

Quantitative Assessment of Submodes of Stress Corrosion Cracking on the Secondary Side of Steam Generator Tubing in Pressurized Water Reactors: Part 1

R.W. Staehle^{†,*} and J.A. Gorman^{**}

ABSTRACT

The work in this article is part of a project to develop a quantitative description of stress corrosion cracking (SCC) on the secondary side of pressurized water reactor (PWR) steam generator tubing based on existing information from operating plants and from laboratory experiments. This work is the second step in developing a predictive model for SCC on the secondary side. The first step involved developing a statistical framework into which dependencies of the various submodes of SCC can be inserted. The results of the present work will lead to quantitative descriptions of corrosion processes that, in turn, will be incorporated into the statistical framework. The chemistry of heat-transfer crevices will then be assessed to determine the proper inputs to the dependencies of the various submodes of SCC, and these will be connected to bulk environments. The modeling here is directed toward predicting the early occurrence of SCC that is too shallow to be detected by nondestructive examination (NDE).

The various submodes of SCC considered in this article include: alkaline stress corrosion cracking (AkSCC), low-potential stress corrosion cracking (LPSCC), acidic stress corrosion cracking (AcSCC), high-potential stress corrosion cracking (HPSCC), lead stress corrosion cracking (PbSCC), low-valence stress corrosion cracking (S^{+} SCC), organic stress

corrosion cracking (OgSCC), doped steam stress corrosion cracking (DSSCC), and low-temperature stress corrosion cracking (LTSCC). To develop a model for each of these submodes, their dependencies on the seven primary variables are evaluated in this article; these variables are: pH, potential, species, alloy composition, alloy structure, temperature, and stress. For some of these submodes, there is broad agreement in the literature on dependencies. In other cases there is some disagreement, but these are mostly related to lack of information or to inconsistencies among experiments that have been conducted under different conditions. To develop a framework for this discussion of submodes, supporting sections are included on designs of steam generators as they affect corrosion, bulk water chemistry as it relates to chemistry in heat-transfer crevices, and the chemistry of heat-transfer crevices. At the end of each section, the state of present knowledge is summarized including the lessons learned from service and laboratory experience. In addition, possible problems for future reliable performance as affected by corrosion are identified.

KEY WORDS: Alloy 600, Alloy 690, Alloy 800, heat-transfer crevices, secondary side, steam generator, stress corrosion cracking, water chemistry

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[†] Corresponding author.

* 22 Red Fox Road, North Oaks, MN 55127.

** Dominion Engineering, Inc., 11730 Plaza America Drive, Suite 310, Reston, VA 20190.

^{Note} References, figures, and tables are sometimes referred to out of order. This has been necessary first to maintain the integrity of sections and second to show connections among previous and succeeding sections.

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1.0 INTRODUCTION

1.1 Background

This discussion is concerned with steam generators (SGs) used in pressurized water nuclear reactors and mainly with the corrosion performance on the secondary side of heat-transfer tubing. The purposes of this discussion are to identify the following:

- the dependencies of various submodes⁽¹⁾ of stress corrosion cracking (SCC) on the primary variabilities of potential, pH, species, alloy composition, alloy structure, temperature, and stress
- important features of heat-transfer crevices that affect the occurrence of SCC⁽²⁾
- important features of design that affect SCC in heat-transfer crevices

The assessments developed in this discussion are used to quantify the parameters in statistical distributions to provide a basis for predicting the probability of SCC and especially the earliest shallow occurrences of SCC.

This discussion is appropriate now since many electric utilities plan to extend the lives of their steam generators beyond the original design lives of about 40 years. Also, this discussion reviews the chronology of corrosion in steam generators together with its lessons and applies these to extending the life of steam generators and assuring their future reliability.

The discussion in Section 1.0 shows how specific design features of steam generators are relevant to corrosion on the secondary side. Failures that have occurred in tubing are described. Types of steam generators are described in Section 2.0, together with the ways that specific features of these steam generators affect corrosion. The evolution of water chemistry is described in Section 3.0. Heated crevices and chemical deposits are described in Section 4.0; these are significant because most of the corrosion that occurs in steam generators is associated with heated crevices and deposits. The dependencies of modes and submodes of corrosion of tubing materials on primary variables are described in Section 5.0. These are limited to those that have occurred in tubing of steam generators and are limited generally to ones that have occurred on the secondary side.

The purpose of this introduction in Section 1.0 is to describe the extent of corrosion that has been observed on the secondary side of steam generators. This discussion could include, as well, the primary side; however, there is only one submode of SCC, low-potential stress corrosion cracking (LPSCC), that occurs on the primary side. Since this submode, in principle, can occur on the secondary side as well, LPSCC is included in this discussion.

The occurrences and study of corrosion in steam generators have produced important advances in the field of corrosion. One is the recognition that SCC can occur in pure water, as well as hydrogenated water, as LPSCC. Before the observation by Coriou, et al., in 1959¹ of what was to become LPSCC, such an occurrence would have been unthinkable since no "specific causative ions" are required for LPSCC to occur. A second advance is the current emphasis on the importance of heat-transfer crevices, which concentrate impurities from nominally pure water to produce high local concentrations of aggressive chemicals; however, concentrations of chemicals related to superheated surfaces have been a well-known phenomenon since the early failures of pressure boilers. A third advance is the recognition of the broad array of submodes of SCC to which a nominally corrosion-resistant alloy, 600MA (UNS N06600),⁽³⁾ is prone.

⁽¹⁾ For clarification, the term "submode" used here refers to the occurrence of SCC, which depends uniquely upon a set of primary variables. For example, while both AkSCC and LPSCC exhibit intergranular SCC, which is sometimes indistinguishable morphologically, these two submodes depend quite differently on pH, potential, species, alloy chemistry, alloy structure, temperature, and stress. Thus, these two domains are different submodes. The array of submodes that apply to Alloy 600MA are described in Figure 82.

⁽²⁾ Throughout this article various abbreviations are used. These are explained on page 993.

⁽³⁾ UNS numbers are listed in *Metals and Alloys in the Unified Numbering System*, published by the Society of Automotive Engineers (SAE International) and cosponsored by ASTM International.

This discussion focuses mainly on the heat-transfer crevices associated with tube supports, sludge, and tops of the tubesheets. Failures in steam generators have occurred mostly at these locations and at superheated surfaces that function much like heat-transfer crevices. Also, at these locations the effectiveness of nondestructive examination (NDE) (e.g., eddy-current tests) may be reduced owing to the complexity of geometries and presence of deposits associated with these locations. General corrosion (GC), SCC, intergranular corrosion (IGC), and pitting have occurred on free-spans; however, the extent of such corrosion is much less than that found in the heat-transfer crevices.

Many of the failures that are described here do not occur in significant numbers in modern pressurized water reactor (PWR) plants. Substantial mitigations have been achieved by improving tubing alloys, water chemistry, and designs. The extent to which these improvements will prevent future failures is considered in each of the major sections here. In fact, the possibilities of future failures in modern steam generators are an important issue in this discussion. Despite the improvements in materials, design, and operation in modern steam generators, there is ample precedent for aggressive corrosion to arise after some delay, as with the delay of the onset of LPSCC in Alloy 600 in operating SGs.

There may be tacit implications in this discussion that SGs that have been produced by all manufacturers have sustained the same corrosion damage. This is not so. For example, most of the SGs fabricated by Siemens have sustained little damage. SGs fabricated by Babcock & Wilcox (B&W) have sustained only modest damage. CANDU plants are not considered in this review.

This discussion does not emphasize corrosion on the primary side, which is mainly associated with LPSCC; however, this submode is of interest to the secondary side owing to the possible lowering of electrochemical potentials by additions of hydrazine (N_2H_4) and because low potentials can be developed in crevices. LPSCC is discussed in Section 5.2.2 as one of the submodes that should be considered on the secondary side. Also, the present discussion does not deal with SCC that has occurred at girth welds in SG vessels.² A brief discussion of corrosion of Alloy 600 in other applications on the primary and secondary sides is included in connection with Table 17 in Section 5.

Typical steam generators of the recirculating steam generator (RSG) and once-through steam generator (OTSG) types are shown in Figure 1, together with typical features that relate to subjects in this discussion. In general, all the manufacturers of PWRs supply, or have supplied, steam generators including Westinghouse and their licensees, Siemens, B&W, and Combustion Engineering (CE) (now part of

Westinghouse). Each of these designs is similar in some ways and different in others. The CANDU steam generators used in pressurized heavy water reactors (PHWRs) are not discussed here since their designs and operating conditions are sufficiently different from the others as to require a separate discussion; however, much of the discussion here is applicable, in principle, to CANDU designs. The Siemens design is generally like the Westinghouse design except that Siemens used different tube supports and alloys for tubing almost from the beginning. These differences are described in Section 2.0. These differences together with Siemens' early and strict control of water chemistry have produced fewer failures than were sustained by SGs of the Westinghouse and CE types. Much of the discussion here concerns failure problems, and these are mostly related to the Westinghouse and CE designs.

It is important to recognize that the present designs of SGs, while improved over the past designs, are still vulnerable to corrosion damage. For example, as will be discussed in Section 5.2, Alloy 690 (UNS N06690) can sustain extensive corrosion under possibly relevant conditions, as shown in laboratory tests. In fact, with the higher chromium concentration, Alloy 690 is inherently a more reactive alloy than Alloy 600. It should be recalled, parenthetically, that virtually every mode and submode of SCC observed in corrosion studies in the laboratory for Alloy 600 (e.g., LPSCC) eventually occurred in operating plants, although some of the corrosion investigations were not undertaken until after failures were observed (e.g., denting). Also, deposits similar to those in drilled hole designs accumulate in modern line contact support designs and provide sites for concentrating chemicals, although at slower rates.

While no SCC has been observed in Alloy 690 in the field, it cannot be assumed that it will not occur. It seems that users of Alloy 690 have become lax in their support of serious inquiries, the incentives for which already are well known from existing data. In the past, serious experimental work on obvious corrosion problems was sometimes not undertaken until after previously predictable failures occurred. With this experience, it would be prudent to identify potentially significant modes of damage early and to initiate serious investigations aimed at quantifying the possibilities for their occurrence, subsequent damage, and ways to prevent or ameliorate such damage.

The reliability of steam generators, from the vantage of the secondary side, is influenced mainly by the characteristics of "heat-transfer crevices" in which dilute species from secondary environments can be concentrated to produce often corrosive environments operating near the hot leg temperature. A "heat-transfer crevice" differs from a "crevice," as it is considered in corrosion technology, by the presence of heat transfer that leads to concentrating chemicals

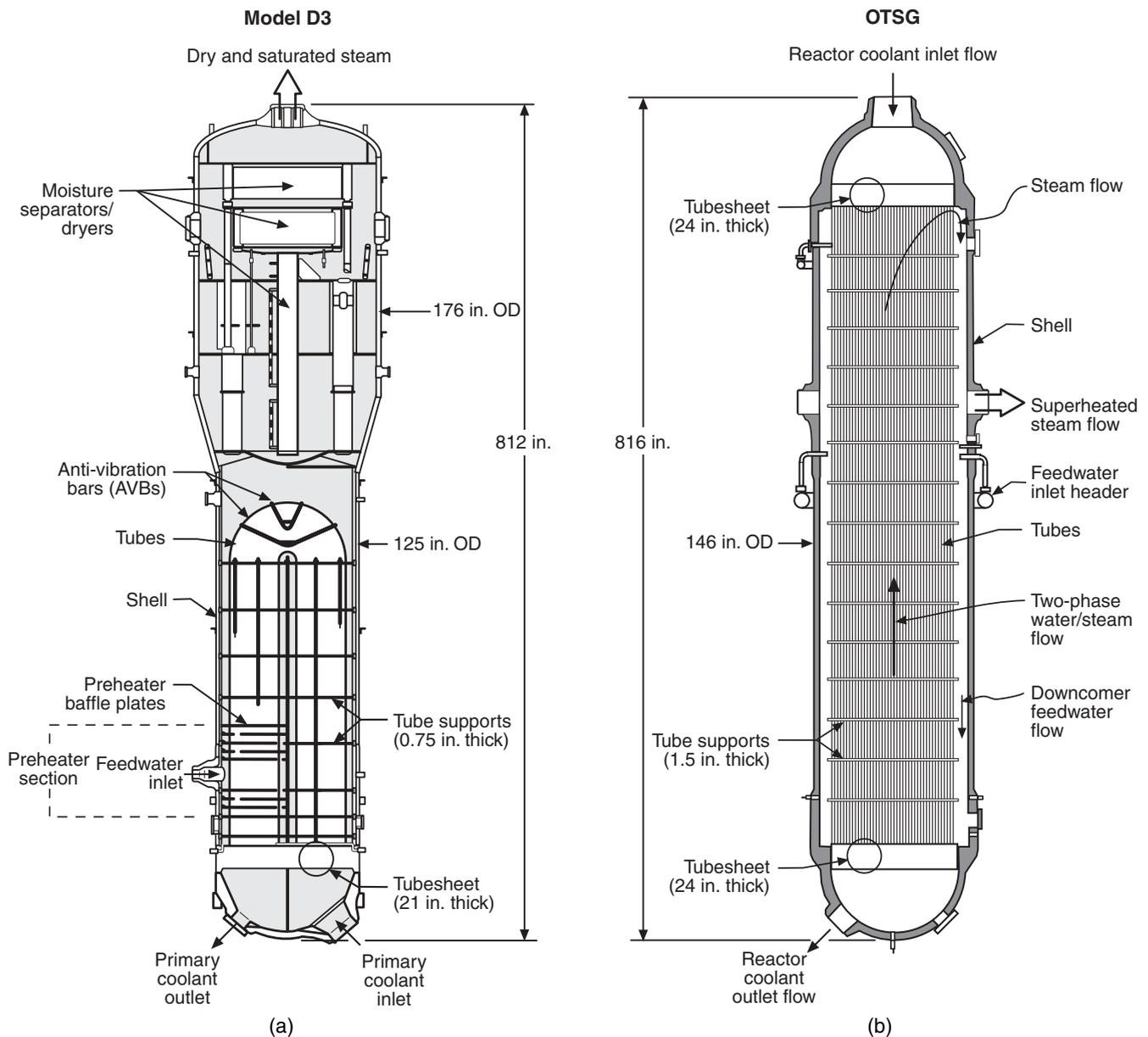


FIGURE 1. Schematic view of (a) RSG (Westinghouse Model D3) and (b) OTSG showing typical features. The RSG design is typical of that of Westinghouse, its licensees, CE, and Siemens. The OTSG design is typical of the B&W SGs.

from adjacent and accessible bulk solutions. Heat-transfer crevices will be discussed in detail in Section 4.0.

Corrosion on the secondary side, as well as the primary side, of steam generators has been considered at many technical meetings. For example, the series of meetings concerned with the subject of environmental degradation in water-cooled nuclear plants began in 1983, and the 10th in this series was the instance where this discussion was given initially.³⁻¹² A similar series has been sponsored in France, held at the Fontevraud Abbey.¹³⁻¹⁶ The materials and corrosion in steam generators have been

described in reviews by Latanision and Staehle,¹⁷ Staehle,¹⁸⁻¹⁹ Theus and Staehle,²⁰ Staehle, et al.,²¹ Gras,²² Cassagne and Gelpi,²³ Smialowska and Rebak,²⁴ Sridhar and Cragnolino,²⁵ and Friend.²⁶ An intensive technical conference was sponsored jointly by EPRI and ANL/NRC in 1995 concerning corrosion problems on the secondary side.²⁷

Corrosion on the primary and secondary sides has been the subject of many EPRI-sponsored workshops²⁸⁻⁴⁷ as well as many topical reports published by EPRI. EPRI published two revisions of a handbook on steam generators⁴⁸⁻⁴⁹ and has sponsored the development and application of software, MULTEQ[†], for quantifying problems in aqueous thermodynamics important to the secondary side.⁵⁰⁻⁵²

[†] Trade name.

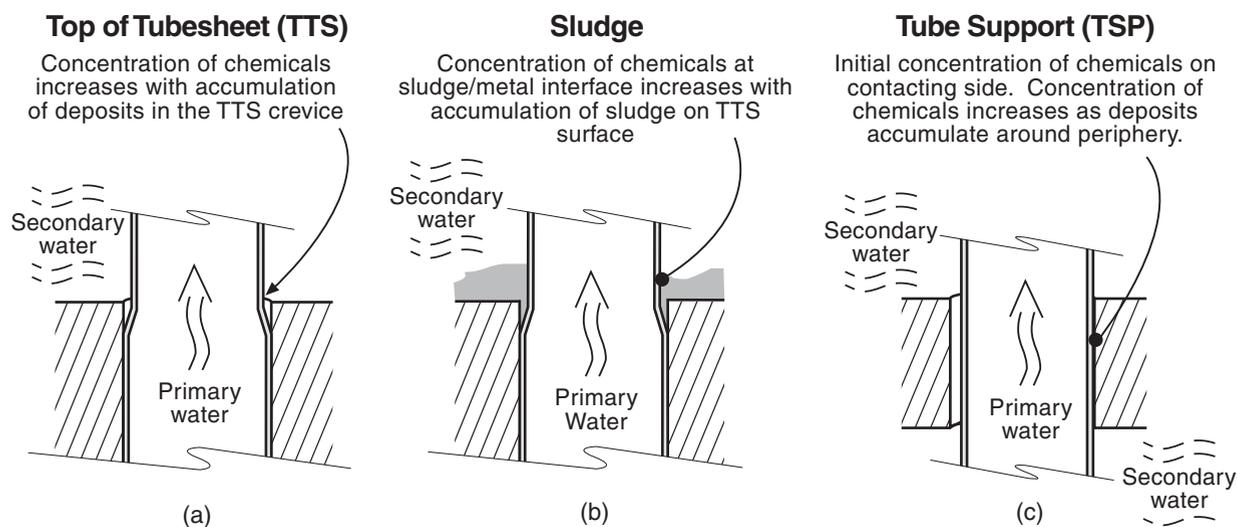


FIGURE 2. Geometries that produce heat-transfer crevices involving tubing in steam generators: (a) top of the tubesheet, (b) sludge, and (c) tube support.

This extensive array of publications, and there are others, attests to the extent of corrosion that has occurred in steam generators.

1.2 Design Features Affecting Corrosion

As background for the discussion of heat-transfer crevices, Figure 2 shows the three types of heat-transfer crevices in steam generators—tube support, tubesheet, and sludge—common to all vertical PWR steam generators together with the important features that affect the performance of such crevices. The locations of the parts with these heat-transfer crevices are shown in Figure 1. For the tube support crevice, there are three types: drilled hole, broached hole, and egg crate, as illustrated in Figure 3. Three designs of tubesheet crevices are shown in Figure 4. One of the early designs was a part-depth roll. This was replaced by a full-depth roll or explosive or hydraulic expansion to avoid possible corrosion in a long heat-transfer crevice. Full-depth expansion is now standard; however, here, there is always a 3-mm to 6-mm crevice near the top of the tubesheet (TTS) to avoid “overexpansion” that would damage the tube. Figure 5 shows the evolution of the types of tube support designs of the four manufacturers of steam generators.

1.3 Materials

Materials of interest to this discussion include those used for tubes and tube supports. Figure 6⁵³ shows a Fe-Cr-Ni ternary diagram with these alloys at their respective compositions together with other Fe-Cr-Ni alloys used in nuclear power. Alloy 600MA was used initially for tubing in the designs of Westinghouse and its licensees, B&W, and CE; later, manufacturers changed to Alloy 600TT and then, generally, to Alloy 690TT. This evolution is described

in Sections 2.5 and 5.1. Alloy 800NG (UNS N08800) has been used by Siemens for the tubing in their steam generators, except for an early plant that used Alloy 600MA. Initially, carbon steel was used for the tube supports by all designers except Siemens; later this was changed to various grades of stainless steel, as shown in Figure 5 and described in Section 2.5. Alloys 600 and 690 are nominally homogeneous and face-centered cubic. However, much of the carbon in these alloys is insoluble and precipitates both on the grain boundaries and in the matrix. The conditions for solubility of carbon in Alloys 600 and 690 are shown in Figure 99. Table 1 shows the compositions of the various alloys discussed here and are shown in Figure 6.

The corrosion behavior of the alloys used for steam generator tubing is dominated by the ranges of their passivity, film-free dissolution, and nobility. Potential-pH diagrams at 300°C for Fe, Cr, and Ni are shown in Figure 7. In addition, diagrams are shown for Cu, Pb, and Ti; these diagrams are included since Cu and Pb are important contaminants on the secondary side of steam generators. Cu often has been used in various heat exchangers in secondary systems and, when it dissolved and reacted with oxygen, can become an unwanted oxidizer. The diagram for titanium is included because it is widely used for condensers and because titanium compounds are sometimes considered for inhibitors in steam generators. Pb is discussed in Section 5.2.5 in connection with the lead stress corrosion cracking (PbSCC) submode. The diagrams shown in Figure 7 should be regarded as approximate since there are, sometimes, disagreements concerning the proper data to use for their construction. Regardless, these diagrams as shown provide good bases for comparisons and estimates of trends.

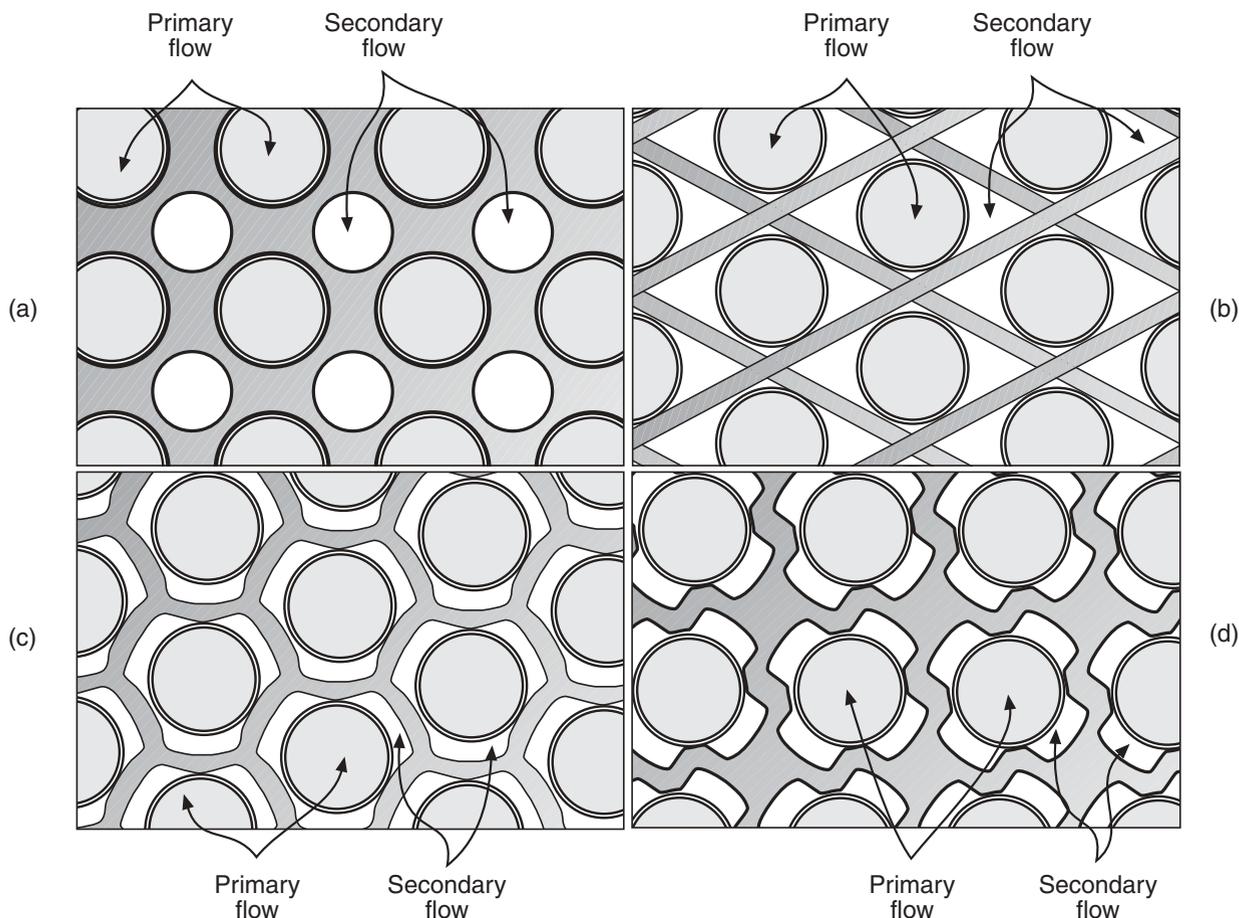


FIGURE 3. Geometries of tube supports: (a) drilled hole typical of early Westinghouse designs; (b) egg crate typical of Siemens and CE; (c) broached trefoil typical of B&W; (d) broached quatrefoil typical of later Westinghouse designs.

With respect to the diagrams of potential vs pH, Figure 7(a) shows that the standard hydrogen equilibrium is at about the same location as the NiO/Ni equilibrium. The low concentration of hydrogen in the secondary environment raises the potential of the H_2O/H_2 half-cell equilibrium to about 250 mV above the standard equilibrium and therefore would accelerate various anodic corrosion processes, as shown in Figures 33, 97, and 123. However, adding N_2H_4 lowers the potential on the secondary side, as shown in Figure 45, because of the low-equilibrium potential for the N_2/N_2H_4 equilibrium as discussed in Section 3.1.2 and shown in Figure 33.

The diagram for iron shows conditions for forming Fe_2O_3 . The presence of Fe_2O_3 inhibits flow-accelerated corrosion (FAC), owing to the low solubility of this higher valence oxide, Fe_2O_3 . Figure 7(c), together with Figure 33, also shows that iron in the presence of hydrazine will remain mainly as Fe_3O_4 , which tends to be nonprotective in high-velocity locations due to its relatively high solubility. Not obvious from this diagram is the metastable extensions of the Fe_3O_4/Fe and Fe_2O_3/Fe_3O_4 lines, which extend passivity into the nominally corrosion domain where Fe^{2+}

is stable. A similar extension applies to the Cr_2O_3/Cr line, which accounts for the stability of chromium in acidic solutions. Important in the chromium diagram is the relatively large region of instabilities of Cr_2O_3 in the alkaline region as CrO_2^- and with increased potentials as soluble Cr^{6+} oxyanions. This is illustrated also in Figure 73.

The Cu diagram shows that Cu cannot be oxidized in fully deaerated water. Further, the diagram shows that Cu can be oxidized by oxygen and that oxidized Cu can oxidize Ni. The Pb diagram shows that Pb is extensively soluble in high-temperature water. The titanium diagram shows that TiO_2 is stable over a broad range of pH and potential, which accounts for its possible application as an inhibitor as discussed in Section 3.1.2.

Important implications of the diagrams in Figure 7 are the following:

1. In a deaerated aqueous system, such as either primary or secondary systems, the upper boundary of potential is determined by the H_2O/H_2 equilibrium as affected by the pressure of hydrogen according to the Nernst equation in Figure 42.

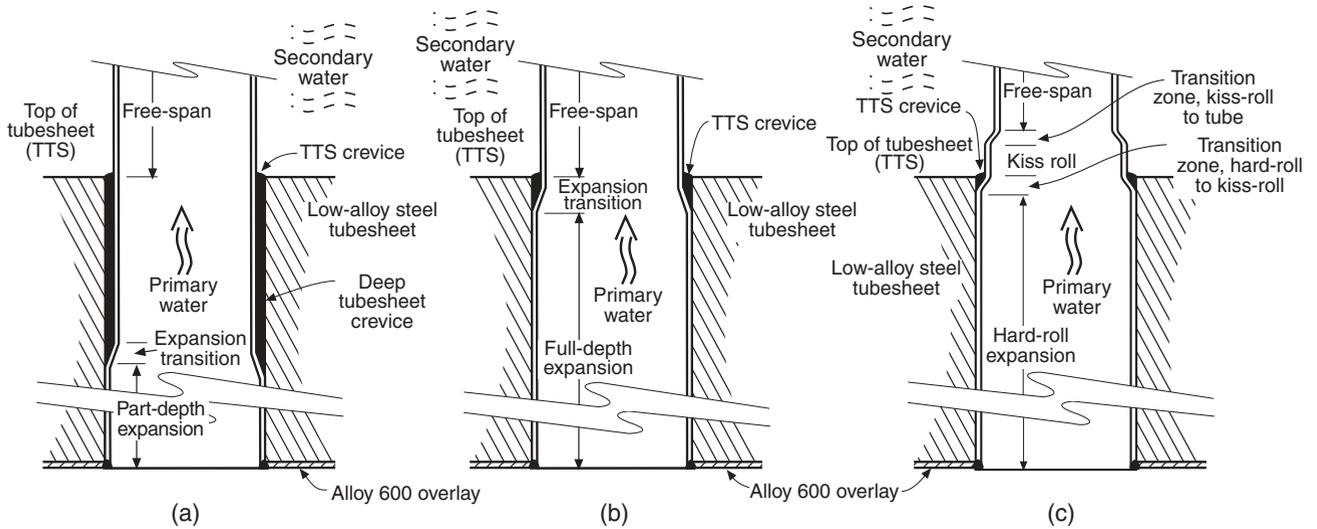


FIGURE 4. Tubesheet geometries: (a) partially expanded, (b) fully expanded, and (c) fully expanded with top "kiss" roll.

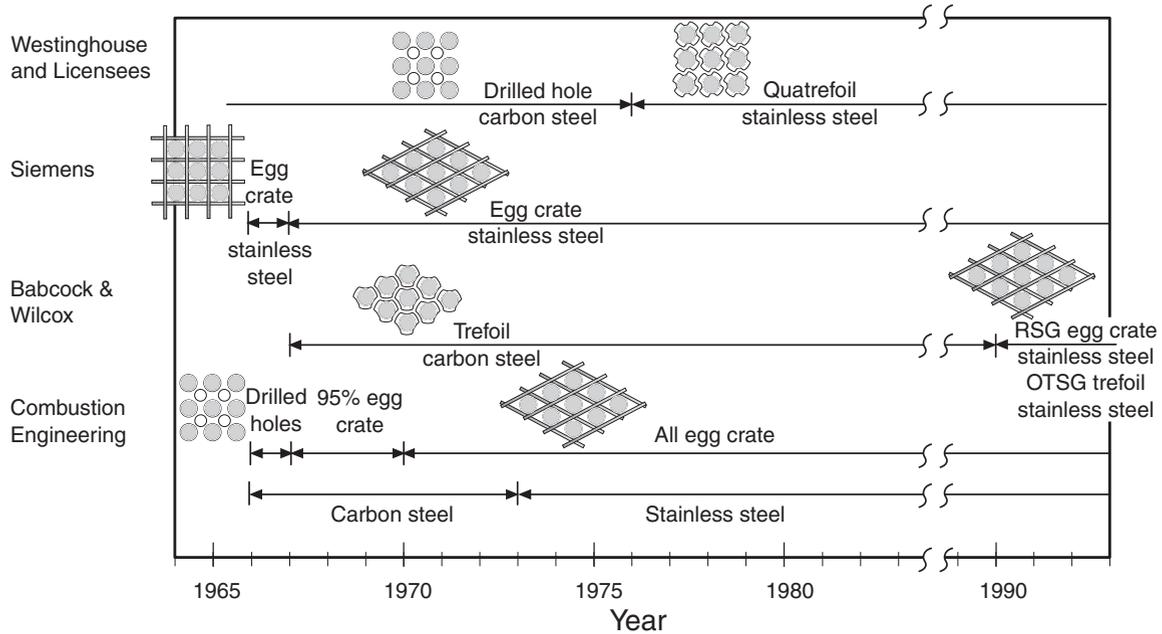


FIGURE 5. Evolution of tube support geometries and materials used by four nuclear steam supply system (NSSS) vendors for steam generators. The geometries of tube supports are shown above the lines, and the materials of the tube supports are shown below. The arrows indicate approximately when manufacturing of the respective designs of tube supports started.

