

## **INDUCEMENT OF IGA/SCC IN INCONEL 600 STEAM GENERATOR TUBING DURING UNIT OUTAGES**

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### **Executive Summary**

The degradation of Unit 4 SG tubing by IGA/SCC has limited both the operating period and end of life predictions for Unit 4 since restart in late 2003. The circumferential IGA/SCC has been most significant in SG4 with substantial increases in both initiation and growth rates from 2005 through the spring of 2007. A detailed review of the occurrence of circumferential OD IGA/SCC at the RTZ in the HL TTS region of Bruce 4 steam generator tubes has led a conclusion that it is probable that the IGA/SCC has been the result of attack by partially reduced sulfur species such as tetrathionates and thiosulfates during periods of low temperature exposure. It is believed that attack of this type has mostly likely occurred during startup evolutions following outages as the result the development of aggressive reduced sulfur species in the TTS region during periods when the boilers were fully drained for maintenance activities. The modification of outage practices to limit secondary side oxygen ingress in the spring of 2007 has apparently arrested the degradation and has had significant affects on the allowable operating interval and end of life predictions for the entire unit.

**Acronyms**

SG	Steam Generator
PH	Preheater
IGA-SCC	Intergranular Attack-Stress Corrosion Cracking
RTZ	Roll Transition Zone
TTS	Top of Tubesheet
EOL	End of Life
AAR	Area At Risk
HTMA	High Temperature Mill Annealed
OPEX	Operating Experience
HL	Hot leg

