Agenda for Summarizing Seminar on Heat Transfer Crevices
Summary session: "Take home" from meeting

1. **Primary scope: Predicting properties of line contact heated crevices with respect to occurrence of corrosion of Alloys 600TT and 690TT.**

2. **Incentives**

   a. **Seabrook shows that line contact geometries with Alloy 600TT can sustain SCC. This occurrence should be considered to be the "first-of-many failure."**
      - **Seabrook SCC driven by exceptional high residual stress in some tubes**
      - **However, concave quatrefoil support developed aggressive chemistry**
      - **Industry does not have technique of identifying equivalent high residual stress elsewhere**
      - **TT location known to have high stresses at expansion transition at supports in other SGs.**

   b. **Lengthen time between inspections based on quantitative criteria.**
   c. **Demonstrated corrosion of Alloys 600TT and 690TT in pH ranges relevant to SGs.**
   d. **Not wait for the "inevitable" failures.**
   e. **Prevent future failures**
   f. **Permit conducting credible operational assessments.**

3. **Demonstrated vulnerabilities of Alloys 600TT and 690TT in relevant environments**

   a. **PbSCC (testing in progress with EPRI/Rockwell**
   b. **S^{y-}SCC**
   c. **Si-Al type SCC (testing done by EPRI and EdF/Framatome**
   d. **AcSCC**
   e. **"Complex environments"**
   f. **AkSCC**

4. **Predicting performance of line contact crevice deposits should include:**

   a. **Rate of crevice deposit accumulation**
   b. **Superheat/concentration factor evolution**
   c. **Low solubility precipitate accumulation**
   d. **Species accumulation**
   e. **Contribution of streaming potential**
   f. **Applicability of information from drilled hole studies.**

g. Effect of diffusion of primary-originated hydrogen into secondary crevices
h. Environment definition, models

5. Critical experiments in model boilers that should be undertaken

a. Rate of development of deposits, solution chemistry.
b. Properties of deposits: superheat condition, concentration of species, geometry of deposits, pH, potential, deposits.
c. Chemical interactions: Reduction of sulfur
d. Effects of flow velocities; streaming potential.
e. Develop methods of detecting fouling in line contact crevices.
f. Bench marking models.

6. Critical experiments in heat transfer lab experiments

a. Facilities available and attributes throughout the world.
b. Present plans for respective facilities.
c. How integrate work?

7. Other experiments

a. Examine tubes
b. NDE detection of critical crevices.

8. Compilation of data sources

a. Cycle averaged chemistry
b. Prompt hide out return
c. Tube examinations: deposits, crack face, corrosion morphology.
d. Chemical cleaning

Panel Discussion Outline

Allen Baum
Bettis Atomic Power Laboratory

- Issues Summary
- Ongoing and Future Work
  - Canada – Peter King
  - France – François Vaillant
  - Sweden – Per-Olaf Andersson
  - U.S. – Keith Fruzzetti
- Corrosion Prediction – Roger Staehle
- Recommended initiatives – Roger Staehle

Issues Summary

- Offensive
  - Tubing materials
  - TSP design
  - Tubesheet joint design
  - Improved chemistry control
  - Fewer lead-bearing components
- Defensive
  - Seabrook evaluation
  - Design concerns
  - Accelerated 600 & 690TT corrosion

TSP Design Issues

- Benefits
  - Lower temps and superhts
    - Lower concentration factor
    - Shorter transport span
    - Less HO & HOR
  - Slower fill for flat configs.
- Liabilities
  - > 2X entrance region
  - Smaller reservoir
  - θ transport
    - Longer cracks
    - Less TSP restraint
  - Flat config. crevices could have more lead

Ongoing/Future SG Research in Canada

Presented by Peter King

<u>B&W Canada</u>

- Long-term expectations for Alloy 690 TT
  - general oxidation testing in AVT
  - detailed passive film evaluations
    - Auger, XPS, SIMS, FESEM, ATEM??
    - other properties
  - evaluate cracking pre-cursors
    - stressed/unstressed
  - compare to 600 MA
    - may include 600 SR, 600 TT, 800
  - formulate expectations for crevice chemistries
    - nature of possible chemistry
    - likelihood of occurrence of various outcomes
  - oxidation testing in crevice environments
    - specific solute influences
    - complex environment
- SCC testing of pre-filmed alloys?

- time to critical crevice deposits
- how to interpret & use extensive field data & relate it to 690 TT expectations

Future SG Work in Sweden

Presented by P. Andersson

R2-repl-89:690 TT
R3-repl-95:690 TT
R4-origin-84:600 MA

1. Transport into/in packed crevices
   - kinetics for HO and HOR
2. Examination of the used crevice
   - deposits physical properties

     chemical properties
3. Repeat earlier HO-tests with Na-24

   Compare with earlier data
   - Does concentrating occur?
   - changes?
4. CGR for 690 TT
   - primary coolant, started
   - secondary??

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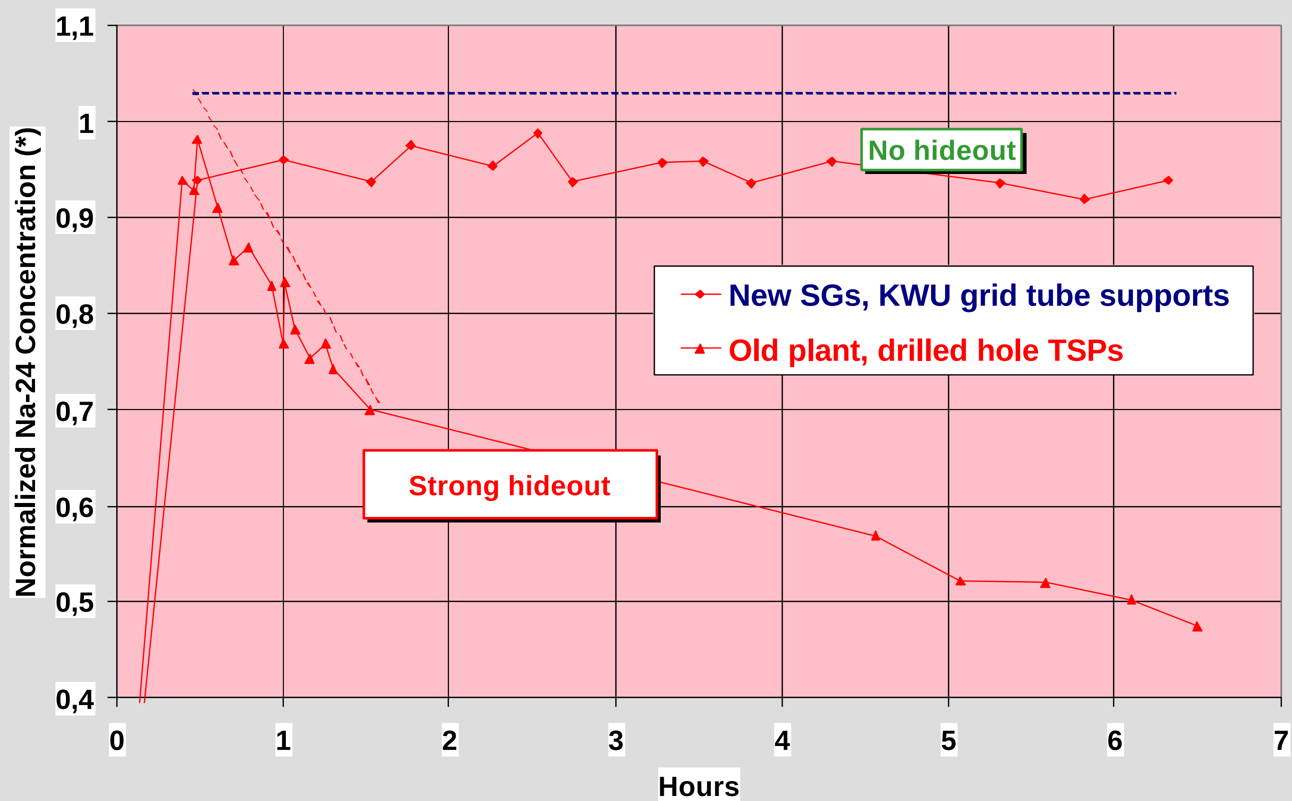
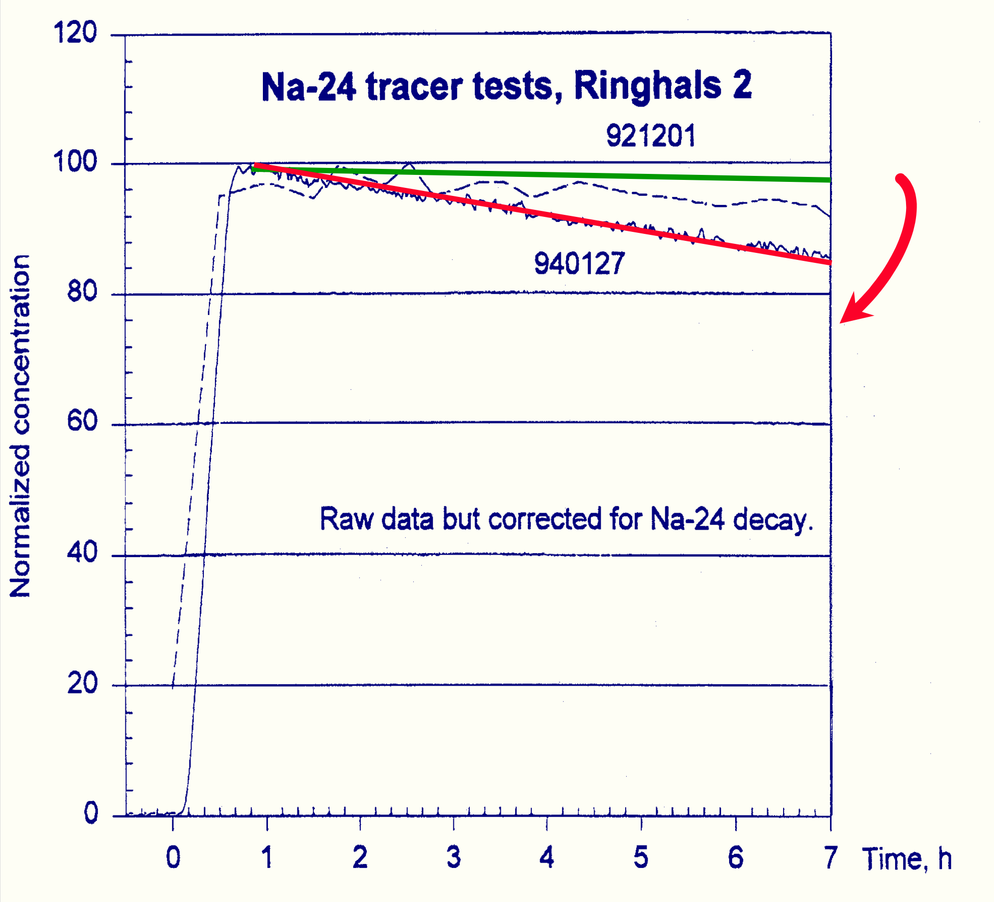
Hydrazine Ratio and Na-24 Hideout Studies