2. The minimum potential for the nickel half-cell equilibrium in an aqueous environment is defined by the NiO/Ni equilibrium, although the presence of chromium and iron may lower this as discussed in connection with Figure 33.
3. The open-circuit potential on the surface of Alloys 600 and 690 lies above the NiO/Ni where the H₂O/H₂ half-cell equilibrium is at low H₂ activities in deaerated environments. A similar constraint applies to the tip of advanc-

ing SCC. If oxygen is present, the open-circuit potential increases according to the effect of oxygen on the potential, as shown in Figures 33 and 41; boundaries of the open-circuit potentials on the outside surfaces become the O₂/H₂O and NiO/Ni equilibria.

4. Below the H₂O/H₂ equilibrium, the amount of hydrogen forming on the surface progressively increases as the potential decreases.
5. The use of hydrazine tends to lower the open-circuit potential owing to its relatively low

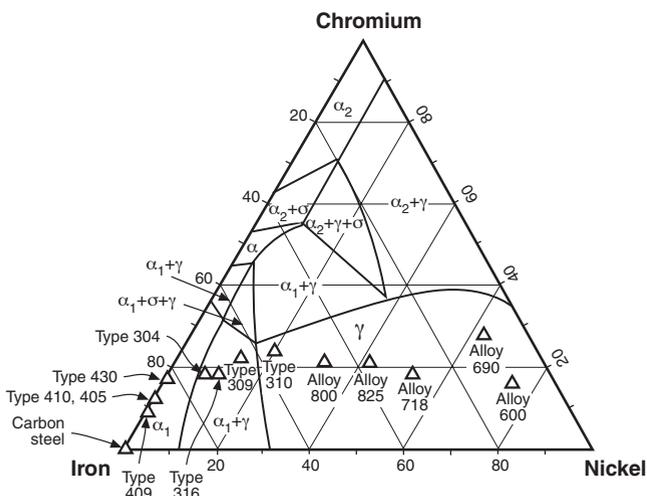


FIGURE 6. Alloys of interest to steam generators superimposed on an Fe-Cr-Ni ternary diagram for 400°C. Fe-Cr-Ni diagram from Pugh and Nisbet.⁵³

half-cell equilibrium potential as discussed in Section 3.1.2 and described in Figures 33 and 41. The use of hydrazine also increases the hydrogen activity on metal surfaces owing to the lower potentials. While the equilibrium calculations show a negative half-cell, direct measurements of the effects of hydrazine show a relatively small decrease, most likely because the exchange current density for the oxidation/reduction of N₂H₄ is low.

6. The minimum solubility of NiO is approximately at a pH in the middle of the NiO region, as shown also in Figure 38. Adjusting the pH to meet this minimum solubility minimizes the general corrosion of nickel-based alloys.

1.4 Heat-Transfer Crevice and Surfaces

Except for LPSCC at highly stressed locations in tubing on the primary side, the concentration of chemicals on the secondary side in heat-transfer crevices produces most of the corrosion in SGs. This concentration of chemicals occurs mainly at heat-transfer crevices but also can occur on all surfaces through which heat is transferred. For example, significant concentrations can be produced on surfaces from boiling or evaporation (e.g., in the upper bundle of OTSGs). Typical regions are shown in Figure 8 together, for comparison, with important features of a nonheat-transfer crevice. The accumulation of chemicals on the heated surface, as illustrated in Figure 8(b), is the same as that found inside the heated crevice in Figure 8(a). In both cases, the accumulated chemistry on the surfaces is not readily diluted. The acceleration of corrosion in the nonheat-transfer crevice in Figure 8(c) results mainly from the relatively high oxygen concentration on the surfaces outside the crevice. Such crevices have been studied extensively; in particular, Pickering and Al-Khamas⁵⁸ have reviewed quantitative features of nonheat-transfer crevices.

Heat-transfer crevices are important for the fact that they concentrate impurities to high ratios of

TABLE 1
Composition of Alloys Used in Tubing and Tube Supports (Maximum w/o, Except Where Noted)

Element	Type 304 ^(A)	Type 316 ^(A)	Alloy 600 EPRI Guidelines ^(B)	Alloy 690 EPRI Guidelines ^(C)	Alloy 800 Nuclear Grade ^(D)	Carbon Steel ASTM A285 Gr C ^(A)	Type 405 ASME SA479 ^(A)	Type 409 ^(A)	Type 410 ASME SA479 ^(A)
C	0.08	0.08	0.025 to 0.05	0.015 to 0.025	0.03	0.28	0.15	0.08	0.15
Mn	2.00	2.00	1.00 max.	0.50	0.4 to 1.0	0.90	1.00	1.00	1.00
P	0.045	0.045	0.015	0.015	0.020	0.035	0.040	0.045	0.040
S	0.03	0.03	0.010 max.	0.003	0.015	0.035	0.030	0.045	0.030
Si	1.00	1.00	0.50 max.	0.50	0.3 to 0.7	—	0.50	1.00	1.00
Cr	18 to 20	16 to 18	15.0 to 17.0	28.5 to 31.0	20 to 23	—	11.5 to 13.0	10.5 to 11.75	11.5 to 13.5
Ni	8.0 to 10.5	10 to 14	>72	Bal. (>58)	32 to 35	—	—	0.50	—
Mo	—	2.0 to 3.0	—	0.2	—	—	—	—	—
Fe	Bal.	Bal.	6.0 to 10.0	9.0 to 11.0	Bal.	—	Bal.	Bal.	Bal.
Cu	—	—	0.50 max.	0.10	0.75	—	—	—	—
Co	—	—	0.015 avg.	0.014	0.10	—	—	—	—
Al	—	—	—	0.40	0.15 to 0.45	—	—	—	—
Ti	—	—	—	0.40	0.60	—	—	6xC to 0.75	—
Other	—	—	—	N = 0.050 B = 0.005 Nb = 0.1	Ti/C ≥ 12 Ti/(C+N) ≥ 8 N ≥ 0.03	—	—	—	—

(A) From ASM Handbook.⁵⁴
 (B) From EPRI.⁵⁵
 (C) From Gorman.⁵⁶
 (D) From Stellwag, et al.⁵⁷

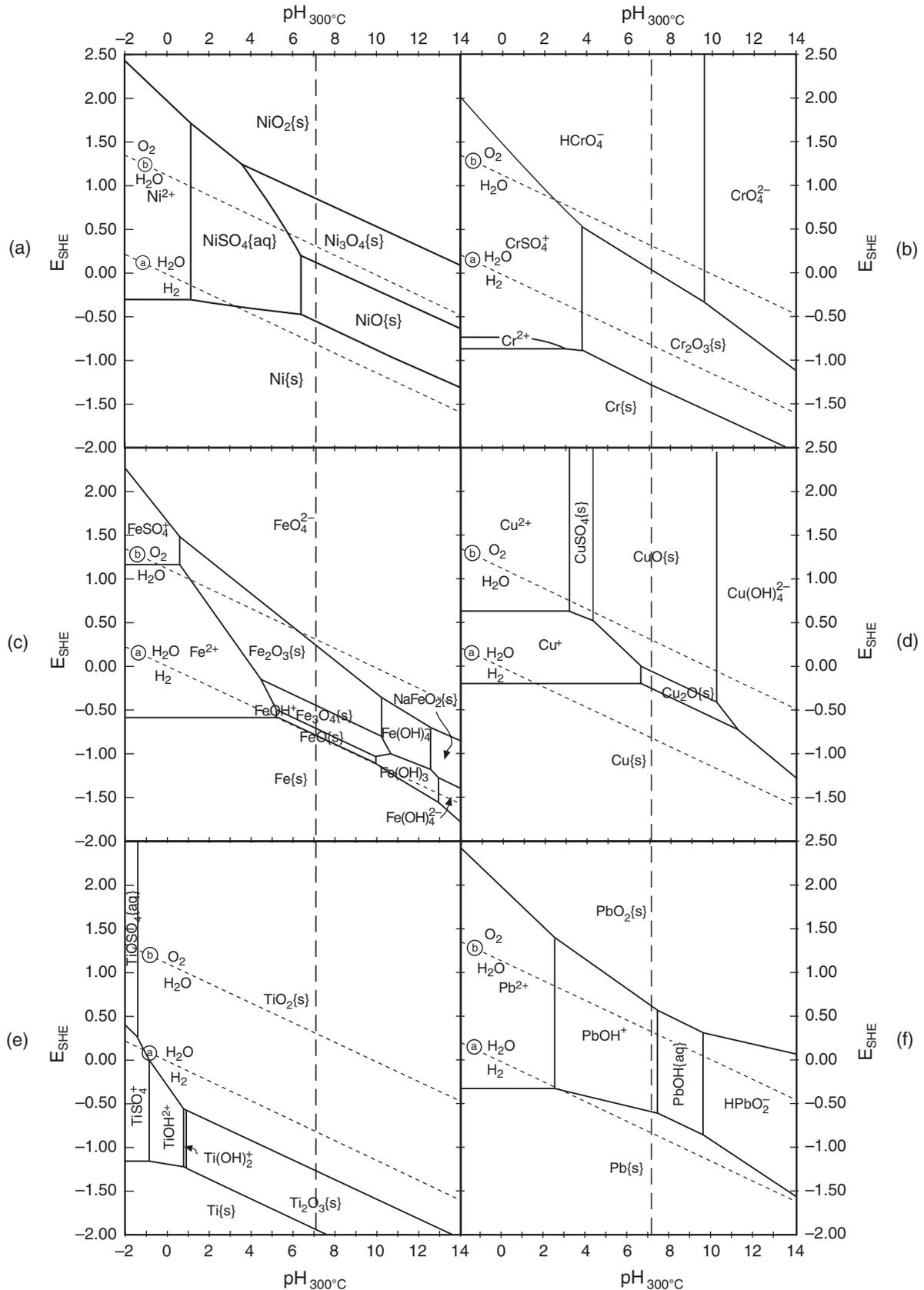


FIGURE 7. Potential-pH diagrams for (a) Ni, (b) Cr, (c) Fe, (d) Cu, (e) Ti, and (f) Pb calculated at 300°C and for 10^{-6} M solubility of the soluble species. Diagrams calculated based on a 0.01-M Na_2SO_4 titrant. The dashed line gives the natural pH of the 0.01-M Na_2SO_4 titrant.

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concentrations inside crevices compared with the respective concentrations of species in the bulk solution. Thus, the corrosion that occurs inside heat-transfer crevices needs to be assessed in terms of these concentrated solutions rather than the dilute bulk solutions.

Heat-transfer crevices are well known in some industries and in the corrosion field. A good illustration of the effects of a heat-transfer crevice is shown in Figure 9 from the work of Warren.⁵⁹ His experiment is shown in Figure 9(d) and is similar to the case for a hot pipe adjacent to wet insulation, which is a common source of aggressive ions. Data from his work are shown in Figures 9(a) through (c). In each of these cases, the hot and concentrating surface is accelerating SCC, just as occurs in steam generators. The problem of heat-transfer crevices and their virulence is not new. Its first manifestation provided the precursor conditions necessary for riveted steam boiler explosions in the 19th century and the first half of the 20th century.

Figure 10 shows a schematic view of a heated crevice at a tube support. Features of the heated crevices at the top of tubesheets and in sludge piles are similar to those of the heated surface that are shown in Figure 8. The important features of heated crevices in steam generators are the following:

- The temperature of the outside diameter (OD) surface of the tube is elevated compared with conventional nucleate boiling heat transfer when it is close to the primary temperature, T_p . The temperature of the secondary water, T_s , is cooler. $T_p - T_s$ is the available superheat.
- The crevice fills with deposits, mainly iron oxide resulting from corrosion of iron-based materials of construction in the secondary system; these deposits are usually bound with silicate deposits. Additional iron oxide is produced by corrosion of the tube support or tubesheet. Eventually, the crevice becomes fully packed with these deposits.
- Owing to the packing of the crevice and its effect on mass flow, the region within the crevice tends to be superheated dry steam with water at the edges at a water-steam interface.
- Compounds and chemistries of deposits that have been observed in heated crevices are discussed in Sections 4.5 and 4.6. Typically, these deposits are complex compounds containing iron, aluminum, silicon, and other species. These deposits also include organic species such as formic and acetic salts.
- Sulfur is often present, initially, as sulfate that can be reduced by hydrazine to produce aggressive lower valence species. Sulfate also can be reduced by Fe, Cr, and Ni in a liquid phase.
- Also, inside heat-transfer crevices there are sharp gradients in electrochemical potential,

temperature, concentration, and fluid density, as illustrated in Figures 10 and 80. These gradients cause substantial changes of the environments in the restricted space of the crevice as described by Baum and Rootham.⁶⁰ The occurrence and magnitude of these gradients depend on the porosity of the deposits.

Such an array of possible conditions inside heated crevices, as implied in Figure 10 and described in Sections 4.3 through 4.6, suggests that there are different environments that could produce corrosion inside these crevices, even within the same SG.

Deposits in heat-transfer crevices accumulate with time, although deposits at the top of the tubesheet are often removed at outages by sludge lancing and occasionally by chemical cleaning. A schematic view of accumulation of deposits at the top of a tubesheet with time is shown in Figure 11. The sludge crevice increases with time, and the associated corrosion does not begin until the sludge reaches some critical depth.

Deposits accumulate in tube support crevices starting with the asymmetric contact shown in Figure 12. SCC starts at such locations. With time, the deposits accumulate around the periphery, as shown in Figures 12(b) and (c), and SCC follows, as shown in Figures 12(d) and (e).

One of the major reasons for using the line contact tube support geometries as shown in Figures 3 and 5 is to reduce the accumulation of deposits as illustrated in Figure 12. However, Figures 13(a) and (b), typical of direct observations, show schematically that deposits gradually accumulate in these geometries. Figures 13(c) and (d) suggest that the local buildup of deposits at line contact geometries leads to thermal gradients in the tubes, which can accelerate SCC. Virtually no work has been conducted to determine whether the conditions of Figure 10 exist for the deposits shown in Figures 13(a) and (b). Further, the smaller volume region associated with the obtuse angle of intersection, as shown in Figure 13(b), can provide conditions for extended accumulation of deposits that are similar to those in drilled hole geometries, as shown in Figure 10. However, to date, and despite the occurrence of accumulations such as those shown in Figures 13(a) and (b), there is little evidence of any serious corrosion in tubing having occurred at these locations. Some cracking occurred in France in Alloy 600MA tubes at Chinon-B1 under broached tube support plates (TSPs) of carbon steel.

A possible consequence of the drilled hole crevice geometry, as shown in Figures 2(c), 3(a), 8(a), 12, and 16, and the corrosive environments that are sequestered, is corrosion of the tube support material adjacent to and around the tube. Such a case is illustrated in Figures 14 and 15. Figure 14 shows the result of corrosion of a carbon steel tube support that produces a relatively large volume of corrosion

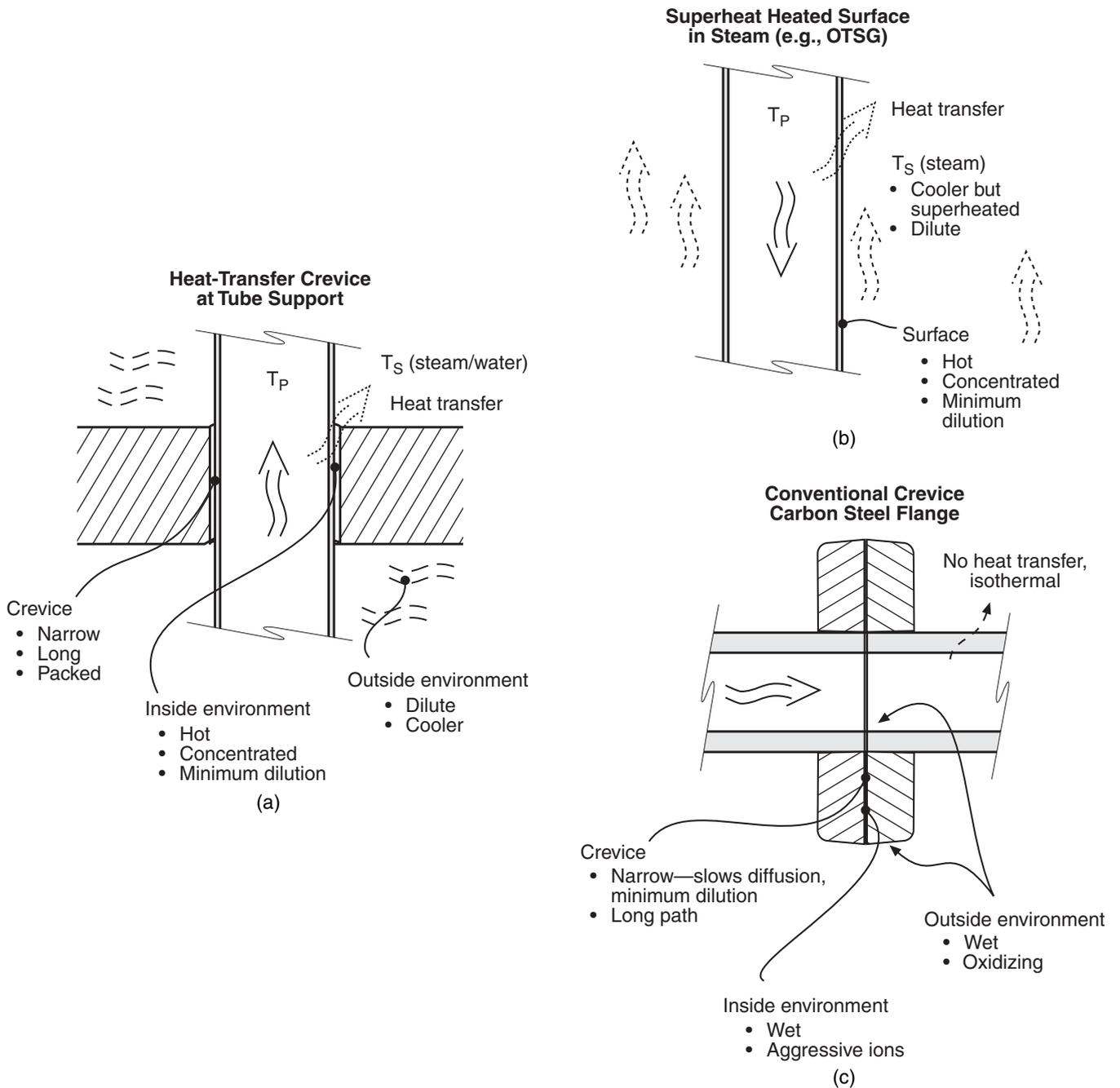


FIGURE 8. Three crevice geometries for producing corrosion: (a) heat-transfer crevice typical of those at tube supports, sludge piles, and top of the tubesheet; (b) heated surface in steam where concentration occurs; (c) nonheat-transfer crevice. The terms T_P and T_S are used for the temperatures of primary and secondary environments.

product. This expanding volume of corrosion products exerts sufficient pressure to distort the tube. The ratio of the specific volume of the oxide to that of the metal is called the "Pilling-Bedworth ratio;"⁶³ for iron oxides, this ratio is about 2.15 and 2.1 for magnetite and hematite, respectively. The corrosion products are typically not fully dense, and the effective ratio of oxide to metal may be in the range of 3 to 4. This expansion partially strangles the tube; when the tube is viewed from the inside, these expanded loca-

tions appear as "dents," and the phenomenon is called "denting." The conditions that lead to denting are described in Section 3.5.3. Another consequence of denting that is imposed on the tube is slow straining. This straining produces and accelerates SCC on both primary and secondary sides in accordance with the well-known effects of slow straining described by Parkins.⁶⁴⁻⁶⁵

In addition to denting that has occurred at tube supports, denting of a lesser magnitude can occur at

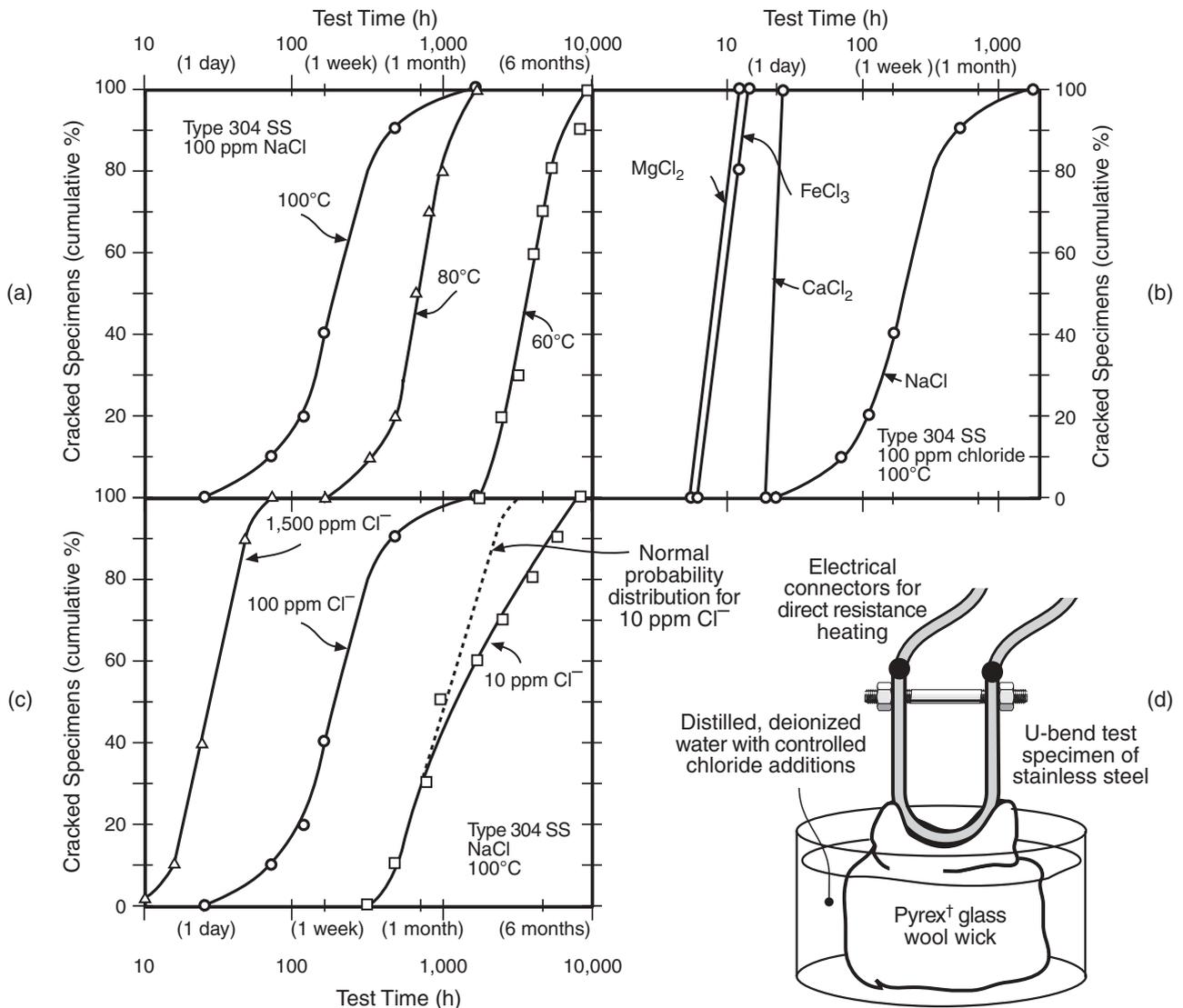


FIGURE 9. Cumulative percent of cracked specimens vs test time considering the effects of: (a) temperature, (b) cation, and (c) concentration of chloride in the bulk water. (d) Experimental arrangement. From Warren.⁵⁹

the TTS crevices as illustrated schematically in Figure 15. This denting is significant since it is not reduced by use of line contact tube support geometries. It is inherent in the use of vertical steam generators. Also, it is inevitable that this crevice remains after fabrication since it is not practical to expand the tube to more than 3 mm to 6 mm from the top of the tubesheet; further expansion above the upper surface of the tubesheet entails risks of producing damaging bulges in the tubing above the top of the tubesheet.

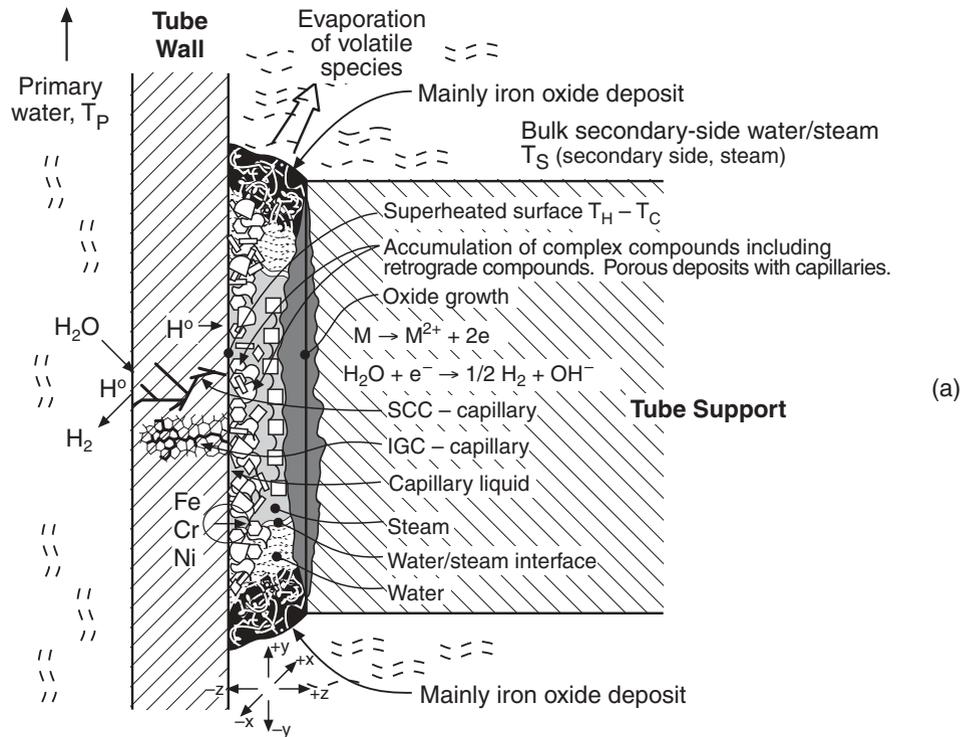
1.5 SCC in Heat-Transfer Crevices and on Free Surfaces

Figure 16 shows a typical distribution of SCC and IGC in a tube at a tube support. The corrosion occurs around most of the periphery, as suggested by Figure 12, and occurs generally about the vertical

midpoint consistent with the most likely location of steam, as shown in Figure 10. IGC here is occurring simultaneously with the SCC, and this is a common occurrence. The SCC is generally longitudinal as is typical of SCC at tube supports.

SCC at TTS locations on the secondary side appears similar to that in Figures 12 and 16, exhibiting both SCC and IGC. However, some circumferential SCC occurs at the TTS location and seems to be influenced mainly by the distribution of stresses that result from tube expansion. Stresses sufficient to produce circumferential stress at the TTS also can be produced by a combination of locking tubes in the TSPs by denting and consequent flexing and buckling.

Figure 17 shows the occurrence of SCC at a free surface on the cold leg. Such a result is surprising both because it occurred on the cold leg and because



Cl	(Cl ⁻ , HCl)	B, Ti, Zn	(inhibitors)
SO _x	(retrograde compounds plus S ⁶⁺ → S ⁴⁺ → S ²⁺ → S ⁰ → S ²⁻)	O ₂	(O ₂ , H ₂ O, compounds)
SiO _x	(SiO ₂ , complex compounds)	H ₂	(H ₂ , H ⁺ , H ⁰)
AlO _x	(Al ₂ O ₃ , complex compounds)	N ₂ H ₄	(N ₂ H ₄ , NH ₃ , N ₂)
Cu	(Cu ⁰ , Cu ²⁺ , CuO)	C	(CO ₃ ²⁻ , organic)
Pb	(Pb ⁰ , PbO _x ^y)	N	(NO _x , organic)
Na, Ca, Mg	(complex compounds)	Fe, Cr, Ni	(Fe ²⁺ , Cr ³⁺ , Ni ²⁺ , complex compounds)
Na ₂ HPO ₄	(retrograde compounds plus H ₃ PO ₄)		

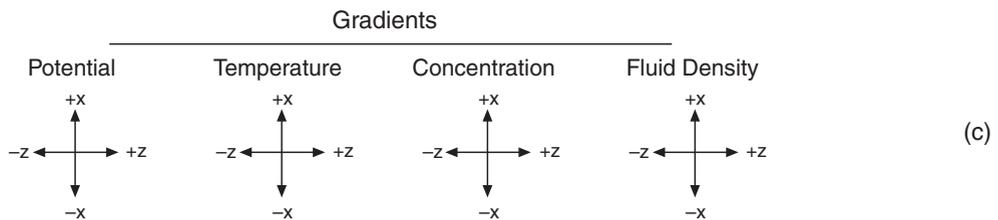


FIGURE 10. Schematic view of heat-transfer crevice at a tube support: (a) geometry, (b) chemicals that accumulate and transform, and (c) types of gradients inside the heat transfer crevice. From Staehle.⁶¹

it occurred on the free surface. SCC at free-spans also has occurred in the upper bundle of OTSGs as noted in Figure 25. Such SCC is associated mainly with the high stresses and cold work produced by scratches that occurred during the insertion of tubes into the steam generator support structure. These SCC incidents are important because, once they were initiated by the scratches, they propagated for other reasons, possibly related to LPSCC on the secondary side. Some of these SCC in Figure 17 propagated through the wall. While incidents of SCC at free-spans, as shown in Figure 17, are not frequent, they

are important in that they show the inherent susceptibility of Alloy 600MA even when exposed to nonconcentrating conditions (i.e., not in heat-transfer crevices but on free surfaces). Table 2 summarizes incidents of SCC at free-spans on the secondary side together with descriptions and possibly contributing factors.

1.6 Temperatures and Concentrations

SCC on the secondary side at tube supports increases with temperatures on the surfaces of tubing and with concentrations of impurities in the crevices.