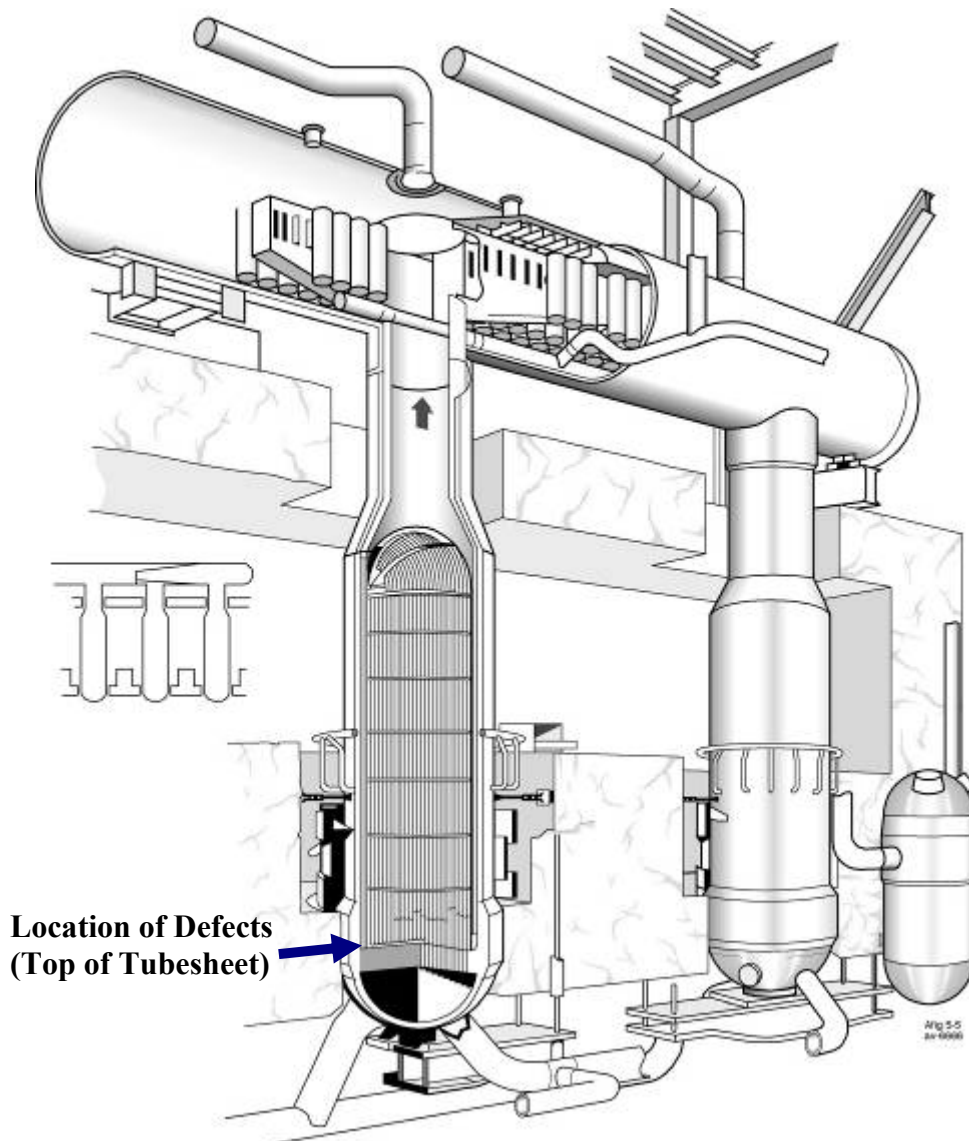
## 1.0 Background

Unit 4 began operation in 1979 and operated until 1997 when it was removed from service. The unit was laid up until restart in 2003. Initial layup practices were far from ideal with the SGs filled with oxygenated water and later drained, leaving them exposed to an oxidizing environment until a condition assessment was performed in 2000. These inspections revealed pitting and IGA in the sensitized I600 SG tubing at and just above the TTS due to the lack of proper layup chemistry. The vessels were then placed in a controlled layup condition until restart of the unit in 2003. The operating history post restart is outlined in table 1.

Interval Start	Duration (Months)
Sept 2003	8
June 2004	9
April 2005	12
June 2006	9
April 2007	5
Sept 2007	6
April 2008	12 (Projected)

**Table 1. Operating History For Bruce Unit 4 Following Restart**

Inspections carried out in 2006 following a 12 month run revealed significant circumferential IGA/SCC in SG4 (16 Defects at the TTS in the HL RTZ, table 2). As a result of these findings, circumferential IGA/SCC defect initiation and growth rates were increased, reducing the subsequent operating interval to 9 months. Nine months later in the spring of 2007 inspections were carried out revealing another increase in both the number and size of circumferential defects, beyond the condition monitoring criteria established following the 2006 inspections. Several of these defects approached the structural limit, where tube rupture could occur under accident conditions (e.g. main steam line rupture). The subsequent operating interval was further reduced to six months. Inspections in the fall of 2007 (following 4.5 months of operation) failed to reveal any circumferential defects in SG4 and the other Unit 4 boilers. This was contrary to the predictions for high temperature damage made after the spring campaign, raising questions regarding the mechanism of attack. Following these findings, a detailed review of the SG operating and layup history was carried out in an attempt find a supportable explanation for this behavior.



**Figure 1. Location of IGA/SCC Degradation ( HLTTS) in the Bruce Steam Generators.**

	SG1	SG2	SG3	SG4	SG5	SG6	SG7	SG8	SG Total
2004	4	0	1	0	0	2	2	0	9
2005	1	0	0	0	0	0	0	1	2
2006	0	2	0	16	0	0	0	0	18
Spring 2007	0	0	0	44	0	0	0	0	44
Fall 2007	0	0	0	0	0	0	0	0	0
Spring 2008	0	0	0	0	0	0	0	0	0
Total	5	2	1	60	0	2	2	1	73

**Table 2. Summary of Detected TTS Circumferential Indications in Unit 4**

The detailed review highlighted a correlation between outage practices and SG tubing IGA-SCC activity. During the 2005 outage 38 tubes were removed in the unit (12 in SG4), and in 2006 another 10 tubes were removed in SG4 and steam drum and boiler secondary side inspections were carried out on the west bank of boilers (encompassing SGs 1 through 4). Efforts to return the secondary side to a controlled layup state were not seen as critical, and the boilers were refilled on a best effort basis. This resulted in the boilers remaining exposed to oxidizing conditions for extended periods of time. Table 3 outlines the secondary side exposure durations for recent campaigns for each of the SGs within the unit. As outlined the SGs either remained in a controlled wet layup, were partially drained (though boiler blowdown) leaving 6-8" water on the secondary side or were completely drained following or in preparation for tube removal activities. Upon review, the duration of the fully drained state correlated with the detection of IGA-SCC in the following inspection outage.