Suat Odar
Framatome-ANP GmbH

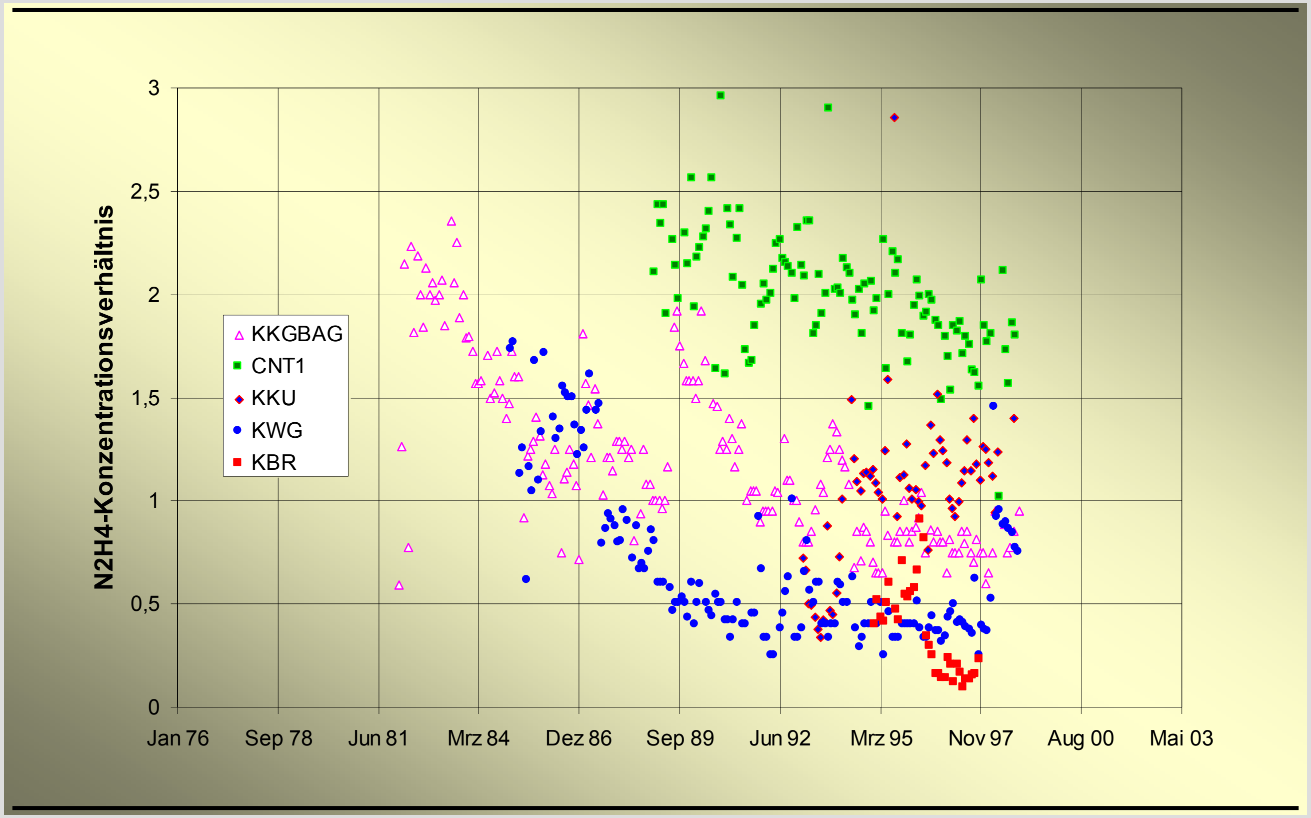
Does Corrosion Risk increase with Increasing Hide-Out?