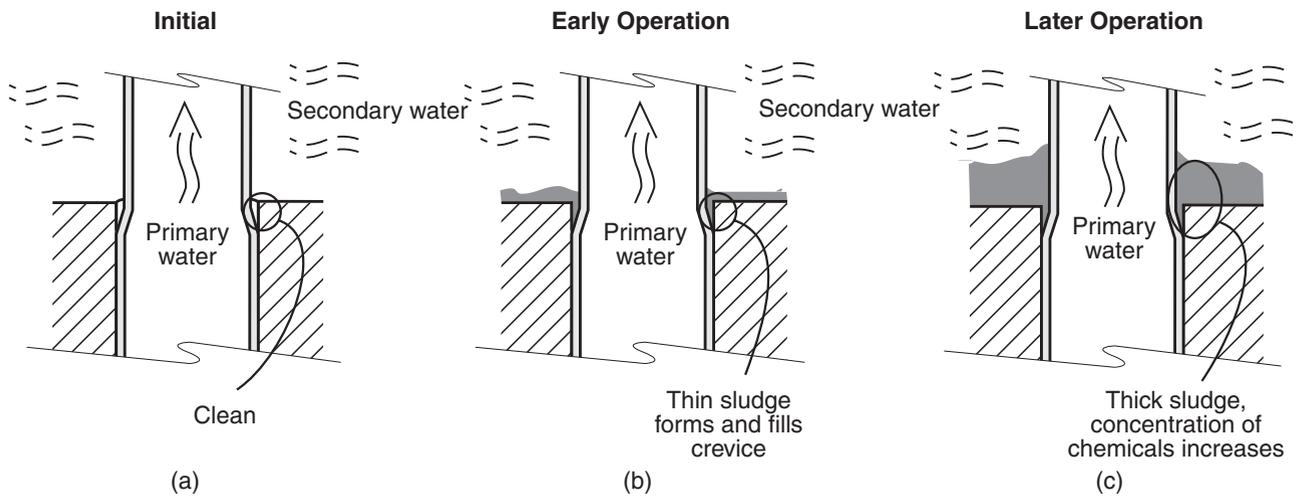


FIGURE 11. Accumulation of deposits on top of the tubesheet with time.

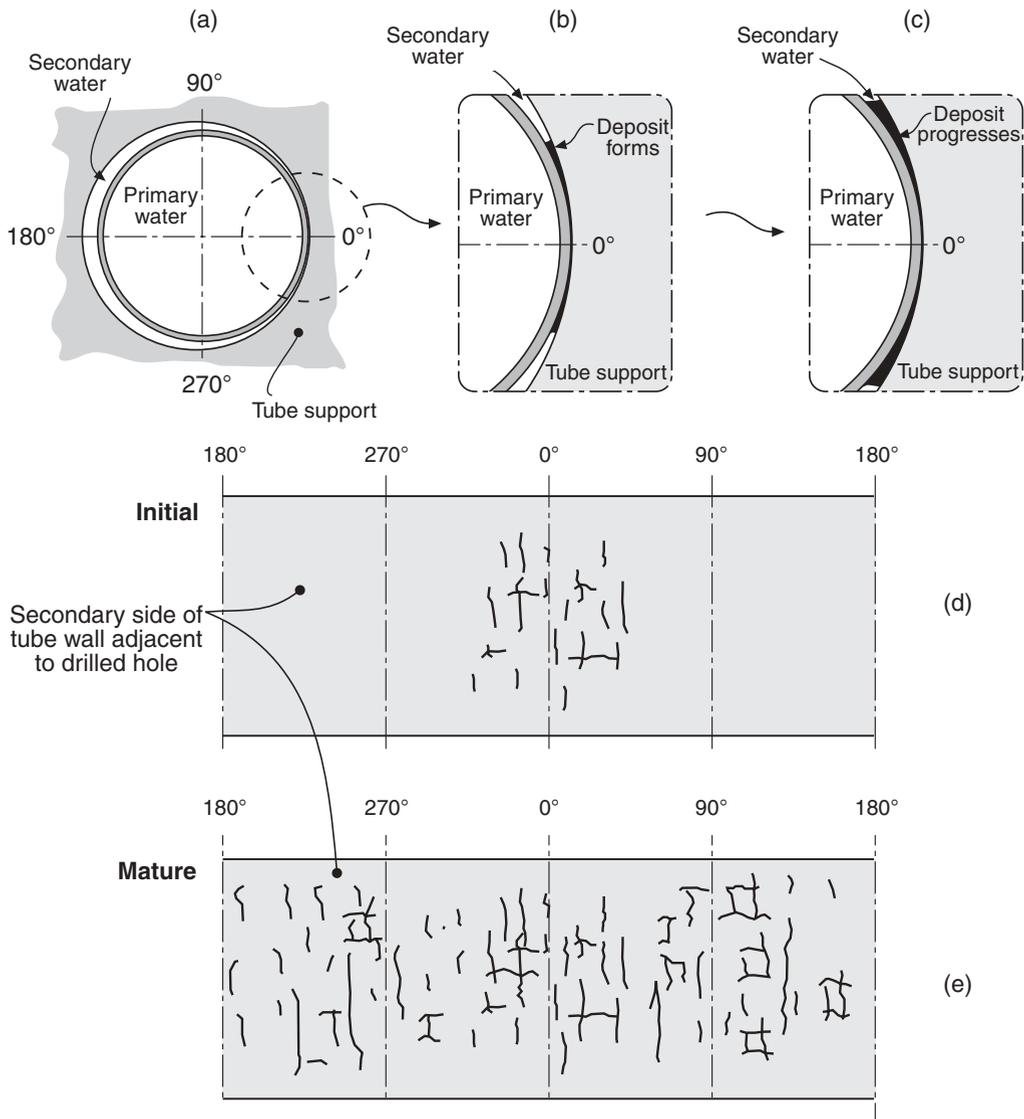


FIGURE 12. (a), (b), and (c) Progressive accumulation of deposits in a drilled hole tube support. Schematic views, based on direct observations, of progression of SCC in (d) and (e) associated with the accumulation of deposits. From Staehle and Gorman.⁶²

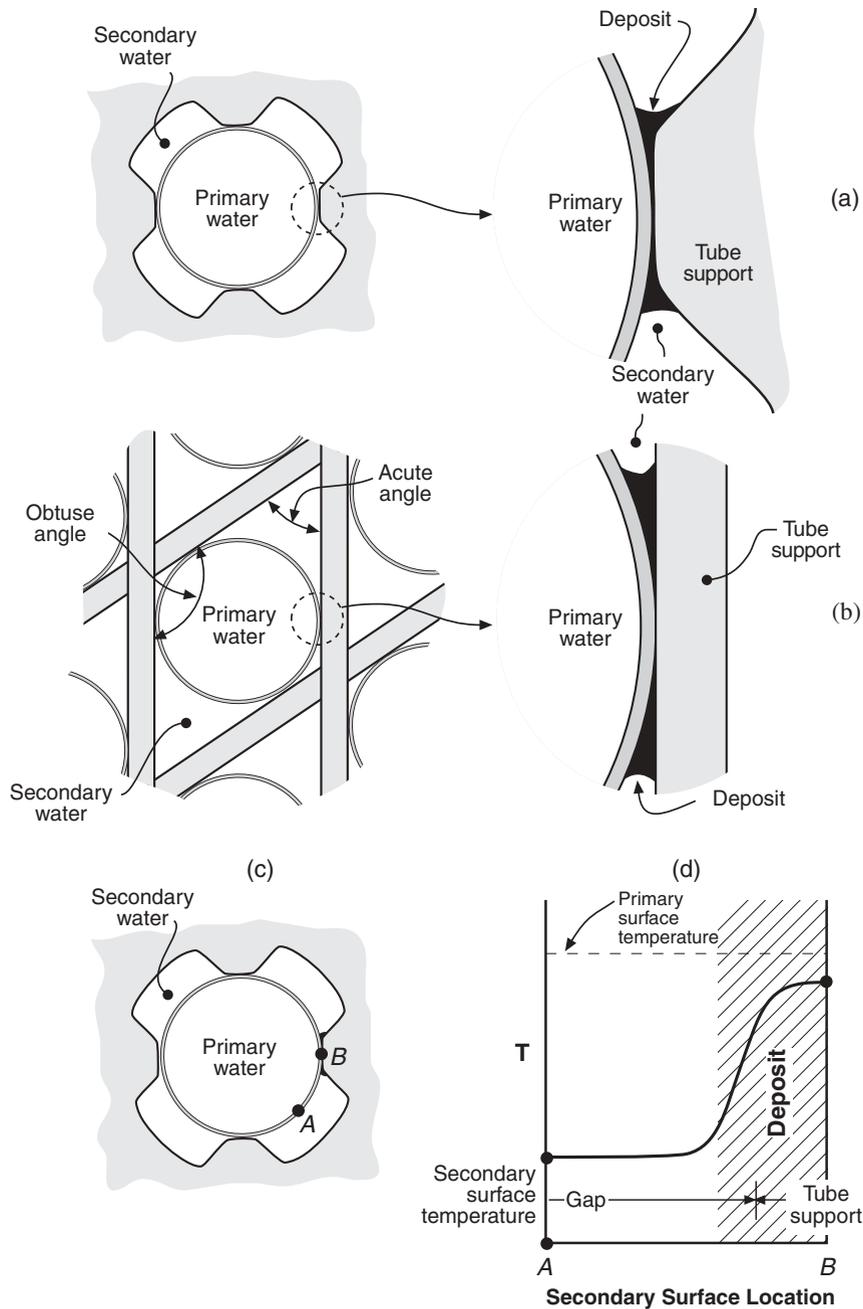


FIGURE 13. Schematic view of accumulation of deposits at line contact crevices associated with: (a) broached holes and (b) egg crates. Example of different temperatures at: (c) open area and (d) deposit-filled contact area.

Figure 18(a), taken from Takamatsu, et al.,⁶⁶ shows the decreasing occurrence of eddy-current indications, presumably due to SCC-IGC, at successively higher tube supports for three different plants. The accumulation of contaminants is decreased with decreasing superheat. With increasing height, T_p is reduced; this reduces both the superheat and the surface temperature. Which of these effects is acting here is not clear. Figure 18(b), taken from Baum,⁶⁷ correlates the T_p with the number of tubes having eddy-current indications. The activation energies

here greatly exceed those of about 20 kcal/mole to 70 kcal/mole for outside diameter stress corrosion cracking (ODSCC), as discussed in Sections 5.2.1.6, 5.2.2.6, and 5.2.3.6. The large difference between the activation energies observed in plants and in laboratory studies suggests that factors other than a direct temperature dependence, such as the effect of available superheat, $T_p - T_s$, are responsible for some of the dependencies on temperature observed in plants. For example, as discussed in this section, as superheat increases, the concentration of impurities in the

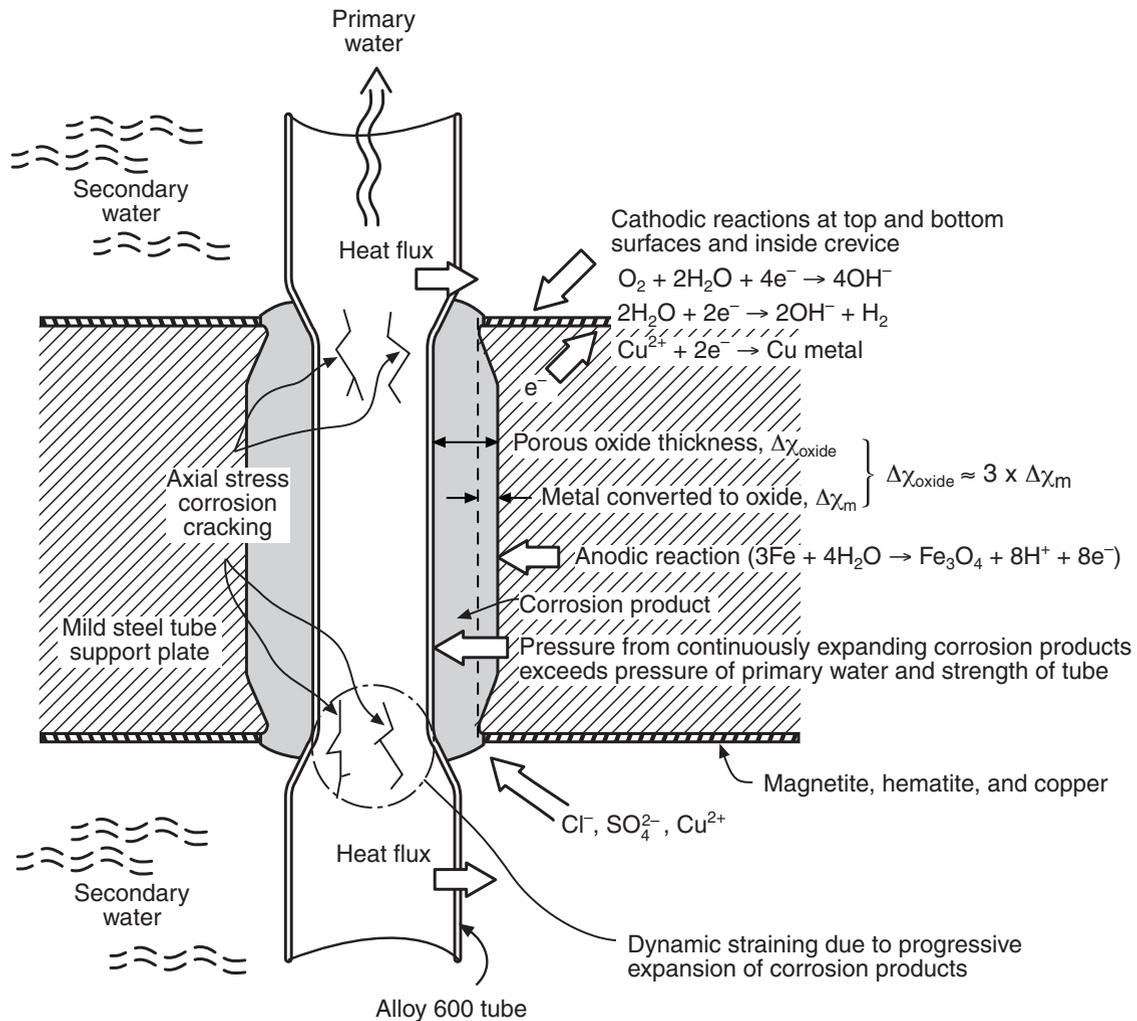


FIGURE 14. Schematic view of denting at a drilled hole tube support location showing the factors involved. From *Steam Generator Reference Book*.⁴⁹

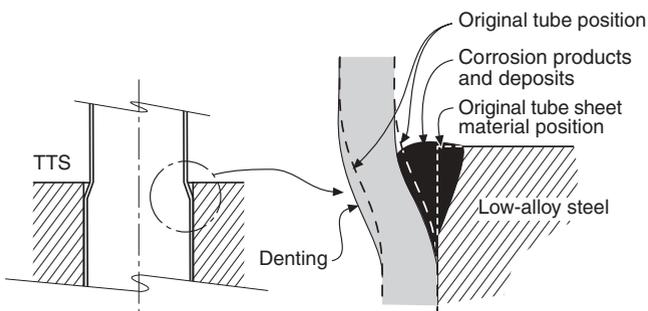


FIGURE 15. Schematic view of denting at the top of the tubesheet (TTS) crevice.

crevice increases and their molar ratios change, possibly increasing the rate of corrosion.

Figure 19 shows the early history of hot leg temperature (T_H) for early steam generators. The low temperatures of early plants protected them from serious corrosion unless other factors were present. For example, SCC due to chlorides or caustics oc-

curred in the Shippingport plant and in the PM3-A on tubes and at locations where stainless steels were used. However, even for some of the early plants with Alloy 600MA and lower temperatures, like San Onofre-1, Conn Yankee, and Robinson-2, alkaline stress corrosion cracking (AkSCC) and alkaline intergranular corrosion (AkIGC) occurred. In addition to the influence of surface temperatures, the concentration of chemical species in crevices from the very dilute bulk concentrations due to superheat contributes significantly to corrosion in heat-transfer crevices. Figure 20 shows the magnitude of concentration that can occur in heat-transfer crevices for various species as a function of concentration in the environment from the work of Takamatsu, et al.⁶⁸ Of note here are the different concentration factors. Na^+ and K^+ concentrate more than Cl^- and F^- , and these concentrate more than B (from boric acid). Such differences are due to the different vapor pressures of these species. The lower concentrating species have higher vapor pressures, which account for their loss

into the steam phase and the apparent differences between the cations and anions. Such patterns also account for some of the variation in compositions in crevices.

1.7 Extent of Failures

Failures in steam generators have been extensive on both secondary and primary sides for recirculating units. Numerous descriptions of the arrays of failures have been published.⁷⁰⁻⁷² Figure 21 shows the many modes of corrosion and failures at various locations that have occurred mainly in recirculating steam generators. Each combination of failure mode and its location is called a “mode-location case.” The nest of tubes where failure occurs and where deposits build up is illustrated by Varrin, as shown in Figure 22.⁷³

Corrosion that is associated with heated crevices in steam generators occurs in all the five modes: GC, pitting, IGC, SCC, and corrosion fatigue (CF). Sometimes, in steam generators, IGC and SCC occur together; the combination is often called intergranular attack/stress corrosion cracking (IGA/SCC). The SCC may be either intergranular or transgranular (IGSCC or TGSCC), although it is predominantly the former in experience with SGs.

Categories of mode-location cases of corrosion that have occurred over time in steam generators using Alloy 600MA, and in some cases Alloy 600TT, are shown in Figure 23. In all, there are or have been 25 specific mode-location cases of corrosion-related failures, as shown in Figure 21, which have occurred on the primary and secondary sides of steam generator tubes. Figure 23 shows that general corrosion was the most frequent form of damage in the early years, but by 1976 denting became important. Pitting and fretting have persisted but have not been dominant. LPSCC on the inside diameter (ID) became a dominant mode by about 1980, and SCC on the OD started to be significant about the same time but continued to increase through 1993; IGA/IGSCC continues to be a dominant concern.

Another view of the rate of failures in RSGs with Alloy 600MA tubes is shown in Figure 24 where the probability of failure vs service time is shown for the Ringhals-4 plant. Here, failures are shown for a set of mode-location cases also identified in Figure 21, although in more detail. Also, in this plot of Figure 24, the individual rates of failure for the respective mode-locations cases are aggregated statistically, and the aggregate plot is shown as the top curve in Figure 24. This shows that the overall probability of failure would reach 0.1 probability, 10% of tubes failed, by about 21 EFPY (equivalent full power years). The evolution of these statistical occurrences of various mode-location failures in steam generators has been described by Staehle, et al.,²¹ and by Stavropoulos, et al.⁷⁴

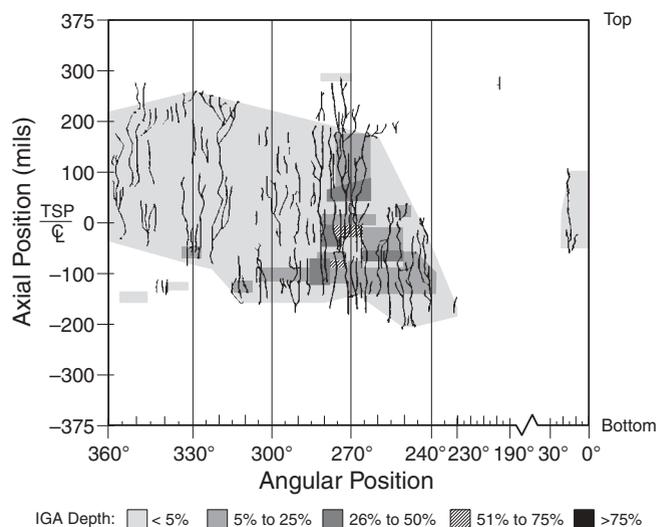


FIGURE 16. SCC and IGC observed in the Catawba-1C steam generator at the third tube support in an Alloy 600 tube. Distance from center line vs angular position from an arbitrary reference point. From Eaker. Courtesy of R. Eaker of Duke Power, Charlotte, North Carolina. Private communication.

With respect to the OTSGs, in contrast to the RSGs, failures in the lower bundle, where the tubes are covered with water, have not been significant. However, significant failures have occurred in the upper bundle where the steam is superheated. Figure 25 shows the water and steam conditions over the height of the steam generator together with the failure of tubes for a single steam generator. Most of the tube failures in OTSGs have been in the higher temperature upper bundle region where the surfaces are superheated. During the early years of plant operation, most of these failures were due to impingement erosion or wear at supports. However, in recent years, the dominant failure mechanism has been IGA and SCC of free-span surfaces, especially at scratches, which are produced during the insertion of tubes.

Probably the best overall assessment of the initial design of drilled hole tube supports in steam generators using Alloy 600MA tubing is shown in Figure 26, where the fraction of steam generators replaced is shown vs calendar years using a median rank with suspended items analysis for 122 plants. The mean calendar time for life is about 25 years and is mostly limited by LPSCC on the primary side. Thus, economic loss due to corroded steam generators has been large, as illustrated in Figure 27.

1.8 Improvements and Amelioration

Failures of steam generator tubing that have occurred over time have incited changes. For example, Figure 28 shows the basic design changes going from the D3 model of Westinghouse-designed plants to the Model F. Here, the tubing alloy, tube

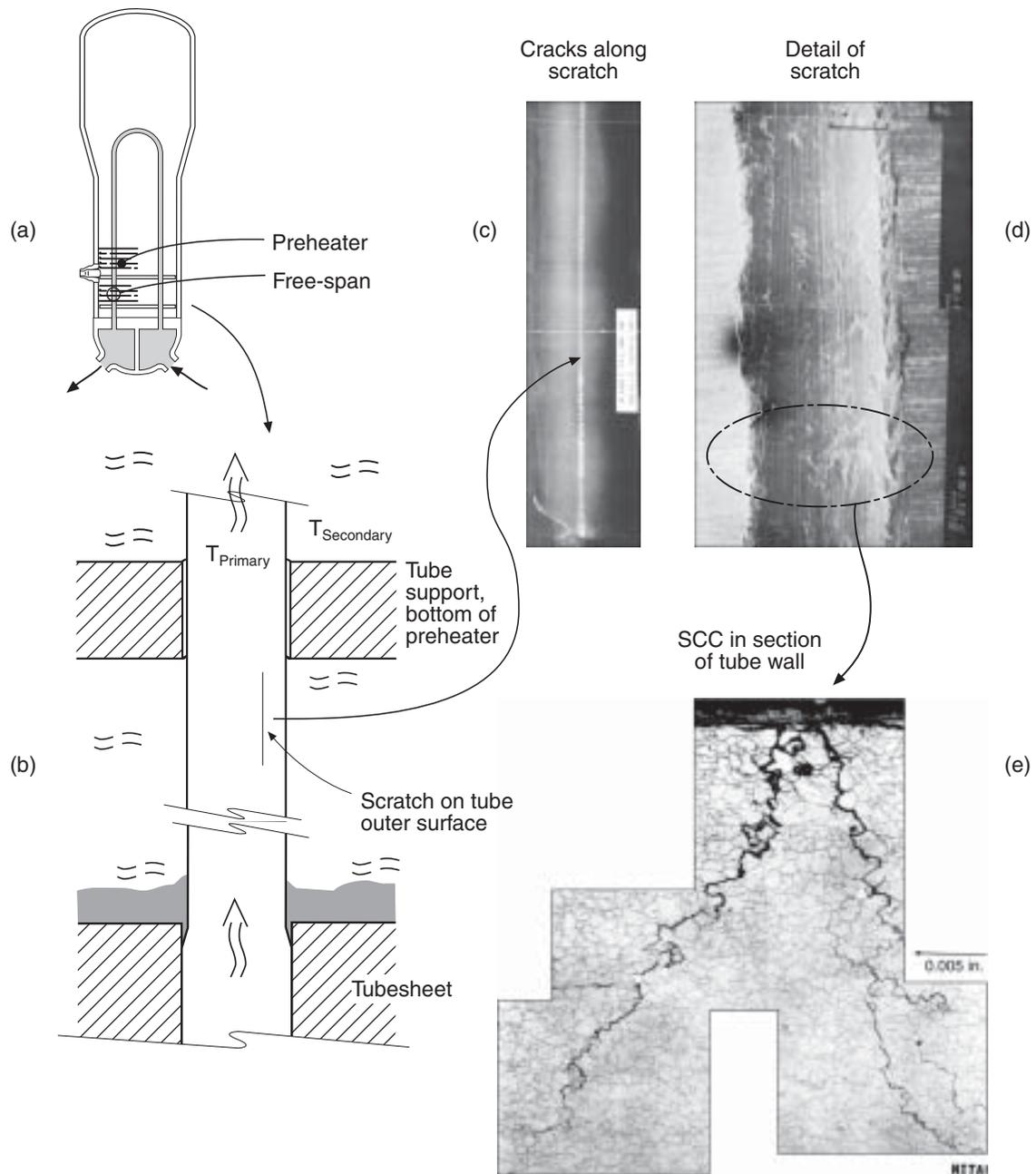


FIGURE 17. SCC at free-span cold leg at McGuire-2 in an Alloy 600MA tube. (a) General location of scratch and SCC; (b) schematic view of location of SCC; (c) OD of tube showing scratch; (d) detail of scratch with SEM; (e) cross section of SCC. From Eaker. Courtesy of R. Eaker of Duke Power, Charlotte, North Carolina. Private communication.

support design, tube support material, stress relief, blowdown, and access were changed.

Overall, for various manufacturers, water chemistry, maintenance, design, and materials were changed (Figure 29).

1.9 Significance

1.9.1 Experience — Experiences summarized in this introductory review are the following:

1. The original design choices for many steam generators did not produce satisfactory perfor-

mance and led to many problems and large costs. Addressing these weaknesses has required many changes, including improvements in steam generator design details, steam generator tube and support materials, and plant water chemistry. Improvements in plant water chemistry have required many expensive changes, such as changes in heat exchanger tube materials and substantial upgrades to water treatment equipment and water quality monitoring instrumentation.

TABLE 2
Incidents of Secondary-Side Free-Span Cracking^(A)

Reactor (Year of Start-up) [Date of Incident]	Steam Generator Type (Tubing Material)	Water Chemistry	Nature of Cracking	Possible Contributing Factors
Ft. Calhoun (1973) [May 16, 1984]	Combustion Engineering Model 67 (600 HTMA)	AVT, boric acid additions	Fish-mouth rupture ~32 mm long, plus series of small fissures ~6 mm long, oriented ~45° to tube axis, all in horizontal portion of tube at intersection with vertical support straps.	Stresses from tube ovalization, thick tube scale, denting of tube from corrosion of adjacent support straps, local "steam blanket" from structural occlusion.
McGuire-1 (1984) [March 7, 1989]	Westinghouse Model D-2 (600 LTMA)	AVT, boric acid additions	Axial crack, ~95 mm long and 10 mm max. opening on cold-leg side at 20th TSP.	Axial abrasions ~1 mm wide and <25 μm deep; surface residual stress ~90 ksi max. No significant surface deposit.
Maine Yankee (1984) [December 17, 1990]	Combustion Engineering Model 67 (600 HTMA)	AVT, boric acid additions	Leaking axial crack ~100 mm long at top of U-bend apex; 10 other tubes plugged for similar smaller cracks.	Tube deformation; adjacent tube support reduces flow in this region, creating local "steam blanket."
Doel-4 (1985) [October 1991]	ACE (Westinghouse) Model E ^(B) (600 LTMA)	Phosphate	Multiple axial SCC; cellular cracks in scratches.	Residual stresses, 20-μm-thick oxide film, "oily film," Pb and Cu in secondary system.
McGuire-1 (1984) [January 16, 1992]	Wesinghouse Model D-2 (600 LTMA)	AVT, boric acid additions	Leaking axial ~25 mm long ~130 mm above 20th TSP; additional cracks between 14th and 15th TSP had depths up to 60% of wall thickness. 94 similar indications were plugged.	Axial grooves similar to those observed in McGuire-1 1989 incident (see above).
Farley-1 (1977) [September 1992]	Westinghouse Model 51 (600 LTMA)	AVT, morpholine (1987); molar ratio control (1990); boric acid (1993)	Three equally spaced axial bands consisting of extended networks of IGSCC; average depth <10% of tube wall; max. depth 24%.	Surface deposits contained >10% Pb and 4% Cu; possible residual stresses.
Palo Verde-2 (1986) [March 14, 1993]	Combustion Engineering Model 80 (600 HTMA)	AVT (hydrazine)	Axial fish-mouth rupture ~65 mm long associated with ODSCC at crevice between adjacent contacting tubes.	Adherent ridge deposit, cold work from prior scratches, susceptible microstructure, increased sulfate from resin intrusion.
Braidwood-1 (1988) [October 23, 1993]	Westinghouse Model D-4 (600 LTMA)	AVT	Leaking axial crack ~33 mm long located above top TSP between two AVBs.	Probable surface defects; no significant surface deposit (?)
Crystal River-3 (1977) [June 1992; April 1994]	Babcock and Wilcox OTSG	AVT	Isolated patches of shallow IGA typically <2.5 mm in diameter and located 20 cm to 46 cm above lower tubesheet secondary face.	Nonuniform tube surface deposits; IGA believed to have occurred early in plant life (reduced S attack) and no longer active.

^(A) From Diercks, et al.⁶⁹ ©1996 NACE International.

^(B) Steam generators replaced with Siemens-Framatome units in 1996.

2. Crevices are the locations in PWR SGs on the secondary side that are most susceptible to corrosion. The risk of corrosion at the crevices increases as their capacity to concentrate impurities increases. Accordingly, design features that minimize crevices and their concen-

trating capacity are important to successful SG design and operation. In this regard, drilled hole tube support plate crevices are especially prone to producing aggressive chemistries, and line contact crevices have performed better. Shallow TTS crevices, rather

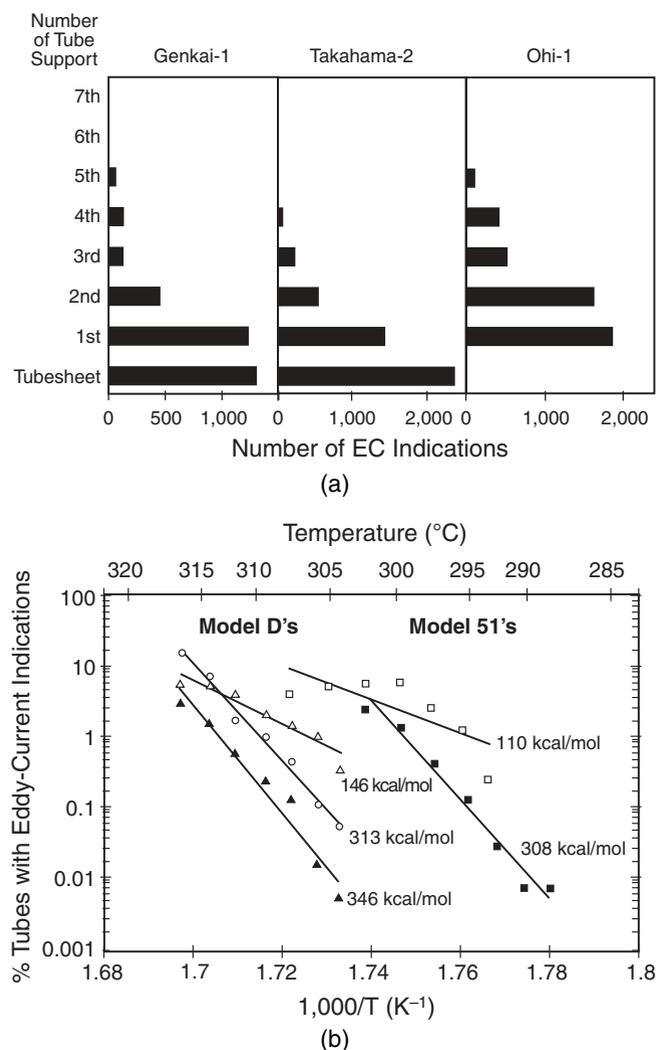


FIGURE 18. (a) Number of indications at successive tube support locations for three plants using Alloy 600MA after about 12 to 15 years service. From Takamatsu, et al.⁶⁶ ©1996 NACE International. (b) Percentage of tubes with eddy-current indications vs $1,000/T$ at successive tube supports for various Model D and Model 51 SGs. Activation energies shown. From Baum.⁶⁷ Courtesy of TMS, Warrendale, Pennsylvania.

than deep tubesheet crevices, are undesirable but seem to be unavoidable; accordingly, the choices of design temperature, processes used for expansion, and the material selected for tubes need to account for this relatively inevitable and vulnerable location.

