LCC6 SPECIAL REPORT

Key Results from Recent Conferences on Structural Materials Degradation in Water Cooled Reactors

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Contents

1 Introduction

Environmentally-assisted degradation of structural materials has presented an economic and safety problem to the water-cooled nuclear reactor industry for 35+ years.

The modes of degradation have encompassed general corrosion, Flow-Accelerated Corrosion (FAC) and a variety of localized corrosion modes associated with, for example, pitting, crevice corrosion, Stress Corrosion Cracking (SCC) and Corrosion Fatigue (CF). These localized corrosion modes have been of particular concern because early detection is difficult and this leads, therefore, to unexpected component failures. These issues have been widely reported in various conferences (Environmental Conferences) and utility workshops, and have been discussed in a series of ANT International Special Topical Reports [Ford, 2006], [Ford & Scott, 2008], [Ford et al, 2010] and [Scott et al, 2011].

Mitigation actions and aging management programmes have been developed to deal with these materials degradation issues. However, isolated incidences continue to occur even in “mitigated” systems, and these may be attributed to the complex interactions between the relevant stress, environment and material conditions that have not always been recognized or controlled during the development of the mitigation actions.

Moreover this oversight has been exacerbated by the reactive management approach that limits the time available to define fully the mitigation actions (Figure 1-1).

Consequently in the last few years attention [Muscara, 2007] and [Pathania, 2008] has focussed on the development of proactive management capabilities for the major degradation modes, whereby degradation events, which are not detectable currently, may be predicted to occur sometime in the future. Such proactive management capabilities would allow for timely decisions associated with, for instance, pre-emptive material replacement, or the definition of more effective inspection techniques and schedules.

The future life prediction capabilities may be developed via an amalgamation of knowledge of the details of past plant incidents, knowledge of the sensitivity of such degradation events to changes in the system conditions (material, environment, etc.), and an adequate understanding of the fundamental mechanism of the specific materials degradation mode (SCC, FAC, etc.). These developments have been discussed in the ANT International reports referenced above.

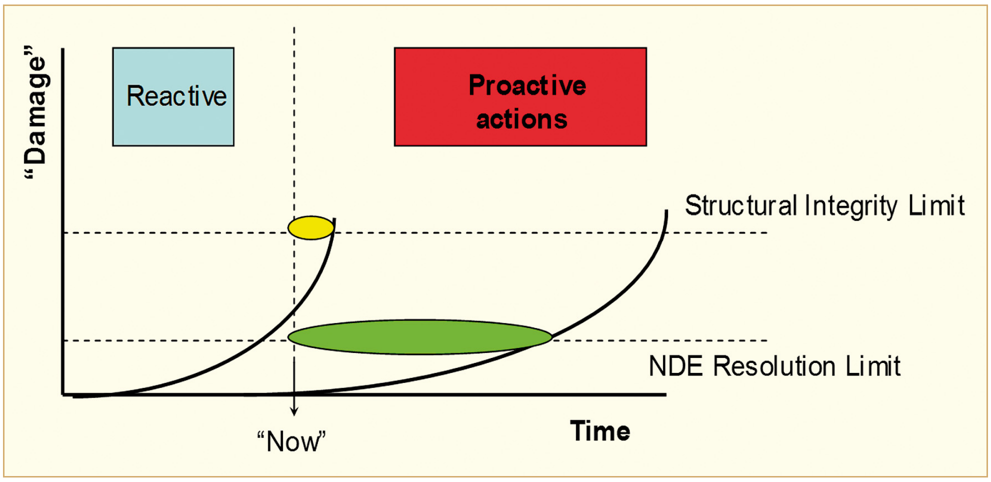

Figure 1-1: Comparison between reactive and proactive management methodologies for addressing materials degradation issues. Note that, in the reactive management approach, action is not taken until the problem has been detected and this leads to a limited time (denoted in yellow) for developing mitigation actions before a structural integrity limit is reached. However there would be a relaxation (denoted in green) in that time limitation if there were a prediction algorithm for the relevant degradation mode that foresees potential problems in the future.

Advances in our understanding of the relevant factors (i.e. the basic science foundation plus knowledge of the parametric interdependencies) that govern the various rates of degradation have been spasmodic and, because of this, it is difficult for someone not intimately involved in such developments to keep abreast of the overall advances in the required knowledge for a proactive management capability. Consequently ANT International decided to review the salient papers presented at recent materials degradation conferences.

The scope of this current report is to review papers published in the following major conferences;

- *"Proceedings of Fourteenth International Conference on Environmental Degradation in Nuclear Power Systems–Water Reactors", held at Virginia Beach USA, August 24-28th, 2009 and published by American Nuclear Society in 2010.*

- *"Contribution of Materials Investigations to Improve the Safety and Performance of Light Water Reactor (LWRs). Proceedings of the International Symposium Fontevraud VII", held in Avignon, 26-30 September, 2010 and published by SFEN (the French Nuclear Energy Society).*

These conferences addressed various degradation modes for a wide range of alloy-type/reactor-system combinations. In order to discuss these issues in a logical manner it was decided to discuss these degradation modes in terms of the impact on the reactor design; that is, materials degradation in Boiling Water Reactors (BWR) is discussed in Section 3 and in Pressurized Water Reactors (PWR) in Section 4.

In addition to these major conferences, a jubilee day took place on 26 January 2010, in honour of Henri Coriou, the French scientist who, 50 years ago, claimed the SCC sensitivity of Alloy 600 in pure water.

- *"Jubilee day: Stress Corrosion Cracking of nickel base alloys at CEA - Coriou Effect - CEA-INSTN, Saclay (near Paris), France - Tuesday January 26, 2010 - EFC event N°333.*

It is interesting, both on a scientific and historical point of view, to understand why this Alloy 600 has been largely used for such a long time, with so many failures that are still occurring in operating units although the weakness of this Alloy 600 had been published as early as in 1959. The presence of senior and retired experts who worked closely with Henri Coriou or had been involved in this controversial issue was a great opportunity to understand all the input data of this story, which is described in Section 2 of this report.

2 Seminar on stress corrosion cracking of nickel base alloys at CEA – Coriou effect – Saclay, France, 26 January 2010 (Francis Nordmann)

2.1 Introduction

For celebrating the 50 years of the 1st publication of Henri Coriou, French Engineer at Commissariat à l'Energie Atomique (CEA) on SCC of Nickel base alloys, at CEA under French Nuclear Energy Society (SFEN) organization, held a Jubilee day on January 26, 2010 where ANT International decided to have one participant for getting the papers.

116 attendees, manly French (45 from CEA) – 4 USA – 2 Japan – 6 Belgium – 4 Sweden – 3 UK – some from Germany, Spain, China, Slovenia, South Africa, etc.

The program of the day was the following:

1) Gérard Pinard Legry. La saga de l'Alliage 600 au CEA.
2) Roger Staehle. Historical views on the stress corrosion cracking of Ni base alloys.
3) Philippe Berge. History and small novels of the SCC of Nickel base alloys.
4) Peter L. Andresen. Understanding and prediction of Stress Corrosion Cracking.
5) E. Herms and I. de Currieres. R&D on initiation of stress corrosion cracking at CEA.
6) Toshio Yonezawa. Coriou's crack experience in Japanese PWRs and its mitigation.
7) Catherine Guerre. R&D on crack propagation of stress corrosion cracking at CEA.
8) Digby Macdonald. Stress corrosion cracking and electrochemistry.
9) Jacques Chêne. Corrosion cracking and hydrogen embrittlement.
10) Philippe Bossis. SCC test facilities at CEA.

The main purpose of this day was to explain why sufficient consideration has not been given to the sensitivity of Alloy 600 (Inconel 600) to SCC in pure water is still lacking. A sensitivity that was pointed out in already in 1959 by the French scientist, Henri Coriou, published [Coriou et al, 1959]. Consequently, many countries and manufacturers selected this sensitive Alloy as construction material, and later on, this has resulted in a tremendous amount of degradation and failures in operating units.

2.2 History of Alloy 600 SCC discovery

2.2.1 History of nuclear reactors

This brief history is based on a complete historical presentation by Roger Staehle of the early times of nuclear energy in the USA [Staehle, 2009]. Admiral Hyman George Rickover (January 27, 1900 – July 8, 1986) was the initiator of the nuclear industry with the Nautilus, at the beginning of military and commercial nuclear power. *USS Nautilus*, SSN 571 was launched on 21st January 1954. The *Nautilus* was powered by the S2W naval reactor, a PWR produced for the Navy by the Westinghouse Electric Corporation. The 60MWe Shippingport plant (Pennsylvania, USA) was the first commercial reactor brought into operation on 26 May 1958. It was designed and built by the Rickover program. The design was based on that to be used for the first aircraft carrier. Naval Reactors program influenced the entire world to the development of safe and reliable nuclear power. Members of the Naval Reactors program, naval personnel, scientists, and members of Rickover's staff became leaders of commercial organizations involved in nuclear power. Rickover developed new materials for nuclear plants including zirconium and hafnium for cladding and neutron control, respectively. He provided a paradigm for quality and technology for the US commercial nuclear program and from there for all other nuclear power programs outside USA. Rickover was known as the "Father of the Nuclear Navy". Rickover contributed to a worldwide legacy of commercial nuclear power in the United States and in other countries.

2.2.2 Why switching from stainless steel to Alloy 600

Stainless steel either 304 or 316 was the initial material for Steam Generator (SG) tubing as it has been the normal material for many applications in Nuclear Power Plant (NPP). But due to its sensitivity to SCC in presence of chloride and the use of nuclear reactor for military naval application, it was important to have an alloy resistant to chlorides from potential sea water inleakage. According to American studies, the known remedy to increase the material resistance to SCC in presence of chloride is to increase the nickel content. U-Bend specimen in boiling 42% $MgCl_2$ solutions show that the time to cracking is close to zero for 10% Ni (content of 304 or 316 Stainless Steel) while the time to cracking increases drastically with up to 40% Ni as shown in Figure 2-1 [Pinard Legry, 2010].