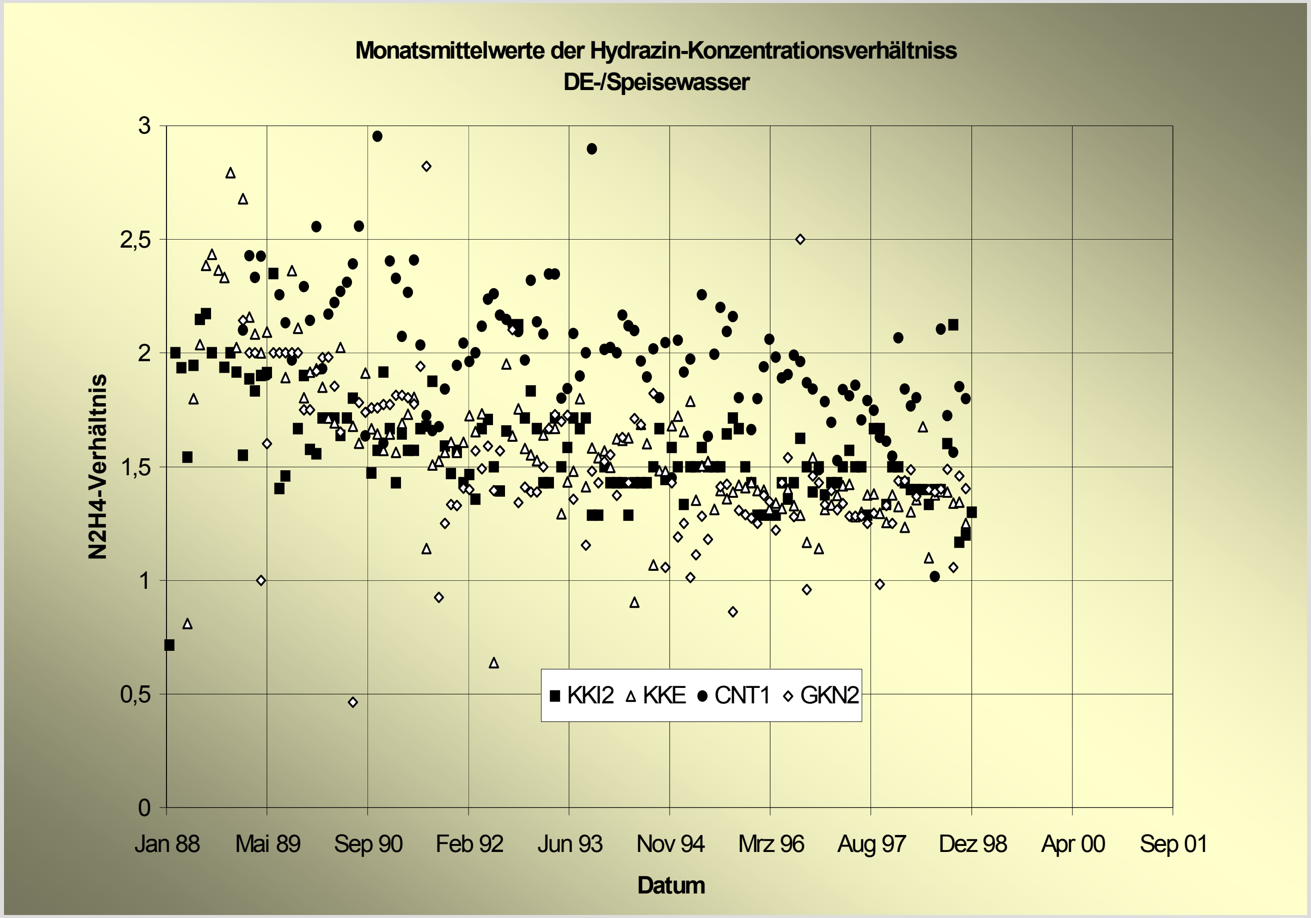
Hide-Out Increase with Operating Time



N_2H_4 Concentration Ratio SG / FW
FW Fe Concentration ~ 1-2 ppb



N_2H_4 Concentration Ratio SG / FW
FW Fe Concentration < 1ppb



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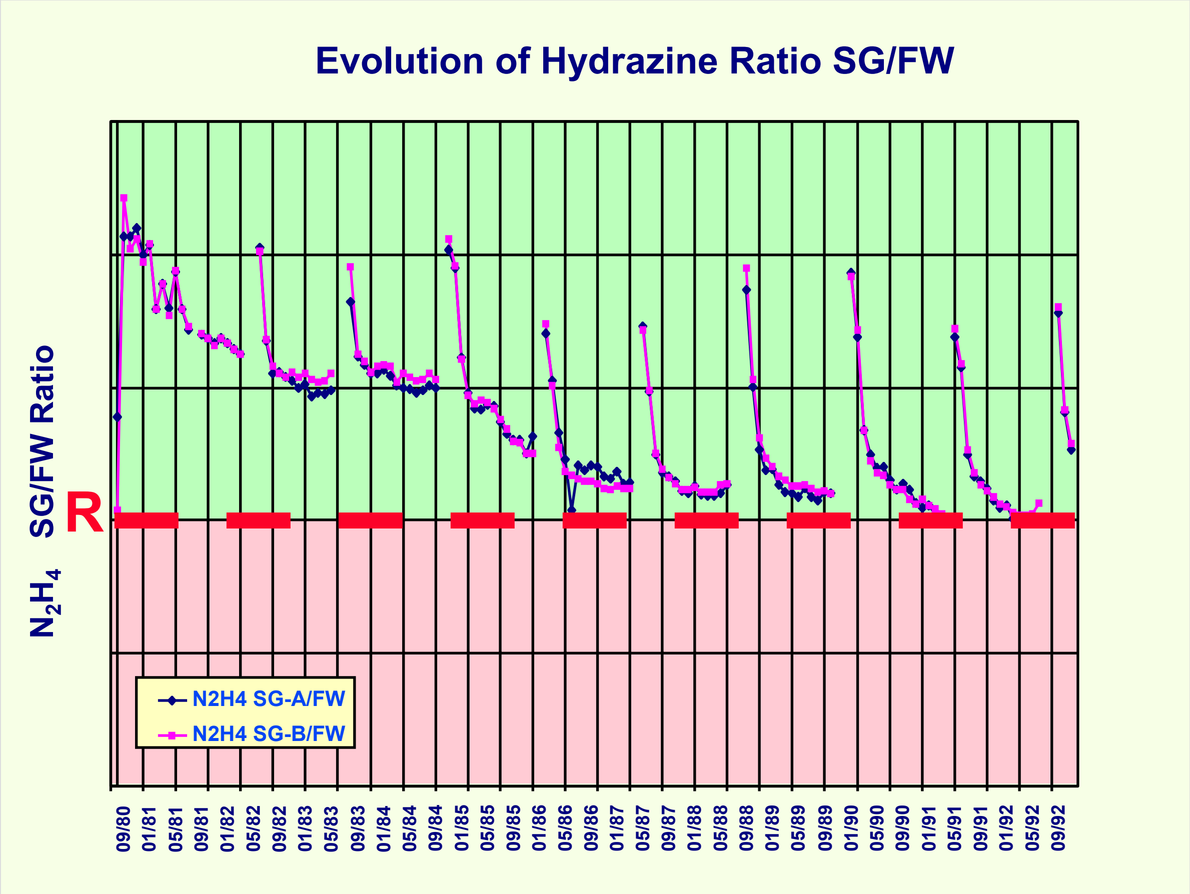
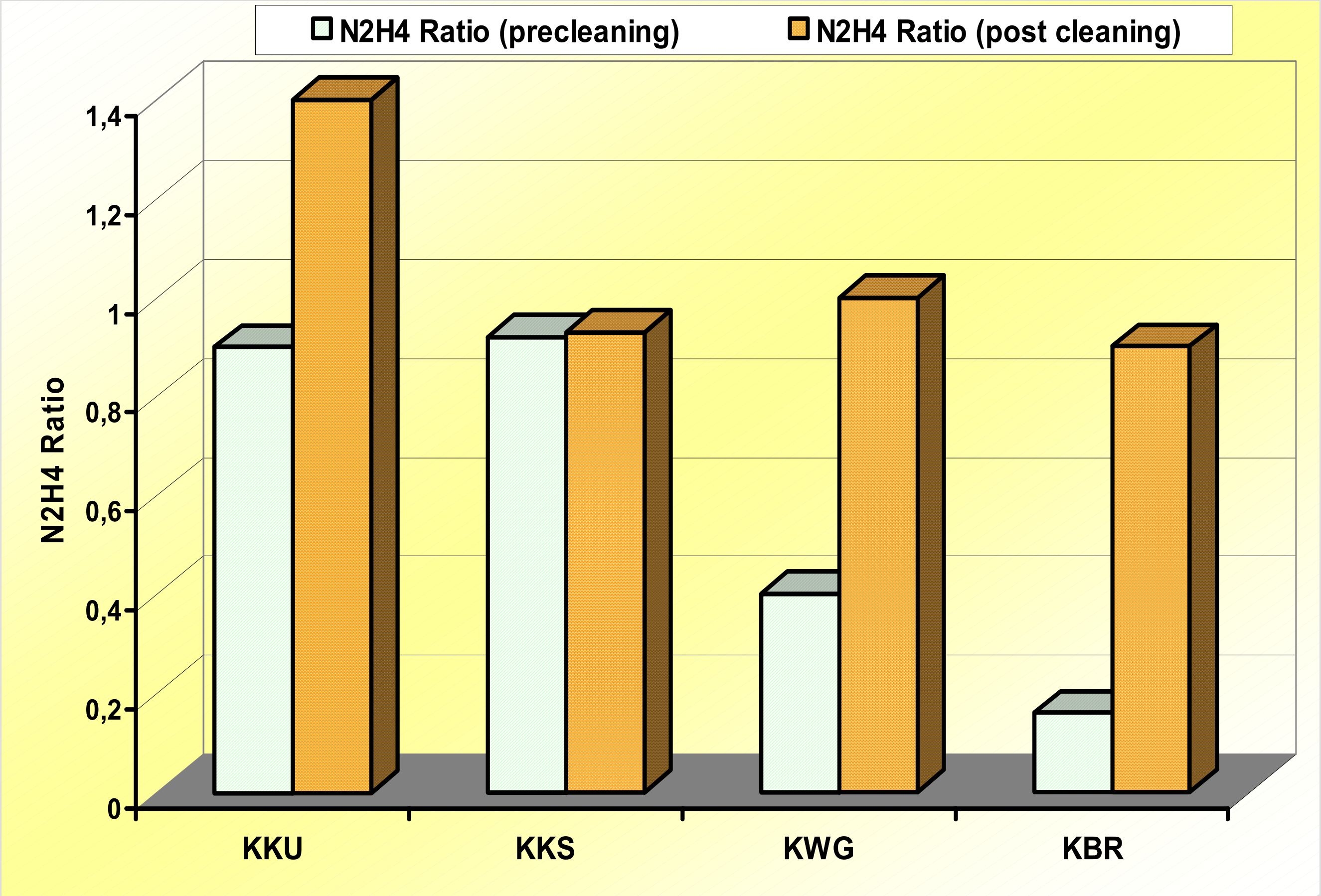
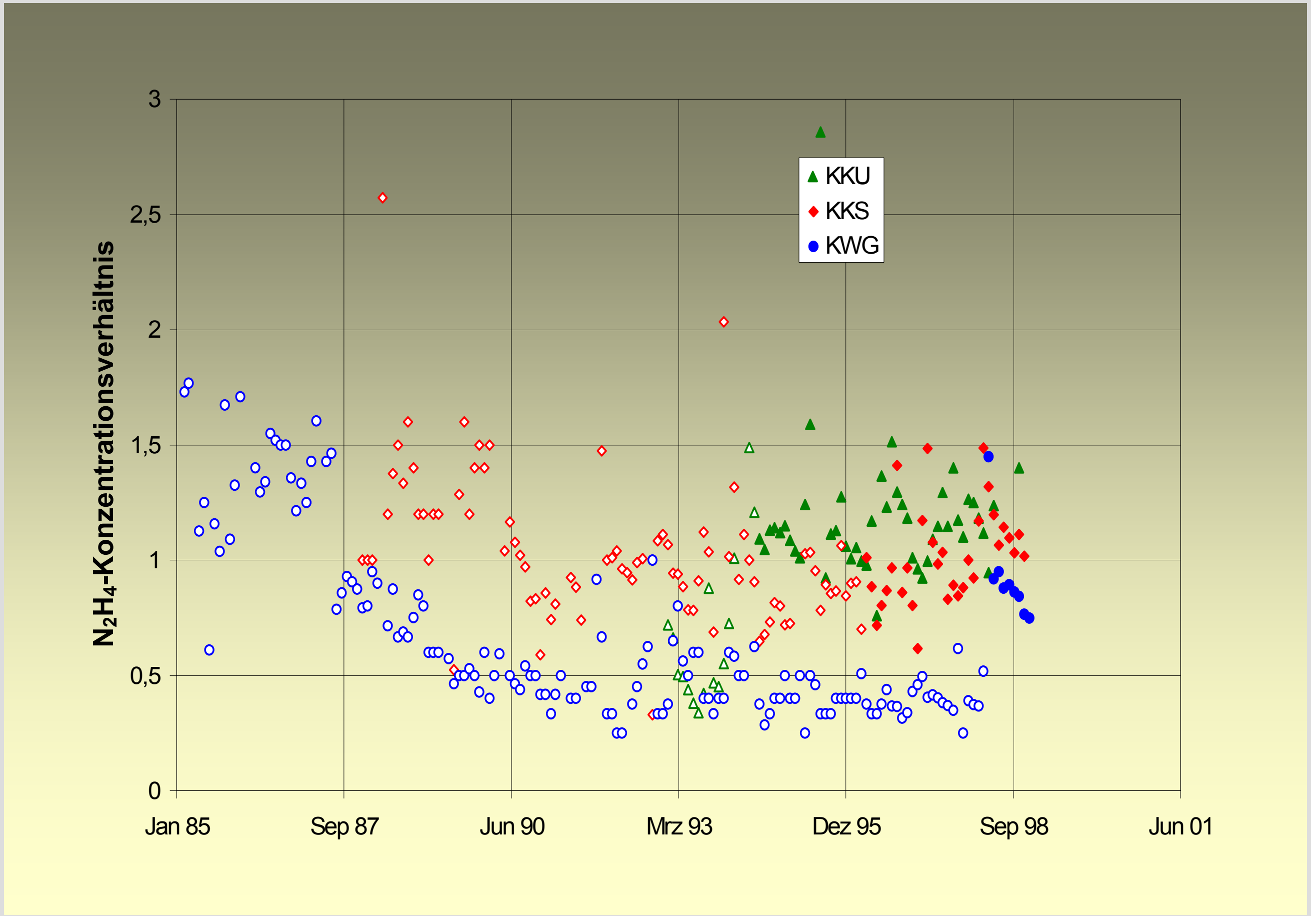
Hydrazine Ratio Time Evolution