3. Alloy 600MA has been found to be susceptible to a large array of corrosion problems in PWR SG service. Other alloys such as Alloys 800NG, 600TT, and 690TT are superior to Alloy 600MA. Despite their improved resistance to corrosion, these nominally improved materials are not immune to corrosion, and SG design and installation, plant design, and plant operating practices need to consider their proneness to corrosion in SG environments.

4. Carbon steel is not a suitable material for tube supports in PWR SGs, especially when used in a drilled hole configuration. Line contact tube support designs together with the use of stainless steel for the support material improve performance.

5. The water chemistry used with PWR SGs must be carefully selected and controlled to minimize corrosion, especially in crevices. In this regard, it has been found that the use of good water chemistry for long times ameliorates corrosion, but does not prevent it.
6. Despite maintenance of very good water chemistry in the SG bulk water, the environments that develop in heat-transfer crevices are very different and can be highly corrosive.

1.9.2 Potential Problems and Approaches — While most of the potential problems for longer operation and new steam generators are discussed in subsequent sections, some deserve special attention in Section 1.0:

1. Despite maintenance of very good water chemistry and use of designs that minimize the presence of built-in crevices, crevices will develop with time as deposits accumulate. Without remedial actions, these crevices may cause corrosion in new SG designs over long times.
2. Heat-transfer crevices have not been eliminated at the top of the tubesheet and in the flat contact tube supports where deposits of a significant extent can accumulate. These two locations require continued attention.
3. In view of the large number of mode-location cases of corrosion that have occurred in the past, the expectation that all of these mode-locations have been eliminated seems unreasonable. In evaluating the likelihood of future failures, the relatively long times required for the occurrence of significant LPSCC on the primary side and for the ODSCC on the secondary side of OTSGs should be considered.
4. While Alloy 690TT has been shown to be better in many ways than Alloy 600MA, the higher chromium concentration makes the alloy intrinsically more reactive. This potential deserves continued attention. Further, as is discussed in Section 5, Alloy 690TT is known to sustain SCC about as intensely as Alloy 600MA in environments that challenge its passivity, such as caustic with lead or reduced sulfur and slightly oxidizing mild (pH 4.5) acids.

2.0 TYPES OF STEAM GENERATORS

This section describes the types of steam generators that have been used in PWRs, the bases for

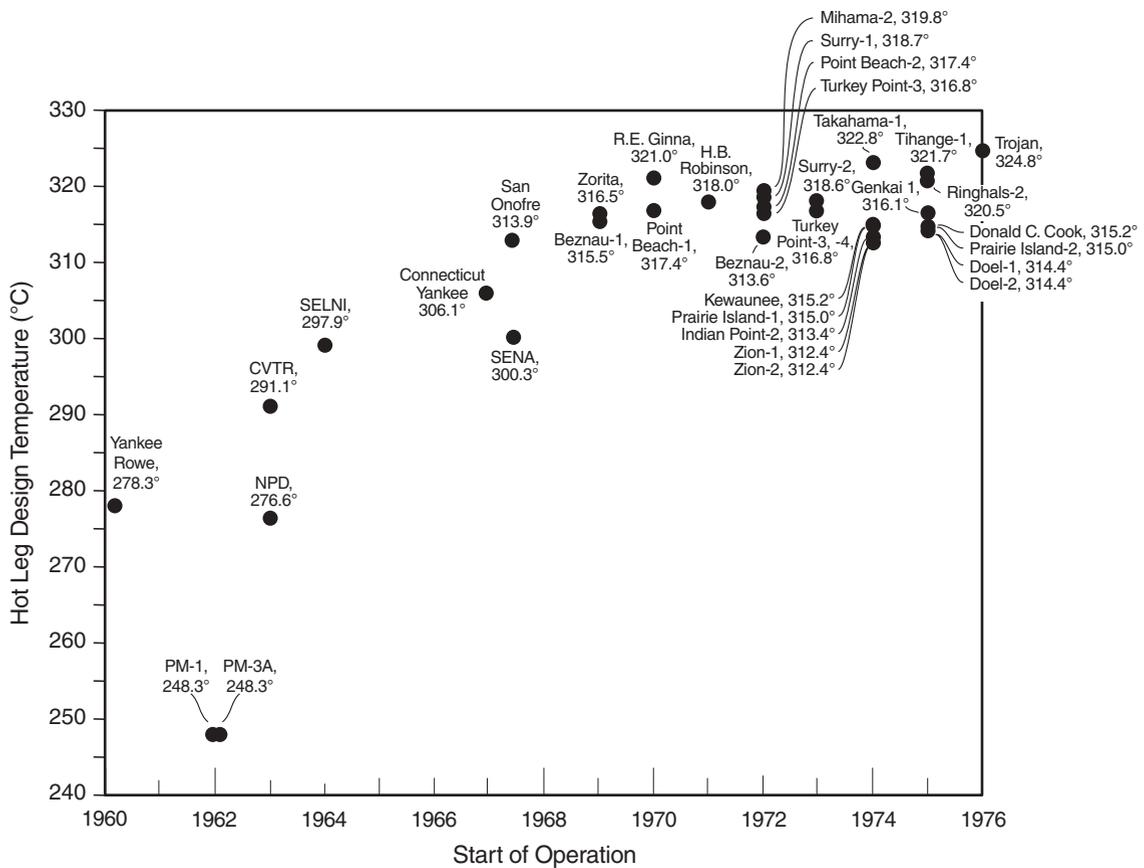


FIGURE 19. Hot leg design temperatures vs starting date of operations of early plants.

selection of various design features, and the significance of the operating experience regarding the desirability of various design features, especially concerning their effects on crevices.

The evolution of steam generator design is summarized in Table 3. Changes that have occurred in each of the main areas in Table 3 are described in this section.

2.1 Hot Leg Temperature, T_H

Hot leg temperatures used in early plants were relatively low, below about 300°C, as shown in Figure 19 in Section 1.6. These low temperatures reflected the experience of the Naval Reactors program and conservative inclinations with respect to avoiding corrosion and other problems such as embrittlement of cast stainless steel. Eventually, competition pressed builders of steam generators to raise temperatures so as to increase thermal efficiencies and power output. Also, during the first few years of operation, some early plants, which operated at low temperatures (Figure 19), did not sustain significant corrosion of SG tubes. In view of this early satisfactory performance, designers increased the hot leg temperatures, during the late 1960s and the 1970s, to the 325°C to 330°C range for plants sold in the mid-1970s. Since corrosion processes are thermally

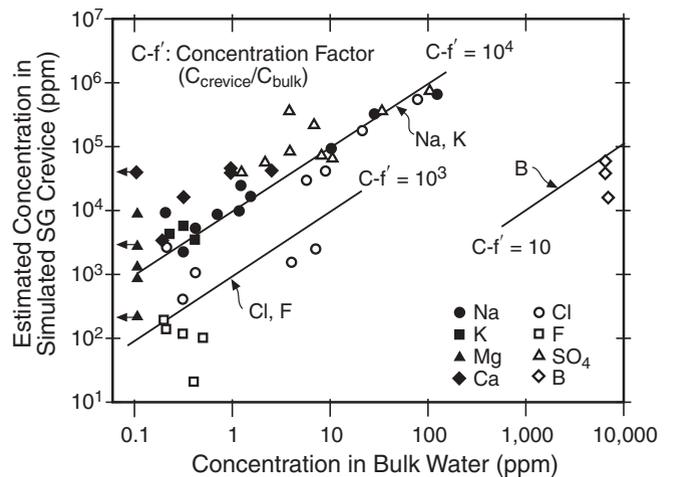


FIGURE 20. Estimated concentration of species in a simulated SG crevice vs concentration in the bulk water. Various concentration factors shown. From Takamatsu, et al.⁶⁸ ©1991 by the American Nuclear Society, La Grange Park, Illinois.

activated, this increase in temperature significantly increased the rates of corrosion (e.g., a factor of 2 increase in corrosion rate for each 10°C increase in temperature for an activation energy of about 50 kcal/mol).

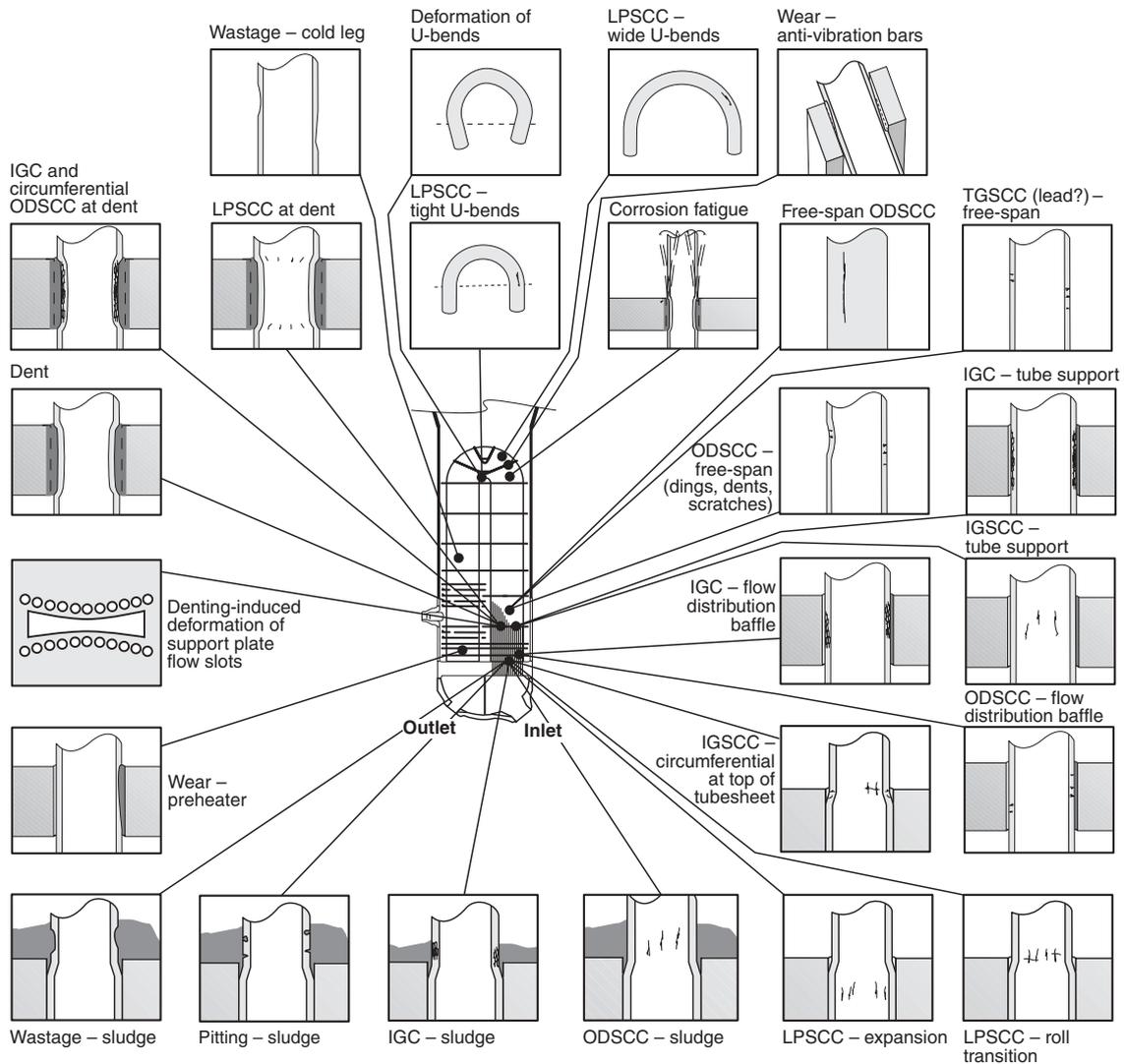


FIGURE 21. Array of modes of failure at various locations (mode-location cases) that have occurred in recirculating steam generators.

2.2 Heat Flux

Average heat fluxes for tube bundles increased from ~40,000 Btu/h/ft² (126 kW/m²) in early plants (1960 vintage) to ~55,000 Btu/h/ft² (173 kW/m²) for plants sold in the mid-1970s. This increase in heat flux occurred for the same competitive reasons as the increases in hot leg temperature. The results were increased rates of boiling and thus increased rates of concentration of impurities, as illustrated in Figure 10 and shown in Figure 20, and higher superheats in crevices that increased the theoretical equilibrium concentration of impurities, as shown in Figure 20 for the crevices illustrated in Figures 2 and 10. Both of these factors increase the likelihood and severity of corrosion in areas of heated crevice.

2.3 Recirculating vs Once-Through Design

Starting with the Naval Reactors program, most PWR steam generators have been recirculating steam

generators (RSGs). The initial reason for using recirculation was to avoid boiling the cooling water to dryness in the steam generator; such boiling can produce rapid accumulation of deposits, high local concentrations of impurities, and accelerated corrosion. However, experience with fossil-fueled plants in the 1960s showed that once-through boilers (with boiling on the inside of the tube) could work satisfactorily if water chemistry was stringently controlled. Based on this experience, B&W developed the once-through steam generator (OTSG) design for PWRs, which minimized crevices and strictly controlled water chemistry similar to that used for once-through boilers in fossil plants. Experience has shown that the OTSGs have improved resistance to corrosion as compared to RSGs of the same vintage but that they are not immune to corrosion problems with Alloy 600MA, especially IGC at scratches in the superheated region shown in Figure 25.

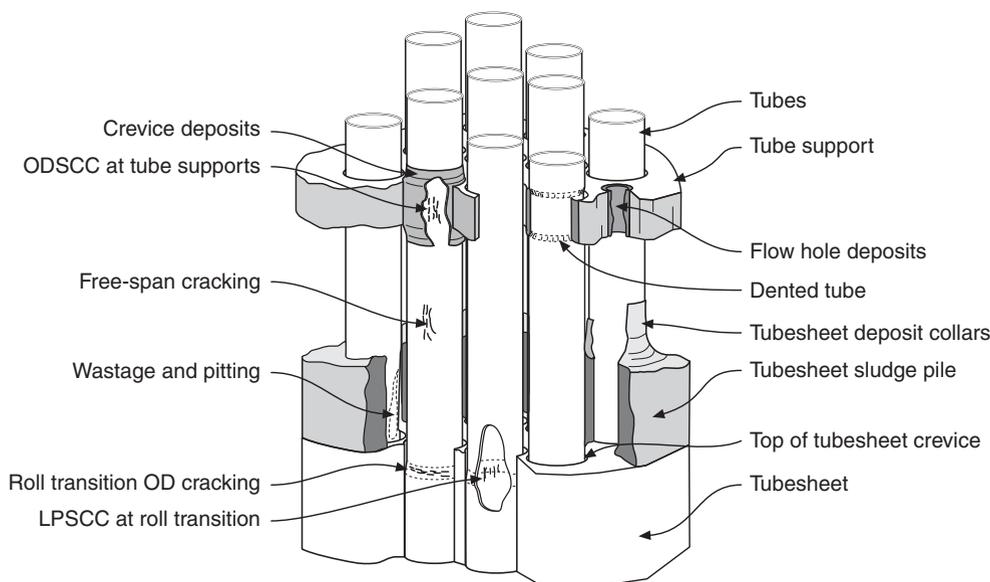


FIGURE 22. Array of tubes at tubesheet and tube support showing locations of deposits and failures. From Varrin.⁷³ Used by permission of EPRI.

2.4 Horizontal vs Vertical Orientation

A number of early PWR steam generators, e.g., those for Shippingport, the N-Reactor, and Indian Point-1, used horizontal tube bundles, a strategy that is maintained in Russian-designed VVER plants. The main advantage of this arrangement is that gravity does not cause sludge to accumulate on the tubesheet, and sludge piles do not occur at the top of the tubesheet as illustrated in Figure 11. However, the horizontal arrangement takes up more floor space and accordingly requires larger reactor buildings with higher costs. For this reason, all modern PWRs use vertically-oriented tube bundles. This has led to accelerated corrosion at crevices between sludge piles and tubes, as illustrated in Figures 21 and 22. Maintenance has been required to minimize the concentrating capacity of this kind of crevice using sludge lancing and chemical cleaning.

2.5 Tube Material

Austenitic stainless steel was used initially for tubing in several early PWRs, but was changed to Alloy 600 as the result of corrosion problems such as caustic and chloride-induced SCC.⁷⁸⁻⁷⁹ However, several other early plants, such as Yankee Rowe, Chooze-A, and Trino Vercellese, used stainless steel with relatively limited corrosion problems, partly due to low hot leg temperatures and attention to condenser integrity, thereby minimizing ingress of cooling water. Based on the decision by Naval Reactors to use Alloy 600, which is discussed in connection with Figure 92, and the problems with stainless steel, subsequent plants in the late 1960s and 1970s used Alloy 600 tubes in the mill-annealed condition

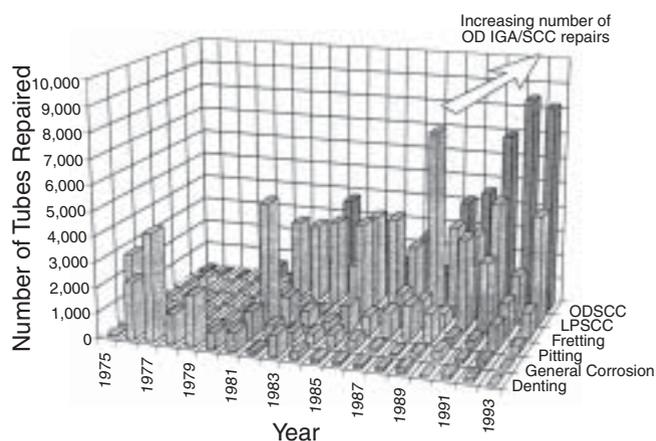


FIGURE 23. Number of tubes repaired vs time and mode of failure. From Varrin.⁷³ Used by permission of EPRI.

(600MA). Siemens decided to use Alloy 800, as discussed in this section and Section 1.3, following bad experience with Alloy 600MA in their lead plant. In the case of OTSGs, the stress relief, which was performed on the entire steam generator (usually called "global stress relief") during manufacture for 10 h at about 600°C, resulted in the tubes being sensitized, which depletes chromium at grain boundaries. On the other hand, residual stresses were relieved by this heat treatment and surface damage from cold work was reduced, which is discussed in connection with Figure 112.

Experience with Alloy 600 in the late 1960s and early 1970s in the early higher temperature large PWRs identified numerous corrosion problems, which are discussed in connection with Figures 21 and 23 in Section 1.7, Figure 92, and Table 17. The

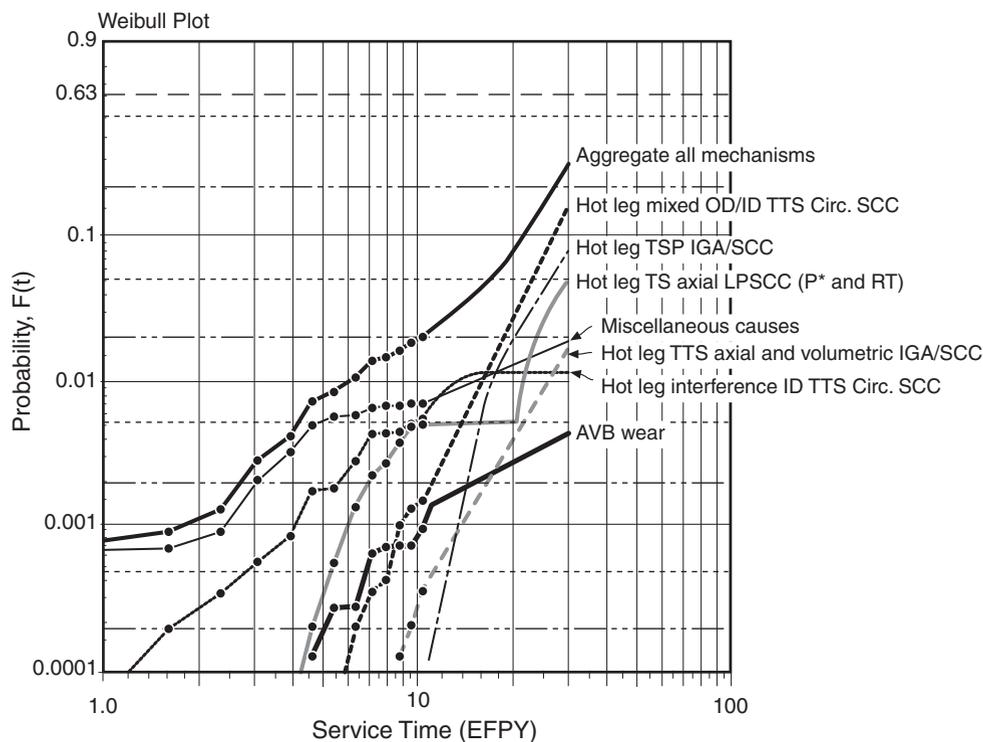


FIGURE 24. Probability of failure vs EFPY for Ringhals-4 PWR for various mode-location cases and the aggregate probability. Alloy 600MA tubing. From L. Björnkqvist and J. Gorman. Courtesy of L. Björnkqvist of Vattenfall and J. Gorman of Dominion Engineering, April, 1999. Private communication.

following problems of the types indicated in Figures 21 and 23 developed in rapid succession:

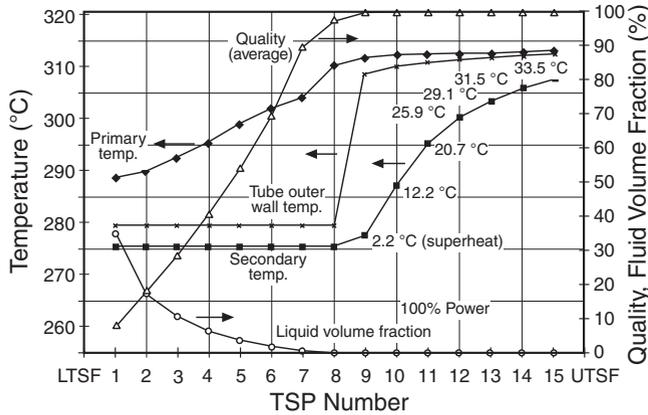
- IGA/SCC occurred in the sludge pile regions on top of the tubesheet in units operating on phosphates with high sodium-to-phosphate molar ratios;
- SCC occurred in the deep tubesheet crevice of a unit (Beznav 1) that operated on an all volatile treatment (AVT) type of water chemistry;
- general corrosion, “wastage,” occurred in several units that used phosphates with a low sodium-to-phosphate molar ratio; and
- rapid denting and LPSCC occurred in several salt water-cooled units with leaking condensers that had switched to an AVT water chemistry.

Research based on the finding of Blanchet, et al., reported in 1973,⁸⁰ as described by Debray and Stieding,⁸¹ showed that the corrosion resistance of Alloy 600 to both AkSCC and LPSCC was increased by a sensitizing heat treatment. This was modified to a “thermal treatment” (TT) of about 15 h at 704°C, the objective of which was to restore the depletion of chromium next to grain boundaries by bulk diffusion. This led to the widescale use of “thermally treated” Alloy 600 (600TT) for many units that started in the 1980s. While some utilities decided to rebuild not yet operational steam generators using Alloy 600TT, many others did not; and many Alloy

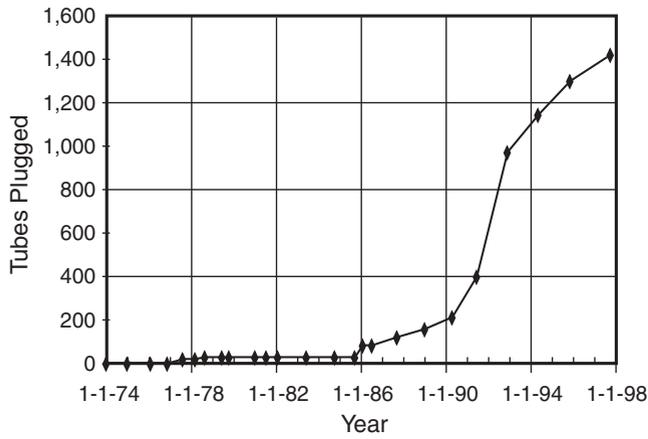
600MA steam generators also came on line in the 1980s.

Although Alloy 600TT was supposed to increase resistance to LPSCC, in fact, the improvements were not great, and Alloy 600TT has sustained significant LPSCC in some applications, especially at the kiss rolls on the primary side, as illustrated in Figure 4(c). Development of a new alloy was underway at the International Nickel Company,⁸²⁻⁸³ at least as early as 1972, and most likely before, according to the tests described in Section 5.3—the result was Alloy 690. Based on this work by alloy suppliers and builders of SGs, many new and replacement SGs, starting as early as 1989, as shown in Figure 92, incorporated Alloy 690TT tubes based on decisions made starting in about 1985.

Siemens took a different course in the choice of alloys for steam generator tubes than did the rest of the industry. Their initial PWR, Obrigheim, used Alloy 600MA. However, in 1967, while Obrigheim was still being constructed, Siemens decided that the risks of intercrystalline cracking were too high with Alloy 600; they switched to a modified or nuclear grade of Alloy 800 (800NG). This decision was based on results of corrosion tests published by Coriou, et al.,¹ as described by Debray and Stieding,⁸¹ which showed that Alloy 600 sustained SCC in high-temperature pure water and at high stress. Siemens placed test specimens of Alloys 600MA and 800NG in



(a)



(b)

SG	Total Plugged	Total Percent Plugged	Plugged for Free-Span IGSCC	Percent Plugged for Free-Span IGSCC
1A	451	2.90	39	0.25
1B	1,453	9.36	128	0.82
Total	1,904	6.13	167	0.54
2A	501	3.23	334	2.15
2B	676	4.35	363	2.34
Total	1,177	3.79	697	2.24
3A	643	4.16	192	1.24
3B	483	3.11	169	1.09
Total	1,126	3.63	361	1.16

(c)

FIGURE 25. (a) Steam generator conditions vs TSP elevation for Oconee steam generators manufactured by B&W. (b) Tubes plugged vs time for Oconee-1B. From Rochester and Eaker.⁷⁵ (c) Comparison of total plugged tubes with those plugged for SCC in the free-span. Figures 25(a) and (c) are courtesy of D. Rochester of Duke Energy, presented at Experts Panel on IGA/IGSCC in Oconee OTSG, July 28-29, 1998.

Obrigheim and found, several years after start-up, that Alloy 600MA sustained SCC while Alloy 800NG did not. Siemens continues to use Alloy 800NG but also uses Alloy 690TT, at the purchaser's option.

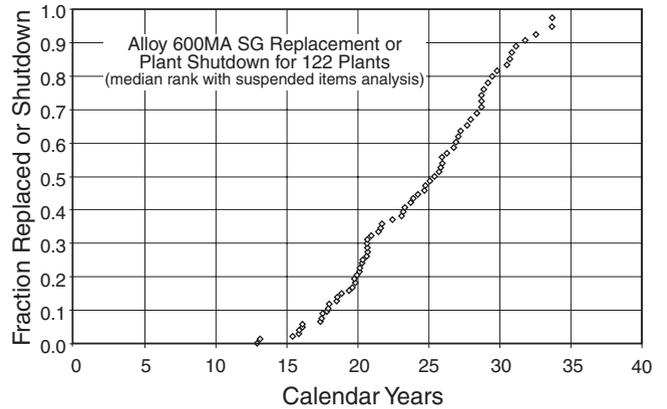


FIGURE 26. Fraction of replaced or shutdown steam generators vs calendar years for Alloy 600MA plants in the world. Data from Steam Generator Progress Report.⁷⁶

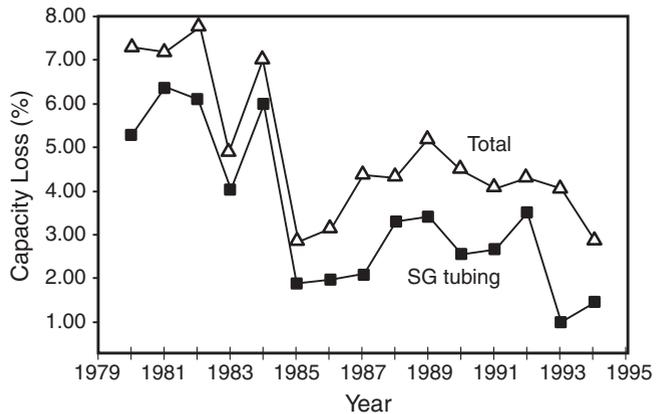


FIGURE 27. Capacity loss vs time for U.S. PWRs due to corrosion, showing the total loss and that only due to steam generator tubing. From Jones.⁷⁷ ©1996 NACE International.

2.6 Support Type and Materials

The purpose of this section is to describe how the alloy used for tube supports of the types described in Figures 3 and 5 affects the performance of the tubes. While one possible effect could involve galvanic corrosion with carbon steel lowering the potential into the LPSCC range for Alloy 600, such an effect has not been observed. The major effect of tube support materials was associated with the rapid corrosion of carbon steel in the presence of concentrated and corrosive chemicals such as Cl⁻ and Cu²⁺ in the heat-transfer crevice. The resulting corrosion products fill the tube-to-tube support gap and exert high stresses on the tubes, causing denting, as described in connection with Figure 14.

Many early steam generators used TSPs fabricated from carbon steel with drilled holes (Figures 3 and 5). After the change from phosphate water chemistry in the mid-1970s described in Sections 3.1.1, 3.4, and 3.5, these early designs operated with an AVT water chemistry that, in contrast to phosphate

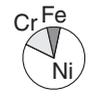
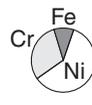
	Model D3, Virgil Summer	Model F
Tube material	Alloy 600MA 	Alloy 600TT or Alloy 690TT 
Heat treatment	Low-temperature mill anneal (LTMA) 	Thermal treatment (TT) 
Tube support hole	Drilled hole, circular 	Quatrefoil 
Tube support material	Carbon steel 	Stainless steel 
Flow holes	Yes 	None 
U-bend stress relief	None 	Yes 
Tube sheet expansion	Mechanical expansion 	Hydraulic expansion 
Blow down	Original design 	Better location, increased capacity 
Secondary-side access	Original design 	Improved 

FIGURE 28. Comparison of Models D3 and F from Westinghouse design plants.

water chemistry, does not provide buffering and therefore allows local chemistries to change at the heat-transfer crevices, shown in Figure 10, to relatively acidic or alkaline pH. As a result, the denting phenomenon developed, as shown in Figure 14, especially in plants cooled with salt water. The denting was attributed to concentration of chlorides and oxidants, such as copper, in the crevices, leading to rapid corrosion of the carbon steel TSP. The Pilling Bedworth ratio⁶³ indicated that the specific volume of the oxide is much greater than the metal in the TSP with the result that the larger resulting volume due to corrosion produced deformation and cracking of the tubes (Figure 14). Designs with carbon steel egg crates, as shown in Figure 3, were found to be less susceptible, but not immune, to this rapid corrosion. In response to the denting problem, alternate alloys were tested for use as support materials and, in the United States, ferritic stainless steels (Types 405 and 409 [UNS S40500 and S40900]) were selected. In addition, broached hole designs were developed to replace drilled hole designs. Designs with stainless steel lattice bars, egg crates, and broached holes, shown in Figures 3 and 5, have not sustained denting.