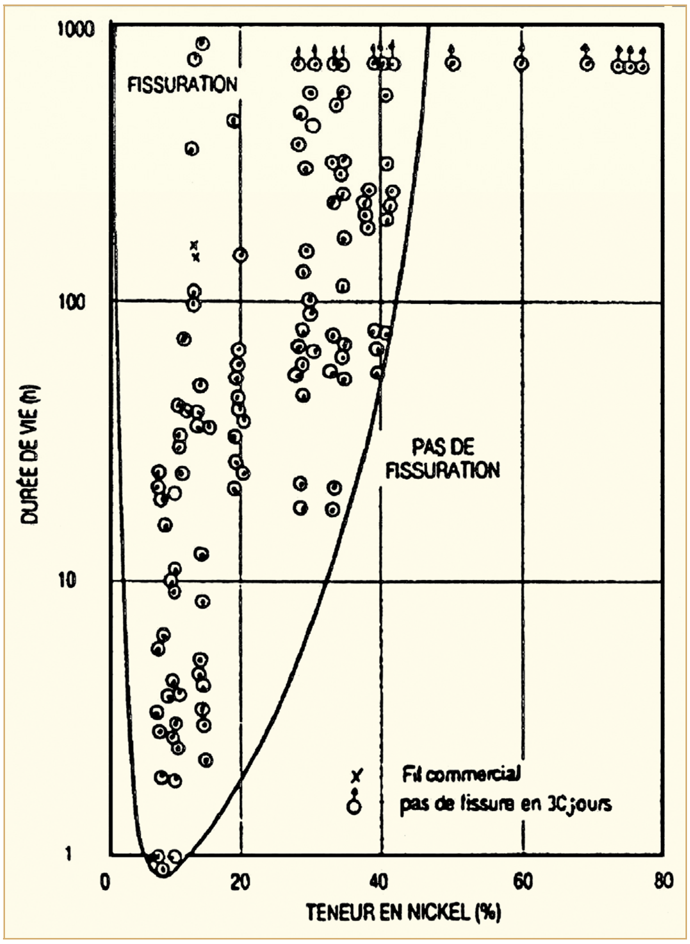

Figure 2-1: Time to cracking (in hours) versus Ni % of the material for U-Bend specimen in boiling 42% $MgCl_2$ solutions at 154 °C [Pinard Legry, 2010].

By 1957, Copson at INCO had demonstrated that SCC of Fe-Cr-Ni alloys in boiling $MgCl_2$ would stop at alloying contents above about 40% Ni.

Consequently, the Alloy 600 (Inconel ™ 600), with high nickel content (>72%) was developed and selected for SG tubing and other materials. Incidentally, Inco, a company selling nickel, developed and sold the Alloy 600, which may explain the company interest for such high nickel contents. In 1960, INCO published a paper saying that Inconel is a material which resists SCC in chloride and alkaline environments. There was interest, therefore, in qualifying this material for NPP application. This required radiochemical, welding and corrosion studies. All of these have been undertaken and all studies have indicated satisfactory behaviour of the Alloy 600. In Conclusion, the results of the study of [Copson & Berry, 1960] indicated that, from a corrosion standpoint, Inconel is an excellent construction material for primary and secondary waters of PWRs.

2.2.3 Sensibility of various alloys to SCC in pure water

A famous curve (Figure 2-2) shows the SCC sensitivity to Cl for Ni < 10-15% and to pure water for Ni > 70% [Coriou et al, 1959].

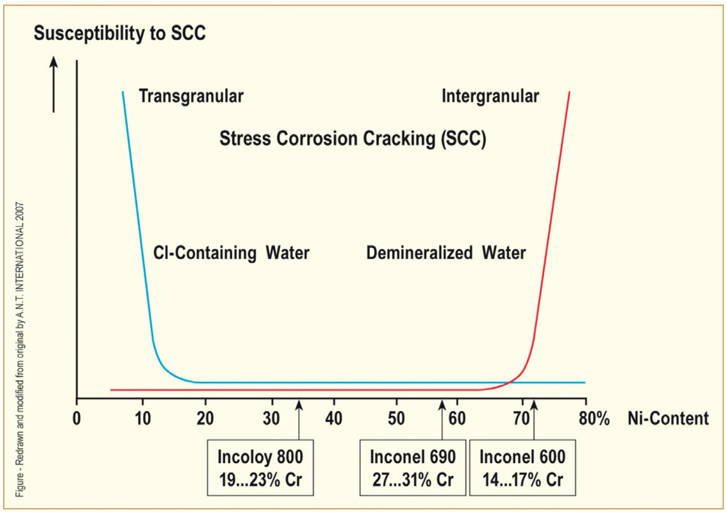

Figure 2-2: Schematic view of SCC intensity vs. Ni concentration for Fe-Cr-Ni alloys used for SG tubes in high temperature pure and chloride containing water, redrawn figure after [Coriou et al, 1959].

Figure 2-2 is supporting the results obtained in 1957, showing the sensitivity of Alloy 600 to SCC in pure water of Figure 2-3.

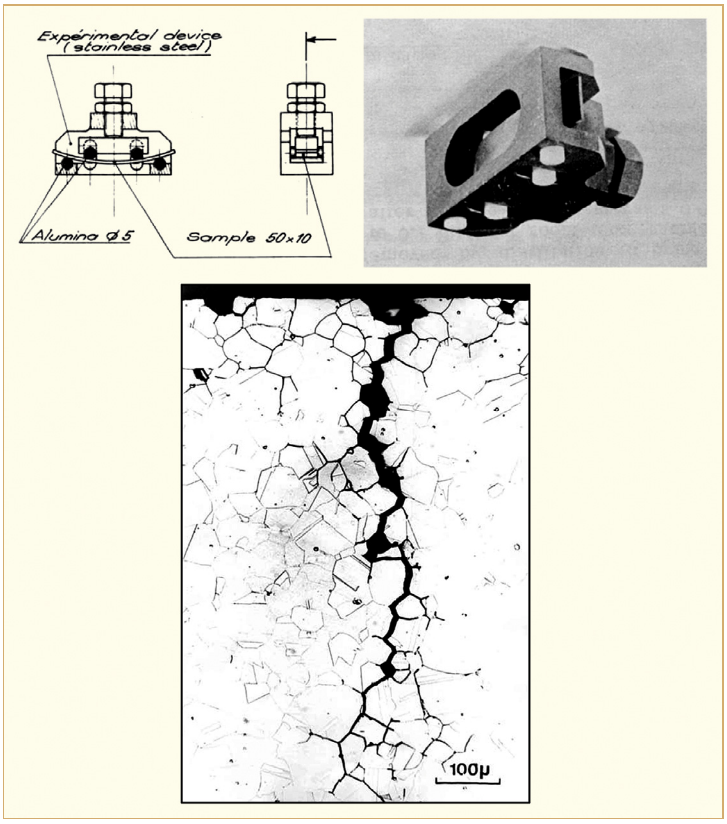

Figure 2-3: Test sample (left part) and crack (right part) obtained on Alloy 600 in pure water by Coriou at Saclay, France, in 1957 and published in 1959 [Coriou et al, 1959].

3 Materials degradation in BWRs (Peter Ford)

3.1 Introduction

Seventysix papers on materials degradation in BWRs were published in the targeted conferences held in Virginia Beach in 2009 and in Avignon in 2010. Because of space limitations only the papers relating to Environmentally Assisted Cracking (EAC) are addressed; discussions of other degradation modes such as FAC and loss of mechanical properties due to thermal aging or irradiation are postponed to a later Annual Report.

Emphasis is focussed primarily on; (a) advances in our understanding of the factors that control EAC i.e. SCC, CF in the structural alloys used in BWRs, and (b) an assessment of the completeness of the mitigation actions for EAC that are either currently used or are in active development.

Each of the alloy classes are treated separately recognizing, however, that there is a common thread in the chronology of degradation in the different alloys in different reactor systems. For example, as discussed in the ANTI reports referenced in Section 1, the progress of a stress corrosion crack may involve a sequence of different phenomena, such as (i) pitting (for carbon steels) or intergranular attack (for austenitic alloys) in high temperature LWR environments, (ii) microscopic crack initiation and (iv) coalescence, followed by (v) crack propagation (or arrest) (Figure 3-1). Moreover, this sequence may be preceded by a “precursor” period (that may be very short or take thousands of hours) during which time the surface material, environment or stress conditions change at a particular locality such that subsequent crack initiation may occur.

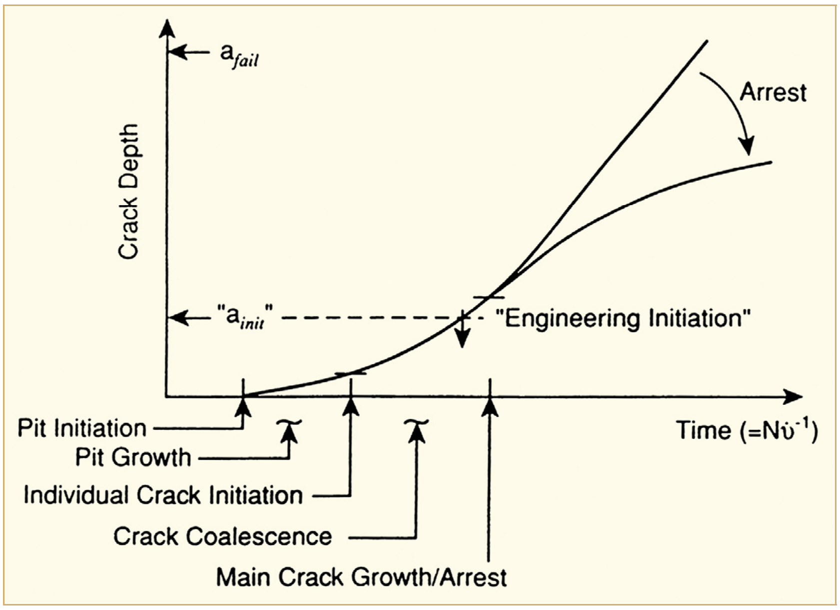

Figure 3-1: Sub-phenomena that can occur during the “initiation” and “propagation” periods in systems sensitive to SCC. Note that this sequence is often preceded by a “precursor” period when the surface conditions may change such that microscopic crack initiation may occur.