Application Results
Thermal Composition of Hydrazine



N_2H_4 Concentration Ratio SG / FW
SG Chemical cleaned PWRs



Ongoing/Future SG Research in France

Presented by F. Vaillant

HELP TO PLANT OPERATION

- [Hydrazine] requirements
  - compatible with risk of SCC by reduced sulfates

    corrosion tests in loop
  - compatible with risk of SCC in (hydrogenated)
  - to avoid FAC

- SCC with shutdowns/start-ups
  - initially tests on Ajax loop
  - simulation in NaOH (static autoclave)

LIFE PREDICTION + FREQUENCY OF NDE

- Based on corrosion model in laboratory:
  - sodium hydroxide
  - neutral to slightly alkaline sulfates
  - complex environments
    (SiO_2, Al_2O_3, organics, phosphates) in AVT

Proposed 2003 EPRI Programs addressing SG Fouling and Crevice Issues

Project Manager: Keith Fruzzetti

Heated Crevice Seminar
October 7-11, 2002
Argonne, IL

TAG 1

EPRI

Heated Crevice Program

Rockwell (Jesse Lumsden)

- Current 2002 Program
  - Evaluating sulfate hideout and formation of reduced Sulfur species by hydrazine
    - Reduced S causes SCC in Alloy 600 and Alloy 690

- Proposed 2003 Program
  - Complex Chemistry
    - Precipitate formation (Raman)
    - Crevice extraction to determine soluble species
    - Hideout return evolution

Evaluation of Line Contact Crevices

iSagacity (Peter Millet)

- Evaluate the susceptibility to tube corrosion at line contact support locations
  - Performed detailed review of experimental and modeling programs (including EPRI projects S118, S119, S121, S180, S192, S133, S134)
  - Develop a qualitative fouling (deposition) model for line contact crevices
  - Rank the susceptibility of various designs to fouling based on experimental and modeling database
  - Determine if a fouling threshold can be obtained from the existing database

EPRI

Multivariable Influence on SG Fouling and FAC

B&W Canada (Peter King)

Project Objective:

- Improve the fundamental knowledge of processes responsible for corrosion, corrosion release and transport, and deposition in the steam generator
  - Improve tools for multivariable analysis to enable preventative strategies against FAC and SG fouling
  - Improve current models for predicting the effect of the chemistry environment on FAC

Multivariable Influence on SG Fouling and FAC

Task 1: Collection and Evaluation of Plant Data and Experiences

- Dissolved and particulate Fe conc's
- pH, Hydrazine, Oxygen, ECP
- Fouling Data

Task 2: Literature Review on fouling and FAC

Task 3: Research into the fundamental role of soluble iron

- FAC & Electrochemistry under soluble iron transport conditions (MTI)
- The role of soluble iron in SG fouling (AECL)

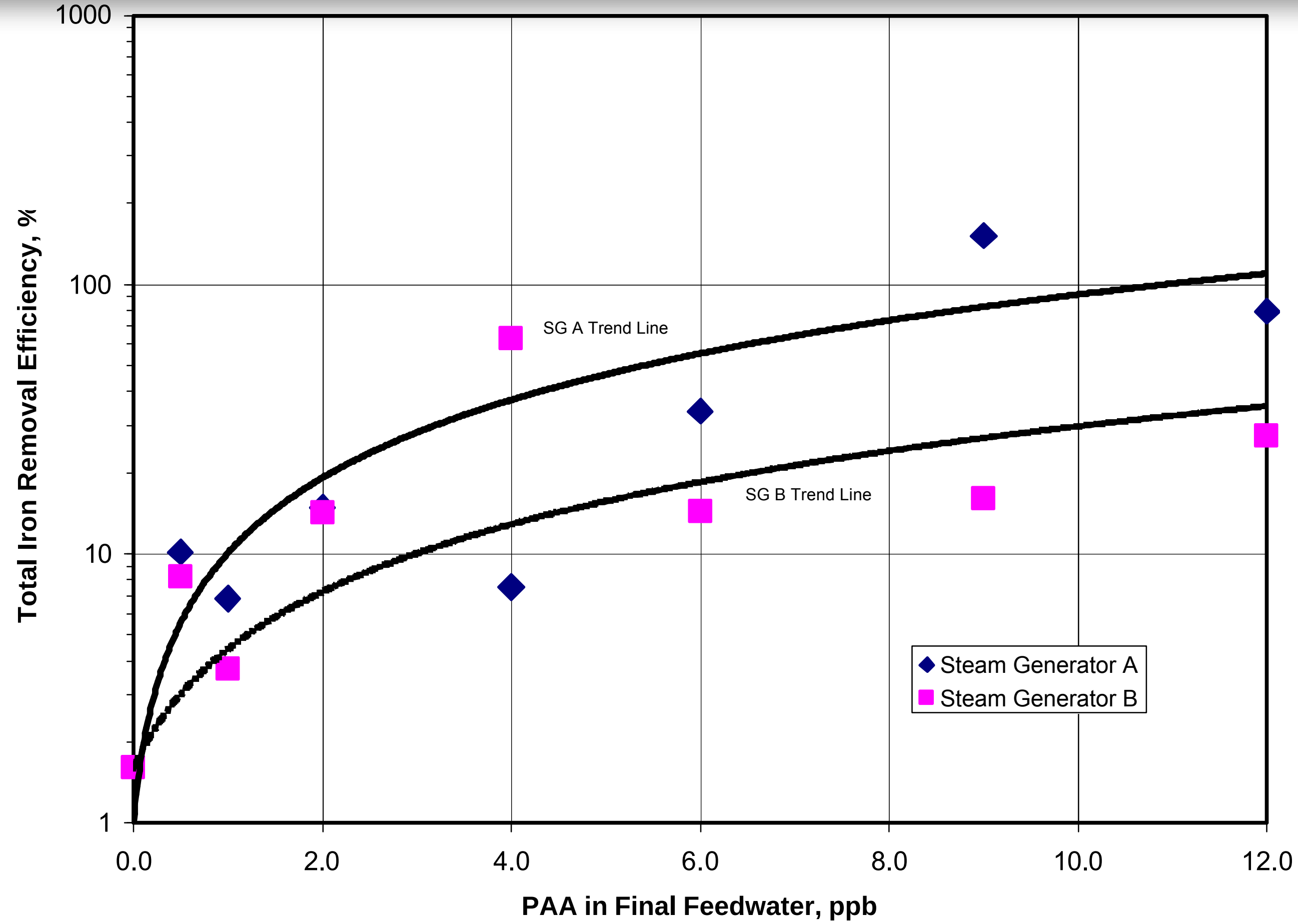
Task 4: Model development for prediction of ECP in the steam cycle

- Facilitate prediction of the operating margin for stability of the protective oxide

Dispersant Program to Mitigate SG Fouling

- Short-Term Trial Completed (ANO-2)
  - Qualification Report completed 3/29/01 (1001422)
  - Trial Report Completed 9/24/01 (1003144)

Short-Term Trial Results



TAG 7

Dispersant Program to Mitigate SG Fouling

Replacement 690 Steam Generators (W and BWC)

- Long-Term Trial Effort in Progress
  - Engineering Assessment for Replacement 690 Recirculating SGs (DEI)
    - Thermal/Hydraulic and thermal margin assessments
    - Separator performance assessments
  - Vendor Reviews in Progress
  - Potential Trial Site Expanded
    - Westinghouse and BWC
    - ANO-2 ready to inject with vendor concurrence

Dispersant Program to Mitigate SG Fouling

Complete Long-term Qualification Work

↓

Vendor Review: Phase 1 (2002 Effort)

Concurrence by Vendors for a Long-term Trial in a Replacement 690 Recirculating SG

↓

Perform Long-Term Trial (ANO-2)
and assess results

↓

Vendor Review: Phase 2 (2003 Effort)

Concurrence by Vendors for Long-term use in fleet of domestic W/CE & BWC Recirculating SGs

2003 SG Secondary Side Management Conference

- 3 Day Meeting: February 10-12, 2003
  - Savannah, GA
- Four Main Topical Areas
  - Deposit Generation and Transport
  - Deposit Control and Mitigation
  - Deposit Consolidation and Removal
  - Short and Long Term Strategic Planning

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Notes from Panel Discussion Session (Friday October 11) by D. R. Diercks

Roger Staehle began by stating that the principal objective of the panel discussion was to "bring things back into focus." He then handed out a proposed agenda for the discussions.

Alllen Baum then opened the first portion of the discussion with a short presentation.

Gorman began the discussion by inquiring about the nature of the cracking seen in the Alloy 600 TT tubes at Seabrook. Mcllree replied that the cracks were short and separated by ligaments. He said that even if they were 99+% throughwall, they would not threaten the pressure integrity of the SGs.

McIlree then noted that if Alloy 600 or 690 TT tubes were properly processed at the mill, in principle there should be no residual stresses present to drive cracking. At Seabrook, however, they found that the tubes were re-straightened after the TT heat treatment, thereby introducing residual stresses. It seems likely that other TT tubing is out in the field with similar residual stresses. He wondered if we could develop NDE techniques to detect residual stresses in tubes in the field or to determine if the TSP lands were becoming active crevices.

Jim Davis (NRC) then commented that the Seabrook tubes appeared to have a MA metallurgical microstructure rather than a TT microstructure. McIlree observed that there is a great variation in TT microstructures, and some in fact resemble the MA microstructure. He stated that one cannot judge SCC resistance from the microstructure. The response of the microstructure to the heat treatment appeared to depend upon the C level. Gorman added that EDF experiments indicate that $\leq$ 0.033 wt. % C was needed to obtain the expected microstructural response to the TT heat treatment.

Davis noted that cracking had also been seen in Alloy 600 TT at a Korean plant, and Baum and Diercks (ANL) said that this was the Kori 2 plant. McIlree added that again the microstructure did not have the appearance normally associated with the TT heat treatment. McIlree said that we must assume that there are other "poor" TT microstructures in the field, and we must control residual stresses and environment in these plants to avoid cracking. He felt that most, but probably not all, of the plants with Alloy 600 TT tubes have low residual stresses.

