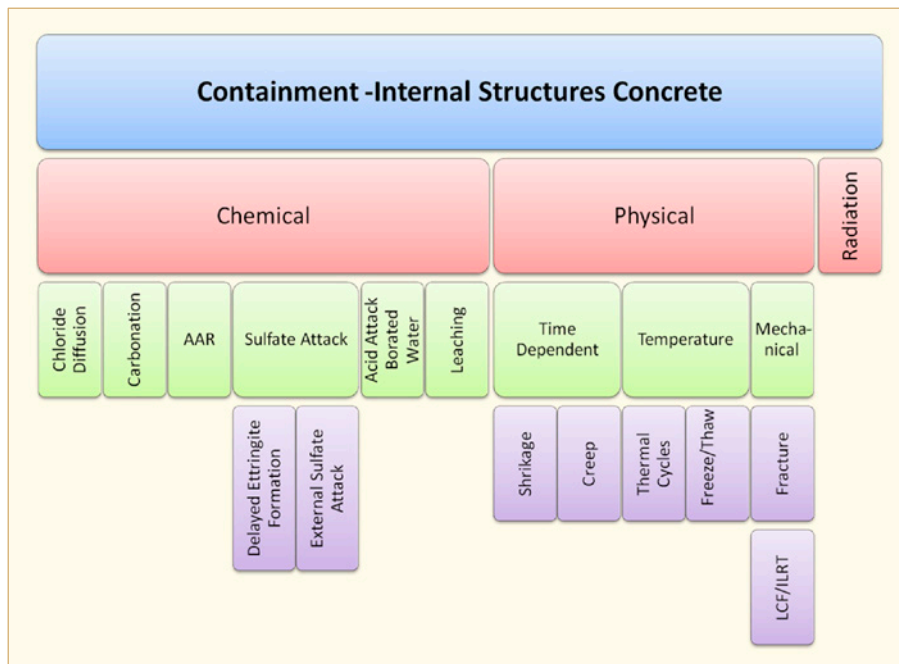


Figure 7-1: Degradation modes in reactor containments: concrete components [Busby et al, 2014].

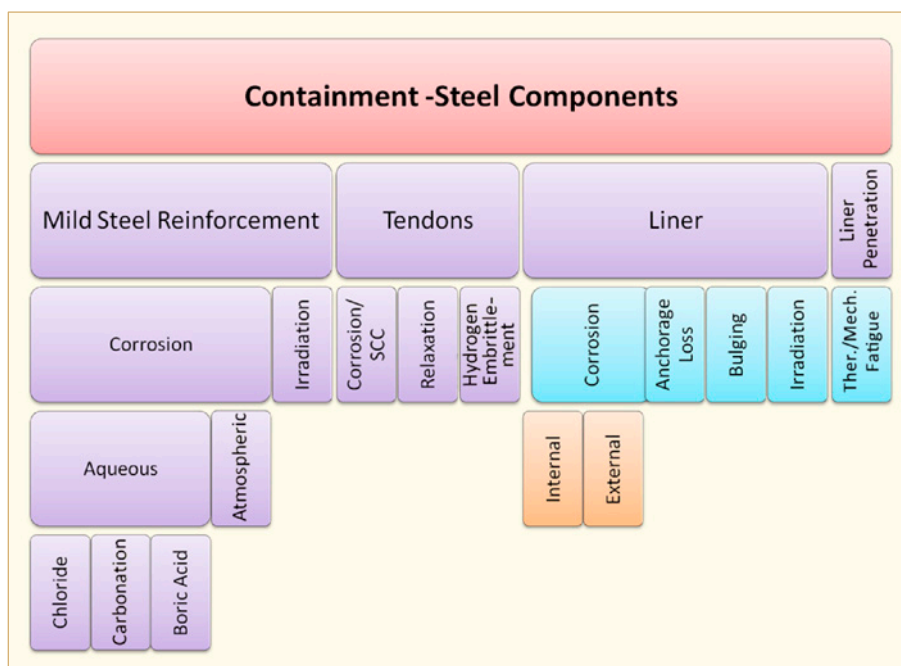


Figure 7-2: Degradation modes in reactor containments: steel components [Busby et al, 2014].