<b>2005 Outage</b>	<b>SG1</b>	<b>SG2</b>	<b>SG3</b>	<b>SG4</b>	<b>SG5</b>	<b>SG6</b>	<b>SG7</b>	<b>SG8</b>
Partially Drained Period (Days)	14	26	26	6	21	10	16	18
Completely Drained/Dry Period (Days)	12	0	0	20	0	11	5	3.5
Wet Layup Period (Days)	7	7	7	7	7	7	7	7
# RTZ Circ Cracks 2006	0	2	0	16	0	0	0	0
<b>2006 Outage</b>	<b>SG1</b>	<b>SG2</b>	<b>SG3</b>	<b>SG4</b>	<b>SG5</b>	<b>SG6</b>	<b>SG7</b>	<b>SG8</b>
Partially Drained Period (Days)	38	38	38	20.5	11	11	11	11
Completely Drained/Dry Period (Days)	0	0	0	17.5	0	0	0	0
Wet Layup Period (Days)	13.0	13.0	13.0	13.0	18.0	18.0	18.0	18.0
# RTZ Circ Cracks Spring 2007	0	0	0	44	0	0	0	0
<b>Spring 2007 Outage</b>	<b>SG1</b>	<b>SG2</b>	<b>SG3</b>	<b>SG4</b>	<b>SG5</b>	<b>SG6</b>	<b>SG7</b>	<b>SG8</b>
Partially Drained Period (Days)	15	15	15	13	15	15	15	15
Completely Drained/Dry Period (Days)	0	0	0	2	0	0	0	0
Wet Layup Period (Days)	8.0	8.0	8.0	8.0	8.0	8.0	8.0	8.0
# RTZ Circ Cracks Fall 2007	0	0	---	0	---	---	---	0
<b>Fall 2007 Outage</b>	<b>SG1</b>	<b>SG2</b>	<b>SG3</b>	<b>SG4</b>	<b>SG5</b>	<b>SG6</b>	<b>SG7</b>	<b>SG8</b>
Partially Drained Period (Days)	0	0	0	0	2	2	2	2
Completely Drained/Dry Period (Days)	0	0	0	0	0	0	0	0
Wet Layup Period (Days)	0	0	0	0	0	0	0	0
# RTZ Circ Cracks Spring 2008	0	0	0	0	0	0	0	0

**Table 3. Summary of Recent Unit 4 SG Planned Outage Layup Exposure.**

A strong correlation was found between the presence of significant IGA/SCC damage in SG4 and the time this boiler spent in a completely drained/dry state. This was the case in the 2006 and spring 2007 outages following the extended exposure of SG4 during the 2005 and 2006 shutdowns. Efforts were made during the spring and fall of 2007 to reduce and/or in the case of the fall, eliminate this exposure, resulting in a null finding for IGA-SCC activity in the 2007 fall and spring 2008 outages. During the last two inspections, in Fall 2007 and Spring 2008, no additional cases of circumferential IGA/SCC at the TTS were detected in any Bruce 4 steam generator. The absence of detected circumferential IGA or cracking at these inspections is not consistent with industry experience with IGA/SCC that occurs during of power operation, nor with the model that had been developed based on that experience.

## 2.0 Mechanism

A proposed mechanism for the circumferential IGA-SCC degradation in Unit 4 was attack by reduced sulfur species occurring during unit shutdowns and subsequent startup evolutions. This mechanism has been documented in US nuclear plants with sensitized Alloy 600 SG tubing. For this to be a viable cause of the SG tubing degradation, specific conditions must be met as follows: an inventory of sulphur compounds within the HL TTS sludge-pile region, exposure to oxidizing conditions and the presence of a susceptible SG tubing microstructure. In addition for IGA/SCC to occur, this chemical attack should be supplemented by mechanical stressors to promote cracking of the material. The morphology of the circumferential defects characterized by metallurgical examination indicates that the IGA accounts for majority of the total wall loss.