3.2 EAC of unirradiated stainless steels

SCC (and CF) of unirradiated stainless steels in BWR environments has received an extensive amount of study since the early observations of cracking in BWR piping and fuel cladding during the 1960s and 1970s. The system parameters that have a bearing on the material, environment and material conditions that control the cracking susceptibility (Figure 3-2) are well recognized. The details of these dependencies are fully reviewed in, for instance [Ford et al, 2010]. Quantitative models of the crack propagation process have been developed and qualified against observations of crack propagation in the laboratory and plant. These qualified models emphasise the varying dependencies of the crack propagation rates on the *combinations* of the materials, environment and stress conditions listed in Figure 3-2.

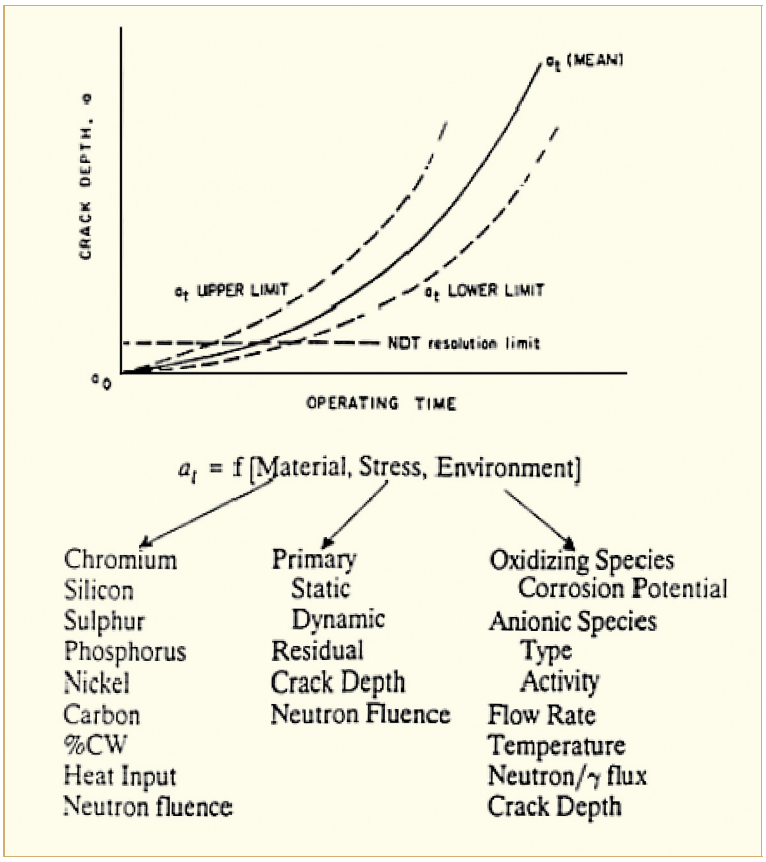

Figure 3-2: Materials, stress and environmental parameters relevant to EAC of stainless steels in BWRs [Ford et al, 2010].

Various mitigation actions have been developed based both on this theoretical knowledge base and the extensive empirical observations of the effect that the system parameters have on EAC of stainless steels in unirradiated environments. Examples of such mitigation actions include the use of L-grade or stabilized stainless steels, the application of higher purity water and “Hydrogen Water Chemistry (HWC)” specifications, and the reduction in stress due to modified welding practices. In many cases *combinations* of these individual mitigation actions are employed.

These mitigation actions have, in the main, been effective but, as mentioned briefly earlier, there have been unexpected failures in recent years [Ford et al, 2010] even in "mitigated" systems (i.e. stabilized or L-grade stainless steels operating in good purity water). These unexpected failures may be associated with, for instance significant effects of surface abuse and strain localization. Further, although there is an adequate understanding of the crack propagation mechanism for relatively deep cracks (i.e.>50µm), there is a lack of quantitative fundamental understanding of the processes relevant to precursor events that lead to microcrack initiation and crack coalescence, (Figure 3-1). This lack of knowledge becomes of importance when assessing design criteria that focus on the behaviour of very shallow cracks encompassed by "crack initiation" or (more pragmatically) "the detection of shallow cracks".

The papers from the 14th Environmental Degradation Conference in 2009 and the Fontevraud-VII conference in 2010 that shed light on possible "unexpected plant events" are now discussed.

3.2.1 Crack initiation

Intergranular stress corrosion crack initiation is conceptually easy to explain for a sensitized stainless steel since it may be associated with intergranular attack of a chromium-depleted, sensitized grain boundary. However, the prediction of "crack initiation" becomes more difficult for a non-sensitized steel, and various hypotheses have been proposed as to the specific surface characteristics that govern the initiation of microscopic cracks in such steels. These hypotheses range from changes in metal structure and residual stress/strain profiles associated with different fabrication procedures, to the morphology and composition of the surface oxide and changes in the corrosion potential associated with the form of that oxide.

Recent investigations of crack initiation in non-sensitised austenitic stainless steels have concentrated on validating (or rejecting) these various hypotheses.

Pre-service grinding of stainless steel reactor components and welds is performed for a variety of reasons ranging from aesthetics to preparation of the surface for non-destructive testing. The original surface of the component formed during initial fabrication is, therefore, often not the final surface microstructure that is exposed to the reactor coolant. This is important since the initiation of cracks may be controlled by this final surface microstructure and the associated local stresses and strains.

Recent investigations [Olszta et al, 2009] have focussed on the characterization of the near-surface microstructure of non-sensitized 316NG stainless steel observed in BWR recirculation piping and laboratory samples, and the relationship between that microstructure and the SCC susceptibility in BWR environments. It was observed that there was a nanocrystalline recrystallized grain layer 0.5-10 µm deep on the metal surface that quickly oxidized with penetration into the underlying heavily deformed subsurface region, which exhibited high dislocation densities, twinning and lath structures. The extent of this surface microstructural heterogeneity was variable in plant components and was dependant on the extent of surface grinding. For instance, samples with only slight surface roughness exhibited a thin (2 µm) recrystallized layer and no strain contrast or grain misorientation in the near-surface region. By contrast, specimens with a larger amount of surface roughness exhibited high strain contrast and grain misorientation, plus a much deeper (10 µm) recrystallized layer. It was concluded that the initiation and progression of surface corrosion cracks in this deformed microstructure is a key issue in the development of SCC precursors.

The first by Mills [Mills, 2010] was directed primarily to the effect of anionic impurities and aerated conditions on SCC of sensitized stainless steels that might occur during transient operations in PWRs., There was a reminder, however, in this paper that SCC can occur at low temperatures (77 °C) in aerated, impure (200 ppb sulphate) water chemistry conditions that might occur under severe upsets in BWR start-up operations. It was observed that under this severe situation, intergranular cracking is observed under dynamic loading, but not under static load[3].

In the second report Huttner discussed [Huttner, 2010] the observation of pitting and transgranular SCC on the sealing surfaces of stainless steel valves. These observations were made on Nb-stabilized low-Mo stainless steels in very specific designs of valves that have pressure-activated closure seating where chloride contamination may accumulate; the source of the chloride was attributed to older asbestos-containing sealing materials.

3.3 EAC of irradiated stainless steels

Irradiation Assisted Stress Corrosion Cracking (IASCC) of stainless steels in BWRs has been recognized since the late1950s with cracking observed in stainless steel fuel cladding relatively soon after reactor commissioning. This was followed in the 1960s by the observation of cracking of core components such as instrumentation tubes, control rod sheaving and handles, etc. that were exposed to different combinations of neutron flux and stress. More recently (i.e. since 1990) there has been increased focus on the cracking of hard-to-replace components such as welded core shrouds. These incidents have been reviewed in ANT International Annual and Topical Reports [Ford et al, 2010] in terms of the dependencies of the cracking susceptibility of core components on, for instance, the neutron flux and fluence, corrosion potential material composition, etc., as well as the working hypotheses for the cracking mechanism. This latter is reviewed briefly below, since it acts as a basis for discussing more recent data.

The SCC susceptibility of irradiated stainless steel components in BWRs respond to the various system (i.e. material, stress and environmental) parameters in much the same manner as is observed for unirradiated stainless steels. For example, the cracking susceptibility in both cases depends on the tensile stress, corrosion potential, anionic impurity concentration and the grain boundary chemistry. Thus, it was reasonable to propose that IASCC was not a new mechanism of cracking in BWRs, but merely mirrored irradiation-induced changes to the rate-controlling parameters in the various phases in crack development described in Sections 3.1, 3.2 and 3.3. Those changes that are driven by neutron flux (i.e. corrosion potential) should have an effect immediately on the cracking susceptibility, whereas fluence-driven changes (such as to material composition, stress relaxation and yield strength) would have an effect over an extended time.

It was hypothesized, therefore, that the slip-oxidation mechanism for crack propagation was the relevant cracking model for irradiated stainless steels in BWRs. Consequently the task was to identify the roles of neutron flux and fluence, and gamma irradiation on corrosion potential, applied stress, the yield stress and grain boundary chemistry.

As a result the *specific* effects of irradiation on the fundamental parameters that control crack propagation in stainless steels were quantified via consideration of the following:

- Corrosion potential and how this changes with radiation flux.
- Irradiation fluence-induced changes in grain boundary composition and especially chromium depletion.
- Irradiation fluence induced-hardening and, for displacement loaded structures, irradiation-induced creep and stress relaxation.

[3] Note that in [Ford et al, 2010] there is some discussion of the possibility of SCC occurring under BWR start-up operations.