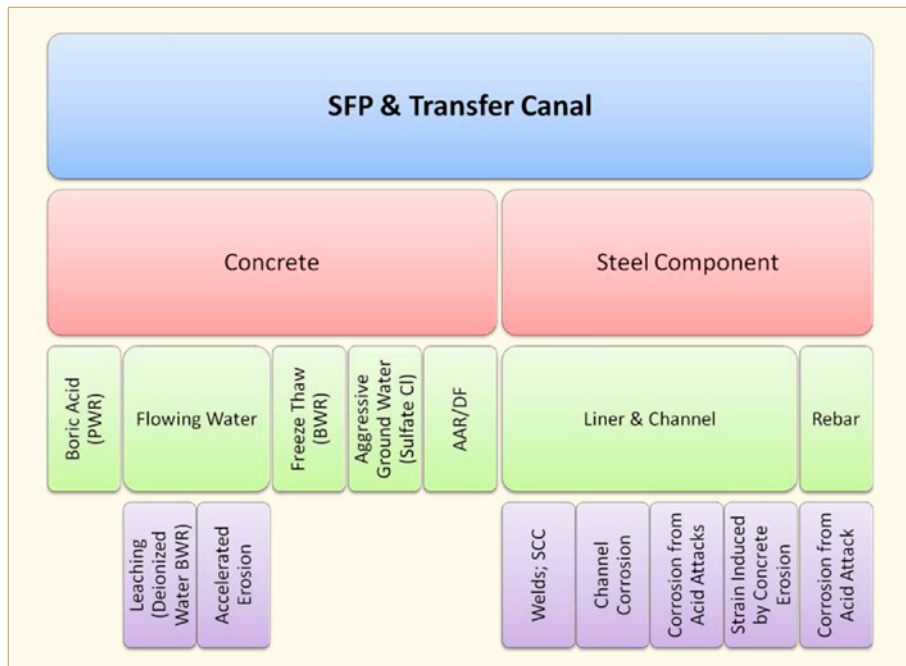


Figure 7-3: Degradation modes in spent fuel pool (SFP) and transfer canal [Busby et al, 2014].

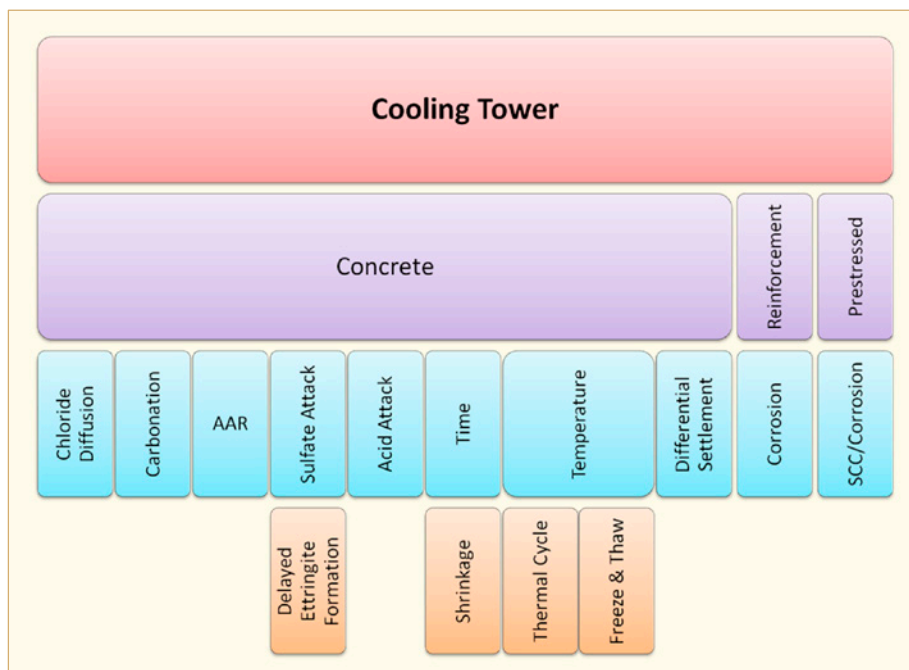


Figure 7-4: Degradation modes in cooling towers [Busby et al, 2014].

One of the areas that require further study is the impact of irradiation on the integrity of concrete structures. There is lack of data at higher neutron fluences as shown in Figure 7-5.

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