The presence of sulfide compounds in the sludge material of the Bruce A boilers has been confirmed through analysis of the sludge deposits and in deposits on removed tubes from the Units. During plant operation sulfides and possibly elemental sulfur accumulate in the sludge and crevices at the TTS as a result of the normal concentration processes that act in this area, particularly in the HL region of the SG. The sources of the sulfur are considered to be the low levels of sulfates that entered the SGs with the feedwater, and also a sulfuric acid (sulfate) ingress event that occurred in 1986. These species remain in a reduced state during normal operation or during controlled wet layup.

During shutdown periods when the SGs were not filled with treated water, e.g., periods when the SGs were drained to low levels of about 15–20 cm (6–8 inches), or drained dry during removal of tubes or other work, the TTS area was exposed to oxidizing conditions for extended periods. The oxidizing conditions that occurred during the fully drained periods caused oxidation of sulfides or elemental sulfur in the TTS area, putting the sulfur into aggressive oxidation states, such as those of tetrathionates and thiosulfates. Detailed assessments by AECL of experience at Bruce Unit 4 during a long outage between December 1997 and late 2003 indicated that reduced sulfur species were present in the TTS region, that they can form aggressive environments in the TTS region when the SGs are drained and exposed to oxidizing conditions [1,2]. These assessments also confirmed this mode of corrosive attack occurred in the Bruce 4 SGs.[1,2] Finally, the oxidizing conditions caused oxidation of copper metal particle surfaces, such this copper oxide could act as an oxidant during the subsequent startup.

The high temperature mill annealed Inconel 600 tubing (600 HTMA) in the Bruce 4 steam generators was sensitized during original construction by exposure to elevated temperatures during the vessel post weld stress relief. The formation of chromium carbide precipitates at the grain boundaries resulting in the reduction in the chromium concentration in these regions increased the materials susceptibility to attack by reduced sulfur species. Laboratory testing of removed tubes from Unit 4 has confirmed that sensitization of the tube material has taken place, though there was no increase in extent in SG4 relative to the other steam generators [3].

The presence of the aggressive sulfur species at the base of the thick sludge piles, oxidizing conditions, and acid pH in the TTS crevice area caused pitting, volumetric IGA, and possibly incipient SCC of the sensitized 600MA tube material during the drained period. However, it appears that stresses were generally not high enough during the

shutdown itself to lead to significant cracking. During startup following the completely drained period, growth of the IGA/SCC occur possibly due to increased temperatures and or stresses that occurred during startups combined with the continued presence of the oxidizing conditions and aggressive sulfur species that had developed during the shutdown. Cracking of the material would be accelerated by the stress risers created by IGA that had occurred during the shutdown. It should be noted that this cracking has been limited to the RTZ where high residual stresses are known to be present as an artifact of the tube to tubesheet rolling operation. The absence of significant cracking during the long 1997-2003 outage, despite the clear presence of aggressive reduced sulfur environments, implies that the stresses at roll transitions during layup may not sufficient to cause significant cracking, only pitting and some significant IGA were observed.

### **3.0 Support For Low Temperature Degradation Model**

The combination of the observations above provide a basis for low temperature attack by reduced sulfur species during unit shutdowns and startup not during normal unit operation as previously assumed. To provide a further technical basis for this failure mode Bruce A observations were supplemented with US industry OPEX with regards to defect location, morphology, and rate of initiation/growth. The main considerations that led to the conclusion that the IGA/SCC experienced at Bruce 4 has been mostly the result of reduced sulfur attack at low temperatures are presented below.