In addition to corrosion of the carbon steel tube support materials, corrosion of the tubes occurs at tube support locations. Corrosion of tubes at TSPs is

most severe with the drilled hole design, but also has occurred in Alloy 600MA tubes at egg crates (e.g., at St. Lucie 1).⁸⁴ One plant with Alloy 600MA tubes in carbon steel broached hole TSPs has sustained corrosion based on examinations by eddy-current testing (ECT) and pulled tubes. This result shows that impurities can concentrate at broached holes, owing to the buildup of deposits (Figure 13). Only one case of confirmed IGA/SCC has been found in Alloy 600TT tubes in broached holes or at egg crates,⁸⁵ although a few ECT indications of possible corrosion also have been observed at several other plants. No indications of corrosion have been reported for Alloy 690TT tubes at tube supports (all of which are stainless steel lattice bars or broached holes), or at any other location.

The Siemens design, using Alloy 800NG tubes and stainless steel lattice bars, has been free from denting and IGA/SCC at support locations.

2.7 Tubesheet Expansion

Some early RSG designs used part-depth rolls (PDR) to fasten tubes to the tubesheet. This left a long crevice, about 18 in. (46 cm) long with a 0.008-in. (0.2-mm) radial gap, at the top of the tubesheet (Figure 4). No problems were experienced in this gap in units operating with phosphate water chemistry, which tends to buffer impurities that develop in crevice areas. However, a unit that started up on AVT in

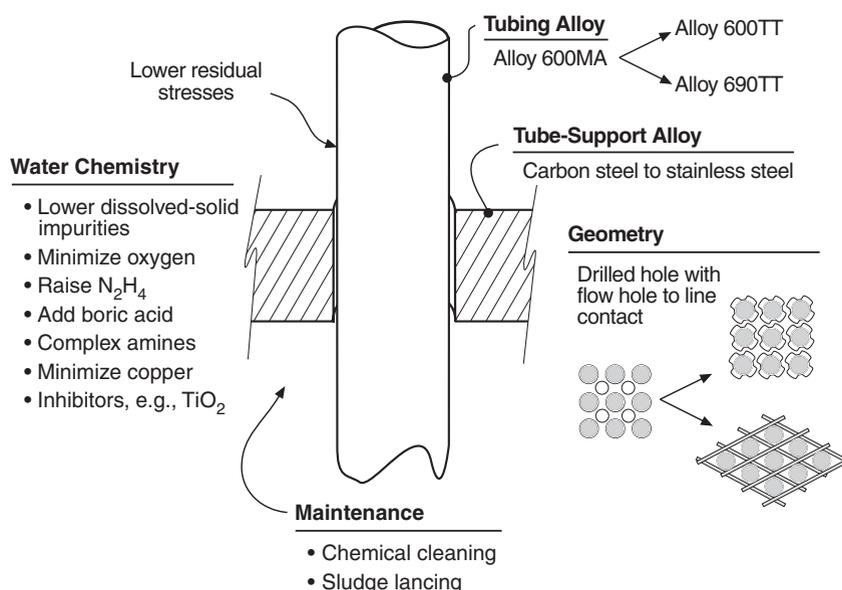


FIGURE 29. Schematic view of changes in maintenance, designs, and water.

the early 1970s (Beznau-1) quickly sustained severe IGA/SCC of Alloy 600MA tubes in the part-depth-rolled crevices; other units that switched to AVT or started up on AVT in the mid to late 1970s also developed severe IGA/SCC in the long crevice. In response to these problems with the part-depth rolling, units that were not yet operational were modified to close the gap using either explosive expansions or full-depth rolls (FDR). Since about 1980, all recirculating SGs have used full-depth expansion, as shown in Figure 4, or have sealed the gap at the top of the tubesheet using an expansion at that location.

CE and Siemens have not used PDRs in their designs. CE has used full-depth explosive (FDE) expansion for all units. Siemens has used multiple hard rolls (e.g., at the bottom, middle, and top of the tubesheet or at the bottom and top) to fully seal the crevice against entry of secondary coolant. Later, Siemens' designs have used hydraulic expansion with supplementary hard rolls at the top and bottom. These designs have been free from OD tube corrosion occurring deep in the crevice.⁸⁶

Use of expansion methods that seal the tube to the tubesheet close to the top of the tubesheet (TTS) have been successful in preventing OD corrosion of tubes deep in the tubesheet. However, it has not been practical to eliminate small crevices completely at the top of the tubesheet. These residual crevices typically have depths of 3 mm to 6 mm, as shown in Figure 4.

Significant corrosion problems have been sustained in Alloy 600MA tubes at these TTS residual crevices, especially in preheater SGs. The crevice conditions at these residual crevices appear to be aggravated by the development of deposit collars, as illustrated in Figure 22, at the tube-to-tubesheet

junction, which enhance the concentration of impurities in crevices.

No OD corrosion problems have been detected at the TTS at units with Alloy 600TT tubes that have not sustained denting at the top of the tubesheet.⁸⁶ Some ODS/SCC was observed at Kori-2 associated with dents at the TTS, as shown in Figure 15, caused by a combination of seawater leaks and the presence of iron shot on the TTS. No OD corrosion has been reported at the TTS, or elsewhere, in units with Alloy 690TT tubes nor, except for one tube, in units with Alloy 800NG tubes.^{76,87}

All operating OTSGs have PDRs at both the top and bottom tubesheets. Few OD corrosion problems have occurred in crevices at the top hot leg tubesheet. Corrosion in the lower cold leg tubesheet crevices also has been limited, apparently because temperatures and superheats are low, thus reducing the severity of corrosion. The fact that the tubes are sensitized and stress-relieved, as well as in axial compression during operation, also may increase their resistance to corrosion relative to Alloy 600MA tubes. It should be noted that the axial compression does not overcome some residual stresses nor does it reduce hoop stress due to internal pressure.

2.8 Flow Distribution Baffles

Early experience has shown that sludge piles tended to develop on the tubesheet in mid-bundle regions where cross flow was low. Experience also has shown that corrosion was accelerated in these sludge piles: IGA/SCC occurred in tubes of Alloy 600MA in units with either high molar ratio phosphate water chemistry or with AVT water chemistry; wastage occurred in both Alloy 600MA tubes and Alloy 800NG tubes in units operating with low molar

TABLE 3
PWR Steam Generator Design Evolution

Model/Lead Plant (First Commercial Operation)	Typical Design T_H (°C)	Avg. Heat Flux for Whole SG (Btu/h/ft ²)	Recirculating or Once-Through	Tube Orientation	Tube Material ^(A)	Support Type	Support Material	Tube Sheet Expansion ^(B)	Flow Distribution Baffle (Y/N)	Preheater (Y/N)
Government Laboratory Designs										
Shippingport (1957)	283	29,000	Recirculating	Horizontal	SS then 600MA	Hour-glass drilled holes	Carbon steel	PDR	—	N
N Reactor (1965)	279	67,000	Recirculating	Horizontal	SS then 600MA	Drilled holes	Carbon steel	PDR	—	N
Westinghouse and Licensee Designs										
Early SS designs (1961)	288 to 302	39,000	Recirculating	Vertical	Type 304 SS	Drilled holes	Carbon steel	PDR	N	N
Early 600MA designs (1968)	310 to 318	43,000	Recirculating	Vertical	600MA	Drilled holes	Carbon steel	PDR	N	N
Intermediate 600MA designs (1977)	317 to 322	44,000	Recirculating	Vertical	600MA	Drilled holes	Carbon steel	FDE, FDR, or FDR-KR	N	N
Later 600MA designs (1981)	323 to 324	48,000	Recirculating	Vertical	600MA	Drilled holes	Carbon steel	FDR or FDR-KR ^(C)	Y	N
Preheater 600MA designs (1983)	326 to 330	49,000	Recirculating	Vertical	600MA	Drilled holes	Carbon steel	FDR or FDR-KR	Y	Y
Model F Type (1983)	326	53,000	Recirculating	Vertical	600TT	Broached holes	Type 405 SS	FDH or FDR-KR ^(C)	Y	N
Preheater 600TT designs (1986)	326	49,000	Recirculating	Vertical	600TT	Broached holes	Type 405 SS	FDH or FDR-KR	Y	Y
Latest replacement designs (1988)	See note ^(D)	44,000	Recirculating	Vertical	690TT	Broached holes	Type 405 SS	FDH or FDR-KR ^(C)	Y	Y
Combustion Engineering and Licensee Designs										
First design (1971)	315	50,000	Recirculating	Vertical	600MA	Drilled holes	Carbon steel	FDE	N	N
Intermediate designs (1973)	313	51,000	Recirculating	Vertical	600MA	Egg crates and drilled holes	Carbon steel	FDE	N	N
Later designs (1983)	321	56,000	Recirculating	Vertical	600MA	Egg crates	Carbon steel	FDE	N	N
Economizer model (1986)	327	52,000	Recirculating	Vertical	600MA	Egg crates	Type 409 SS	FDE	Y	Y
Latest Korean model	322	47,000	Recirculating	Vertical	690TT	Egg crates	Type 409 SS	FDE	Y	Y

Continued

TABLE 3 (Continued)
PWR Steam Generator Design Evolution

Model/Lead Plant (First Commercial Operation)	Typical Design T_H (°C)	Avg. Heat Flux for Whole SG (Btu/h/ft ²)	Recirculating or Once-Through	Tube Orientation	Tube Material ^(A)	Support Type	Support Material	Tube Sheet Expansion ^(B)	Flow Distribution Baffle (Y/N)	Preheater (Y/N)
Babcock & Wilcox Designs										
Prototype plant design (1963)	271	—	Recirculating	Horizontal	Type 304 SS	—	—	—	—	N
Once-through design (1973 and later)	317 to 320	33,000	Once-through	Vertical	600MA + sensitized	Broached holes	Carbon steel	PDR	N	N
Replacement RSGs (1992 and later)	See note ^(D)	50,000	Recirculating	Vertical	690TT	Lattice bar	Type 410 SS	FDH	Y	N
Siemens										
Lead plant design (1969)	312	61,000	Recirculating	Vertical	600MA	Lattice bar	SS	Rolled at top, middle, and bottom	N	N
Second-generation design (1972)	314 to 319	68,000	Recirculating	Vertical	800NG	Lattice bar	SS	Rolled at top and bottom	N	N
Preheater design (~1983)	329	57,000	Recirculating	Vertical	800NG	Lattice bar	SS	Rolled at top and bottom	Y	Y
Replacement design (1989)	See note ^(D)	51,000	Recirculating	Vertical	800NG or 690TT	Lattice bar	SS	FDH with roll at top and bottom	—	N

^(A) MA = mill annealed, TT = thermally treated, NG = nuclear grade.

^(B) PDR = part-depth roll, FDR = full-depth roll, KR = kiss roll, FDE = full-depth explosive, FDH = full-depth hydraulic.

^(C) Variety of full-depth tubesheet expansion designs used in Japan.

^(D) T_H of replacement SGs is usually lower than that of original SGs.

ratio phosphate water chemistry. To minimize corrosion associated with these sludge piles, flow distribution baffles have been used in most units that started operating in the early 1980s or later. A flow distribution baffle is a plate that is installed relatively close to the top of the tubesheet, which directs flow coming from the downcomer across the tubesheet. The use of flow distribution baffles reduces the depth and extent of sludge piles but does not eliminate them.

2.9 Preheater

PWR suppliers determined that the thermal efficiency of the plants could be improved by making a portion of the steam generator act as a preheater, which is also known as an “economizer.” With this design, the feedwater is directed to the cold leg where it is heated by primary coolant inside the tubes until it reaches the saturation temperature, at which time it is introduced into the full tube bundle. This is in contrast to a feeding unit, where feedwater is intro-

duced into the downcomer above the tube bundle and where it mixes with recirculating water coming from the moisture separators.

Corrosion in Westinghouse preheater units has been significantly worse than in feeding units with similar hot leg temperatures. This accelerated corrosion has occurred especially at the TTS, but also at TSPs. This difference is attributed to the lower subcooling, and therefore greater available superheat in preheater units, which leads to more severe conditions in crevices at the TTS and at supports. In a feeding unit, the water in the downcomer is significantly subcooled (e.g., 22°C) since it is a mixture of cold feedwater and recirculating water. In contrast, in a preheater unit, the water in the downcomer has only limited subcooling (e.g., <3°C), due to the static pressure head and a small fraction of the feedwater that is introduced into the top of the steam generator (called the bypass flow, often about 10% of the feedwater flow).

No confirmed OD corrosion problems have been reported in preheater units with Alloys 600TT, 690TT, or 800NG tubes. However, because of the poor experience with preheater units with Alloy 600MA tubes, all replacement SGs have been of the feedring type, even where the original plant design used preheater SGs. Larger numbers of smaller diameter tubes have been used to increase the heat-transfer area of the replacement SGs, thereby compensating for the lower efficiency of the feedring design.

2.10 Significance

2.10.1 Experience — The main experiences from the design and operation of steam generators are the following:

1. The increased rates of corrosion that have occurred in steam generators with Alloy 600MA tubes, as higher operating temperatures and heat fluxes were used, have shown that such increases in temperature and heat flux should be approached cautiously. They should only be implemented together with suitable selection of design features and materials that are demonstrated to prevent significant corrosion at these temperatures and heat fluxes for the design lifetime.
2. Heat-transfer crevices in steam generators are locations where impurities concentrate and cause corrosion of tubes and structural materials. To minimize these problems, designs should eliminate crevices to the extent practical. Crevices that cannot be eliminated should be designed so as to minimize their concentrating power, e.g., by keeping them as shallow as practical (e.g., for TTS crevices) and by using line contact arrangements, rather than surface to surface contact (e.g., for tube supports).
3. The designs and materials chosen for the steam generator and for the whole plant, as well as the choice of water chemistry, should be directed toward minimizing the accumulation of deposits in the steam generator. Experience demonstrates that deposits cause crevices that concentrate impurities and accelerate corrosion. Typical design features directed toward eliminating deposits have included:
 - eliminating copper alloys from the secondary system so that high-pH AVT water chemistries can be used that minimize iron ingress into the steam generator, as well as minimizing copper ingress
 - using flow distribution baffles to minimize sludge accumulation on the tubesheet
 - selecting design features that produce high circulation ratios in the steam gen-

erator so that flow velocities are adequate to minimize deposit formation

- selecting design features that facilitate removal of deposits by sludge lancing and chemical cleaning (e.g., provision for appropriate service ports for sludge lancing at the tubesheet and at support elevations and selecting materials resistant to corrosion in likely chemical cleaning solutions)
 - using better blowdown design
4. Choosing a tubing alloy and its heat treatment should be based on demonstrated corrosion resistance in any of a credible range of crevice environments for the expected design life time (e.g., 60 years). Based on available test data and service experience, the most suitable choices for PWR steam generators are 690TT and 800NG.
 5. Choosing alloys for tube supports should assure that undesirable corrosion will not occur in crevices over the design lifetime. Based on available test data and service experience, any of the normal stainless steels or corrosion-resistant nickel-based alloys are suitable so long as similar materials are not used as the wear couple (e.g., Types 304 [UNS S30400], 316 [UNS S31600], 405, 409, or 410 [UNS S41000] stainless steel and Alloys 800 and 600).
 6. Use of preheaters should be approached cautiously since the lower subcooling in these designs at the hot leg TTS and lower hot leg support locations increase tendencies for corrosion. However, the successful use of preheaters in Siemens units with Alloy 800NG tubes for about 15 years or more indicates that preheaters may operate satisfactorily for long times if suitable tube materials, support materials, and design features are chosen that minimize crevices and deposit accumulation. Preheaters also have been used successfully in CANDU plants.

2.10.2 Potential Problems and Approaches —

1. The location where egg crates form acute angles, as shown in Figure 13, should be studied carefully as a possible geometry where deposits can accumulate and can produce conditions that can concentrate aggressive chemistries.
2. Several of the corrosion processes, as described in Section 5.0, did not occur immediately, and some time (10 to 20 years is not unusual) elapsed before significant corrosion occurred. The same pattern is likely to apply to the use of Alloy 690TT, especially because it is an inherently reactive material if its passivity is breached.

3. The crevice at the top of the tubesheet persists for all tubes. In addition, the occurrence of denting at this location should be considered in view of the tubesheet being low-alloy steel. Since this crevice can become packed and is a location of the highest temperature on the primary side as well as a location of high stress, possible corrosion of the tubes and tubesheet should be considered and supporting experimental work should be carried out.

3.0 SECONDARY ENVIRONMENTS

This section describes how control of secondary environments in PWR steam generators has evolved over time and how the bulk secondary environments affect the development and chemical conditions of heat-transfer crevices. This section emphasizes the bulk water chemistry while Section 4.0 describes chemistry inside the heat-transfer crevices. Most of the changes in bulk water chemistry have been required to ameliorate corrosion, shown in Figures 21 and 22, which is associated with local chemistry within heat-transfer crevices.

While minimizing corrosion damage to tubing on the secondary side has been a main objective of controlling water chemistry of the secondary side of PWRs, minimizing corrosion damages to other equipment in the secondary system, such as turbines, condensers, feedwater heaters, moisture separators, and piping, also have been important. Water chemistry limits that have been developed to protect steam generators generally have resulted in satisfactory limits for turbines. However, in some cases, limits on water chemistry relative to steam generators have resulted in less than optimum conditions for secondary system heat exchangers with copper alloy tubes and steel piping components; accordingly, some compromises have been required. The main problems and compromises are the following:

1. A relatively high pH_T in the secondary system is desirable to protect the steam generator from corrosion and to minimize iron ingress into the steam generator from the secondary system. Corrosion rates of copper heat exchanger tubing increase as the pH_{RT} is increased above about 8.8, while corrosion rates of carbon steel piping and other components continue to decrease as pH_{RT} is raised, at least to a pH_{RT} of 10. Thus, if copper alloys are used in the secondary system, pH_{RT} has to be selected on a compromise basis at about 9.2 so as to not increase too much the corrosion of either the copper alloys or the carbon steel. Of course, secondary circuits without copper can and have exploited the advantage of a higher pH in terms of reducing the amount of iron-based sludge reaching the steam generator.

2. Protection of the steam generators against corrosion requires that oxidizing conditions be avoided. Achieving this goal is facilitated by maintaining low oxygen concentrations in the feedwater and by using concentrations of hydrazine adequate to scavenge any dissolved oxygen. However, low oxygen and high hydrazine can increase rates of flow-accelerated corrosion (FAC) of carbon steel piping and steam generator internals. For this reason, hydrazine concentrations are selected considering both of these goals (i.e., high enough to ensure protection of the steam generators while not so high as to significantly aggravate tendencies toward FAC).

3.1 General Approaches

Corrosion in steam generators is generally minimized by maintaining low oxygen, low impurities, and moderately alkaline pH. Two general approaches to water chemistry in steam generators have been used, and these have been directed primarily toward minimizing corrosion in heat-transfer crevices.

3.1.1 Phosphates — Additions of phosphates was the first approach to treating water on the secondary side of steam generators. This approach was based on the many years of successful experience with treating water in fossil boilers. The addition of phosphates was aimed at inhibiting the general corrosion of steel, buffering local solutions to prevent pitting, and producing a sludge that could be readily removed. This choice, which may have seemed reasonable at the time, was based on designs of fossil boilers where heat-transfer crevices were not present as they are in PWR steam generators, as shown in Figures 3 and 11 through 15. Further, there was no geometry in fossil boilers similar to the top of the tubesheet where heat transfer can lead to sludge piles (Figure 2[b]). The evolution and later rejection of phosphate treatments is a commentary on the utilization of a water treatment that worked well in one application with one set of geometries but did not work well in another. Further, based on early work with locomotive steam boilers, the buildup of deposits in heat-transfer crevices was knowingly inevitable. The evolution of localized general corrosion related to phosphate treatment is described in two references.⁴⁹⁻⁵⁰

This section describes, first, some of the important chemical properties of aqueous phosphate solutions. Next, experiences with general corrosion associated with the application of phosphate water chemistry to PWR steam generators are discussed. Finally, interpretations of corrosion due to the phosphate water chemistry are discussed.

Important properties of aqueous phosphate solutions are shown in Figure 30. Figure 30(a) shows the potential-pH diagram for phosphorous and water at 25°C. This figure shows the bases for molar ratios of

phosphates starting with the alkaline end: trisodium phosphate with $\text{Na}^+/\text{PO}_4^{3-} = 3$; disodium phosphate with $\text{Na}^+/\text{PO}_4^{2-} = 2$; monosodium phosphate with $\text{Na}^+/\text{PO}_4^- = 1$; phosphoric acid with H_3PO_4 . These ratios have been used extensively to describe the boundaries of phosphate-containing additives and are used in this discussion.

The fact that phosphate environments exhibit special properties of solubility with temperature was noted by Schroeder, et al.,⁸⁹ as shown in Figure 30(b). Here, they show that the solubility of Na_3PO_4 first increases with increasing temperature and then decreases as the temperature increases further. Such behavior is the basis for retrograde solubility of phosphates and also suggests that substantial attention should be given to the existence and properties of various phases of phosphates.

Similar to the trend in Figure 30(b), Figure 30(c) from Panson, et al.,⁹³ shows that the solubility of phosphate solutions decreases with increasing temperature as well as with increasing molar ratio. Figure 30(d) from Economy, et al.,⁹⁵ shows that solubility decreases rapidly as a molar ratio of 3.0 is approached at 300°C. Further, Figure 30(e), where the solubility of disodium phosphate is presented as a binary phase diagram including hydrated phases, shows a similar pattern in the occurrence of a retrograde reaction at about 220°C. Thus, the generally retrograde solubility occurs over a broad range of Na/PO_4 ratios, and decreasing Na/PO_4 ratios exhibit increasing solubility.

Figures 30(c) and (d) suggest that the nature of the solubility is changing with molar ratio, and this is also important in the discussion of Figure 31(b). This condition has been investigated by Marshall,¹⁰¹ as shown in Figure 30(f). Here, evidence is shown for two liquid phases with a miscibility gap. The dotted line for the molar ratio of 2.0 shows that there is a liquid with about 12% dissolved solid and another with about 59% dissolved solid. At molar ratios above about 2.16, the miscibility gap phenomenon ceases. This observation by Marshall provides important insights into conditions that produce corrosion as discussed in connection with Figures 31 and 32.

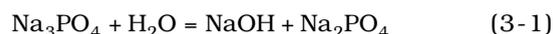
Phosphate chemistry has involved two variations: "Coordinated Phosphate Water Treatment"¹⁰²⁻¹⁰³ and "Congruent Phosphate Treatment."¹⁰²⁻¹⁰³ The bases for these treatments are shown in Figures 31(a) and (b).

Coordinated phosphate water treatment is a standard water chemistry for fossil-fired boilers with steam pressures in the same range as PWR steam generators and was used initially for most early PWRs. Coordinated phosphate water treatment involves using two or more types of phosphate, such as disodium and trisodium phosphate, combined with an oxygen scavenger such as sodium sulfite or hydrazine. The objective is to maintain a high-pH and

low-oxygen environment in which the corrosion rates of carbon steel are low.

The term "coordinated" means that the pH of the boiler water is kept high enough to minimize corrosion, but still is controlled by additions of phosphate chemicals at concentrations such that free caustic is not produced, the criterion for which is shown in Figure 32(a). This type of water chemistry works well in conventional boilers with boiling on the inner surfaces of the tubes and with mud drums for the collection of sludge, since there are no crevices. The phosphate not only keeps the pH in the desired range but retards the formation of hard scale by combining with elements such as calcium and magnesium to form precipitates that are easily removed.

Trisodium phosphate and water react as in Equation (3-1):



Coordinated phosphate control maintains the pH below the value produced by Na_3PO_4 ($\text{Na}/\text{PO}_4 = 3$) at the measured phosphate concentration. The basis for this treatment was that any pH higher than this value is due to some other source (free caustic). If the molar ratio of sodium to phosphate is too low, then corrosive concentrated phosphate solutions, which may be acidic,⁴⁹ can develop. The formation of locally concentrated alkaline and acidic environments have occurred, in fact, in PWRs, and are related to the rapid general corrosion, as well as SCC, shown in Figures 21 and 23.

Congruent phosphate treatment is based on Figure 31(b) from Economy, et al.,⁹⁵ Here, a congruent composition for phosphate precipitation occurs at approximately a Na/PO_4 ratio of 2.8. This is the typical ratio of phosphate that precipitates from a phosphate-containing solution in a heat-transfer crevice. If the original solution has a ratio of Na/PO_4 higher than this congruent composition, solid with a ratio of 2.8 will precipitate, and the solution will be enriched in Na. Such a continued process would produce a concentrated alkaline solution with the inevitable general corrosion, AkSCC, and AkIGC. However, if the molar ratio of Na/PO_4 is less than 2.8 in solution, the precipitate will contain two solid solutions, I and II, as shown in Figure 31(b). Solid Solution I has an invariant composition of about 2.13 and that of Solid Solution II is 2.8. As the concentration of the solution is decreased, the amount of Solid Solution I is increased. When the composition of the solution is decreased below the concentration of Solid Solution I, the composition of the solution and that of the solid solution are congruent. Thus, to avoid both acidic and alkaline conditions, the Na/PO_4 molar ratio should be maintained between 2.8 and 2.13.

Localized general corrosion, unfortunately colloquially called "wastage" in the industry, associated

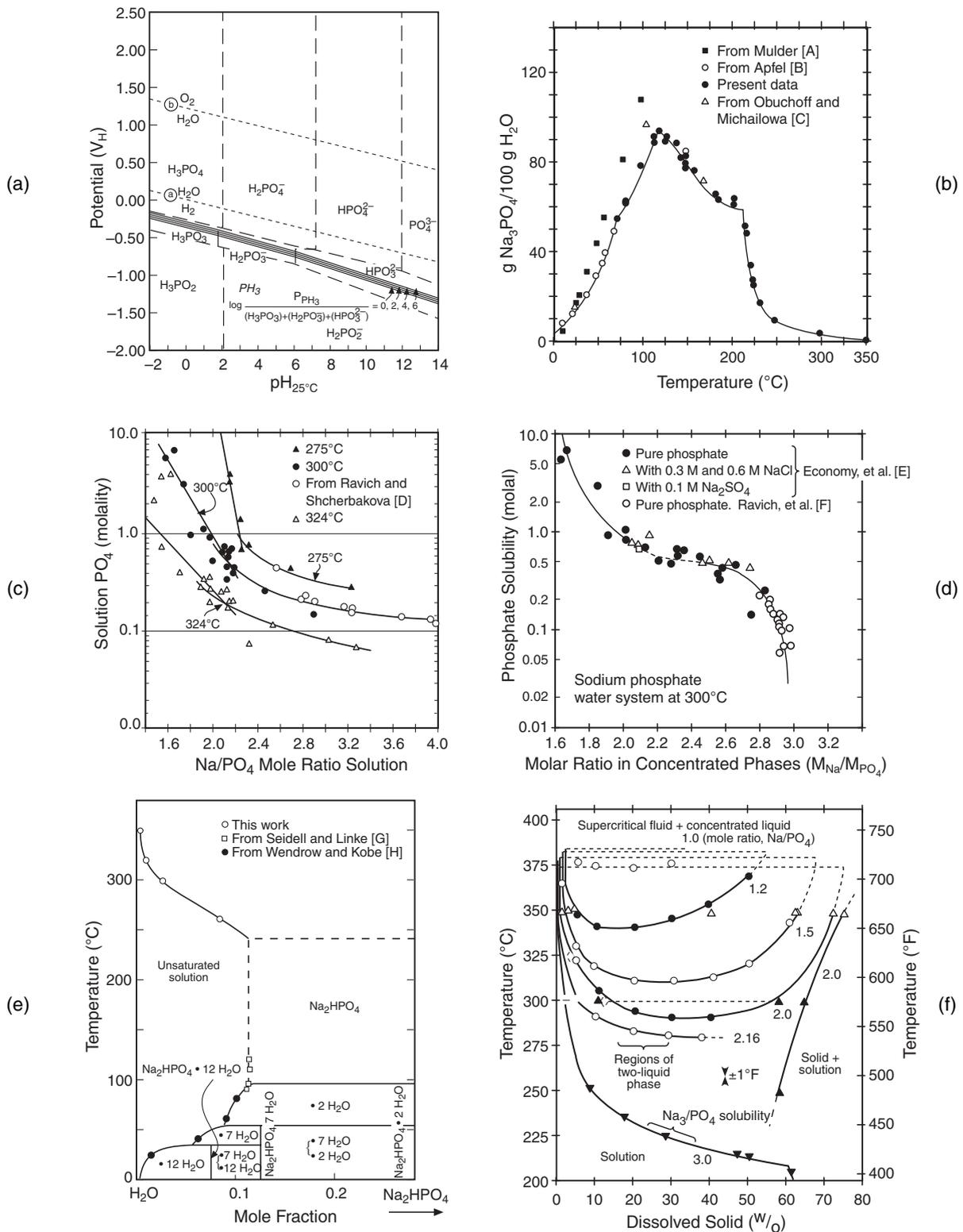


FIGURE 30. (a) Potential vs pH for the P-H₂O system at 25°C. From Pourbaix.⁸⁸ Courtesy CEBELCOR, Brussels. (b) Solubility of Na₃PO₄ in water vs temperature. From Schroeder, et al.,⁸⁹ including [A] Mulder,⁹⁰ [B] Apfel,⁹¹ and [C] Obuchoff and Michailowa.⁹² Reprinted in part with permission from American Chemical Society. (c) Solubility of PO₄ vs Na/PO₄ mole ratio in solution for three temperatures. From Panson, et al.,⁹³ including [D] Ravich and Shcherbakova.⁹⁴ Courtesy Electrochemical Society. (d) Phosphate solubility vs molar ratio in concentrated phases for sodium-phosphate water system at 300°C. From Economy, et al.⁹⁵ Courtesy Electrochemical Society. Including [E] Economy, et al.⁹⁵ [F] Ravich, et al.,⁹⁶⁻⁹⁸ (e) Temperature vs mole fraction of Na₂HPO₄ in water showing phase fields. From Panson, et al.,⁹³ including data from [G] Seidell and Linke,⁹⁹ and [H] Wendrow and Kobe.¹⁰⁰ Courtesy Electrochemical Society. (f) Temperature vs dissolved weight for ratios of Na/PO₄ from 1.0 to 3.0 and temperatures from 200°C to 400°C. From Marshall.¹⁰¹

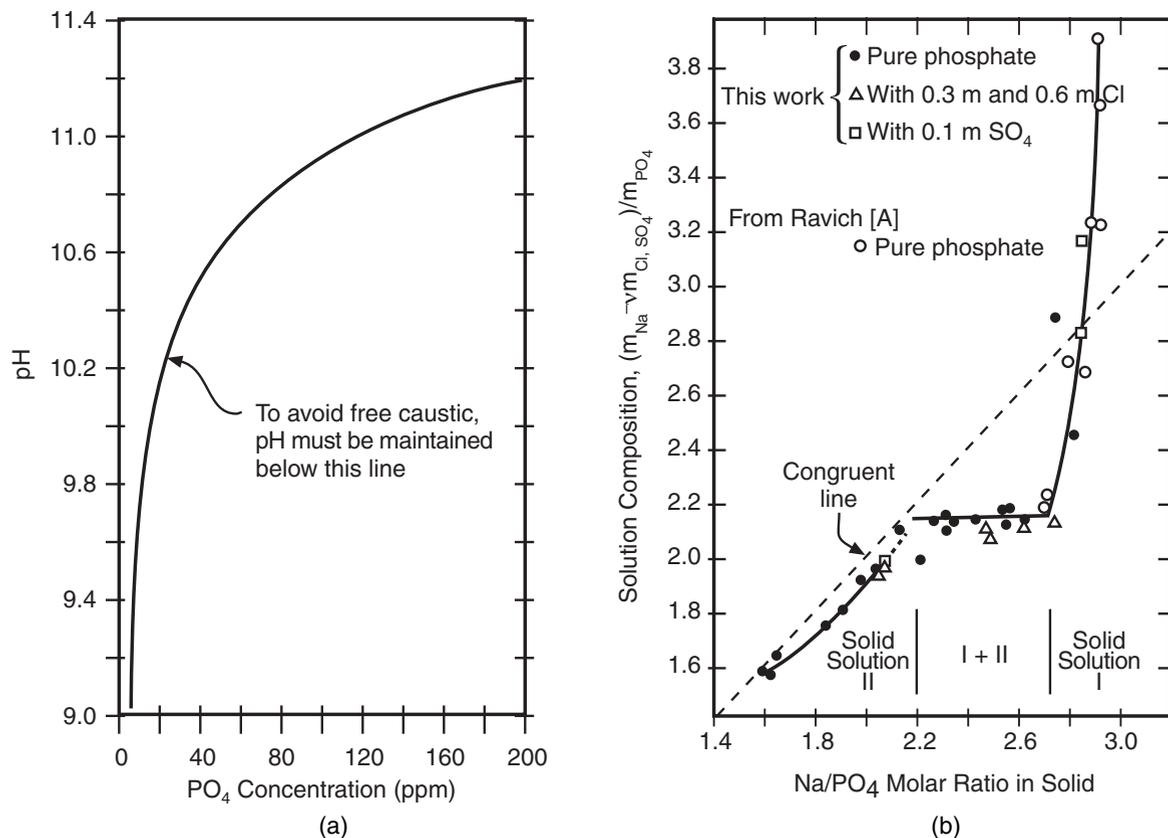


FIGURE 31. Bases for coordinated and congruent phosphate water chemistry. (a) pH value vs concentration of trisodium phosphate for avoiding free caustic. From Whirl and Purcell.¹⁰⁴ The information presented was part of a paper presented at the 3rd Annual International Water Conference, which took place in Pittsburgh, Pennsylvania, 1942, and Marcy and Halstead.¹⁰⁵ (b) Composition of solution vs. Na/PO₄ as molar ratio in the solid. From Economy, et al.,⁹⁵ including data from [A] Ravich referenced in Economy, et al.⁹⁵ The information presented was part of the 1975 International Water Conference.

with the use of phosphate water chemistry, was first observed in about 1972 after the molar ratio of Na/PO₄ was reduced to a molar ratio of about 2.0 from the previous ratio of about 2.8 or above. The higher ratio had produced AkIGC and AkSCC; lowering the Na/PO₄ ratio was intended to obviate such corrosion. However, about a year after lowering this ratio, general corrosion associated with phosphates began to occur in the hot leg sludge pile in Westinghouse plants and in the vicinity of anti-vibration bars of CE units.⁴⁹⁻⁵⁰ To prevent the general corrosion that was presumed to be associated with the molar ratio of 2.0, the molar ratio was increased to a tight range from 2.3 to 2.6 with the phosphate concentration in the range from 10 ppm to 80 ppm. However, this did not slow the general corrosion significantly. Corrosion rates as high as 0.005 in./y were observed. Figure 32(a) illustrates the extent of corrosion in a tube that was pulled after sustaining wastage. Here, the depth of penetration is shown relative to distance from the top of the tubesheet.