Staehle observed that the highest stresses in SG tubes are typically at the top of the tube sheet. He suggested that perhaps our focus on chemistry at the line contact region of the tube sheets might therefore be somewhat misplaced. If we could somehow reduce the residual stresses from tube manufacture and SG fabrication, we might not have to worry so much about the operating environment. However, this does not seem likely. He agreed with McIlree that perhaps the use of NDE techniques to detect residual stresses in tubes in the field should be looked into further.

Duncan suggested that we should determine if there is a small population of high stress tubes in service that need to be followed more closely. Baum said that his earlier model boiler work focused on tube sheet joints, where the residual stresses were highest. Nevertheless, that one incident of ODSCC in Alloy 600 TT tubing was in a TSP crevice rather than a TS crevice.

Staehle then suggested that another important consideration is to figure out how to lengthen the time between inspections. He also noted that, in laboratory tests, both Alloy 600 TT and 690 TT can be cracked in environments not greatly different from those in service. He wondered why they are not cracking in service in significant numbers. Finally, he stated that we must be proactive in anticipating failures in Alloy 690 TT. We need to do the research now so that we are in a position to prevent failures in the field. As a part of this, we need to do research to enable us to conduct credible operational assessments.

Duncan stated that in addition to lengthening the time between inspections, we must make decisions about appropriate inspection intervals in the case where we have an existing crack. We therefore need crack growth rate data under the relevant conditions.

Muscara said that regulators must take the position that undetected cracks exist in the field, and these cracks must be properly dealt with. He also noted that should not totally shift our emphasis away from the TSP region, since conditions for mild denting exist there and even this mild denting can lead to cracking.

Gorman agreed that crack growth rate data are essential for doing a proper operational assessment. He also noted that the subject of mild denting at the top of the tube sheet was considered at a secondary side workshop in 1995. He expects to see more such denting in the future, leading to tube cracking.

Muscara noted that in terms of crack evolution and operational assessments, we must consider when to use ligament correlations for existing cracks and when to use a planar crack correlation.

Baum then offered one last comment on the Seabrook cracking, noting that while the greatest concern is on the mechanical aspects (i.e., residual stresses), there is also a chemical aspect to the problem. He noted that Seabrook had low silica (5-10 ppb) in the bulk water chemistry compared to other plants, and he wondered if this was significant.

Peter King then briefly reviewed ongoing SG research in Canada (Denise is typing up his handwritten overheads).

King noted that the precursors for cracking do not appear to form in Alloy 690 like they do in Alloy 600, at least not in reasonable times. He felt that the major questions to be answered were the time to critical crevice deposits and how to interpret and use the extensive available field data and relate it to our expectations for Alloy 690 TT.

Chambers then made several points. He first noted that residual stresses from manufacture are unavoidable. In addition, one would expect operating stresses in tubes from temperature cycling. He also suggested that *in situ* monitoring of autoclave and heated crevice test (e.g., Raman spectroscopy) could be very enlightening. Finally, he wondered how crevice chemistry might change under zero-power hot conditions.

King responded by first noting the work of Lumsden and others on monitoring crevice chemistry. With respect to the effect of zero-power hot conditions on crevice chemistry, he noted that people are working on this problem. He added that it is very difficult to get crack growth rate data under field conditions. He also agreed that we must assume that tubes, in general, have residual stresses from manufacture, and we must also assume that operations will produce aggressive environments somewhere in the SG. Therefore we must have the most crack resistant tube material possible, and this is why the industry is going with Alloy 690 TT at present. He stated that BWC is still interested in model boiler testing and crevice monitoring, but these tests are very expensive. He expressed the hope that future work at ANL can address these areas.

Per-Olaf Andersson then briefly reviewed ongoing SG research in Sweden. He noted that Sweden has only three PWRs, namely Ringhals 2, 3, and 4. Units 2 and 3 have replacement SGs, and there have been no problems with these. Unit 4 still has the original Westinghouse Model D-3 SG with Alloy 600 MA tubes from 1984, and this unit has experienced very little tube cracking.

Odar supplemented Andersson's presentation by showing results obtained from Na-24 hideout studies conducted in the Ringhals 3 plant simulator before SG replacement. Significant Na hideout was observed. The new KWU SG with grid tube supports showed virtually no hideout. Tests conducted three years later showed some hideout, indicating sludge buildup. The hydrazine concentration ratio in the SG feedwater for plants with higher Fe (1-2 ppb) decreased with time. For lower Fe (<1 ppb), the decrease was less rapid, thus suggesting increased sludge loading.

François Vaillant then summarized ongoing SG research in France. He reviewed work on life prediction and frequency of NDE inspections as well as on hydrazine requirements and SCC under plant shutdown and startup conditions.

Keith Fruzzetti reviewed EPRI-sponsored work on SGs.

Staehle then continued the discussion by noting that in order to predict cracking in a specific environment, we must have sufficient relevant data. However, almost no relevant data exist on cracking associated with reduced S species. With respect to silica effects, he cited a 1985 paper by Berman, who found a specific region in the silica-alumina system where cracking occurs in Alloy 690 MA. He felt that acid SCC had been pretty well characterized, as had alkaline SCC, though both remain issues. Cracking in complex environments has not been well defined, but relevant work is being conducted in France. Overall, he felt that there was much room for serious work to determine the dependencies for several of these submodes. He added that we cannot predict the behavior if we do not know the dependencies.

Gorman stated that, with respect to sulfate chemistry and complex environments, he has difficulty in understanding the process for attack since the species involved are not liquid at the superheats present in drilled hole crevices. He felt that further testing and/or modeling was needed.

Lindsay made an impromptu presentation on the subject of predicting the performance of line-contact crevice deposits, noting that, on the molecular scale, the tube surfaces and the TSP lands look flat. We must consider things on this scale. When things are considered on this scale, it is clear that chemical reactions in solution within the various types of crevices are not different from each other or from reactions that can take place in bulk water. The differences among crevice types are most likely due to their differing susceptibility to fouling. Lindsay added that he was not aware of any significant engineering science relating to crevice fouling under boiling heat transfer conditions. Good engineering science research is needed in this area. Staehle concluded that the important question is what remains to be done in this area and what resources are available to do it.

On the subject of critical experiments in model boilers, Staehle noted that there are only one or two model boilers available at present. How do we best use them and what alternative experimental techniques do we have? Duncan agreed that model boiler tests are needed, since codes and modeling cannot answer all of the questions that must be addressed. He said that model boiler experiments should emphasize multiple variable validation experiments. He felt that a different model boiler design from that being developed at ANL was needed to understand the fouling process at the crevices. Muscara noted that the ANL model boiler was not designed for such studies, but rather to study the evolution of crevice chemistry and how it leads to crack initiation.

Baum added that MULTEQ is a useful tool. But model boiler experiments were nonetheless needed to determine crevice chemistries with confidence. Duncan contended that model boiler results cannot reliably predict absolute cracking rates—they can only provide relative rates.

Staehle then stated that he felt that items 6-8 on the panel discussion agenda had already been thoroughly considered in the previous sessions, and, in view of the short time, they would not be further discussed here.

Muscara observed that, with respect to the data base, a tremendous amount of work was still needed. He suggested that perhaps an international group should be constituted to coordinate this work, and he suggested that perhaps the NRC and EPRI could pull this together.

Staehle suggested that discussions be ended on that note, and he thanked all of the participants.

Questions/Answers and Comments

October 8 (Tuesday) Topic: Corrosion in Crevice Geometries

An Overview of Recent French Studies of Possible Secondary Side Crevice Environments Causing IGA/IGSCC of Mill Annealed Alloy 600 SG Tubes by P. M. Scott and F. Vaillant [Keynote Talk]

Question to Francois Vaillant/Peter Scott by Jeff Gorman

1 Re the plant with TGSCC in the TTS area, did it also experience significant SCC in TPSs? If not, why not? (Why did lead only affect TTS area, if this is the case?)

2. In France, were some TT 600 tubes restraightened after TT? If so, were they re-TT'd?

3. What are the pH and potential dependence of CGR & crack initiation in complex environments?

4. Has anyone done tests in doped superheated steam at 320°C (with various levels of hydrogen)? ANS – last slides – tests in process – C-rings – 600 MA will crack.

5. Could the gap size – BPE effect allow more complex solutions to stay liquids (?), e.g., wall based solutions?

Response to Jeff Gorman by Francois Vaillant

1. TGSCC was found in one plant, with lead near the crack. I am not sure that investigations (of cracks and deposits) were performed in other parts of the tubes … and on other tubes.

2. I am not aware of restraightening of heat-treated tubes in Alloy 600.

3. Most of the results (at least in EDF) in complex environments were within the range 5 to 6.5. the influence of pH was not investigated, but the ratios of the different species or the addition of same species (carbonates) which could modify the pH are the matter of the ongoing program in EDF. The influence of the potential is still to be clarified.

4. The influence of hydrogen in superheated polluted steam could be investigated in future programs, and not yet scheduled.

5. I don't know at now, but yes in the principle.

Question to Peter Scott by Peter King

1. You talk about both brittle, non-protective chromium hydroxide gel. Could you please comment further on the use of these two terms: are they referring to the same thing but at different physical points (i.e., dry at ambient temp. vs. at operating temp in aqueous system) or are they different?

2. In your presentation the occurrence of IGA/IGSCC in complex environments is related to the breakdown of the passive film by formation of alumino-silicates, etc. In these conditions, you have said that 600 TT is more resistant and that 690 TT is immune.

    a. Why would the passive film on 600 TT be better than that on 600 MA?

    b. Is 690 TT truly immune or is testing too short? Have passive films on 690 TT been shown to resist becoming non-protective?

3. Arguments for dryout apply to deposit blocked TSP locations. How do they apply to deposits on free tube surfaces?

Response to Peter King by Peter Scott

1. The two terms are used interchangeably to describe the same thing. The implication of the word "gel" is that molecules of water are incorporated into the molecular structure. No effect of temperature between the two terminologies is implied.
2. Our interpretation of the comparative behavior of alloy 600 MA and TT is primarily an effect of grain boundary structure on propagation rates. Obviously no intrinsic difference in passive film behavior between the two having the same composition is expected. However, alloy 690 with its higher chromium content appears to resist formation of the non-protective chromium hydroxide "gel". Whether long term exposure could change that situation is an open question, although I doubt it in these environments. The presence of minor amounts of lead (Pb) could be expected to degrade completely the protective qualities of the passive film so there may be the possibility of synergism between Pb and the alumina-silicates.
3. We have not observed such dense deposits on the free span; they are only seen in the crevices and then primarily at the entrance and exit to the crevice.