As reviewed earlier [Ford et al, 2010] these modifications were quantified and validated in separate effects tests, and led to the prediction of the locations in the reactor core where IASCC of stainless steel components might be expected, as well as the magnitude of the degradation (Figure 3-14).

Although there is a reasonable agreement between the average observed and predicted crack depths, it should be noted that are considerable uncertainties in not only the observed crack depth (because of difficulties in non-destructive testing) but also in the predicted value (because of uncertainties in the definition of the cracking system). This latter uncertainty is primarily related to a lack of factual knowledge of the initial residual stress profile in these complicated welded structures, but some uncertainty is undoubtedly associated with the assumptions regarding the cracking mechanism.

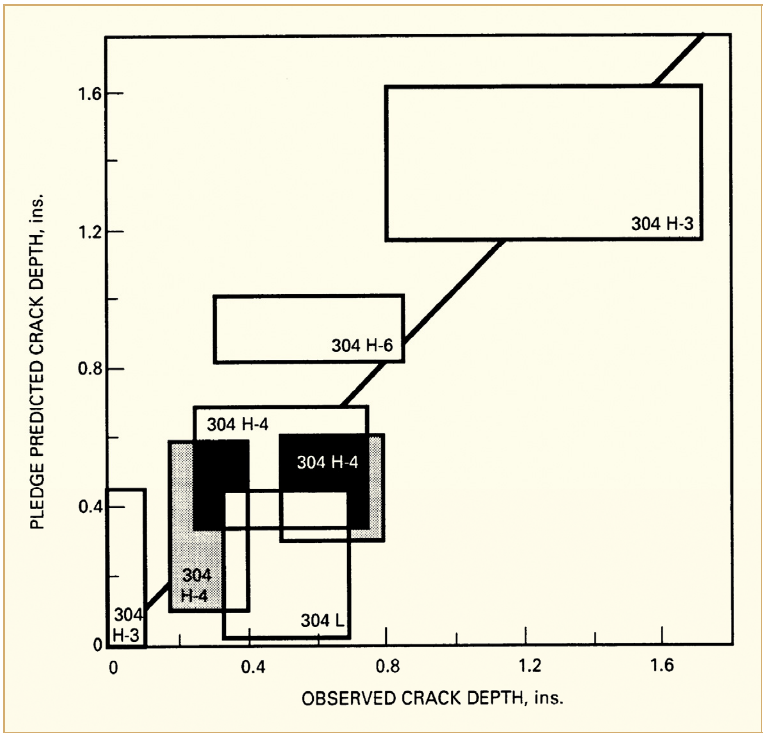

Figure 3-14: Comparison between the predicted and observed extent if cracking adjacent to various welds in core shrouds of BWRs [Ford et al, 2010].

Some of these aspects are discussed below in the light of recent data, which are discussed in terms of (a) parametric dependencies, (b) advances in fundamental understanding, and (c) new plant observations.

Parametric dependencies for IASCC of stainless steels

Several investigators have proposed in recent years crack propagation rate vs. stress intensity factor relationships for irradiated stainless steel. Such "disposition" relationships would be used for predictions of the crack advance during the next operating cycle, should a crack be detected during a routine inspection. Pathania [Pathania et al, 2009] discussed the known effects of neutron fluence on irradiation hardening, stress relaxation and grain boundary sensitization and concluded that disposition relationships could only be quoted for specific neutron fluence "bins". Pathania and colleagues also recognized that, because of various data quality issues associated with the demanding experimental conditions with irradiated specimens that some screening of the data would be required and, as a result, the amount of data upon which the disposition relationship is based is limited.

An example of such a depleted data base is given in Figure 3-15. This data was used to define (Figure 3-15a) the suggested disposition relationship for Type 304 stainless steel irradiated to between 5 and 30 $x10^{20}$n/cm^2 (E> 1MeV)[4] in reactors operating under the EPRI water chemistry guidelines for "NWC" is;

Eq. 3-2 $$da/dt = 8.21 \times 10^{-8} K^{2.5}$$

In this example the relationship line (where the propagation rate is in units of inches/hour and K in units of ksi√in) bounds 76.6% of the data.

Steels exposed to the same fluence band, but operating under EPRI water chemistry guidelines for "HWC", would (Figure 3-15b) would be evaluated by the relationship;

Eq. 3-3 $$da/dt\ 2.72 \times 10^{-8} K^{2.5}$$

where the $K^{2.5}$ relationship is assumed because of lack of data.

By comparison, the relationship for fluence levels below 5.0 x 10^{20}n/cm^2, the disposition relationship accepted by the USNRC (Figure 3-10b) [Hazelton & Koo, 1988] for stainless steels operating in rather impure (0.3-0.5 µS/cm) water at 288 °C is;

Eq. 3-4 $$da/dt = 3.59 \times 10^{-8} K^{2.161}$$

(where the propagation rate is in units of inches/hour and K in units of ksi√in).

This relationship is not necessarily relevant to the cracking of stainless steels exposed to current BWR because of the poor water quality conditions under which the data were obtained, and the fact that, currently, water conductivities <0.1 µS/cm are regularly maintained during operations. However this disposition relationship is often quoted as a reference line denoted as "NUREG-0313" (referring to the [Hazelton & Koo, 1988] report number).

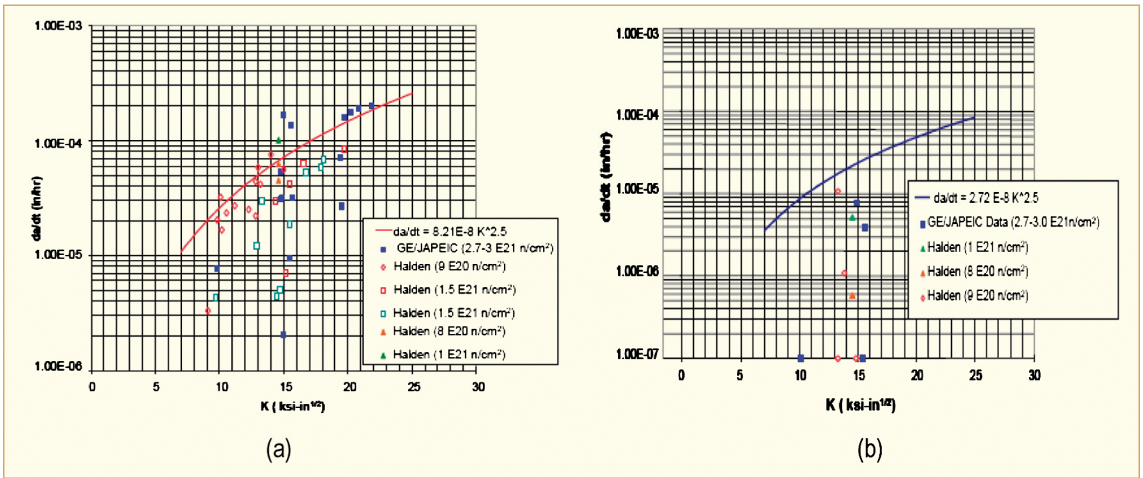

Figure 3-15: Summary of screened crack growth data under (a) NWC conditions and (b) HWC conditions [Pathania et al, 2009].

[4] Note that in the region of the high flux regions of a BWR core shroud, the fluence accumulation is of the order of 0.5 to 1x 10^{20} n/cm^{-2} per effective full power year. Thus for a 40 year life the relevant end-of-life fluence is 2 to 40 x 10^{20}n/cm^{-2}.

Material, temperature and strain rate conditions that lead to discontinuous yielding have for many years been associated with increased SCC susceptibility in a wide range of alloy environment systems, including SCC of low alloy steels in BWR environments. Recently Ivanchenko and colleagues have observed discontinuous yielding associated with dynamic strain aging for weld metals 182, 82, 152 and 52. at 300 °C and strain rates of 10^{-5} and $10^{-6}s^{-1}$. This work is ongoing in assessing the role of these observations in predicting crack initiation and propagation in both PWR as well as BWR environments.

3.5 EAC of low alloy steels

There have been extensive collaborative investigations over the last 35 years on the whole issue of EAC of carbon and low alloys steels in both BWRs and PWRs, starting with the question of the formulation of the disposition curves for CF crack propagation and initiation (i.e. the ASME XI disposition and III design criteria) and broadening to cover SCC and strain assisted cracking of statically loaded and dynamically loaded structures. These developments have been reviewed elsewhere [Ford & Scott, 2008]. The points worth noting at this stage, which serve as background to current concerns are as follows:

- The slip oxidation model for crack propagation serves as a skeleton for supporting life prediction algorithms for CF crack initiation and propagation.
- The model sets out the rationale for the SCC disposition relationships (for BWR "normal" and "hydrogen" water chemistry conditions) that are acceptable to the USNRC.
- There is sufficient theoretical and experimental knowledge to understand why SCC is a relatively rare occurrence in BWRs, and to define the conditions (combinations of dynamic straining and or high stresses, oxidizing conditions, material chemistry and yield stress, and anionic impurities) under which SCC has occurred in operating plant. These conditions all relate to the requirements for maintained crack propagation that rely on a sustained crack tip strain rate and a crack tip environment that has a critical concentration of chloride and/or sulphur anions. If these cannot be maintained then crack propagation will arrest.

Parametric dependencies for EAC in low alloy steels

Four papers have been published recently [Kubo et al, 2009], [Kumagai et al, 2009], [Peng et al, 2010] and [Ritter et al, 2010] that address an issue similar to that addressed above for the propagation of stress corrosion cracks in 316L plate into a 316L stainless steel weld. In this case, however, the concern is for the continuing crack propagation from an Alloy 182 attachment weld into the underlying low alloy steel pressure vessel steel. Such welds are associated with core shroud support in various BWR designs and would be expensive to replace or repair because of the restricted access to this subassembly.

All four investigations used similar approaches to the investigation and came to similar conclusions, namely:

- High hardness values were observed in the dilution (1.8mm) zone adjacent to the fusion boundary, indicative of high residual stress (c.f. Figure 3-26) [Peng et al, 2010] and [Ritter et al, 2010].