Despite raising the Na/PO₄ ratio to the 2.3 to 2.6 range, it became clear that there was something inherent in the phosphate chemistry that could not be

controlled in the heat-transfer crevices; and the Westinghouse and CE design SGs switched to AVT water chemistry by 1975. At the same time, the KWU SGs retained the phosphate water chemistry with their Alloy 800 tubes; but the phosphate was in the range of 2 ppm to 3 ppm with a Na/PO₄ ratio of 1.8 to 2.0. This procedure continued, and only moderate amounts of phosphate-related corrosion were observed in KWU SG units.

Early explanations for the general corrosion related to phosphate, which somehow produced an acidic environment resulting from the precipitation of phosphate phases. However, from the work shown in Figures 30 and 31, it appears that it is not reasonable physically that an acidic solution (e.g., H₃PO₄) could form. Rather, it appears that corrosion due to phosphates in heat-transfer crevices is related to more complex aspects of the precipitation chemistry of the phosphate compounds.

The work of Pessell, et al.,¹⁰⁷ showed that corrosion in various concentrations and molar ratios of Na/PO₄ solutions exhibited a parabolic behavior (Figure 32(b)) with a parabolic dependence and rate constant A, as shown in Equations (3-2) and (3-3). These

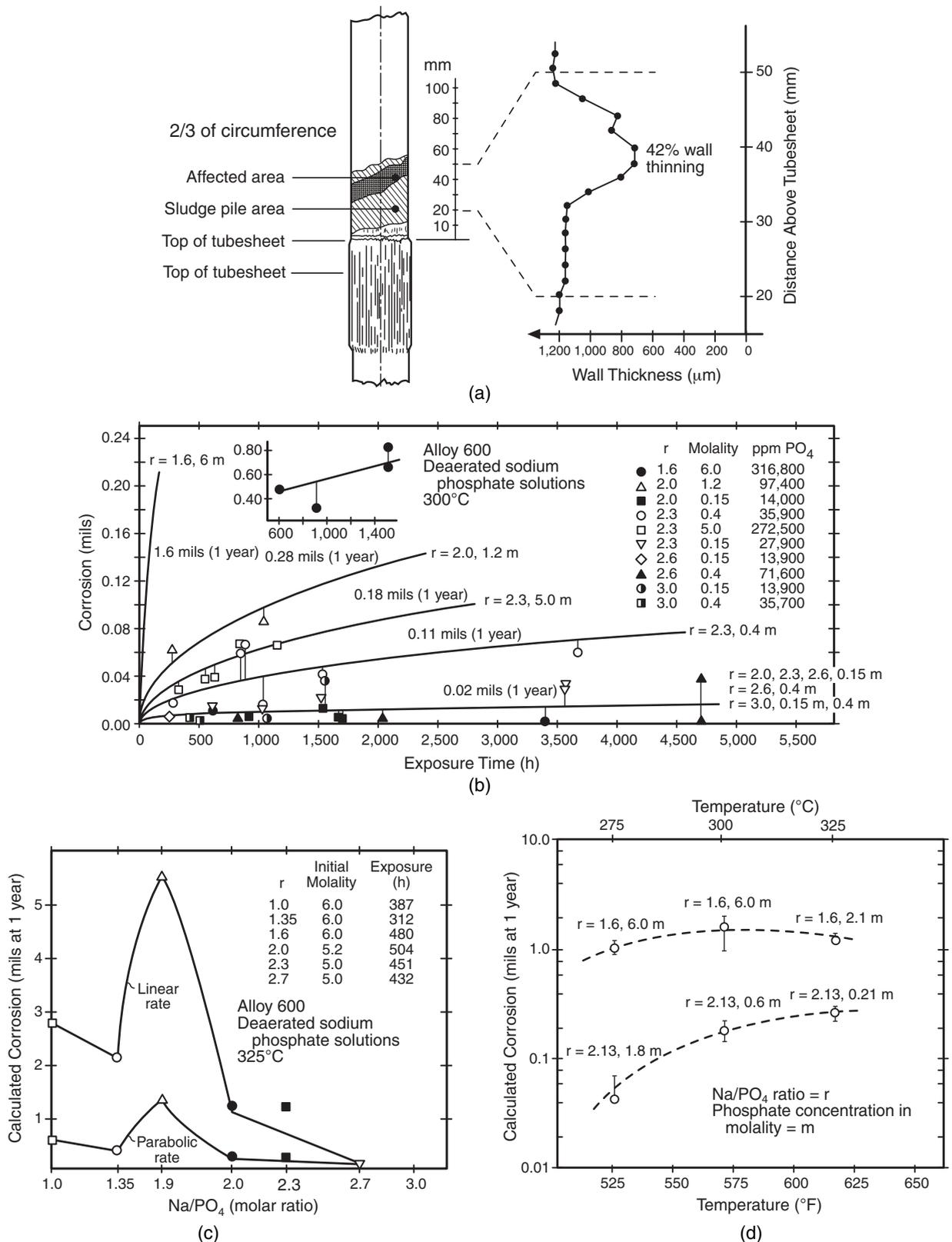


FIGURE 32. (a) Depth of localized general corrosion in Alloy 600MA in deaerated solutions containing sodium and phosphate exposed at 300°C for 7 years relative to distance from a tubesheet based on an examination described by Odar.¹⁰⁶ (b) Corrosion depth vs exposure time for specimens fully immersed having various Na/PO₄ ratios and various molalities. Data fitted by least squares. From Pessell, et al.¹⁰⁷ © 1997 NACE International. (c) Calculated corrosion in 1 year for Alloy 600MA at 325°C in deaerated solutions vs Na/PO₄ ratio using linear and parabolic extrapolations. From Pessell, et al.¹⁰⁷ © 1997 NACE International. (d) Calculated corrosion after 1 year vs temperature for Alloy 600MA tubing exposed to deaerated solutions near saturation conditions for Na/PO₄ ratios of 1.6 and 2.13. From Pessell, et al.¹⁰⁷ © 1997 NACE International.

data showed that the maximum rate was associated with a Na/PO₄ ratio of 1.6, where the rate was similar to that observed in operating SGs.

$$D = A t^{1/2} \quad (3-2)$$

$$A = 0.02 + 0.26 m \quad (3-3)$$

where D is depth of corrosion, mils; t is time, years; A is parabolic rate constant; and m is phosphate molality.

Figure 32(c) shows that there is some special feature of the Na/PO₄ ratio of 1.6 that produces especially accelerated corrosion. Published discussions⁴⁹⁻⁵⁰ speculate that this elevated rate is associated with one of the concentrated immiscible liquids indicated by Figure 30(f). It appears that there is some special chemical complex formed by this liquid that increases the solubility of nickel. Pessel, et al.,¹⁰⁷ also studied the corrosion behavior of Alloy 690, Alloy 800, Type 304 stainless steel, and Croloy (2.25Cr-1Mo-bal. Fe) and found high corrosion rates comparable to those of Alloy 600.

Figure 32(d) shows the effect of temperature on the calculated corrosion of Alloy 600 due to Na/PO₄ solutions of different ratios for concentrations near saturation in deaerated solutions. It appears that the pattern of the effect of temperature here is altered both by the reaction rate and the solubility of the compounds with these influences producing opposite effects.

It appears that the processes by which corrosion is produced by phosphate solutions are still not understood. Some of the explanation may lie in the observation by Marshall from his work¹⁰¹ in Figure 30(f), where he demonstrates the existence of two immiscible liquids with one possibly being corrosive via the formation of soluble complexes between nickel and phosphate species. Another aspect of this complexity may result from the reaction between phosphate and magnetite where stable compounds containing iron and phosphate are formed with the release of sodium, as described by Economy, et al.⁹⁵ Regardless, there appears to be no simple means for preventing localized general corrosion due to the presence of phosphates, especially at the concentrations used in the Westinghouse and CE steam generators between 1972 and 1975.

3.1.2. All Volatile Treatment (AVT) — AVT typically involves using ammonia (NH₃) to raise pH and N₂H₄ to minimize oxygen; AVT does not involve solids that can concentrate in heat-transfer crevices. Other variations of AVT in common use involve the addition of organic amines. The basis for using hydrazine is shown in Figure 33. Here, the N₂/N₂H₄ half-cell equilibrium is shown to be quite negative relative to the H₂O/H₂ half-cell equilibrium and still more negative relative to the O₂/H₂O half-cell equilibrium. From

such a position, hydrazine might be expected to be reducing and should lower the corrosion potential significantly. However, general experience has shown that the hydrazine reaction is somewhat sluggish, and the corrosion potential is not lowered to near the N₂/N₂H₄ equilibrium. Nonetheless, hydrazine has been shown to be effective in reducing the concentration of oxygen and in lowering the potential as discussed in connection with Figure 45.

The equilibria in Figure 33 also show that hydrazine will reduce S⁶⁺ to S²⁻. This is important since the lower valences of sulfur produce rapid SCC in most high-nickel alloys and since sulfate impurities, as well as sulfur-bearing resins from water treatment, are usually found in feedwater.

While producing no deposits, AVT provides no buffering capacity, and its use requires that the feedwater be very pure to minimize concentration of impurities on heat-transfer surfaces, especially in crevices; otherwise, locally corrosive conditions are produced in heat-transfer crevices. This type of water treatment was developed for fossil boilers as higher temperatures and pressures were used and as once-through designs evolved.¹¹⁰

Even with very high water purity, the prevention of corrosion in heat-transfer crevices is difficult because of the efficient boiling-driven "hideout" mechanism for the concentration of less volatile impurities. As a result, the addition of a chemical buffer, boric acid, is now being used in many PWR plants, and a few plants are using nonvolatile titanium inhibitors for the same purpose. Figure 34 illustrates the effect of adding boric acid on the failure rate of several plants. While failures of tubes are not prevented, they are somewhat mitigated by adding boric acid.

The application of titanium inhibitors with AVT treatment has been developed and is based on the work of Lumsden, et al.,¹¹¹ shown in Table 4. Here, the addition of titanium compounds to alkaline solutions is shown to provide the best inhibition over the broadest range of applied potentials. The fundamental basis for the effectiveness of titanium is derived from the broad insolubility of the titanium oxides, as shown in Figure 7(e).¹¹²

In recent years, "advanced amines" such as morpholine and ETA, have been used increasingly in place of ammonia for pH control. Figure 35(a) shows the amount of several different advanced amines required to produce a pH_T of 6.6 at the moisture separator/reheater in a PWR. Figure 35(b) shows the effect of ETA in reducing the concentration of iron in solution. Consideration of the use of these alternative additives has been stimulated mainly by the desire to reduce corrosion rates of carbon steel parts in the secondary system without risk of undue copper alloy corrosion, and to reduce, thereby, the buildup of deposits in steam generators as well as minimize wall

thinning problems in the secondary system. With respect to the data in Figure 35(a), the use of 15 ppm of NH₃ is close to a practical upper limit. Typically, 0.25 ppm of NH₃ is necessary to give pH 9 in feedwater at 25°C.

In addition to the use of advanced amines, the use of molar ratio control and high hydrazine were introduced in the early 1990s in the U.S as a result of the continued worsening of corrosion at crevices in plants with Alloy 600MA tubes. These initiatives are discussed in Section 3.5.5. Figure 36(a) shows the progression among U.S. PWRs from 1985 in the use of advanced amines, molar ratio control, hydrazine, and boric acid. Figure 36(b) shows the progress of plugged/sleeved tubes vs time related to modifications of water chemistry from Japanese plants. The overall chronology of water chemistry control in Japan is shown in Figure 37. Figure 37 also shows the actions that were taken to improve water chemistry in Japanese plants over time.

3.2 Chemical Thermodynamics and Electrochemical Conditions in the Secondary Side

Corrosion on the secondary side of steam generators is well understood to reside in a thermodynamic framework. This understanding exists in both boiling water reactors (BWRs) and the primary and secondary sides of PWRs. This framework is useful for correlating corrosion studies with operating conditions and for assessing schemes for controlling water chemistry. This facility with thermodynamics and electrochemistry has been a major achievement of the nuclear industry. Specific features of thermodynamics and electrochemistry that are part of this broad framework are the following:

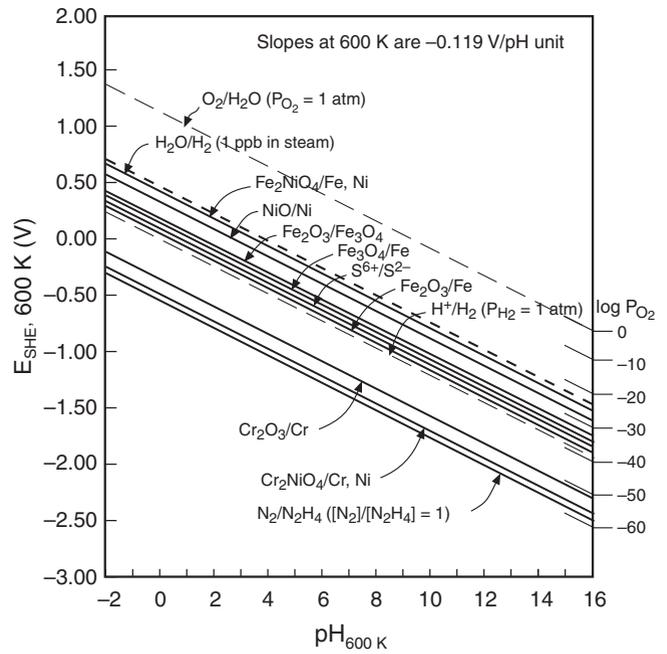


FIGURE 33. Potential vs pH calculated at 600 K (327°C) for various half cells including those involving protective oxides and water. This diagram is incomplete since competing solubilities such as the dissolution of metal ions are not shown. Data from Pourbaix,⁸⁸ Chen, et al.,¹⁰⁸ and HSC Chemistry.¹⁰⁹

1. In general, the secondary side is fully deaerated, and the tubing material is nickel-based. Therefore, at steady state, the electrochemical potential is dominated by the H₂O/H₂ equilibrium, as shown in Figures 7, 33, 39, 41, 42, and 43, and in the discussions associated with Figures 82 through 85. This results from the NiO/Ni and the standard H₂O/H₂ equilibria

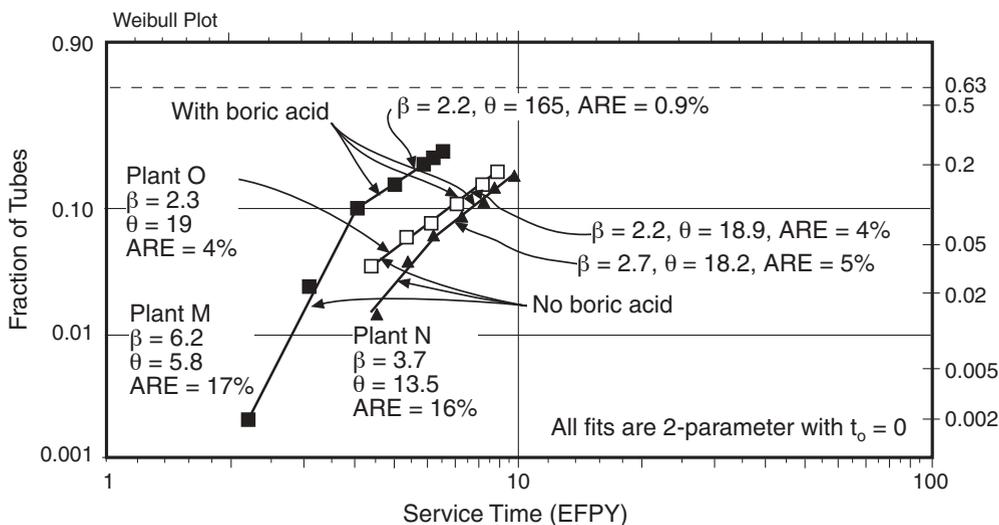


FIGURE 34. Probability vs service time for the failure of Alloy 600 tubes in three plants where boric acid has been added. From Staehle, et al.²¹ ©1994 NACE International.

TABLE 4
Effect of Inhibitors on SCC of Alloy 600MA in Alkaline Solutions^(A)

Potential, mV _{EC} ^(B)	0	100	150	200
Reference (50% NaOH)—2 days	N ^(C)	N	Y	Y
Reference + 5% H ₃ BO ₃ —2 days	N	N	Y	Y
Reference + 15% H ₃ BO ₃ —2 days	N	N	Y	Y
Reference + 0.1% Ca(OH) ₂ —1 week	N	Y	Y	Y
Reference + 2% Na ₂ ZrO ₃ —1 week	N	Y	Y	Y
Reference + 2% ZnO—1 week	N	N	N	Y
Reference + 2% Zn(PO ₄) ₂ —1 week	N	N	N	Y
Reference + SAT H ₂ TiO ₃ —1 week	N	N	N	Y
Reference + 0.5% SiO ₂ TiO ₂ —1 week	N	N	N	Y
Reference + 1% TiO ₂ (TiLAC)—1 week	N	N	N	N
Reference + 0.8% TiO ₂ (TiLAC)—1 week	N	N	N	N
Reference + 0.5% TiO ₂ (TiLAC)—1 week	N	N	N	N
Reference + 0.1% TiO ₂ (TiLAC)—1 week	N	N	N	Y
Reference + 1% TiO ₂ (TiOE)—1 week	N	N	N	N
Reference + 1% Anatase—1 week	N	N	N	Y
Reference + 1% TiB ₂ —1 week	N	N	N	N
Reference + Amorph TiO ₂ —1 week	N	N	N	N
Reference + Rutile—1 week	N	N	Y	Y
Reference + ZnTiO ₃ —1 week	N	N	Y	Y
Reference + 2% Ce(Ac) ₃ —1 week	N	N	N	Y
Reference + 2% Ce(Cl) ₃ —1 week	N	N	N	Y
Reference + 2% Ce(NO ₃) ₃ —1 week	N	N	N	N

^(A) From Lumsden, et al.¹¹¹ ©1995 NACE International.

^(B) Potentials taken relative to deaerated open-circuit potential.

^(C) N = no SCC, Y = SCC occurred.

being virtually close; the exchange current for the H₂O/H₂ reaction is much greater than for the NiO/Ni reaction. This disparity leads to the corrosion potential being almost the same as the H₂O/H₂ half-cell equilibrium, depending on the pH and the pressure of hydrogen gas.

- In the shutdown mode, sometimes oxygen is admitted to the secondary system and the oxidizing potential is increased. The magnitude of this increase is bounded by the O₂/H₂O half-cell equilibrium. However, owing to the sluggishness of the exchange current density, the practical upper limit of the potential in oxygenated environments is about 200 mV below the standard O₂/H₂O half-cell equilibrium.
- The presence of oxygen often oxidizes copper in the secondary system to Cu²⁺, which is soluble and mobile depending on the pH. The potential-pH diagram for copper in water is shown in Figure 7(d). While the availability of oxygen as an oxidant is limited, substantially greater oxidizing capacity can be sequestered by the soluble copper. Soluble copper, for example, was found to be an important contributor to the conditions that were critical for denting, as identified in Figure 14 and discussed in Section 3.5.3.
- In the secondary-side steam, hydrogen concentration is considered to be in the general range of 0.1 ppb to 10 ppb owing to the boiling action of the water and the removal of

noncondensable gases at the condenser. According to the applicable Nernst equation, shown in Figure 42, 1 ppb H₂ corresponds to an equilibrium potential that is about 250 mV above the standard H₂O/H₂ equilibrium half-cell, at secondary temperature, above the standard hydrogen equilibrium, as shown in Figure 33. This potential is the maximum oxidizing condition that can occur in the fully deaerated secondary side aside from the presence of any sequestered Cu²⁺ or other oxidizers such as Fe³⁺. Only species that have been oxidized by oxygen ingress, such as CuO and Fe₂O₃, can raise the potential beyond that due to very low hydrogen. Further, as plants have reduced the inventory of copper, the presence of iron oxides contributes to the oxidizing capacity. The Fe₂O₃/Fe₃O₄ equilibrium is significantly oxidizing, as shown in Figures 7(c), 33, and 40(b). The oxidizing capacity of oxygen can be sequestered as it oxidizes iron to the Fe³⁺ state (i.e., Fe₂O₃) during times of oxygen leakage into the secondary system. Thus, as Cu is less available, Fe³⁺ can still contribute to the oxidizing capacity in deposits and sludge.

- The action of hydrazine is understood in terms of its N₂/N₂H₄ half-cell equilibrium shown in Figure 33. Hydrazine should reduce oxygen to water because of the large difference in electrochemical potential between the respective half-cells. Hydrazine also should lower the

electrochemical potential on the surfaces of tubes, according to the mixed electrode condition, although the evidence for this action is ambiguous except in the overall measurements of open-circuit potentials in secondary systems, as shown in Figure 45(a). In this regard, hydrazine reduces the electrochemical potential to the prevailing H_2O/H_2 potential, but not so low as the N_2/N_2H_4 equilibrium, probably because of kinetic limitations. Hydrazine also should reduce sulfate to sulfide according to the relative locations of the half-cells that are shown in Figure 33. While hydrazine, because of its nominal reducing capacity, should reduce species and reduce electrochemical potentials, such reactions are slow or sluggish and, in practice, there is little lowering of the electrochemical potentials. This sluggishness may be due to the multiple steps required for the decomposition of N_2H_4 .

6. The much-reduced hydrogen concentration that results from intense boiling on the secondary side produces an opposite trend in potential from that associated with the addition of hydrazine. These trends are clear from Figure 33 as well as in the discussion with Figures 42 and 43; however, this complexity has not been explored experimentally.
7. The solubility of oxides depends on both the pH and potential according to thermodynamic predictions, as shown in Figure 7. Maintaining the pH with ammonia or other amines derives its rationale from the need to minimize the solubility of iron oxides, which is defined in thermodynamic terms and illustrated in Figure 38. In fact, much of the purpose of inhibitors involves adjusting the pH to the point at which the solubilities of oxides are at their minimum, e.g., SCC typically is minimized when the solubility of protective oxide is minimized. Such a pattern is evident in the SCC of Alloy 600 as discussed in Section 5.1.1 for Alloy 600 in Figure 82 as well as for steels as demonstrated by Parkins¹¹⁶ and Congleton, et al.,¹¹⁷ in Figure 83. Such a minimum in SCC for Alloy 600 is shown also in Section 3.5.5.6 in Figure 46.
8. The actions of phosphates in both minimizing localized corrosion through buffering reactions and producing accelerated general corrosion is understood in a thermodynamic framework as illustrated in Figures 30 through 32.
9. The occurrences of submodes of SCC as well as of pitting are bounded and related to a thermodynamic framework as discussed in Sections 5.1 and 5.2 in Figures 82 and 84.
10. Chromium is expected to be depleted from nickel-based alloys because of the high solu-

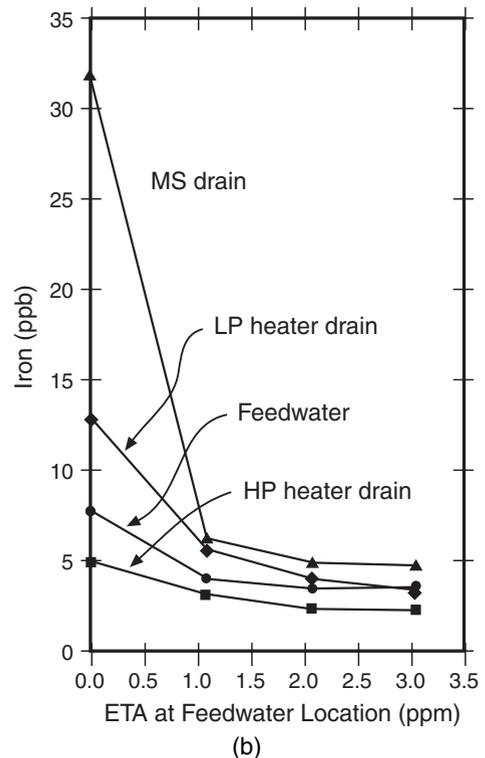
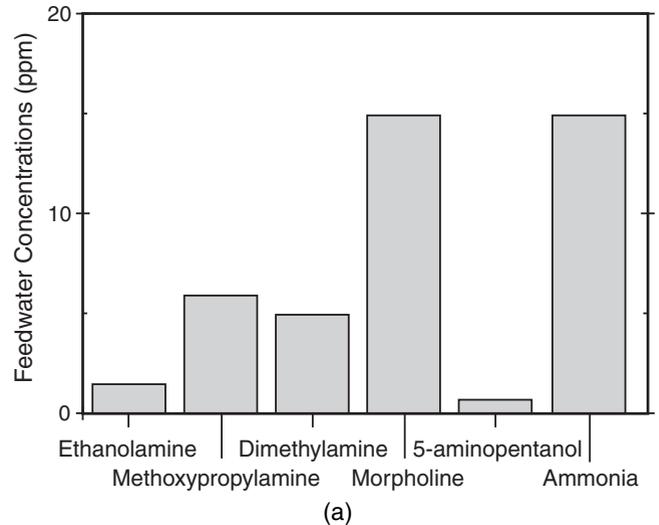


FIGURE 35. (a) Amount of amine required to produce a pH_T of 6.6 at the moisture separator reheater of a PWR. From Millett.¹¹³ (b) Concentration of iron vs the concentration of ETA in the feedwater. From Higuchi, et al.¹¹⁴

bility of chromium in alkaline solutions as evident from Figure 7(b) and from discussion of Figure 73.

As a clear indication of the value of chemical thermodynamics for analyzing corrosion problems in nuclear power, EPRI has committed a significant effort toward developing a software package, MULTEQ, based on chemical equilibrium, for the purpose of calculating equilibrium properties of solutions as determined by pH, potential, and saturation conditions.⁵¹⁻⁵³

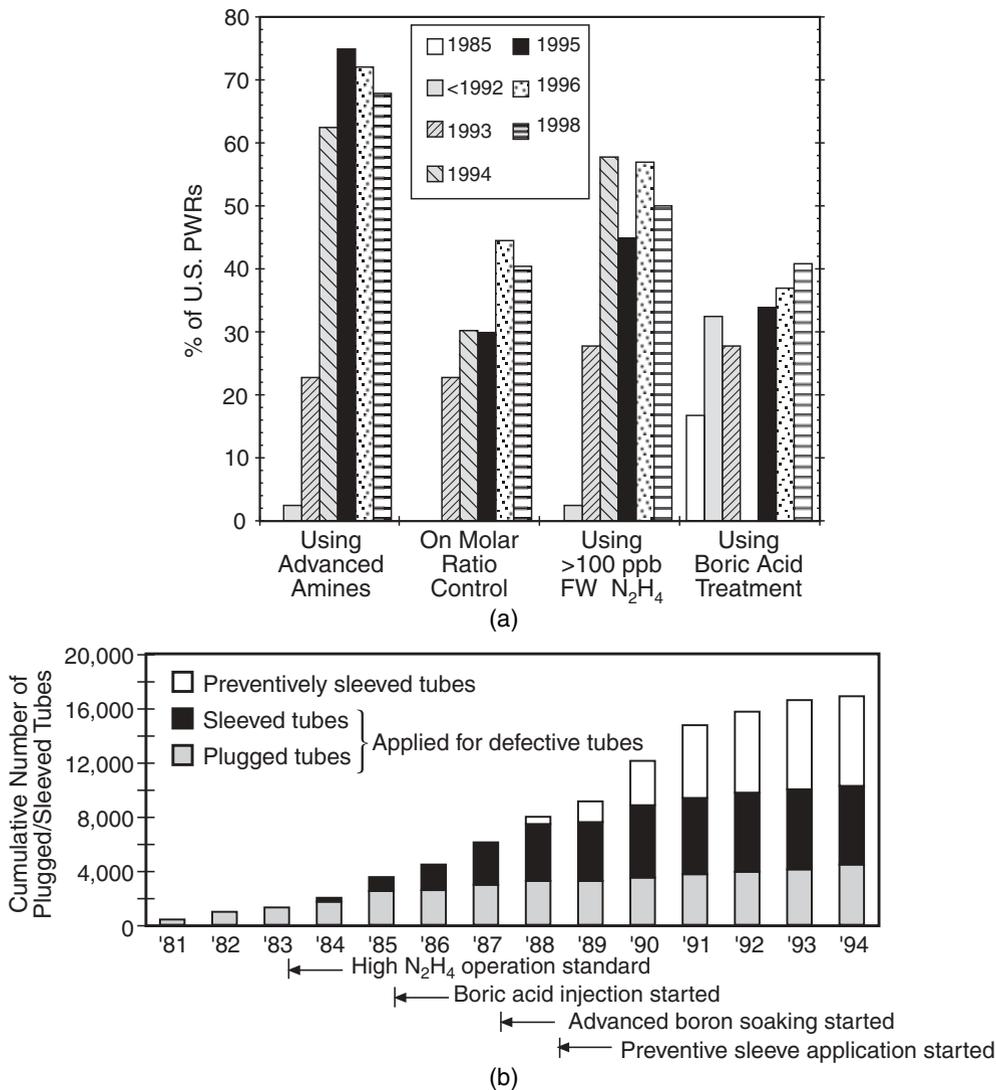


FIGURE 36. (a) Percent of U.S. PWRs using different strategies for water treatment from 1985 through 1998. From Millett and Wood.¹¹⁵ (b) Cumulative number of plugged/sleeved tubes vs time relative to various water chemistry treatments and repair actions. From Takamatsu, et al.⁶⁶ ©1996 NACE International.