#### **3.1 Degradation Progression Based on PWR Experience**

As shown in Table 2, during the 2006 and Spring 2007 inspections, 16 and 44 tubes with circumferential OD IGA/SCC at the roll transition zone at the TTS were detected in the most severely affected SG (SG4). Following normal industry practice for PWRs, this degradation was modeled as increasing with increasing service time, measured in effective full power years (EFPY). Based on this modeling, it was predicted that the numbers of affected tubes in SG would increase rapidly as service time increased. A null result in the Fall 2007 and Spring 2008 inspections contradicts the models put forth. It should be noted as well that the occurrence of volumetric indications in the HL RTZ markedly decreased as control over the exposure durations was established in the Spring of 2007.

#### **3.2 Correlation Between Severity of Degradation and Duration of Exposure to Uncontrolled Layup Conditions**

An evaluation was performed of whether the occurrence of circumferential IGA/SCC in the Bruce 4 steam generators correlated with factors such as length of partially, completely drained or wet layup periods. The only significant correlation was that shown in Table 3, which implies that long dry periods of over about 12 days may correlate with occurrence of the IGA/SCC, while short periods correlate with no IGA/SCC. In addition, the facts that, prior to the last two inspection intervals, there were either very short (2 day) or no completely drained periods and that there was no subsequent detected IGA/SCC tend to confirm the correlation between the length of the dry period and the occurrence of IGA/SCC.

### **3.3 Lab Scale Test Results**

Laboratory tests and PWR plant experience indicate that sensitized Alloy 600 tubes of the type in the Bruce 4 boilers are susceptible to IGA/SCC in environments with reduced sulfur species, low pH, and oxidizing conditions at low temperatures [4]. Crack growth rates in sensitized Alloy 600 can be very rapid in these aggressive reduced sulfur species environments, such that significant crack growth, in the range of magnitudes observed in steam generator 4, is plausible during a short startup period.

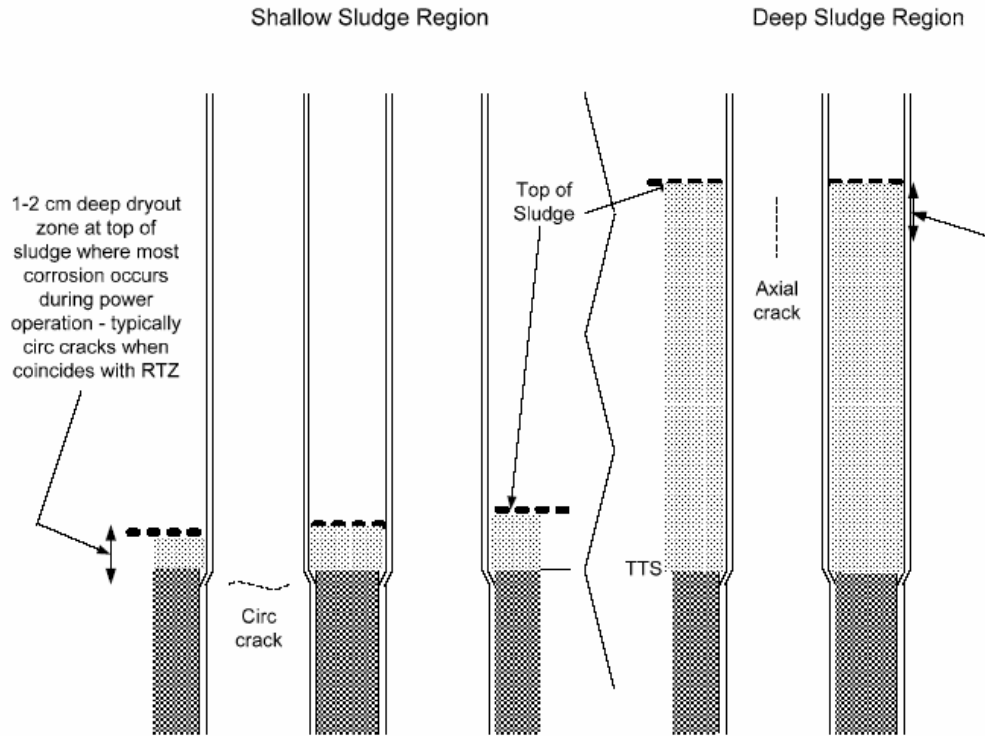
### **3.4 Observed Crack Growth Rates Relative to PWR OPEX**

The crack growth rates determined for SG4, assuming that they occurred at high temperature and high power, are about 10 times higher than expected based on PWR experience. However, they are consistent with the rapid crack growth rates observed in low temperature attack of sensitized material in the presence of reduced sulfur species coupled with low pH and oxidizing conditions [5].