- All stress corrosion cracks in Alloy 182 in high purity 288 °C water arrested at or near to the weld fusion line when the stress intensity factor was below 60 MPa√m [Ritter et al, 2010]. At that point the crack tip blunted into a pit or oxide plugged crack (Figure 3-26). These arrested cracks could be reactivated from the pit however in 2 ppm oxygenated water with an increase in sulphate to 20 ppb or in 0.25 ppm oxygenated water with an increase in sulphate to 400 ppb [Peng et al, 2010][9].

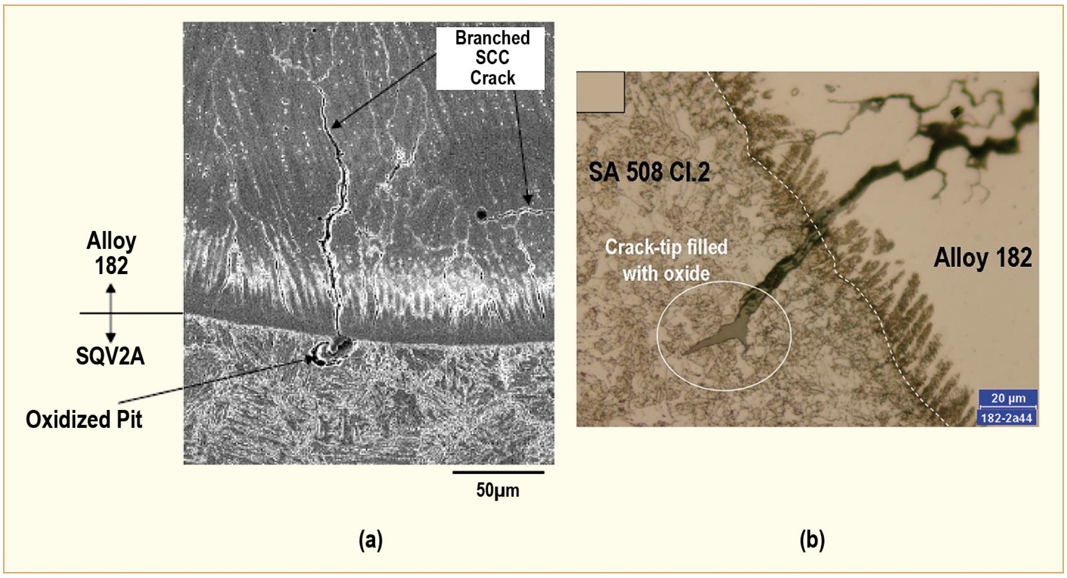

Figure 3-26: Arrested crack at, or near to the interface of Alloy 182 weld and low alloy steel [Kubo et al, 2009], (a) [Peng et al, 2010] and (b) [Ritter et al, 2010].

- As might be expected, there were combinations of stress intensity and sulphate concentration that lead to sustained crack growth as the crack advanced into the A53B low alloy steel. Tentative combinations of stress intensity factor and sulphate concentration that would sustain crack propagation in the low alloy steel at rates >1x 10^{-8} mm/s are shown in Figure 3-27a and corresponding threshold combinations for chloride concentrations are given in Figure 3-26b, [Kumagai et al, 2009]. Very similar threshold condition for chloride was observed by Ritter and his colleagues (Figure 3-28). It is noted that these threshold combinations are similar to those observed in classical SCC tests where the starter crack is introduced by cyclic loading. In other words the fact that the "starter crack" is provided by a crack in Alloy 182 does not introduce a complicating factor, such as a galvanic effect.

  It is apparent from comparisons of Figure 3-27a, Figure 3-27b and Figure 3-28 that chloride impurities are more deleterious than sulphate anions, to the point that >5 ppb Cl^- may, depending on the stress intensity factor, give sustained propagation into the low alloy steel.[10]. This is to be expected based on the separate SCC investigations by Seifert and Ritter [Seifert & Ritter, 2008] but *it should be pointed out, however, that the reason for this anion specificity is not understood at this time.*

[9] It should be noted that under normal operating conditions BWRs operate currently with <5 ppb sulphate impurity levels.
[10] It should be noted that BWRs currently operate with less than 0.5 ppb Cl^-.

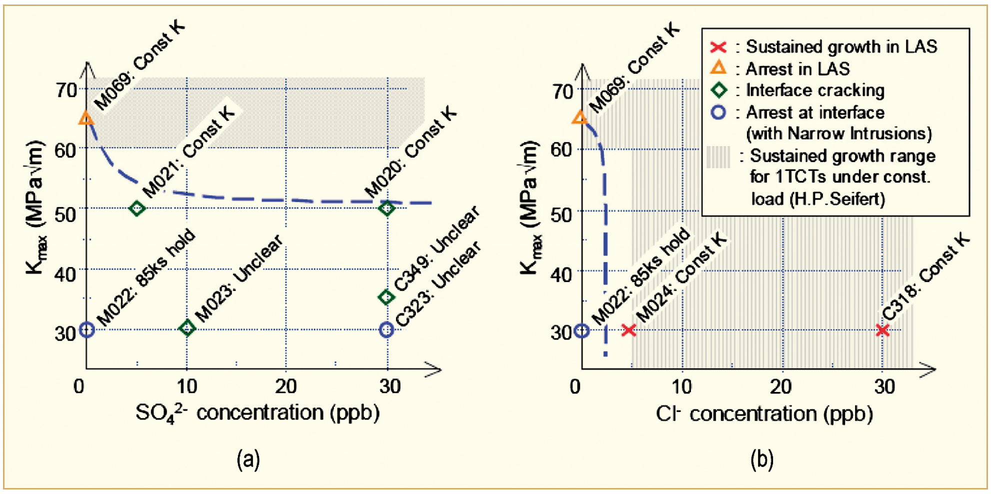

Figure 3-27: Tentative threshold conditions for crack propagation into low alloy from Alloy 182 [Kumagai et al, 2009]. Also shown by the shaded regions are the combinations of stress intensity and anion concentration that lead to sustained propagation in the absence of a "starter crack" in Alloy 182 [Seifert & Ritter, 2008].

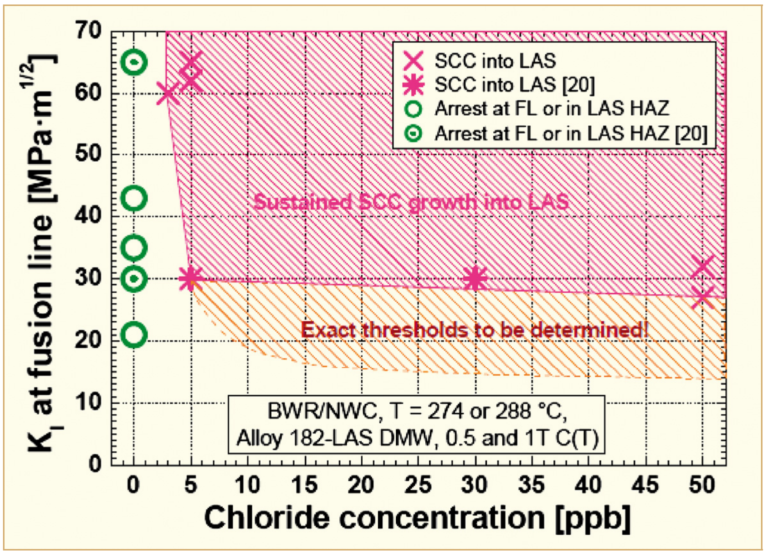

Figure 3-28: Combinations of Stress intensity factor and chloride concentration for sustained crack growth into the low alloy steel under BWR "normal water" chemistry conditions [Ritter et al, 2010]. Reference (20) in this Figure refers to the data from Kumegai et al. in Figure 3.14(b). (FL refers to "fusion line").

4 Materials degradation in PWRs (Pierre Combrade)

4.1 Introduction

A total of about 115 papers related to PWR plants were presented at the 14th International Conference Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors held in Virginia Beach (USA) in August 2009 and at the Fontevraud VII conference held in Avignon (France) in September 2010. This total does not include the presentations devoted to irradiation-induced modification of pressure vessel steel microstructure and mechanical properties. It is obviously impossible to summarize all of them individually and the following is an attempt to describe comprehensively the main topics that were tackled, the main new degradation events that were reported, and the main research progress that has been made. In the following text, many extracts of the authors' papers have been reproduced and, except for possible (but unintentional) omissions, they are inserted between quotation marks.

It must be noted that the Environmental Degradation Conferences are usually mainly devoted to research progress on structural materials and the Fontevraud Conference is mainly oriented to field experience and related failure analyses and improvements. However, no severe generic material degradation problem has been reported in the past few years, and the majority of presentations in Fontevraud VII conference were devoted to laboratory studies, particularly in the scope of the long lifetimes expected for existing and future plants.

In the past, the most important material degradation was due to IGSCC of nickel base alloys. This occurred initially in both the secondary side of steam generators (IGA-SCC of Alloy 600 SG tubes in sludge piles on top of tubesheet and inside SG tube to tube support plate crevices) and in the primary circuit (Alloy 600 tubes in the roll transition zones of mechanically expanded tubes and in the small radius U-bends. Later the primary side IGSCC extended to Alloy 600 pressurizer nozzles, Alloy 600 CRDM nozzles and related Alloy 182 J-welds, Alloy X-750 guide tube pins, etc.). These forms of degradation were extensively covered in earlier "Environmental Degradation of Materials in Nuclear Systems – Water Reactors" and "Fontevraud" conferences as well as in many others.

Primary Water Stress Corrosion Cracking (PWSCC) of nickel Alloys has been analyzed in detail and, even if not fully understood mechanistically, has been mitigated practically since Alloy 600 and related weld metals have been replaced by alloys with higher chromium contents, Alloys 690, 152 and 52(M) for new plants and for replacement components (steam generators, pressurizers, pressure vessel heads). In addition, methodologies of prediction and monitoring for existing plants have been developed to optimize NDE strategy as well as for planning component replacement. Consequently, there is presently no more extensive degradation reported at Fontevraud VII conferences.