Typical operating conditions on the secondary side are identified in the Ni-H₂O equilibrium diagram of Figure 39, which is drawn for 300°C. This location is consistent with the equilibria identified in Figure 33, for the H₂O/H₂, NiO/Ni, and N₂/N₂H₄ equilibrium half-cells. Following the equilibria of Figure 33, which are drawn for 327°C, the upper boundary of this secondary-side region in Figure 39 corresponds with the potential for the 1-ppb H₂ concentration in steam as identified in Figure 33.

Since hydrogen is removed rapidly by boiling on the secondary side, its concentration tends to be low, in the range of about 1 ppb, which sets the half-cell equilibrium potential about 250 mV above the 1 atmosphere hydrogen line. LPSCC decreases and becomes negligible at potentials more than 100 mV above the NiO/Ni line, as shown in Figure 106. Without significant concentration due to the effects of

heated crevice, shown in Figures 8 and 10, the free-span region is not expected to sustain significant SCC. However, some SCC and corrosion have been observed on the free-span, as shown in Figures 17, 21, and 71 and Table 2. Tests indicate that the free-span is a region of relatively much lower susceptibility to SCC and other modes of corrosion since the potential is generally in a nonaggressive range. Thus, free-spans on tubes without deposits that are exposed to bulk secondary coolant are expected to be relatively free from corrosion as long as the stresses are not high.

Chemical thermodynamics provide a framework within which kinetic processes can be bounded and understood. Figure 40 shows three examples of open-circuit potentials that are bounded by equilibrium half-cells. Figure 40(a)¹²¹ shows the effect of the concentration of dissolved oxygen on the corrosion

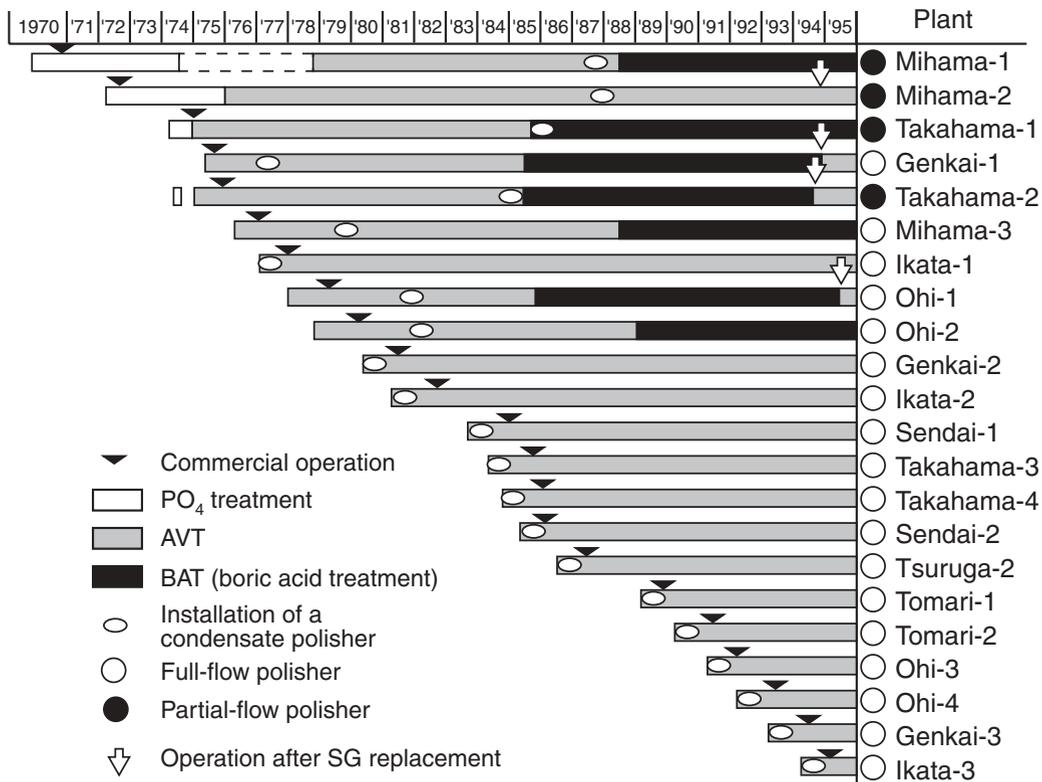


FIGURE 37. Chronology of control of water chemistry in Japanese PWRs. Figure taken from Takamatsu, et al.⁶⁶ ©1996 NACE International.

potential for five temperatures. These trends are consistent with thermodynamic boundaries as shown in Figure 33 but are dominated by the metal-oxygen mixed electrode, which accounts for the large change in potential from 1 ppb to 100 ppb. Figure 40(b) shows the corrosion potential of Alloy 600 associated with the presence of certain solid additions to the environment.¹²² These display patterns that are consistent with their oxidation state and their respective equilibrium half-cells shown in Figure 33. Figure 40(c) shows the effect of hydrogen pressure on the SCC of Alloy 600 exposed to 40% NaOH to which CuO has been added.¹²³ Such an addition of CuO places the Alloy 600 directly in the AkSCC submode, as shown in Figure 97. As hydrogen is added, following the thermodynamic direction for increasing hydrogen shown in Figure 33, the surface of the Alloy 600 moves out of the AkSCC regime and to a range where minimum AkSCC occurs, although alkaline IGA may still be active. The location of AkSCC in potential-pH space is shown in Figures 82, 84, and 85.

Electrochemical thermodynamics also has provided important frameworks for bounding models of corrosion on the secondary side of steam generators. For example, Figure 41 shows how the E-pH diagram of nickel defines the possible ranges of potentials for stress corrosion cracks and crevices in aerated and deaerated cases. For the deaerated case of PWR

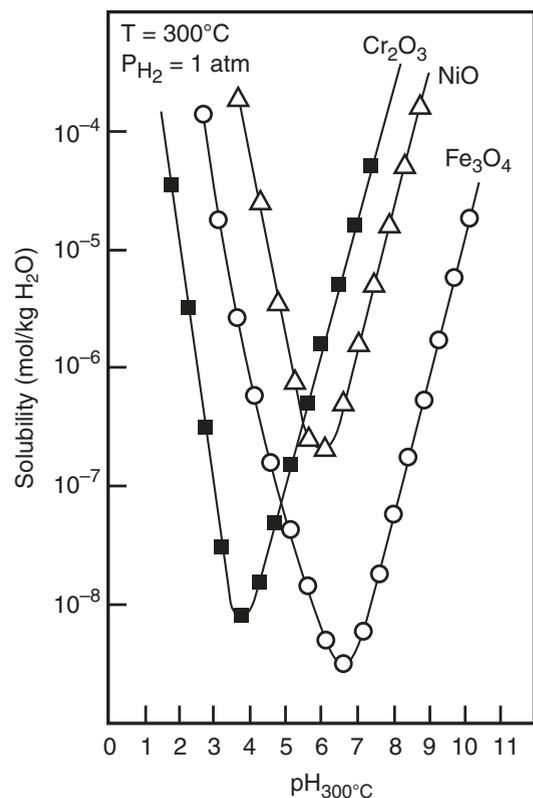


FIGURE 38. Solubility of Ni, Fe, Cr oxides vs pH at 300°C. From Sumitani, et al.¹¹⁸

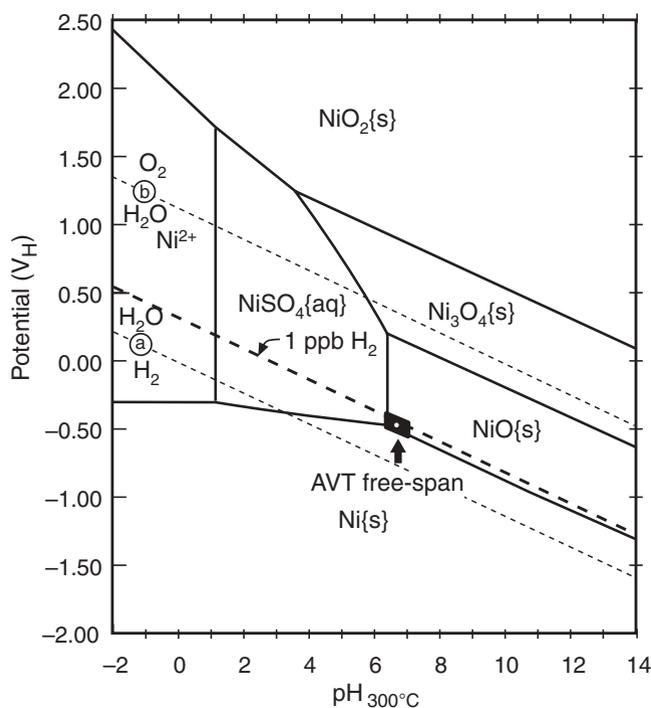


FIGURE 39. Potential-pH diagram for Ni-H₂O system at 300°C. Dissolved species activities of 10⁻³. From Kishida.¹¹⁹ Typical conditions for free-span on secondary side shown in the box. Line A is drawn for 1 ppb of hydrogen dissolved in water, which is the estimated value for the secondary side. Diagram calculated based on a 0.01-M Na₂SO₄ titrant. From Millett.¹²⁰ Used by permission of EPRI.

steam generators, the lowest potential on the outside surface is defined by the NiO/Ni equilibrium, and the highest is defined by the H₂O/H₂ equilibrium evaluated at 1 ppb dissolved H₂. In fact, this is a narrow range as shown in Figure 41. The same range of potential applies to the tip of an advancing SCC regardless of the submode and the external environment, as shown in Figure 41(a). Thus, there is little gradient in potential from the outside surface to the tip of the crack for the case of a nickel-based alloy in a deaerated solution, although a shift in the pH of the crack tip relative to the surface can increase or decrease the gradient. With the presence of oxygen or other oxidizers like CuO, a higher potential can occur on the external surface, and the crack tip will remain close to the range defined by the mixed electrode consisting of the H₂O/H₂ and NiO/Ni half-cell equilibrium potentials. In the case of a crevice, a similar deduction is available a priori, as shown in Figure 41(c). The inside of the crevice exists at a potential defined at the lowest value by the NiO/Ni equilibrium while the outside is defined by the same H₂O/H₂ equilibrium. Deviations from this condition may result from the presence of hydrazine on the outside surface, which lowers the potential on the surface. A difference in pH on the outer surface relative to the inside also will affect the gradient, depending on the

local chemistry or the presence of oxidizers such as cupric or ferric ions.

Figure 42 provides another set of insights using the bounding conditions of electrochemical thermodynamics. Here, the conditions of an operating SG are compared with an autoclave used for corrosion testing and with the conditions of potential and pH from a potential-pH diagram. The essential patterns here are the following:

1. The potential in the primary system is close to the standard hydrogen equilibrium owing to the intentional addition of hydrogen.
2. The potential on free-spans in the secondary side are about 250 mV above the standard hydrogen equilibrium owing to the efficient removal of any hydrogen by the boiling process. This trend is countered somewhat by the intentional addition of hydrazine.
3. An autoclave that is static may sustain a buildup of hydrogen and therefore a lowering of potential unless some action, such as a palladium valve, is taken explicitly to remove the hydrogen. This condition, then, differs from that in the secondary side of a steam generator and may render experimental work inapplicable because of the differences in potential between the test (where hydrogen accumulates) and the operating secondary side of a steam generator (where hydrogen is efficiently stripped). For example, in Figure 97 of Section 5.2.1.2, the potential associated with low hydrogen may produce rapid AkSCC in an operating plant; whereas, experiments that operate at the low potentials associated with high hydrogen may produce no SCC.

Finally, Figure 43 shows a third framework provided by electrochemical thermodynamics applicable to steam generators. Here, important equilibria from the Ni-H₂O and the Fe-H₂O systems are shown. Arrows and dots correspond to inferences that can be derived for ranges of potential and pH associated with various conditions and geometries in the secondary side of a steam generator. A diagram such as Figure 43 permits good estimates to be made of potential and pH at various geometric locations inside a steam generator. For example, the open-circuit potential in deaerated conditions can be specified exactly as lying in the region between the H₂O/H₂ (1 ppb H₂) and NiO/Ni half-cell equilibria, which is specified as Environment 1a in the diagram. When oxygen is added to the solution, or any oxidizing species such as Cu²⁺ is added, the possible range of potential is raised according to that defined by the equilibrium of the additional environmental half-cell; examples of which are shown in Figure 40. For oxygen, this upper limit is the O₂/H₂O equilibrium, although, in practice, this limit is not reached because of the relative sluggishness of the oxygen reduction.

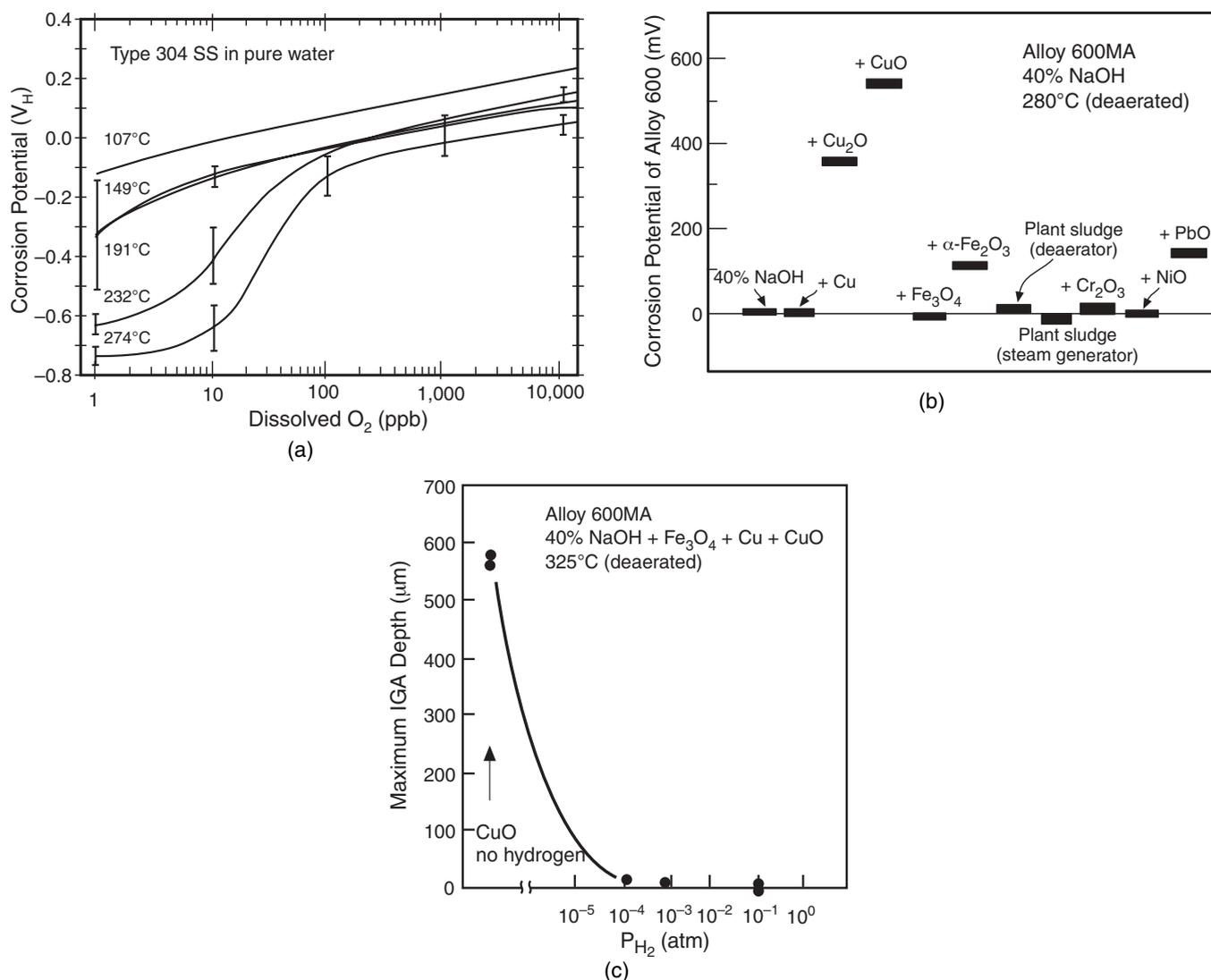


FIGURE 40. (a) Corrosion potential vs concentration of dissolved oxygen for Type 304 stainless steel in pure water exposed at five temperatures. From Andresen.¹²¹ ©1993 Elsevier Science. (b) Corrosion potential of Alloy 600 exposed in 40% NaOH deaerated solutions at 280°C plus various additions. From Kishida, et al.¹²² Courtesy of TMS, Warrendale, Pennsylvania. (c) Maximum IGA depth vs hydrogen pressure for Alloy 600MA in a solution of 40% NaOH+ Fe_3O_4 +Cu+ CuO at 325°C tested for 200 h using pre-cracked C-ring specimens stressed at 30 kgf/mm². From Nagano, et al.¹²³ ©1990 NACE International.

Similarly, effects of increasing hydrogen and hydrazine can be estimated. Further, the electrochemical conditions inside acidic and alkaline crevices can be estimated, as shown in Figure 43.

3.3 Evolution of Secondary Water Chemistry in PWRs

Table 5 shows how secondary chemistry and corrosion at the majority of plants have evolved with time; there have been some exceptions to these trends for individual plants. Both the regime of chemistry and the associated problems are shown.

3.4 Phosphate Water Treatment

Most early PWRs used the coordinated phosphate treatment, the basis for which is shown in Fig-

ures 31 and 32. The possibility of alkaline corrosion occurring in crevice areas, including under deposits, was recognized from fossil boiler experience. To minimize this possibility, initial programs with coordinated phosphate treatment in PWRs used a molar ratio of sodium to phosphate of 2.6 to 1. Nevertheless, AkSCC and AkIGC occurred relatively rapidly in early PWRs as in San Onofre-1 and Conn Yankee in about 1970. This was attributed to more severe dryout conditions in the heated crevices of PWRs than in fossil boilers where this treatment had been used successfully for many years. The heated crevices of PWR steam generators, as shown in Figures 2, 8, and 10, are not found in fossil boilers. In fossil boilers, the hot surfaces are on the outside surfaces. On the inside surfaces of tubes where boiling occurs

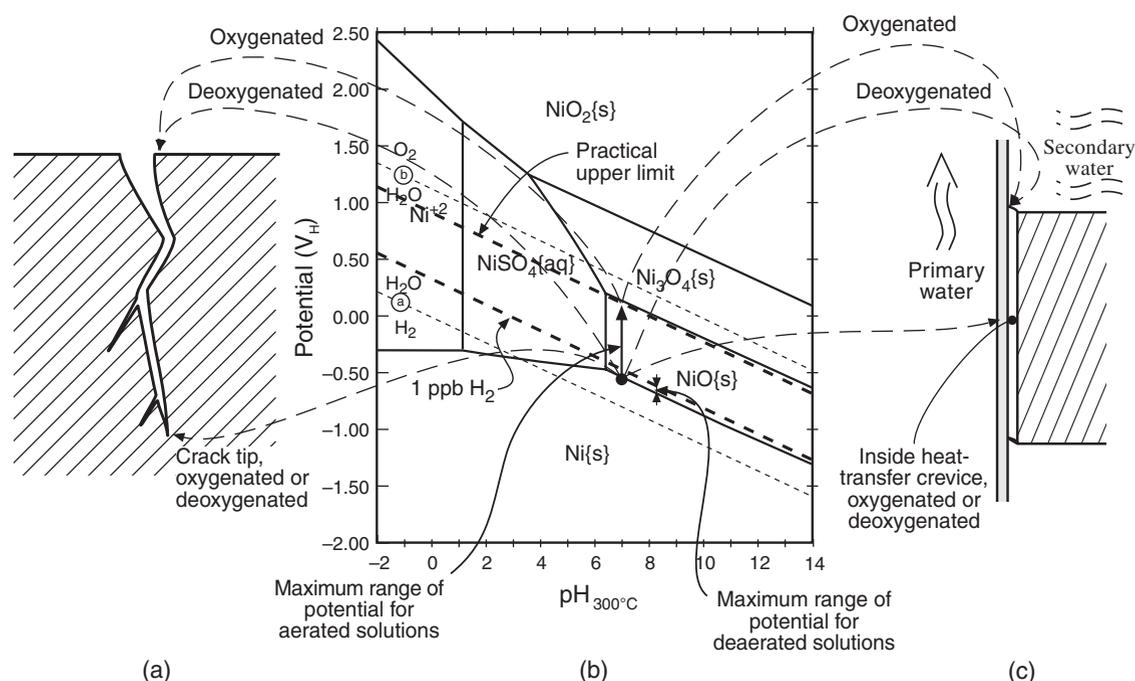


FIGURE 41. (a) Schematic view of an advancing SCC relative to possible ranges of potential, being aerated or deaerated, defined by the Ni-H₂O potential-pH diagram. (b) Potential-pH diagram for Ni-H₂O at 300°C from Figure 7(a). Diagram calculated based on a 0.01-M Na₂SO₄ titrant. (c) Schematic view of a heat-transfer crevice relative to possible ranges of potential defined by the potential-pH diagram for Ni-H₂O.

in fossil boilers, there are no crevices, and impurities concentrate only in relatively thin surface deposits where diffusion limits the magnitudes of chemical concentrations.

As a result of the AkSCC that occurred with the coordinated phosphate treatment, congruent phosphate treatments, the basis for which is shown in Figure 32(b), were instituted in the early 1970s. In the congruent treatment, the molar ratio of sodium to phosphate was reduced to about 2.3. This alleviated the caustic cracking; however, it was then found that GC, often called "wastage," occurred at heat-transfer crevices and especially in the hotter sludge crevice shown in Figures 2(b), 11, 21, and 22. This GC was attributed to the development of corrosive phosphate solutions due to reactions associated with the retrograde solubility, as shown in Figure 31, that produced local concentrations of aggressive species. It is likely that phosphate hideout due to retrograde solubility could make staying within the desired Na/PO₄ limits difficult. As a result of these problems, most plants, except in Germany where Alloy 800 tubes are used, switched to AVT in about 1974, although even in Germany the transition was made in the 1980s.

Some PWRs continued to use phosphate water chemistry. For example, San Onofre-1 continued to use phosphate water chemistry until its shutdown in 1992, and H.B. Robinson used it until its steam generators were replaced in 1984. At both plants, severe

corrosion of tubes was sustained, which may have been caused partly by the phosphate water chemistry. However, these plants did not sustain the severe denting found in some plants that switched to AVT. Several Siemens PWRs continued to use phosphate water chemistry, at low concentrations, until the late 1980s.¹²⁵ They used a low molar ratio of sodium to phosphate of 2.0 to 2.6 to avoid AkSCC; however, they sustained some GC. They continue to use Alloy 800NG rather than Alloy 600 tubes, but this difference is not significant for GC.

The GC at Siemens plants was limited to the sludge pile region at the top of the tubesheet since these plants used egg crate tube supports, as shown in Figures 4 and 5, which are less susceptible to caustic hideout. They also controlled GC by thorough sludge lancing together with improvements in water chemistry on the secondary side, which were directed toward reducing the input of sludge-forming materials and other impurities and by use of chemical cleaning to remove sludge. The relative success of Siemens plants using phosphate water chemistry is probably related to the low concentrations of 2 ppm to 6 ppm relative to the 10 ppm to 80 ppm used initially by other plants. Nevertheless, because of their successes with use of AVT in other plants, all Siemens PWRs switched to AVT by 1989.¹²⁵

The phosphate water chemistry used previously by Siemens provides some significant advantages as compared to the AVT used in the U.S. These advan-

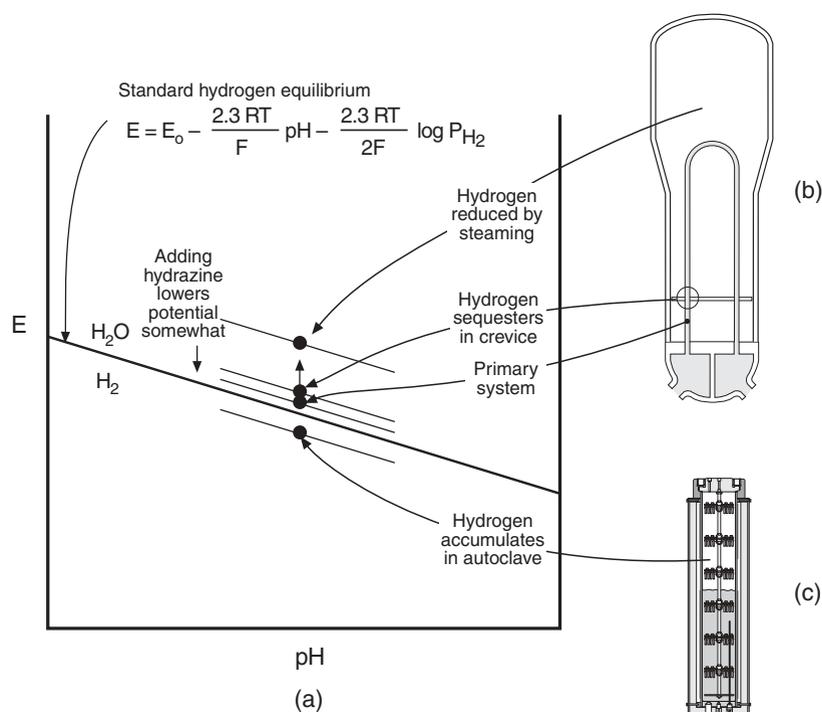


FIGURE 42. (a) Schematic view of potentials associated with conditions of hydrogen in operating steam generators. (b) Locations in a steam generator where hydrogen concentrations are different. (c) Autoclave for corrosion testing where hydrogen is not vented.

tages have been described by Siemens.¹²⁶ Phosphates provide a chemical buffer that protects against the development of acidic conditions that lead to denting and pitting, making their SGs more tolerant of chemical upsets such as condenser tube leaks or accidental ingress of acidic species. Siemens found it was possible to avoid GC with rigorous attention to minimizing sludge piles and by using line contact tube supports as shown in Figures 3 and 5.

3.5 All Volatile Water Treatment (AVT)

3.5.1 Early Recirculating Steam Generators (RSGs) — AVT was used at several early PWRs with RSGs (e.g., Indian Point-1, Beznau, and Obrigheim). Their experience is summarized as follows:

1. Indian Point-1¹⁰⁴

Indian Point-1 relied on AVT and comprehensive monitoring of condensate and feedwater using automatic equipment to prevent undetected ingress of impurities. They also maintained a relatively low hot leg temperature of 271°C. However, some SCC of its Type 304 stainless steel steam generator tubes still occurred.

2. Obrigheim¹²⁷

Obrigheim started in 1969 with AVT water chemistry and continued its use after steam generators were replaced in 1983. Both primary- and secondary-side cracking of its 600MA tubes were sustained within a few years of initial operation. The primary-side cracking eventually required replacement of the

Alloy 600MA steam generators with Alloy 800 tubes. Secondary-side cracking, which occurred mainly in the tubesheet sludge pile region, affected about 100 tubes per generator but did not affect the life of the steam generators.

3. Beznau¹²⁸

Beznau-1 started in 1970 using AVT water chemistry with Alloy 600MA. SCC was sustained in tubesheet crevices during the first fuel cycle. This was attributed to a lack of a chemical buffer, and phosphate treatment was instituted. However, because first SCC and then GC occurred, AVT was resumed by about 1974 but with increased attention to minimizing ingress of impurities.

3.5.2 Once-Through Steam Generators (OTSGs) — Because all incoming feedwater is boiled dry in OTSGs, they have been designed for AVT and have always used it.¹²⁹⁻¹³⁰ Use of AVT is necessary with OTSGs to minimize the deposition of chemical solids on tube surfaces, since such deposits would interfere with heat transfer or fluid flow and could cause corrosion. The same considerations have required that the feedwater meet high standards of purity. As a result, all secondary systems for OTSGs use condensate polishers. In addition, the use of copper in the secondary system was discouraged by B&W to minimize the introduction of difficult-to-remove copper into the OTSGs and to allow use of a higher pH in the secondary system, thus minimizing the introduction of iron corrosion products. OTSGs have sustained

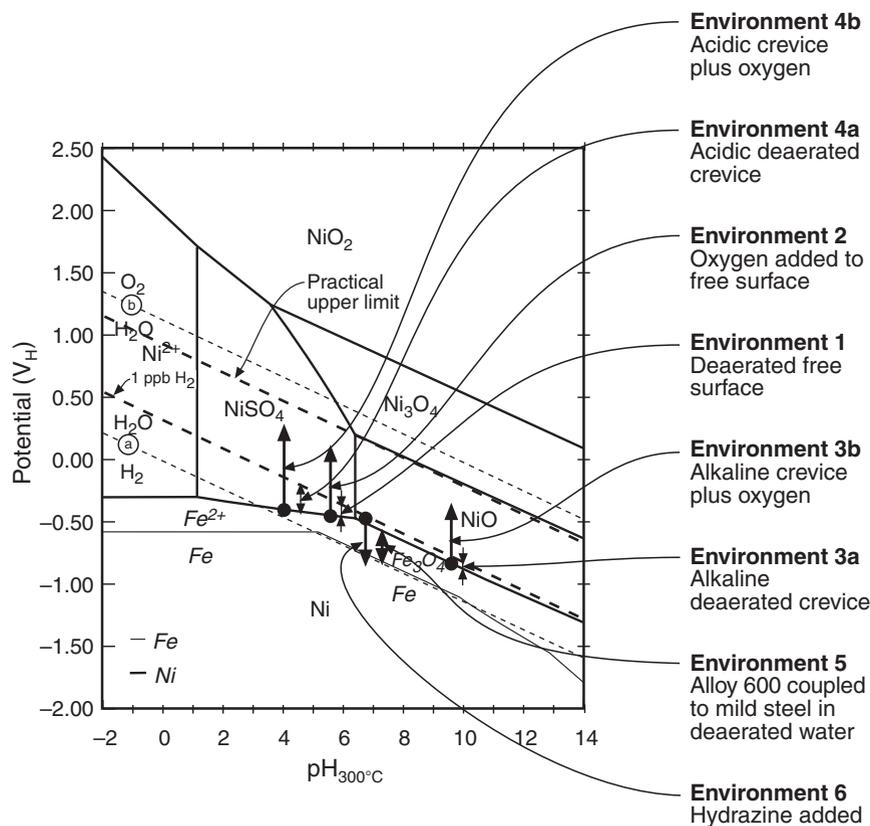


FIGURE 43. Equilibria for the Ni-H₂O and Fe-H₂O systems at 300°C relative to inferences that can be drawn for the conditions of potential and pH in steam generators. Diagram calculated based on a 0.01-M Na₂SO₄ titrant. From Staehle.¹²⁴

less rapid corrosion of Alloy 600 than RSGs of similar vintage and hot leg temperatures, although the alloy is sensitized by the stress relief treatment applied to this type of SG.