Question to Francois Vaillant by Peter King

1. You reported circumferential cracking <u>under</u> the top surface of the TS. Normally, this would be taken to mean the expansion-transition region. Could you please comment further on your use of the term "under the top surface of TS?"

2. Please comment further on the observation that cracking seems to be more predominant in low yield stress material (your graph of degradation vs YS) and the relation of this to threshold stress (EDF caustic model). I would interpret this to mean that some amount of plastic strain is necessary for cracking to proceed.

Response to Peter King by Francois Vaillant

1. The observed circumferential cracks were located in the crevice at the roll-transition, the low contact between tubes and the tube sheet being under the surface of the tube sheet.

2. The relationship between the high susceptibility of tubes with low YS (field experience) and the low value of the threshold stress in laboratory, could result from the fact that most of the tubes at the TSP level may have similar stress levels (residual stresses and operating stresses). At a given stress level, the difference between the stress and the threshold stress is the highest on the tubes with low YS, inducing severe degradation. Some plastic deformation is likely, even if the threshold stress is slightly lower than YS (σ_s = 0.55 YS + 82 for the average line).

Approach to Predicting Corrosion of SG Tubes Based on Quantifying Submodes of SCC in a Statistical Frame Work by Roger W. Staehle [Keynote Talk]

Question to Roger Staehle by Peter King

I continue to have difficulty with the idea that there is a fundamental basis for the value of β. β is an empirical fit parameter for the failure process, not the direct consequence of fundamental processes. In this context, I think that a search for a functional relationship for β is problematic.

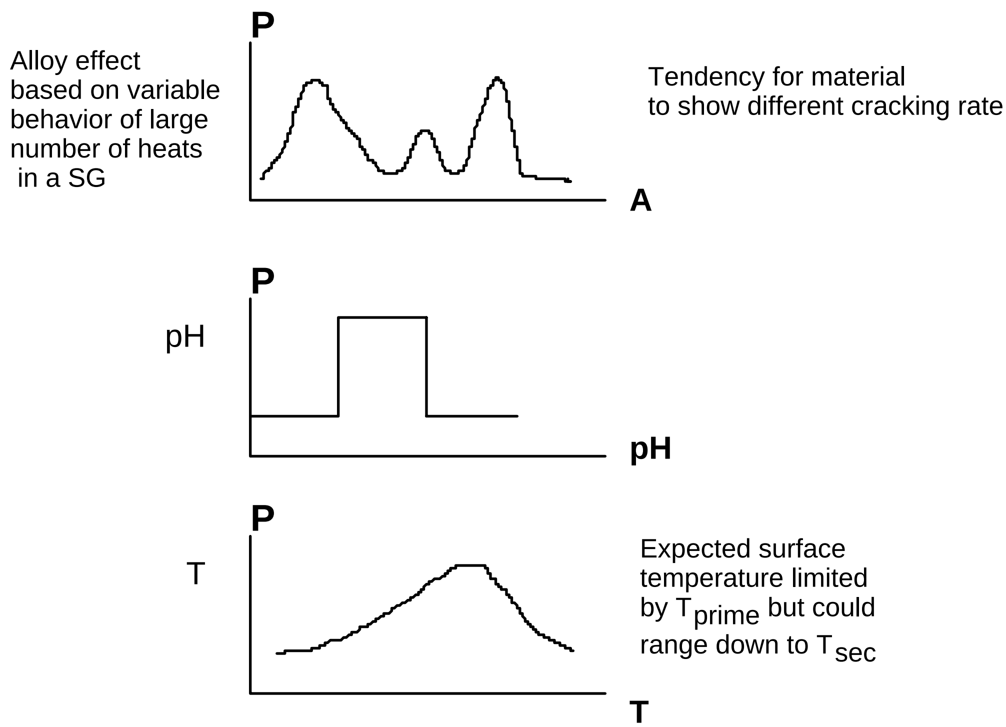
I do not argue that a Weibull distribution is an appropriate representation of a real world failure distribution. However, I believe that the value of β is the consequence of stochastic variability of the input factors, rather than being a consequence of the absolute value of the input factors. In other words, β is a function of the uncertainty in the input data, not the inputs themselves. This would be true for both laboratory experiment and field data.

I would suggest that a better approach to obtaining a final β would be to apply Monte Carlo techniques to a deterministic model of cracking. Stochastic variability (probably not a Weibull distribution) in the various input parameters will yield a distributed outcome. I suspect that said outcome would be well fitted by a Weibull distribution.

For instance:

$$x = \left[H^{+}\right]^{n} [\gamma]^{p} \sigma^{m} e^{\left(\frac{E - E_{o}}{b}\right)} e^{\left(\frac{Q}{RT}\right)} t^{q} .$$

is deterministic. However, if each of the parameters are represented by probability functions (i.e., allow for parameter uncertainty), then the outcome x is a distributed function,



Response to Peter King by Roger Staehle

The shape parameter is fundamental to the physical processes. Surface controlled processes give characteristically $\beta \approx 1.0$. Propagation controlled processes give $1.6 < \beta < 10$ depending on the system. There has never been an effort to explain β except for this first order difference. Rather than β representing an arbitrary stochastic representation, it is rather the result of the mechanistic process. Aside from mechanistic interpretation β is affected by the aggregation or data as explained in my TMS paper.

Question to Roger Staehle by Jiaxin Chen

It is very good to start to study the corrosion problem through a fundamental approach such as the one started by Prof. Staehle.

My questions basically concern:

1. Probability theory as used by Prof. Staehle is feasible if those “submodes” probability functions can be considered as independent from each other. The independency should mean that each involved probability function could stand alone without being necessarily connected to some parameters that are used simultaneously for other “submodes.” This is the start-up point when one could write the “total probability” function.

2. Probability theory is generally more useful when the 'event" involves physical processes. When chemical processes are involved, it has problem. The problem is caused by the fact that, in such a case, it is not the quantity" it changes, but probably the "nature" it changes. In other words, the total probability function may not be a continuous function (mathematically) of the "submodes" probability functions which are likely non-continuous function, too.

Therefore, interpretation of the "total probability" function may be difficult. But, the above-mentioned problems should not hinder further refinement of the thinking which may lead us to a better understanding of the mechanisms.

I look forward to reading more about Prof. Staehle progress in this exciting field as he is exploring now.

Response by Roger Staehle

I appreciate your comments.

October 8 (Tuesday) Topic: Experimental Methods

Heated Crevice - Design, Experimental Methods, and Data Interpretation by Jesse B. Lumsden and Keith Fruzzetti [Keynote Talk]

Question to Jesse Lumsden by Francois Vaillant

Are SCC experiments scheduled (as model boilers?) to verify the hypothesis that oxidizing period during layup and start-ups could be relevant to explain IGAICC in plants?

Response to Jesse Lumsden by Francois Vaillant

Yes, electrochemical noise will be monitored while the tube is pressurized. The noise will detect the initiation and propagation of SCC.

Experimental Simulation of Crevice Chemistry Evolution by C. B. Bahn, S. H. Oh, and I.S. Hwang

Question to Chi Bum Bahn by Peter King

Your graphs of axial temperature profiles show temperatures on the secondary side well below saturation temperature. Can you comment on these measurements and on the inlet temperature to the crevice vessel?

Response by Chi Bum Bahn

The secondary water was charged by a diaphragm pump and ejected through a back pressure regulator. Flow rate of the secondary system was maintained at 4 L/hr. The secondary pressure was adjusted automatically by a PID-controlled back pressure regulator at 5.50 ± 0.04 MPa with a saturation temperature of 270°C. The inlet temperature of the crevice vessel was not controlled. Therefore, it was affected by the primary water temperature. To maintain the constant secondary temperature, a preheater can be installed before the inlet line.

Question to Chi Bum Bahn by Zhongquan Zhou

Compared with internal reference electrode, what's the advantage to choose external reference electrode for monitoring ECP?

Response to Zhougquan Zhou by Chi Bum Bahn

As I know, internal-type Ag/AgCl (water) electrodes have worked very well at high temperature/high pressure aqueous environments. But the AgCl solubility at about 300°C is around 10^{-3} mol/kg and that at room temperature is around 10^{-5} mol/kg. The concentration gradient in the case of internal-type electrode is higher than that of the external-type electrode. For both internal and external-type Ag/AgCl (water), high purity water was used as the filling solution in which Cl^- ion activity can be established and maintained at the solubility of AgCl even with the sustained leakage for a long period. But, although I did not compare two electrodes in the same environment, the life of internal-type Ag/AgCl (water) will be shorter than that of external-type Ag/AgCl (water) because of higher diffusion rate.

Question to Chi Bum Bahn by Jiaxin Chen

I wonder the type and distance of the thermocouples to the tube (primary).

Response to Jiaxin Chen by Chi Bum Bahn

They are k-type and distance is 0.5 mm

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Experience of Heated Crevice Experiments at Studsvik by H-P Hermansson, A. Molander and P-O. Andersson

Question to Jesse Lumsden, Chi Bum Bahn, Anders Molander, and Hans-Peter Hermansson by Jeff Gorman

I. What do we really want to be able to measure in heated crevice tests to help predict corrosion? Are they:

1. pH?
2. potential relative to Ni/NiO stability point?
3. when become liquid filled?
4. All of the above as a function of bulk water chemistry?

I. What are the plans for test design improvements directed at obtaining answers to the above questions?

Response to Jeff Gorman by Chi Bum Bahn

I. As you know, the real crevice environments are very complicated. Therefore, various factors should be considered simultaneously. But I'd like to emphasize the surface film formed on the heated crevice tubes. The oxide and deposit characteristics and the interaction between oxide and surface deposit as a function of time and water chemistry can be studied.