Nevertheless, unexpected events still occur, like PWSCC of divider plates in several French steam generators that was covered in two Fontevraud VII presentations. This shows the need for improved prediction tools and additional data. In addition, the longer life of new plants and the life extension of existing plants also require a better capability for predicting long-term behaviour of materials, including those like Alloy 690 and related weld metals that have not experienced any environmental degradation to date. These studies were an important part of the presentations of both conferences, particularly those on Alloys 690, 152 and 52(M). These presentations also included studies of chemical and mechanical mitigation techniques.

Degradation of the secondary side of steam generator tubes has been a huge problem in the past but only a few new cases were reported at Fontevraud VII: these include circumferential cracks in the tube to tube support plates crevices in Bugey 3 (Alloy 600 MA tubes and Carbon steel tube support plate) and fatigue cracks due to vibrations in Cruas 4. This last event was due to the formation of hard deposits that impair fluid flow in broached tube support plates. This phenomenon affects several plants and was described (as well as its consequences) in two presentations. Concerning material degradation of steam generator tubes, four presentations were also devoted to chemical aspects, i.e. the role of lead and sulphur. It must be remembered that the replacement of most steam generators originally with Alloy 600 MA tubes is one of the main causes of the decreasing number of SG degradation events.

FAC is also an important issue for PWR secondary circuits and has sometimes caused human casualties in several NPPs. This phenomenon is now well understood and predictive models have been developed both on a mechanistic basis (French BRT-CICERO model, British Energy model) or on more empirical basis (Checkworks). Nevertheless, new models are still being built and existing models are being improved by improving field experience data bases, thermodynamic data and, very importantly, detailed knowledge of local thermo-hydraulic conditions in zones particularly prone to high mass transfer conditions and subsequent FAC. Several presentations were devoted to FAC both in the 14th ED and Fontevraud VII conferences.

Other problems of material degradation are being increasingly addressed although they have not caused extensive damage up to now but may constitute major problems in the future. The most important issues are clearly the increasing risk of degradation under neutron irradiation that increases with operating time. This includes degradation of pressure vessel steel mechanical properties (that are not in the scope of this summary) and the IASCC of stainless steels, which is due to the progressive modification of stainless steel microstructure under irradiation. Studies of this problem have been undertaken over long time periods and they constituted a very important part of the presentations at both conferences. A large number of presentations, mainly on laboratory studies were devoted to irradiation effects in PWR plants, in both conferences, i.e. 13 papers in 14th ED and 45 papers in Fontevraud VII.

Stress corrosion of cold worked stainless steels has not caused extensive damage in PWRs to date but occurs occasionally on heavily cold worked components. This is at the origin of a significant number of studies, particularly those oriented to understanding the effect of cold work on deformation processes because strain localization is assumed to be a major consequence of cold work.

The effect of environment on fatigue resistance is also an issue that requires some attention, particularly in the case of stainless steels, that seems unexpectedly more susceptible to environmentally enhanced degradation in PWR than in BWR coolants. However, there were only two presentations on this topic at the Fontevraud VII conference.

4.2 Environmentally assisted cracking (EAC) of nickel-based alloys in PWR primary water (PWSCC)

4.2.1 Alloy 600 and related weld metals

4.2.1.1 Field experience and related studies

Only five presentations were devoted to Alloy 600 field experience and the few PWSCC events that were reported tend to show that this problem is now under reasonable control.

Hwang et al. [Hwang et al, 2009] reported the analysis of PWSCC cracks in a SG drain nozzle, i.e. in a dissimilar metal weld region in a PWR plant in Korea. The hot leg nozzle is built with SCC resistant Alloy 690 and, thus, cracking occurred only on the Alloy 600 cold leg drain nozzle. The Alloy 600 microstructure was considered as “good” and not be very susceptible to PWSCC. Cracking was attributed to the presence of high residual stresses that were confirmed by finite element analysis.

Four papers from EdF were devoted to cracks found in pressure vessel head CRDM nozzles and in several divider plates of French steam generators. Two described crack observations and related analyses, and two presented R&D studies aimed at predicting the propagation of the cracks observed in Steam Generator (SG) divider plates:

- Duisabeau et al. [Duisabeau et al, 2010] presented an extensive inspection program performed by EdF on more than 800 Alloy 182 J-welds from decommissioned pressure vessel heads using automatic dye penetrant testing. **No SCC had initiated on the surface and no propagation from weld defects was found on pressure vessel heads with operating times up to 170 000 h**. However, in a very few instances, cracks that had initiated in base metal propagated into Alloy 182 welds and, in one single instance, a crack initiated at the root weld pass that was in contact with primary water due to a leaking through-wall crack. In J-welds of SG tube drain nozzles, the weld root pass is in contact with primary water and one SCC event leading to water leakage was identified in the 2008 and 2009 outages. This nozzle was plugged and maintenance policy of similar zones was reinforced.
- Déforge et al. [Déforge et al, 2010a] summarized the analyses performed on Alloy 600 crack events that had occurred in pressurizer nozzles, in CRDM nozzles and, since 2002, in SG divider plates. Figure 4-1 shows the replacement ratio of these three components as a function of time. The recent problem of SG divider plates has affected twelve 900 MWe SGs out of over 87 inspected but no 1300 MWe SGs are affected. All cracks are located in the hot side of the stub runner attached to the primary side of the tube sheet and no crack has been detected in the divider plate itself. Cracks are superficial (depth < 2 mm in most cases), mainly located in two lines parallel to the weld axis (Figure 4-2) and in the centre of the stub (i.e. no cracks were found near the junction between stub and SG channel head). No propagation was detected after successive observations. All cracks occurred on stubs with low mechanical characteristics (the important parameter is claimed to be the yield strength difference between stub and divider plate) and they are located in zones that underwent plastic strain during fabrication. Material microstructure also played a role: “the more susceptible materials are characterized by a rather low rate of elongation during hot-rolling (around 2.5%), a carbon level above 0.05% and a final heat treatment above 900°C”.

  Comparison with past analyses of cracked CRDM nozzles showed strong similarities;

  - **a strong material effect is observed.** In both cases, the most susceptible materials are those with a high carbon level and whose final heat treatments were performed in the upper temperature range allowed,
  - **significant prior-deformation occurred on components that developed cracking,**
  - **significant surface cold work is present due to the manufacturing process.** Cold work is due to grinding and machining (including planing of divider plates and stubs and counterboring of vessel head CRDM nozzles). In both cases, the materials are deformed after surface cold work, which promotes the development of high tensile residual stresses in the cold work layer.

  Déforge at al. insisted on the fact that the recent events that were not predicted by the so-called “indices model” for PWSCC susceptibility thus supporting the need for an improvement of this empirical model by taking into account the effect of complex strain paths (see hereafter Léonard et al. presentation (Section 4.2.1.2)).

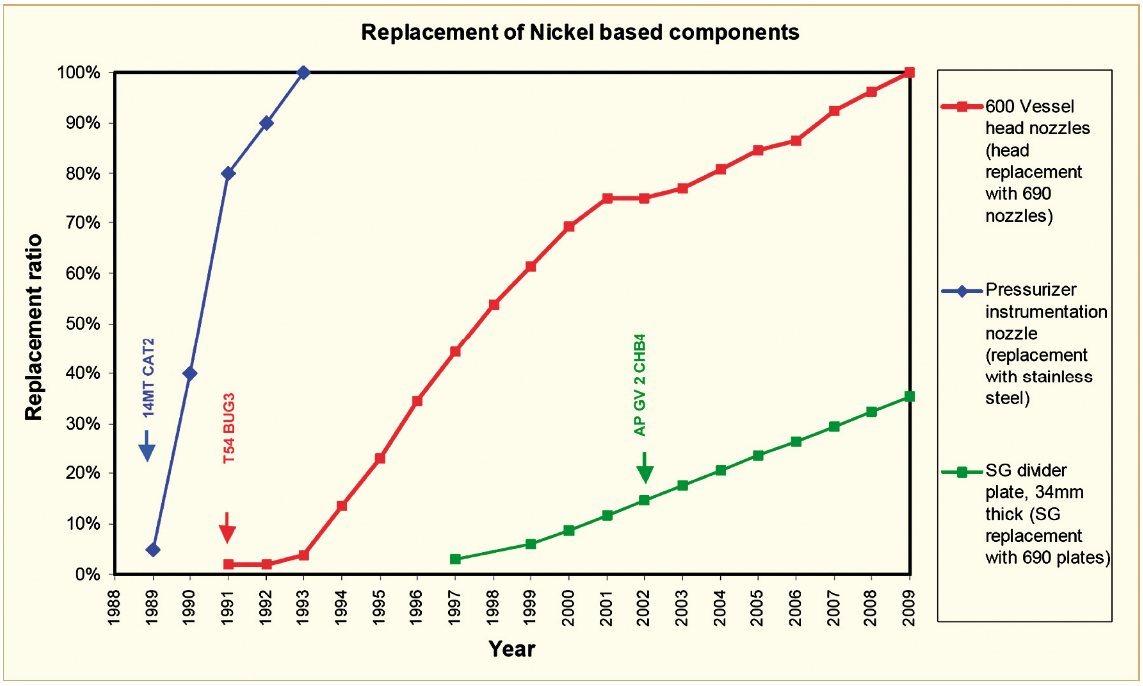

Figure 4-1: "Replacement ratio of Alloy 600 components due to SCC in France. Vertical arrows indicate the first case of SCC reported for each component", after [Déforge et al, 2010a].