### **3.5 Location of Flaws Within the HL Sludge Pile Region**

The locations of the circumferential OD IGA/SCC in Bruce steam generator 4 are not consistent with the locations observed in PWRs where attack is believed to have occurred at high power. The defect location between the Bruce SGs and the PWR SGs is shown in figure 2. In this regard, model boiler tests and PWR plant experience indicate that, at high power, impurities concentrate in the top 1 to 2 cm ( $\frac{1}{2}$  to 1 inch ) of the sludge, which acts as a dryout zone, and that IGA/SCC tends to occur in this region. For this reason, circumferential cracks at PWRs tend to be concentrated near the edges of the sludge pile where the 1 to 2 cm dryout zone corresponds to the roll transition zone [6]. In the central deep sludge pile region IGA/SCC tends to occur at the top of the sludge and to be mostly axial cracking. In contrast in Bruce 4 SG4 the circumferential cracks and IGA are concentrated in the center of the sludge pile. They also are all close to the TTS, i.e., they are not near the top of the sludge. In other words, the degradation pattern at Bruce 4 is consistent with startup attack but not full-power attack.

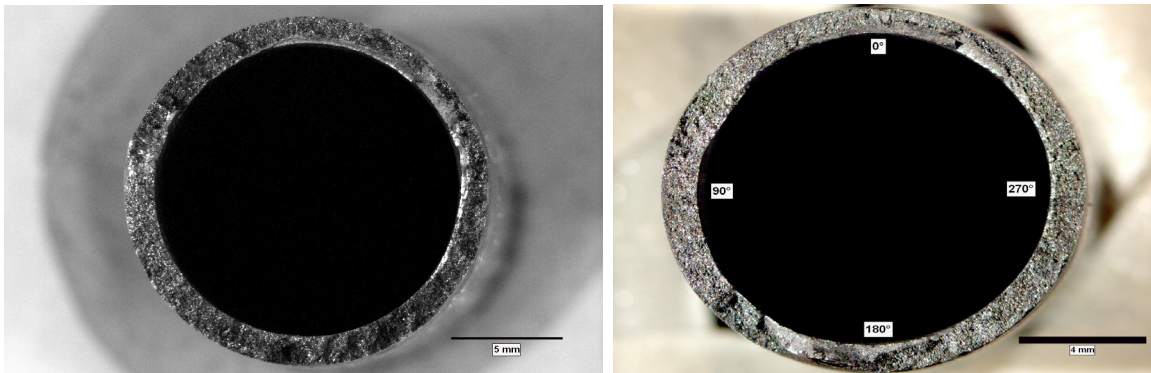




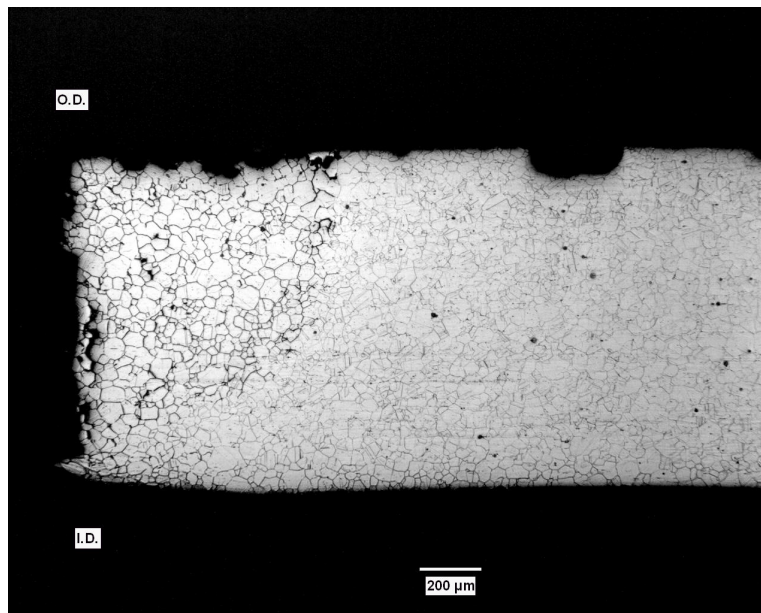
**Figure 2. Location of Typical Defects at the TTS Within PWR Boilers [6].**