II. I do not have any specific plan to the heated crevice testing. Briefly speaking, microstructural analysis for corroded tubes in a model boiler by using SEM, AES, TEM, etc. will be helpful to get answers to heated crevice environments.

General comment by Jiaxin Chen

We have discussed a lot about the impact of crevice chemistry on the precipitation inside crevice. We should also remember that corrosion process inside crevice region actually produces corrosion products inside the crevice. This produced corrosion products fill up the space inside the crevice and will continue to fill the dense crevice so that the density of crevice packing will increase with time. It increases the stress on the oxide layer constantly.

High Temperature Ph Probes for Crevice/ Crack Tip Solution Chemistry Applications- A Preliminary Study by R. Srinivasan and Y. Takeda and T. Shoji
[Not attending paper included in proceedings]

October 9 (Wednesday)   Topic: Results from Experimental Studies

Limits to Crevice Concentration Processes by Allen Baum [Keynote Talk]

Question to Allen Baum by Peter King

1. Please comment further on your distinction that sulfates adsorb but phosphates precipitate.

2. Can you comment on the degree of surface coverage of sulfate that might have occurred in your tests.

Response to Peter King by Allen Baum

1. We tested ammonium sulfate and sodium phosphate. This was infrared in the statement, but was not explicit. Had we tested ammonium phosphate and sodium sulfate, we would likely have reported different results.

3. No. Because of the large radial gradients in the concentrations, it would be difficult to assign the appropriate surface area.

Question to Allen Baum by Jeff Sarver

By what mechanism do you feel that sodium increases the transport of Pb into the crevice?

Response to Jeff Sarver by Allen Baum

Because of its low solubility, lead would either precipitate at the tube surface while the crevice is filling with corrosion products (after which it would be in a superheated steam environment or it would precipitate at the edge of the novice. Sodium increases the transport of lead to the crevice interior in three ways.

1. Soluble lead in the bulk water may concentrate together with sodium in the crevice, with the lead remaining soluble in the concentrated caustic solution where it would then migrate to the crevice interior.

2. Caustic may dissolve lead that has precipitated near the crevice periphery and carry it to the interior of the crevice.

3. Likewise, caustic may dissolve lead that had originally precipitated at the tube surface and carry it further into the interior.

Question to Allen Baum by Jiaxin Chen

> Comment on slide Nr. 31 where interpretation of "activation energy." I think one should not attribute the activation energy to any particular corrosion process. This is actually an "apparent activation energy" which shows the temperature dependence of the measured parameter as the data are obtained.

Response by Allen Baum

> I agree. I did not mean to suggest that any corrosion process would produce such a high activation energy, but rather some mechanism unrelated to a conventional corrosion process must be responsible for the very strong observed temperature variation. The Crevice chemistry Test results suggest that the progressive volatilization of anions with increasing temperature and superheat causes the environment to be more aggressive at the hottest crevice locations.

<u>My Conclusions after 25 Years of Model Boiler Testing by Jacques Daret (CEA) [presented by A. Baum]</u>

<u>Inferences Regarding PWR SG Crevices from Plant Operating Experience by Jeffrey A. Gorman [Keynote Talk]</u>

<u>Inferences Regarding PWR SG Crevices from Model Boiler Results by Jeffrey A. Gorman [Keynote Talk]</u>

Question to Jeffrey Gorman by Steve Chamber

> In Jeff's first presentation (plant operating experience), Jeff mentioned some Case 5 (Japanese) plants started up on PO4. Question: Did use of PO4 lead to any benefit or detriment in re IGA/SCC at TS or TSPs, either while on PO4 or after switch to AVT?

Response to Steve Chamber by Jeffrey Gorman

> There were no reports of IGA/SCC being experienced at Japanese units while on PO4, although wastage did occur at Mihama 1. With regard to post PO4 experience, the units that had prior operation on PO4 (Mihama 1 and 2 and Takahama 1) seem to have experienced IGA/SCC at a somewhat less severe rate than most of those that started up on AVT immediately after those units (Takahama 2, Genkai 1 and Ohi 1). However, the situation is made less clear by the fact that other non PO4 units (e.g., Ohi 2, Mihama 3, Ikata 1 and 2, Genkai 2) also experienced IGA/SCC at low rates or avoided it altogether. The Japanese concluded that the prior phosphate experience

was not a major factor (see the paper by Kishida, et al., "The Causes and Remedial Measures of Steam Generator Tube Intergranular Attack in Japanese PWR," Proceedings of the Third International Symposium on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, p465-471, TMS, 1988).

Laboratory Experiments on Steam Generator Crevice Chemistry by P. V. Balakrishnan and G. L. Strati

Question to Gina Strati by Francois Valliant

What is the % de sulfate which could be adsorbed by magnetite, and an alloy 600 or S steel?

Response to Francois Vaillant by Gina Strati

In the one test we reported on, 30% of the sulfate that passed through the magnetite-packed bed adsorbed on the bed. At saturation, the surface concentration amounted to 12 $\mu g/m^2$ (based on a surface area of 1.2 $\mu g/m^2$ and the assumption that the surface of all the particles was available).

From our experiments, we cannot say how much adsorption there will be on Alloy 600 a stainless steel. (There will be no adsorption beyond the point of saturation of the magnetite surface.)

The Hideout and Return in a Sludged Ringhals TSP Crevice by P-O. Andersson, A. Molander, J. Chen, and P. Gillen

Question to P-O. Andersson by Feron Damien

These results have to be compared to the results obtained by Brumel & Campan on Clairnette loop. They also studied the hideout of ^{24}Na on 2 plant crevices. With one of these two crevices, they found the same results as those presented; sodium was accumulating in the crevice under boiling . But with the other crevice, no accumulation was found. These results have been published as in EPRI report (and presented in some conferences).

Response by P-O. Andersson

The presented data is not from a systematic study. It is a study of a given condition, crevices from the retired R3 SGs, in order to verify if sodium will accumulate or not.

Comments for general discussion by Jiaxin Chen

About Pb in crevice:

Pb presence at the metal/oxide interface, if so, could it be the similar phenomenon as Pt marked in corrosion study in general, when the inward/outward diffusion processes are to verify? In other words, Pb presence from the beginning and with corrosion outward diffusion of metal atoms makes the Pb stay at the metal/oxide interface.

As Pb has higher concentration at the metal/oxide interface, there is indeed no reason why they could not diffuse, via grain boundaries, to the crack tips as observed.

Experimental Study of Concentrated Solutions Containing Sodium and Chloride Pollutants in SG Flow Restricted Areas by D. You, S. Lefevre, D. Feron and F. Vaillant

Question to Damien Feron by Jeff Gorman

Please explain in more detail what you mean by saying that hideout return is not the opposite of hideout. Do you mean that the timing of the return is different?

Response by Damien Feron

It means that, if you have some precipitates which have been formed during the hideout

- the timing of return will be different if these precipitates are soluble

- the quantities and nature of chemicals which will return, will be different if these precipitates are insoluble under hideout return conditions

These differences between hideout and hideout return data have been illustrated by the trisodium phosphate study presented at the last Studsvik seminar on crevice chemistry in 1998.

Some Effects of Steam Generator Deposits on Crevice Chemistry by Chuck Marks

Evaluation of the Effect of Startup Oxidants on a Crevice Filled with Deposits by Jesse B. Lumsden and Al McIlree

October 10 (Thursday) Topic: Modeling

Mechanisms for Concentrating Impurities at Line Contact Tube Support Crevices in PWR SG's by Peter Millett and Dennis Hussey [Keynote Talk]

Predicting Crevice Chemistry in PWRs based on Hideout Return Chemistry by Steve Sawochka [Keynote Talk]

Question to Steve Sawochka by Jim Davis

Is it possible that species observed in the vicinity of the crack tip could come from the alloy rather than the solution? Roger Staehle and I did work on SCC on high purity alloys and did not observe species in the vicinity of the crack tip. With commercial alloys with the same nominal composition species were observed in the vicinity of the crack tip.

Response by to Steve Sawochka

Certainly, some elements that are present in the base material could concentrate along the crack face during the corrosion process. However, this effect should not be of importance relative to species believed to control solution chemistry in the crevice, e.g., sodium, chloride, potassium, sulfate, etc.

Question to Steve Sawochka by Tina Gaudreau

Have observations of hideout return from plants that have added chloride indicated that those efforts have been successful in modifying the crevice chemistry?

Response by Steve Sawochka

Detailed studies performed at several plants have demonstrated that crevice chemistry can be controlled by chloride injection, i.e., molar ratio control.

Status of EPRI Software Tools for Evaluating Crevice Chemistry by Tina Gaudreau

Question to Tina Gaudreau by Peter King

Could you please expand on the similarities and differences of MULTEQ and CREVSIM. I understand that CREVSIM is built on MULTEQ but it is not clear which tool would apply to what circumstances. For example, if I run a laboratory test with flowing water in which hideout is occurring (and hideout rates may be different) and I want to estimate crevice chemistry, would MULTEQ or CREVSIM be the better tool, and what factors determine the choice?
I think that this issue is relevant to the consideration of integrating chemical and CFD models, for instance.

Response by Tina Gaudreau

In this example MULTEQ is a better choice. While CREVSIM will model the hideout, the user must enter an approximate 'hideout fraction' so the result is an estimate.
In MULTEQ, the amount of impurity left behind with each step in the concentration model is determined based on the physical properties of each species. So nonvolatile species will accumulate to a higher concentration factor than volatile species as the solution is boiled.

Question to Tina Gaudreau by Zhongquan Zhou

1. What database does MULTEQ use? The thermodynamic data in the database is dependent on the low temperature data. For the high temperature calculation, the data are extrapolated from the low temperature. Are the results reliable? How would one check it and improve it?

2. Compared with other commercial available software, what's the advantage of MULTEQ?

Response by Tina Gaudreau

1. The MULTEQ code used a database of thermodynamic information for each species. The database is controlled by a committee of experts who approve each change or addition to the database. In as many areas as possible, the database entries are based on high temperature testing. Continual updates allow for inclusion of new test results as they are available.