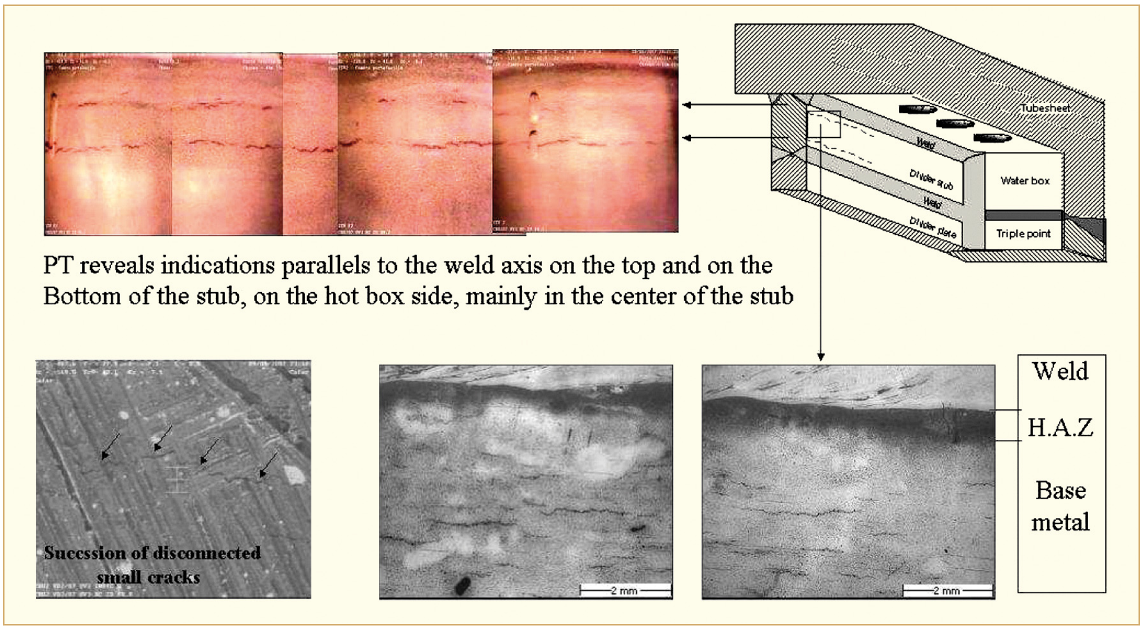

Figure 4-2: Location of cracks on divider stub, after [Déforge et al, 2010a].

4.3.4 Atmospheric SCC

In the Koeberg NPPs, atmospheric SCC has affected safety injection system piping (RIS) and the Refuelling Water Storage Tank (RWST) fabricated from Type 304 and 304L stainless steels and exposed to a marine atmosphere. Two papers were presented on this subject to the Fontevraud VII Conference:

- Doubell presented for Alexander et al. [Alexander et al, 2010] a mechanical characterization of the toughness of cold worked and mill annealed materials removed from the plant in order to facilitate the assessment of critical defect sizes and to provide an assessment of the overall integrity of a defective tank.

- Doubell et al. [Doubell et al, 2010] presented a technique to seal the leaks by a Type 316 L Weld OverLay (WOL) deposited using a Laser Beam Welding (LBW) technique. The procedure had been designed and validated on mock-ups. It allows repair of leaking tanks that are still filled with liquid. The presence of boric acid in the leaking water has been shown to be acceptable. “Potential benefits identified were:

  – Repair at drastically reduced time and cost as opposed to component replacement.

  – Repair activities could be accommodated by existing outage schedules.

  – Repairs without draining the fluid contents will be of great logistical benefit.”

4.4 IASCC of stainless steels

No new plant events involving IASCC were presented in the two conferences, but Pokor et al. [Pokor et al, 2010a] summarized EdF field experience on baffle to former bolts of CP0 units as follows:

- “Significant differences in the behaviour between units were observed as shown in Figure 4-47.

- “The cracked bolts almost exclusively belong to the four lower formers (96% of the cracked bolts are located on these formers).

- Analysis of the inspection results shows that there is a cracking threshold dose, slightly higher than 4 dpa.

- The phenomenon of cracking is more or less significant according to the units (Figure 4-47).

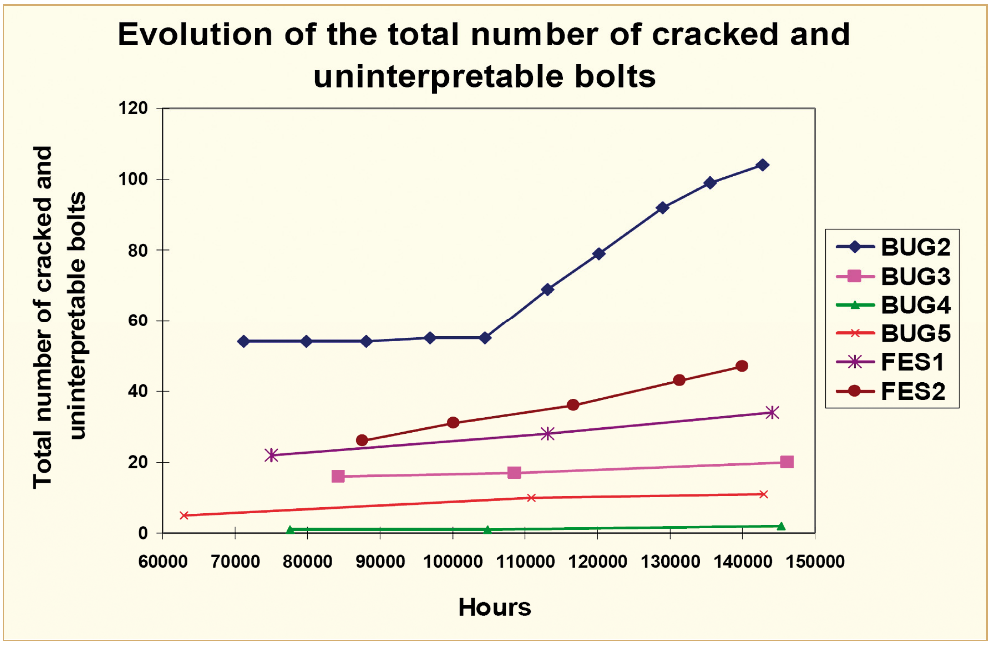

Figure 4-47: Summary of the number of baffle to former bolts classified as defective in EdF CP0 type units, after [Pokor et al, 2010a].

- The propagation of cracks is fast. A bolt detected with indication during a visit is detected cracked at the next visit (CGR is on the range of 3.10^{-10} $m.s^{-1}$).
- The intensity of the neutron flux seems to play a role on the life time of the bolts: the greater the flux, the sooner the bolts crack.
- On the four lower formers, dependence between the level of the stress estimated by simplified method and the number of cracked bolts is observed.
- Cracking time seems to be correlated with the temperature through an Arrhenius law. This relationship only applies on the bolts of the lower formers.
- No definitive conclusion could be made on the influence of the chemistry of the heats on the number of cracked bolts for each unit."

Regarding the causes of IASCC, it is commonly assumed that susceptibility to IASCC arises from several factors including:

- Grain boundary segregation: Cr-depletion has no significant importance at low corrosion potential in PWR primary water but Si segregation could be very deleterious.
- Hardening and associated strain localisation.

A large number of studies devoted either to characterizing microstructure and/or mechanical properties of irradiated materials or to improving the determination of thresholds for IASCC are on-going. These studies used both materials irradiated in service, mainly from baffles bolts, internals and flux thimble tubes, as well as materials neutron irradiated in research reactors (Halden, BOR-60) or proton irradiated in laboratory.

4.4.1 Irradiation effects on microstructure and properties

4.4.1.1 Microstructure

Takakura et al. [Takakura et al, 2009b], Yang et al. [Yang et al, 2009] and EdF laboratories ([Renault et al, 2009], [Renault et al, 2010] and [Garnier et al, 2010]) carried out TEM microstructural examinations of irradiated stainless steels, with and without applied stresses.

In particular, Takakura et al. and EdF characterized materials with very high doses coming from baffle bolts and flux thimble tubes [Takakura et al, 2009b] or from irradiations in the BOR-60 fast reactor (EdF).

All results confirm that changes in microstructures and mechanical properties due to irradiation saturate for doses from 10 to 20 dpa, except for the helium concentration which continuously increases with dose. In a stainless steel equivalent to Type 321 used in VVERs, Belozerov et al. [Belozerov et al, 2010] also observed a continuous increase of Helium concentration with dose but, for high irradiation doses, the increase of helium concentration does not cause any further increase in material strength.

Stress applied during irradiation seems to modify the distribution (number and size) of Frank loops but, at first sight, the results reported by [Renault et al, 2010] and [Garnier et al, 2010] do not seem fully consistent. However, the important result is that the presence of stress does not cause any large anisotropy in the distribution of Frank loops. “Cluster dynamics modelling of irradiation defects incorporating a physically based bias for defect/loop absorption triggered by an applied stress is able to reproduce accurately the small anisotropy observed experimentally by TEM between different families of loops”. Thus, “the stress induced preferential absorption of point defects on loops is unable to account for the macroscopic [creep] strain rate. Other mechanisms must be envisaged to explain this discrepancy, keeping in mind that the dislocation loop behaviour is properly described by the current model. Two possible routes may be envisaged, either via a bias of other defect/microstructural feature interactions (such as pre-existing dislocation networks or grain structure) or via a destabilisation of dislocation structures induced by irradiation”.

The use of atom probe tomography is being generalised in the study of SCC in LWR environments and it has also been applied to irradiated stainless steels by Jiao et al. [Jiao et al, 2009] and by Etienne et al. [Etienne et al, 2010]. Both studies identified irradiation induced Ni and Si segregation into clusters which are likely to be associated with irradiation defects (Figure 4-48), i.e. defect clusters or dislocation Frank loops. Jiao et al. also identified grain boundary segregation of Ni and Si as well as B and P in commercial alloys.