### **3.6 Flaw Morphology Based on Metallurgical Examination**

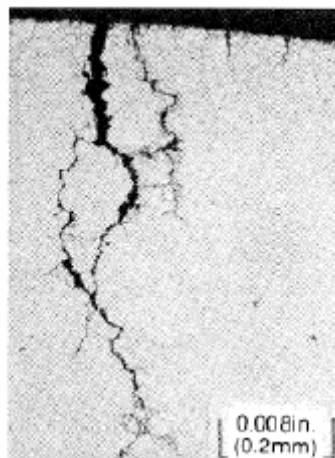
The flaw morphology exhibited by the circumferential OD IGA/SCC in Boiler 4 is characterized by very broad bands of IGA in the roll transition area (figures 3 and 4), rather than by narrow IGA with leading fingers of SCC (figure 5), and thus is more consistent with IGA caused by exposure to reduced sulfur species at low temperature than it is with IGA/SCC at high temperature.



**Figure 3. SG4 fracture surfaces from tube removals during the 2007 spring outage. (Left Tube R33C61 SG4 2007 (PDA at 84%), Right Tube R31C57 SG4 2007,(PDA 75%))**



**Figure 4. Micrograph of tube R28C5 Unit 4 SG4 displaying fracture surface, on the left hand side and the adjacent extensive IGA field.**



**Figure 5. Typical high temperature SCC crack morphology from PWR SG Tubing Failure [7].**

#### **4.0 Prevention and Path Forward.**

The approaches being used to minimize occurrence of circumferential OD IGA/SCC and also volumetric attack in the TTS sludge pile region are based on the conclusion that most of the flaw growth has been of the low temperature type associated with the presence of reduced sulfur species coupled with oxidizing conditions. The preventive approaches include the following:

- 1) The development of aggressive reduced sulfur species in the TTS region is being avoided or minimized by eliminating exposure to partially and fully drained conditions. The intent is to minimize exposure to oxidizing and dryout conditions. Oxidizing conditions tend to develop aggressive sulfur species and to acidify the crevices—and also to oxidize copper particle surfaces (which tend to act as oxidants during subsequent startups), while dryout tends to concentrate chemicals to more aggressive levels [6].
- 2) Following any drained periods, the duration of exposure to treated low-oxygen water is maximized to the extent practical before commencing startup. The intent is to reduce the concentration of aggressive sulfur species by allowing time for diffusion out of crevice areas, increase the pH in crevice areas by allowing impurities to diffuse out and pH agents such as morpholine to diffuse in, and reduce copper oxides by the action of hydrazine before startup and operating stresses and temperatures are applied.

#### **5.0 Conclusions**

The aggressive IGA/SCC was observed in the Unit 4 SG tubing is a result of reduced sulfur species attack. This attack is as result of the oxidation of sulfur compounds to more aggressive species during uncontrolled layup conditions in the SGs during outage campaigns. The rate, location and morphology of the attack of the sensitized Alloy 600 SG tubing is consistent with this hypothesis and differs substantially from that observed for PWRs during high temperature operation. Controlling and or eliminating these exposure durations is critical to maintaining extended operating intervals and end of life plans for Bruce A Unit 4. Favorable inspection results from the upcoming 2009 Unit 4 Outage will serve to confirm the stated mechanism conclusions hopefully showing approximately 2 years of operation with no significant observed IGA/SCC degradation.

## References

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