2. The database described above is the advantage of MULTEQ.

<u>Modeling and Analyses Supporting Argonne Model Boiler Design by Kenneth E. Kasza and et. al.</u>

<u>Multidimensional CFD and Thermal Modeling of the SG Tube and Support Plate Crevice Region by Stephen Bajorek and Donald Helton</u>

<u>Application of Chemical Equilibrium Model to the Evaluation of Magnetite-Packed Crevice Chemistry by C. B. Bahn, I. H. Rhee, and I. S. Hwang</u>

Comments to Chi Bum Bahn by Jiaxin Chen

General comment on the use of thermodynamics in crevice chemistry:
Crevice environment is an open system where not only mass transport in and out occurs but also heat transfer in and out proceeds constantly. In Bahn's treatment, you have even included some solid phases whose interaction with liquid environment is also a slow process (kinetically controlled). In such a system (crevice area) none of the thermodynamics pre-requests are satisfied, there is a great risk that any calculation and its implication may mislead us.

Response to Jiaxin Chen by Chi Bum Bahn

As you commented, the crevice environment can be far from thermodynamic equilibrium. To consider kinetically controlled process, the reaction rate constant for each reaction should be introduced. This is very difficult work.
I think, at high temperature, the thermodynamic equilibrium assumption is a first approach we can do.

Question to Chi Bum Bahn by Zhongquan Zhou

Have you performed any experimental tests to verify the calculation used in your model? Without experimental confirmation of the thermodynamic equilibrium phases, it is not very ensuring by just inputting data and getting outputs from software calculations.

Response to Zhongquan Zhou by Chi Bum Bahn

I did not conduct any experimental test to verify the calculation. At first, I tried to use MULTEQ®. But MULTEQ could not manage some soluble iron species such as FeO_2^-. This is the reason why I did not use MULTEQ®. As you commented, the verification of model is very important especially for the thermodynamic database. HSC Chemistry® uses the revised HKF model to calculate the Gibbs energy of soluble species. The revised HKF model originated from geochemical field is published at available journals.

<u>The Conditions Known to Produce Crevice Corrosion by the IR Mechanism and Those Yet to Be Investigated by Howard W. Pickering</u>
<u>[Not attending paper included in proceedings]</u>

October 11 (Friday) Panel Discussion

Post-Seminar Comments and Thoughts from Jeffrey Gorman

Thanks for letting me participate in the heated crevice meeting. It was interesting and thought provoking. I want to briefly describe with my main thoughts based on the meeting regarding prediction of IGA/SCC at heated crevices:

The steps involved in the likely occurrence of IGA/SCC in new SGs with 600TT or 690TT tubes seem to be pretty well understood, as discussed at the meeting, and include:

(1) Gradual fouling of the initial as-built crevices at TSPs and TTS areas, with the superheat at these areas increasing steadily as deposits build up and densify.

(2) As the superheat at crevices increases, the concentration of dissolved species in them increases, and the aggressiveness of the under deposit environment increases. In addition, as the thickness and density of deposits increases, the environment at the tube surface may become superheated steam, at least for early parts of the fuel cycle.

(3) Because the tubes mostly have low stresses and since the superheats in the newer design SGs are relatively low (with resulting low concentration impurities), corrosion is likely to often involve a long slow IGA phase.

(4) When the IGA gets deep enough such that either the stress intensity reaches a threshold value, or the crack tip chemistry conditions reach some critical state, IGSCC is likely to start, with significantly increased growth rates after the IGSCC starts.

(5) Possible aggravation of IGA/SCC, with resulting bursts of corrosion, may be caused by episodic exposure to startup oxidants. Another similar possibility was hinted at by Chambers related to attack by chemicals released at shutdown, but I don't know enough about this possibility to discuss it here.

My comments regarding the knowns and unknowns involved in the above steps, and ways to best address the unknowns, are as follows:

(1) I think that, for the immediate future, the best way to deal with the deposit buildup step it to do what Fruzzetti's presentation indicated that Millett is contracted to do for EPRI, i.e., develop an empirical model based on available plant and test data and experience. In this regard, the 30 year history at Siemens SGs should not be ignored, since they have geometries and materials similar to those of our new SGs and also have had, at least for plants that started up in 1982 and later, low iron ingress rates similar to those now seen in the USA. In the longer term, it would be desirable to quantify models that show how plant chemistry parameters affect deposition rates (parameters such as iron ingress rates, feed water pH, pH control agent used, presence or absence of boric acid). Some of this information may already be available, such as from AECL testing and from Japanese investigations regarding

effects of hydrazine on deposit densification. Some additional testing may be needed, but I would first see what the current EPRI/Millett project develops.

(2) A lot of work has been done on identification of the various types of environments that might develop in heated crevices and attack 600TT and 690TT, as discussed in Roger's NRC report. It seems to me that the main tasks that still need to be worked on include:

a) For all of the chemistry environments, a systemic gathering and quantification of the rates of IGA and then IGSCC for both of the alloys as a function of superheat (i.e., concentration), stress, temperature and potential. This needs to consider the range of compositions and thermal histories of the alloys, which apparently is quite a large range for 600TT. I believe that Roger's work is addressing this aspect, and that he will be identifying the areas that need additional testing.

b) For some (possibly all) of the potentially important environments it is not known whether IGA/SCC can occur in superheated steam at 325C or not. The answer is important since, if the attack can occur in superheated 325C steam, it takes very little impurity to result in an aggressive environment under the deposit. If this is the case, then the main defense has to be by control of the mix of the impurities, and not on the amount of impurities. In other words, ALARA chemistry is not good enough, and plant chemistry control also should include molar ratio and/or other impurity composition controls. Model boiler tests might be a useful way to address this question, but I think that autoclave tests using 325C doped superheated steam (with controlled and varied levels of hydrogen) should precede them.

c) Related to the above item is the need to determine if some of the identified aggressive environments actually develop at tube surfaces in heated crevices. This is especially true for the sulfate, complex, and alumino-silicate environments. Model boiler tests seem to be a useful way to address this question.

d) The aggravating effect of startup oxidants, including hematite washed in from the secondary system (particularly during early power operation following startup), is not well characterized. Tests in caustics have been shown to be very sensitive to the oxidizing nature of the sludge, with both copper oxide and hematite (unpublished MHI test) leading to very large increases in crack initiation and growth rates. The effects of oxidizing corrosion products on many of the other aggressive environments are not as well characterized and warrant detailed study. This is another area where model boiler tests may help, although most of the tests probably should be autoclave tests.

e) As was discussed at the meeting, lead is very aggressive but seems to mostly be tied up by other species. Understanding this in depth, so that utilities can avoid conditions that release the lead that is present in their crevices, is important.

(3) Verification of any predictive model is important to make sure that it provides reasonable and reliable results. The industry events and the model boiler results I summarized at the meeting may provide a start at identifying cases that can be used to verify and adjust predictive models. However, these cases would first need to be fleshed out with detailed supporting information such as chemistry histories, pulled tube examination results, etc. In addition, and probably more importantly, these cases should be reviewed by an expert panel to make sure that both the data and conclusions are reasonable and correct. I will provide Keith with a suggested list of cases that could be pursued for this purpose.

NRC FORM 335
(2–89)
NRCM 1102,
3201, 3202

U. S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

(See instructions on the reverse)

1. REPORT NUMBER
(Assigned by NRC. Add Vol., Supp., Rev., and Addendum Numbers, if any.)
NUREG/CP-XXXX
EPRI 1009355
ANL – 03/24

2. TITLE AND SUBTITLE
Proceedings of the USNRC/EPRI/ANL Heated Crevice Seminar

3. DATE REPORT PUBLISHED

MONTH	YEAR

4. FIN OR GRANT NUMBER
W6487

5. AUTHOR(S)
Jangyul Park, Keith Fruzzetti, Joseph Muscara, and Dwight R. Diercks

6. TYPE OF REPORT
Conference Proceedings

7. PERIOD COVERED *(Inclusive Dates)*
October 7-11, 2002

8. PERFORMING ORGANIZATION – NAME AND ADDRESS *(If NRC, provide Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address; if contractor, provide name and mailing address.)*
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

9. SPONSORING ORGANIZATION – NAME AND ADDRESS *(If NRC, type "Same as above": if contractor, provide NRC Division, Office or Region, U.S. Nuclear Regulatory Commission, and mailing address.)*
Division of Engineering Technology
Office of Nuclear Regulatory Research
U. S. Nuclear Regulatory Commission
Washington, DC 20555-0001

10. SUPPLEMENTARY NOTES
J. Davis, NRC Project Manager

11. ABSTRACT (200 words or less)

An international Heated Crevice Seminar, sponsored by the Division of Engineering Technology, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, Argonne National Laboratory, and the Electric Power Research Institute, was held at Argonne National Laboratory on October 7-11, 2002. The objective of the seminar was to provide a working forum for the exchange of information by contributing experts on current issues related to corrosion in heated crevices, particularly as it relates to the integrity of PWR steam generator tubes. Forty-five persons from six countries attended the seminar, including representatives from government agencies, private industry and consultants, government research laboratories, nuclear vendors, and electrical utilities.

The seminar opened with keynote talks on secondary-side crevice environments associated with IGA and IGSCC of mill-annealed Alloy 600 steam generator tubes and the submodes of corrosion in heat transfer crevices. This was followed by technical sessions on (1) Corrosion in Crevice Geometries, (2) Experimental Methods, (3) Results from Experimental Studies, and (4) Modeling. The seminar concluded with a panel discussion on the present understanding of corrosive processes in heated crevices and future research needs.

12. KEY WORDS/DESCRIPTORS *(List words or phrases that will assist researchers in locating this report.)*
Steam Generator
Tubes
PWR
Crevice
Corrosion
Stress Corrosion Cracking
Alloy 600, Inconel 600
Alloy 690

13. AVAILABILITY STATEMENT
Unlimited

14. SECURITY CLASSIFICATION
(This Page)
Unclassified
(This Report)
Unclassified

15. NUMBER OF PAGES
646

16. PRICE