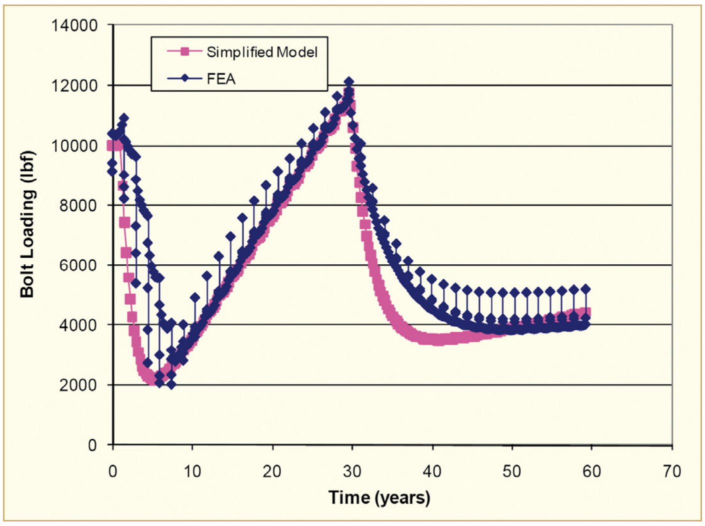

Figure 4-62: Comparison of simplified model and full FEA model for a bolt irradiated to ~75 dpa, after [McKinley et al, 2010]. Note that at 30 dpa, the core loading was changed to low leakage in the calculation and this gives rise to the discontinuity shown in the figure.

4.5 Low alloy and carbon steels

This section includes the effects of irradiation on pressure vessel steels that have been a very important topic in Fontevraud VII Conf., EAC of carbon and low alloy steels (that is not a serious problem in PWRs) and FAC of carbon steels.

4.5.1 Pressure vessel steels

The effect of irradiation on the mechanical properties of pressure vessel steels has been one of the main topics of Fontevraud VII conference. The main issues are related to life extension of NPPs, which requires an update of the evaluation of the microstructural and mechanical evolution of the pressure vessel steels under neutron irradiation.

This includes:

- fundamental studies aimed at understanding and predicting these evolutions,
- improvement of master curves and techniques of evaluation of embrittlement,
- implementation of surveillance programmes able to cover life extension,
- improvement of the approaches necessary to evaluate the safety margins in case of an accident,
- improvement of the procedures and regulations for assessment of life extension.

4.5.2 Low alloy and carbon steels

Arioka et al. [Arioka et al, 2009] presented to the 14th Env. Deg. Conf. a paper on the mechanisms of IGSCC of cold worked carbon steel in high temperature water. Three main points were outlined:

- IGSCC in high temperature, hydrogenated water at 280 to 360 °C is possible on cold worked carbon steel at moderate stress intensity factors and by creep cracking in gaseous atmospheres.
- Cavities were observed ahead of crack tips both in high temperature hydrogenated water and in air. They are believed to be the results of vacancy diffusion and coalescence and to act as crack embryos.
- A specific experiment with Ni plating was designed to demonstrate the diffusion of vacancies under a stress gradient.

4.5.2.1 FAC of C-steels

Two presentations directly related to plant applications were presented in Fontevraud VII and three presentations in the 14th ED Conference were devoted to FAC.

Trevin et al. [Trevin et al, 2010] presented an upgrade of the EdF BRT-CICERO software now recognized by the French safety authority and used in all EdF plants for FAC surveillance. The presentation described the improvements introduced in version 3 of the software. These include improvements of the thermodynamics database (from the IAPWS 1997 release), chemistry calculations (using MULTEQ 4.0 version), two phase flow (with a steam void calculation module based on the Chexal-Lellouch model), and many geometrical factors for components that have been updated in order to take into account NPP the analysis of operating experience. This version also includes the beneficial effect of Cr in order to avoid over-conservative results and the beneficial effect of dissolved oxygen. The validation steps of this new BRT-CICERO version were also presented and the comparison between measured and calculated thickness values, based on more than 14 000 measurements were presented that show that 93.3% of predictions are conservative (Figure 4-63).

The development of a technique using digital radiography coupled to a software platform to follow up FAC in PWR secondary circuits was presented by EdF and GE Sensing and Inspection Technologies [Koetz et al, 2010].

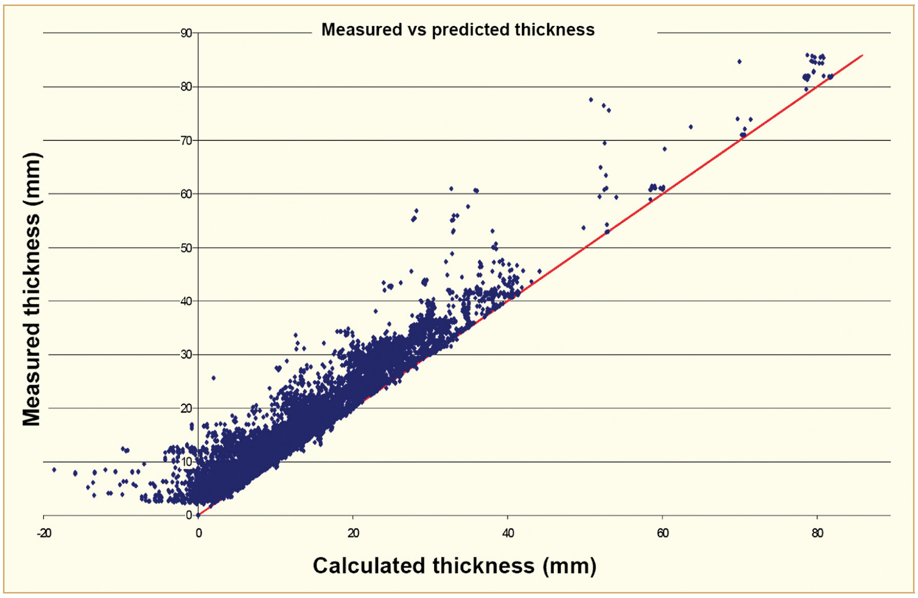

Figure 4-63: Measured versus calculated component thicknesses using BRT-CICERO™ 3.1.b version, after [Trevin et al, 2010].

The presentation of Garud [Garud, 2009] reviewed recent dramatic FAC events (at Mihama 2 and at a fossil plant IATAN-1). He noted that, even though old events like that at Surry-2, which triggered major efforts to institute systematic monitoring and management programs, the reduction of FAC events in the last 20-25 years was, in his opinion, not really satisfactory and "there is room for improvement in the assessment and prevention/mitigation of the FAC events in operating plants". He also noticed that, in both the cases of Mihama-2 and IATA-1, current models significantly underpredicted wall thinning. In particular, he presented a comparison of measured versus predicted wall thinning according to the Chexal-Horowitz (EPRI) model that clearly exhibits a very large uncertainty in the results and a wide range of underestimated values (Figure 4-64).

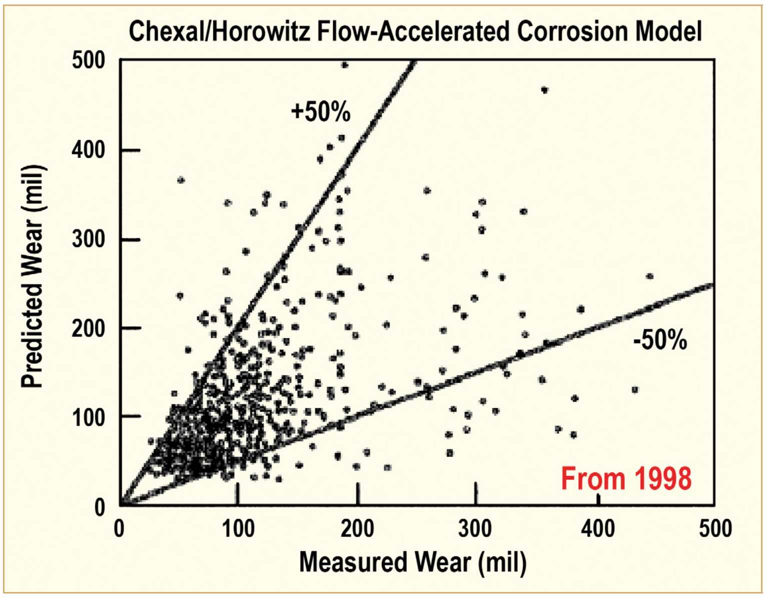

Figure 4-64: Measured versus calculated component wall thinning according to the Chexal-Horowitz model, after [Garud, 2009].

Garud attributed this poor reliability of predictive models part to an oversimplified estimation of temperature effects (the temperature range for FAC is probably underestimated and several peak temperature may possibly exist) and, more importantly, to a lack of reliable evaluation of mass transport in complex geometries.

The main conclusions included the following points:

- "Control of FAC through optimized and well controlled water chemistry, while essential, does not appear sufficient to eliminate the likelihood of serious events, even in single-phase flow".

- *Curiously*, Garud claims that "partial replacement with FAC-resistant (high chromium) material in an otherwise susceptible line should be evaluated more carefully, <u>if not discouraged</u>". *The technical basis for this remark is not evident.*

- "Effort should be made to reduce the uncertainty in FAC rate estimation and to explicitly account for various sources of uncertainty in the inspection prioritization and the final structural integrity assessment. These sources include model inputs, empirical parameters, modelling relation(s), and the NDE measurements" "This effort is likely to be benefited through a round-robin effort, starting with an updated state-of-the-art survey, and systematic CFD results with benchmarking."

- "Given the current state-of-the-art, the prevention or minimization of FAC damage is likely to be more certain and cost effective, considering that the monitoring and measurements needing accuracy and prediction reliability are not easily achievable."

Two Japanese papers presented at the 14th ED Conference were devoted, respectively, to developing a FAC model and to a parametric study of FAC.

Uchida et al. [Uchida et al, 2009b] presented the development of a model involving a detailed analysis of mass transfer at the component surface, and coupled models of "static" electrochemistry (Evans diagrams) to calculate local corrosion potentials in regions of hydrazine injection at the inlet of steam generators, and a "dynamic" model of oxide growth. Comparison of the results of the model with a few data points revealed reasonable agreement regarding the effect of mass transfer and pH.

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