3.5.3 Industry Switch to AVT — As a result of the SCC and GC sustained with phosphate water treatment, the use of AVT water treatment was initiated at most PWRs with RSGs and Alloy 600MA tubing in about 1974. Within a few years, a new problem—denting of tubes at tube support plates—appeared.⁴⁹ This was caused by the progressive expansion of iron corrosion products in the tube support plate crevice, mainly in drilled hole geometries, as shown in Figure 14. Because the atomic volume of the metal oxides is about three times the volume of the metal,⁶³ the expansion of corrosion products constricts and deforms the tube. From the perspective inside the tube, the tube is seen to have “dents.” The chronology of denting is shown in Figure 23.

Data for the rate of denting are shown in Figure 44, and a schematic view of denting at a tube support is shown in Figure 14 and at the top of a tubesheet in Figure 15 in Section 1.4. In Figure 44, the denting grows rapidly when a bulk solution of CuCl₂ is concentrated by boiling in an experimental crevice. Most importantly, the denting continues at about the same rate when the chloride in the experi-

mental environment is removed. It should be noted that chloride is not a prerequisite for denting. The denting also interferes with eddy-current inspection of the narrowed tube. Further, the slow but inexorable straining produces SCC on both primary and secondary sides as well as cracking of the TSP (Figures 14 and 21). The occurrence of denting required that steam generators be replaced at Surry and Turkey Point and has led to extensive tube plugging and other problems at several other plants, such as primary-side cracking at supports and U-bends and secondary-side cracking at tube supports. The occurrence of denting demonstrated the need for significantly upgrading secondary system designs and water chemistry practices.

As a result of the problems sustained with denting, the water chemistry in PWRs was significantly improved in the middle to late 1970s and early 1980s. The amounts of impurity and oxygen ingress were reduced by improving condenser integrity and by eliminating leakage of air into the secondary system. Instrumentation for monitoring chemistry was improved; in some cases, condensate polishers were added or additions of boric acid were initiated. The tube supports in new steam generators also were changed from carbon steel to stainless steel to reduce the corrosion rate. These changes by various

TABLE 5
Evolution of Secondary Water Chemistry

Time Period (approx.)	Westinghouse and CE PWRs		B&W PWRs		Siemens PWRs	
	Chemistry	Problems	Chemistry	Problems	Chemistry	Problems
Up to 1974	Phosphate water chemistry	SCC then wastage	High-purity AVT	None detected (limited operation)	Phosphate water chemistry	None detected
1975 to 1980	Low-purity AVT	Denting	High-purity AVT	Limited corrosion fatigue, impingement thinning	Phosphate water chemistry	Wastage in sludge piles
1980s	AVT of increasing purity	Pitting, IGA/SCC	High-purity AVT	Limited corrosion fatigue, impingement thinning, IGA	Gradual switch to high pH AVT	Wastage in sludge piles—resolved by cleaning and by switch to high pH AVT
1990s to present	High-purity AVT coupled with initiatives such as high hydrazine, molar ratio control, and advanced amines	Continued IGA/SCC, slow denting-induced LPSCC	High-purity AVT coupled with initiatives such as high hydrazine and advanced amines	Upper bundle free-span IGA/SCC, limited impingement thinning, IGA	High pH AVT	None reported

manufacturers are indicated in Figure 5 for both materials and geometry of the tube supports. As a result of these improvements, denting largely has been controlled, although some minor denting still occurs. Despite these improvements in water chemistry, other secondary-side corrosion problems have occurred.^{49,131} These include IGA and IGSCC in tubesheet crevices, sludge piles, and TSP crevices, and pitting in sludge pile regions. In response to the continued occurrence of these modes of corrosion, limits on impurities and oxygen have been progressively tightened and the other changes have been initiated, as described in Sections 3.1.2 and 3.5.5. These trends are discussed in Sections 3.5.4 and 3.5.5.

3.5.4 Trends in AVT Water Chemistry Limits — The chronology of typical AVT water chemistry limits for U.S. PWRs with RSGs is shown in Table 6.

3.5.5 Current Use of AVT and Recent Initiatives — All large PWRs in the U.S. now use AVT water chemistry because it provides the best protection against corrosion damage on the secondary side when combined with strict controls on ingress of impurities and oxygen. However, because corrosion problems with Alloy 600MA tubes have continued despite strictly controlled AVT water chemistry, the following initiatives gradually have been instituted by many plants:

1. Copper

Most plants that initially used copper alloy heat exchanger tubes already have replaced, or are in the processes of replacing, these tubes with non-copper alloy materials (e.g., stainless steel or titanium). Copper has been found to be undesirable or deleterious for several reasons:

- Its presence limits the maximum practical values of pH_{RT} in the secondary system to the range of 9.2 to 9.4. The instability of copper at higher pH and at higher potentials is shown in Figure 7(d). The limit on pH increases rates of iron transport to the steam generators and also increases problems with FAC throughout the carbon steel parts of the secondary system.
- Copper ions carried into the steam generator are oxidants and aggravate corrosion problems such as denting, pitting, and IGC/SCC. Copper that is oxidized during shutdowns is an oxidant and can accelerate corrosion during subsequent operation.

2. High Hydrazine

Starting in 1992, EPRI water chemistry guidelines have recommended high concentrations (e.g., 100 ppb) of hydrazine in the feedwater¹³⁷ to minimize oxygen and electrochemical potentials in crevices.

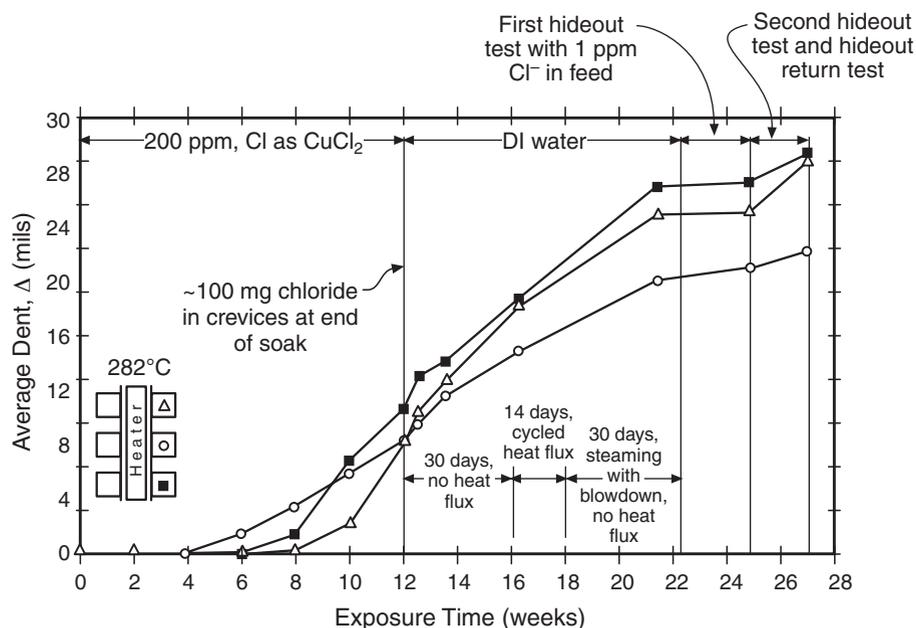


FIGURE 44. Average dent, Δ , vs exposure time. Experiments conducted in autoclave with three experimental tube supports. From Lindsay.¹³²

TABLE 6
Typical AVT Water Chemistry Limits for U.S. PWRs with RSGs—Power Operation

Parameter (ppb)	Year				
	1975 ^(A)	1982 ^(B)	1987 ^(C)	1992 ^(D)	1999 ^(E)
Sodium in blowdown	<50	<20	<20	<20	<5
Chlorine in blowdown	—	<20	<20	<20	<10
Sulfate in blowdown	—	—	<20	<20	<10
Oxygen in final condensate (CDO)	<5	<10	<10	<10	<10
Hydrazine in final feedwater	$5 \times O_2$	$>3 \times O_2$ in cond.	>20	>100	≥ 8 (CDO), 20 min
Copper in final feedwater	<5	<2	<2	<1	<1
Iron in final feedwater	<10	<20	<20	<10	<5

(A) From Sawochka.¹³³

(B) From Bell, et al.¹³⁴

(C) From PWR Secondary Water Chemistry Guidelines.¹³⁵

(D) From Interim PWR Secondary Water Chemistry Guidelines.¹³⁶

(E) From Millett.¹²⁰

A high hydrazine-to-oxygen ratio (≥ 8) has been found to be necessary to maintain low ECP values in the final feedwater and to minimize the fractions of iron in the feedwater and blowdown that are not fully reduced. The data in Figure 45 as well as the thermodynamic calculations in Figure 33 provide bases for the concentrations of hydrazine that should be added. In addition, some Japanese plants have used even higher concentrations, in the range up to 600 ppb, for many years without apparently causing undesirable side effects. Millett¹³⁸ has reported the effects of hydrazine on the corrosion potential in secondary circuits.

In fact, the real effect of N_2H_4 on potential is not clear; the results of Figure 45(a) also could result from lower oxygen. There is, unfortunately, a lack of direct measurements of the electrochemical kinetics

of the reactions associated with N_2H_4 . The effect of dissolved N_2H_4 on the potential at $288^\circ C$ is shown in Figure 45(c) from the work of Staehle, et al.¹³⁹ While these data are taken with a stainless steel base, they are indicative of the behavior for Alloy 600. Further, they are the only available data giving a direct effect of hydrazine on potential.

3. Corrosion Product Control

The occurrence of IGA/SCC in steam generators is often correlated with deposits in sludge piles and at tube supports. Several initiatives have been taken to reduce these deposits. First, the concentrations of corrosion products in the feedwater continuously have been reduced over the years to reduce the sources of the deposits. This has been accomplished mainly by eliminating copper alloys, increasing pH, and using advanced amines (Figures 35[a] and 36[a])

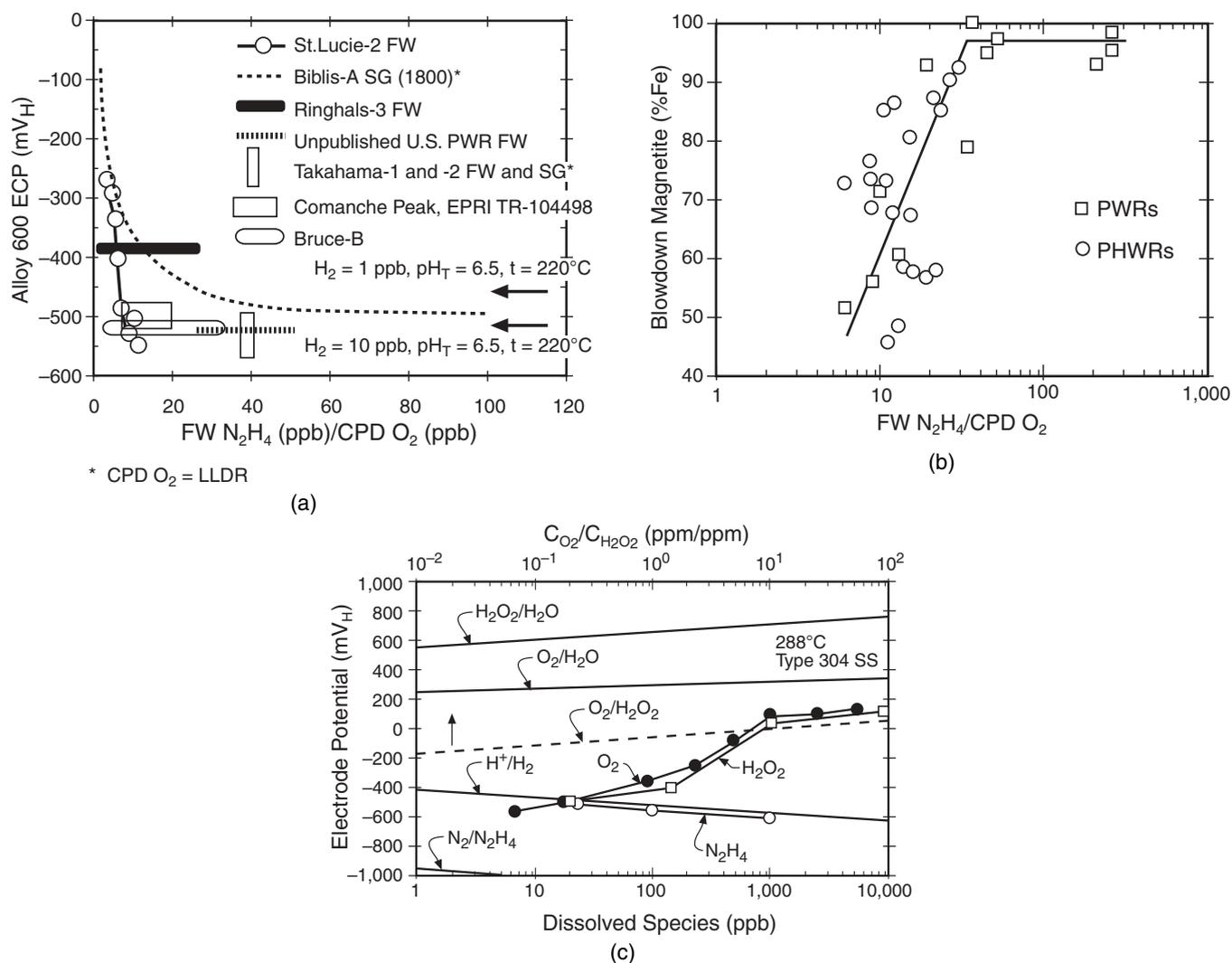


FIGURE 45. (a) Electrochemical potential on Alloy 600MA vs the ratio of feedwater hydrazine to condenser oxygen. Potentials corresponding to hydrogen at 1 ppb and 10 ppb at 220°C and pH_T 6.5. From Millett.¹³⁸ (b) Percent of iron as magnetite relative to total iron as magnetite plus hematite vs the ratio of feedwater hydrazine to condenser oxygen. From Millett.¹³⁸ (c) Corrosion potentials of Type 304 stainless steel at 288°C in high-purity water containing dissolved O₂, H₂O₂, or N₂H₄ shown in relation to the thermodynamic stability lines of H₂O, H₂O₂, and N₂H₄. From Staehle, et al.¹³⁹

as well as by increasing attention to cleanup during start-ups. Also, deposits are removed by improved sludge lancing and more extensive applications of chemical cleaning.

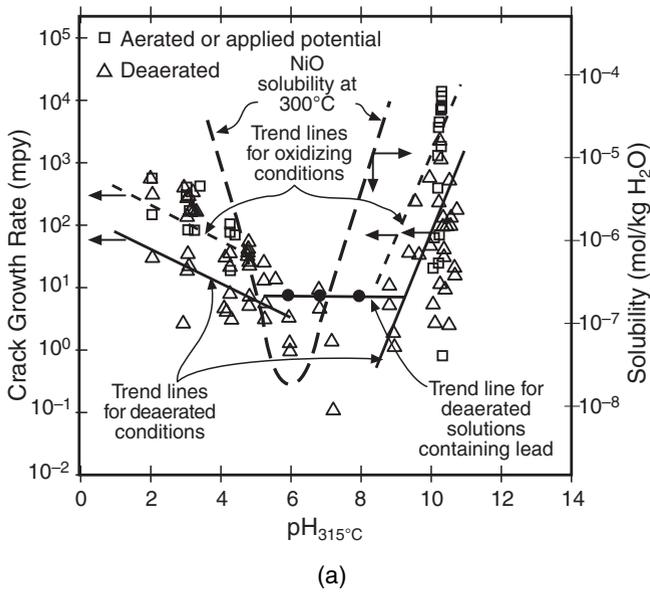
4. Advanced Amines

The use of advanced amines, such as morpholine and ETA, has increased steadily starting in the late 1980s and is continuing through the 1990s. Figure 36(a) shows their usage as of 1998. The main objective of using advanced amines has been to reduce iron transport to the steam generators. This is accomplished by using amines that are less volatile than ammonia and thus distribute more to the water phase, thereby raising the pH of steam drains in the secondary system but without side effects on the corrosion rate of copper alloys. The increase in pH reduces general corrosion of steel

surfaces in the steam drain system, which is a major contributor of iron to the feedwater. Increasing pH in the steam drain piping also reduces rates of FAC, which is an increasing problem throughout the industry; FAC is quite likely aggravated by the use of N₂H₄.

5. High pH

Higher pH is being used to decrease iron transport to the steam generators and to reduce FAC. Raising pH with ammonia requires that copper alloys be eliminated from secondary system heat exchangers. The main limitation to raising pH, once copper alloys have been eliminated, is the effect of amines on resins in condensate polishers and blowdown demineralizers since their use at the concentrations needed to achieve the desired pH_{RT} (e.g., 9.8 to 10) can rapidly exhaust the resins due to the decomposi-



- bypassing both condensate polishers and blowdown demineralizers during normal operation, except during start-ups and upsets, which requires tight condensers and high-quality makeup water
- use of resins in the amine form

6. Molar Ratio Control

Figures 46(a) and (b) show that the crack growth rates (CGRs) of SCC in secondary systems are relatively low at intermediate pH; thus, it is desirable to control pH in heat-transfer crevices in this intermediate range. The relative minimum in the SCC shown in Figure 46(a) corresponds approximately with the minimum in solubility of NiO, as shown in Figure 38 and superimposed here. This minimum in solubility also corresponds to the minimum in anodic SCC of steels, as shown in Figure 83.

Some researchers have questioned the reasonability of the crack growth rates shown for near-neutral pH on Figure 46(a), indicating that the crack growth rates are too high to be realistic.¹⁴⁰ However, the crack growth rates of Figure 46(a) are consistent with the measured crack growth rates shown in Figure 46(b). In addition, many plants with TSP crevices, which are believed to have near-neutral environments, have sustained thousands of detectable cracks, with depths of 10 mils or more, after about 10 years, indicating that a crack growth rate of about 1 mpy at a typical secondary-side temperature of 290°C is realistic. Using an activation energy of 50 kcal/mole to adjust this to 320°C indicates that crack growth rates of 10 mpy at 320°C are realistic. Another check of crack growth rates in near-neutral environments are those measured in Ohi-1 side-stream boilers with near-neutral crevices; a crack growth rate at 280°C of 0.17×10^{-5} mm/h was measured, corresponding to 0.38 mpy.¹⁴¹ Adjusting this to 320°C using an activation energy of 50 kcal/mole results in a crack growth rate of 8 mpy; this is somewhat higher than that shown in Figure 46(a) but confirms that the crack growth rates in the figure are not unrealistically high. Regardless of the absolute value of the crack growth rates in near-neutral solutions shown in Figures 46(a) and (b), the basic point remains that crack growth rates are minimized at pH values near neutral.

An important method now being used by the industry to maintain the pH of heated crevices in the desired near-neutral range of pH is to utilize "molar ratio control." The basis for this approach is that the molar ratio of strong cations (sodium and potassium) to strong anions (chloride and sulfate) in the crevice liquids is believed to be the parameter that controls the pH in the heated crevices during operation. Since the concentration of these species in crevices cannot be measured during power operation, attempts to estimate these concentrations during the previous operating cycle is performed using hideout return

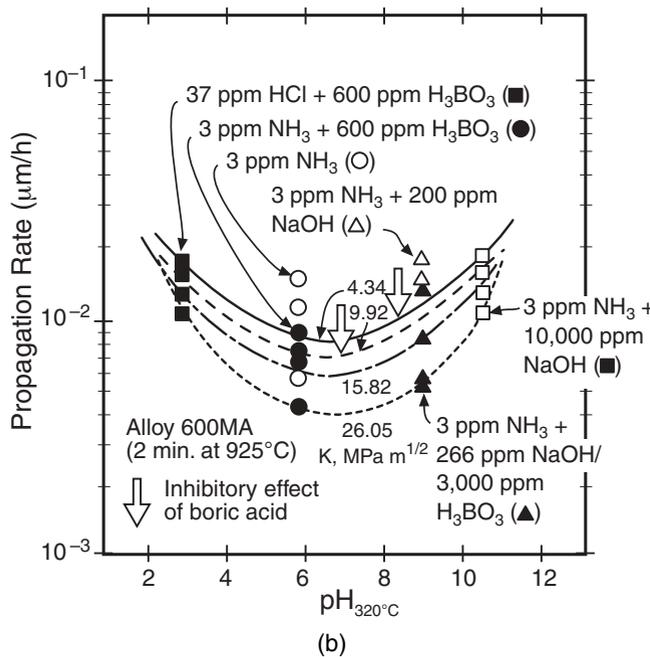


FIGURE 46. (a) SCC growth rate vs pH at 315°C for Alloy 600MA for relatively high stress intensities in the range of 10 MPa m^{1/2} or greater. From EPRI PWR Molar Ratio Control Application Guidelines.¹⁴³ Note: There are not sufficient data to develop the trend lines for oxidizing conditions. Accordingly, they were drawn parallel to the nonoxidizing condition trend lines. Used by permission of EPRI. (b) Propagation rate vs pH for constant values of stress intensity. From Kawamura, et al.¹⁴⁵ ©1985 NACE International.

tion products of organic acids. Various approaches are being used to address this limitation:

- bypassing condensate polishers during normal operation and using a moderately high pH such as 9.7 or 9.8 that does not require too frequent regeneration or replacement of blowdown demineralizer resins

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analyses immediately after plant shutdown. These analyses involve measuring the concentrations of impurities returning to the steam generator bulk water by diffusion after the plant is shut down. Based on these analyses, the chemical balance for the next operating cycle is adjusted, often using the “Molar Ratio Index” (MRI) approach discussed in connection with Equation (3-4).

It is thought that controlling the MRI of strong cations to strong anions in the prompt hideout return to a value close to 1.0, where the MRI is calculated in accordance with Equation (3-4), maximizes the probability that the pH inside heat-transfer crevices will stay within the desired range of pH_T 5 to 8.¹⁴²

$$\text{MRI} = \frac{[\text{Na}] + [\text{K}]}{[\text{Cl}] + [\text{excess SO}_4]} \quad (3-4)$$

In Equation (3-4), [Na], [K], [Cl], and [excess SO_4] are in equivalents, with [excess SO_4] being defined as SO_4 in molar excess of calcium. The values used are those from the “prompt” hideout return measured immediately after shutdown before cooldown.

Many plants are using molar ratio control in which concentrations of impurities are controlled in accordance with Equation (3-4) with the objective of achieving near-neutral values of pH in crevices. For example, if at the end of an operating period, hideout return analyses indicate that the MRI was significantly above 1.0, then the chloride concentration might be increased during the next operating cycle. Achieving an MRI close to 1.0 during hideout return often involves having a molar ratio of sodium to chloride in the steam generator blowdown during operation in the neighborhood of 0.3. Experience in Japanese plants, where a type of molar ratio control has been used for more than 20 years, indicates that using low sodium-to-chloride molar ratios during operation correlates with relatively low rates of IGC/SCC, although it does not prevent its continuation once it has initiated.¹⁴³

Control of the MRI in U.S. plants is achieved usually by injecting concentrations of NH_4Cl in the ppb range. Implementing this control usually involves the following:

- performing careful analyses of hideout return at each shutdown
- analyzing returned impurities to determine the MRI for the crevices during the previous period of operation
- adjusting the balance of impurities (if necessary) during the next cycle, e.g., by injection of NH_4Cl

The extent to which molar ratio control is used in U.S. plants has increased since 1993, and the trend is shown in Figure 36(a).

As discussed by Sawochka, et al., control of crevice pH is difficult and subject to many uncertain-

ties.¹⁴⁴ A major uncertainty concerns what fraction of the impurities, measured during hideout return, actually comes from the crevice areas that are of interest. Another problem concerns the wide range of crevice conditions (superheats, deposit morphologies) in any steam generator, so that the hideout properties and ratios of species also vary widely. In addition, results from analyses of hideout return are dominated by the operating history just before plant shutdown and provide little information about chemistry conditions for periods early in the previous operating cycle. For these reasons, molar ratio control cannot be depended upon to provide high assurance of continuously maintaining crevice chemistry in the desired range. Also, using MULTEQ for interpreting data is not entirely reliable.

7. Boric Acid

Tests indicate that boric acid inhibits denting and inhibits alkaline IGC/SCC, when added in sufficient quantities to neutralize the alkalinity in heat-transfer crevices. In addition, operating experience indicates that boric acid reduces the rate of corrosion of TSPs as indicated by reduced rates of hydrogen released to the secondary system. Service experience supports the idea that boric acid inhibits denting, although ascribing a clear benefit to using boric acid is obscured by other chemical processes. Additions of boric acid also may decrease the rate of increase of IGC/SCC, as shown in Figure 34. Based on these results, many plants affected by denting and/or IGC/SCC use on-line additions of boric acid.

8. Titanium Oxide Inhibition

Lumsden, et al., have studied the effects of TiO_2 , as shown in Table 4, as an inhibitor for SCC in caustic solutions.¹¹¹ Their results in Table 4 indicate that titanium oxide can inhibit caustic-induced IGA/SCC if crevices are clear of deposits so that titanium can reach the metal surfaces in the crevice. Evidence that such crevices can be cleaned before adding the titanium inhibitor must be demonstrated. TiO_x in the anatase form (not rutile) successfully inhibits IGA/IGSCC in Alloy 600 in model boiler tests but not if the crevices are packed with sludge. Additional work by Molander, et al.,¹⁴⁶ shows that when crevices are full of deposits, entry of the titanium oxide (even if originating from titanium acetate) is impeded. Based on the results from Lumsden, et al., in Table 4, several plants are adding titanium to the secondary system. Plants that have added titanium continuously since chemical cleaning sustain relatively low rates of increase of IGC/SCC. For this reason, using titanium inhibitors is recommended for consideration for any plant where IGC/SCC is occurring and where chemical cleaning is implemented.

9. Start-up Oxidant Control

Laboratory and model boiler tests indicate that increases in electrochemical potential associated with the presence of oxidants increase the rate of IGC/

SCC in many environments, including both alkaline and acidic. This is evident from the effect of potential on the occurrence of AkSCC shown in Figures 82 and 97. During normal power operation it appears that present water chemistry controls adequately ensure that aggressive high-potential conditions do not develop in steam generators. Preventing high-potential conditions particularly during start-ups and immediately after start-ups is sometimes difficult since the systems are sometimes open to the atmosphere or oxidized impurities can enter the secondary system inadvertently. For these reasons, increased attention is being given to controlling oxidants during shutdowns and start-ups by maintaining high hydrazine concentrations in the ppm range until power increases to more than several percent of full power.

10. Chemical Cleaning

Chemical cleaning is used at plants relatively routinely where thermal performance has been reduced by surface deposits and/or where serious IGC/SCC is occurring; however, chemical cleaning is expensive. In addition, a few plants have been chemically cleaned to prevent IGC/SCC by removing deposits that have formed in crevices that can lead to concentration of impurities and corrosion. There have been several chemical cleanings that were followed by rapid IGC/SCC (e.g., ODSCC at the TTS of Doel-4 following a 1992 chemical cleaning and ODSCC at the TTS and IGC/SCC at TSPs at Byron-1 following a 1994 chemical cleaning). Since that time, experience following chemical cleaning has been good at about 10 units, with relatively low rates of IGC/SCC subsequent to chemical cleaning. There is extensive practical experience with chemical cleaning, but a discussion here is not included. It appears that chemical cleaning, if properly performed, can reduce rates of IGC/SCC, with relatively low risks of subsequent accelerated attack.

3.6 Significance

3.6.1 Experience —

1. AVT treatment combined with low impurities, relatively high hydrazine, and high pH_{RT} has emerged as the optimum chemistry for the secondary side. This treatment is combined with eliminating copper alloys from the secondary side and minimizing ingress of oxidants during shutdowns. Further, this treatment is complemented by line contact tube supports and improved alloys in modern replacement SGs. This integrated approach avoids earlier problems of denting; much of the SCC and IGC has been avoided by advanced alloys for tubing and improved crevice geometries for supports.

2. Phosphate water chemistry is not suitable for the secondary sides of SGs because of the AkSCC and/or general corrosion that results as a consequence of the retrograde solubility of phosphate compounds and the associated formation of aggressive

environments that produce nonprotective conditions. The possible benefits of phosphate treatments for controlling off-chemistries are not balanced by the associated corrosion caused by additions of phosphates.

3. Molar ratio control and advanced amines may have produced some benefits in minimizing localized corrosion and in reducing the formation of deposits of iron compounds.

3.6.2 Potential Problems and Approaches —

1. While AVT water chemistry together with line contact stainless steel tube supports and Alloy 690TT have exhibited negligible SCC or IGC for up to 14 years in modern replacement SGs, it should be recognized that many of the corrosion phenomena associated with Alloy 600 did not occur until after periods of ten years or more. For this reason, the corrosion performance of Alloy 690TT tubing should continue to be monitored carefully.

2. Alloy 690, as described in Section 5.0, sustains SCC in high-pH lead and low-valence sulfur environments, and in mildly acidic environments. Such occurrences are consistent with the fact that, if its passivity is compromised, Alloy 690 is an inherently more reactive alloy than Alloy 600, owing to the higher Cr. The specific conditions under which these submodes of SCC as well as IGC occur should be defined with respect to operating conditions and their evolution over time.

3. While the crevices in line contact tube supports appear to be less prone to produce aggressive concentrated environments such as were common with the drilled hole design, these designs nevertheless can produce extensive deposits as have been observed in operating plants. Deposits have even been known to bridge free-span tubes in zones of unusually high steam quality with consequent IGA/IGSCC. Such deposits could readily produce the same concentrations as observed in drilled hole geometries, although possibly longer times might be required. Such possibilities should be investigated.

4. The electrochemical effects of hydrazine and hydrogen as they are relevant to the secondary side should be investigated. There are limited data, for example, on the rates of oxidation of hydrazine in realistic complex conditions and the rates of reaction between hydrazine and high-valence sulfur or oxygen. These reactions may be primarily heterogeneous so that the tendency to cleaner systems may reduce reaction rates involving O_2 and H_2 . Further, there are no bases for predicting the properties of the mixed electrode involving hydrazine. Since hydrazine is so important to present plants, such information is important.

5. Since reduced sulfur species combined with caustic have been found to produce SCC in Alloy 690, strategies for minimizing the formation of such species due to the hydrazine reduction of sulfates

should be explored. Attention should be paid to the role of potential catalysts such as magnetite.

6. While there is uncertainty in the behavior of N_2H_4 , it is reasonable to speculate that the extensive use of hydrazine could lower the potential and could place it into a range where LPSCC could occur. While Alloy 690 has been shown generally to resist LPSCC, such work has been conducted only in primary and relatively pure water. The possible occurrence of LPSCC on the secondary side due to the combination of hydrazine and concentrated impurities typical of secondary-side crevices should be investigated.

7. While copper alloys have been eliminated in many cases from the secondary side, the possible oxidizing influences of Fe^{3+} formed during shut-downs should be investigated relative to corrosion at deposits.

8. The chemical conditions required to produce corrosion in crevices at the tops of tubesheets should be investigated.

9. The possible existence of a steam phase in deposits at line contact deposits or in drilled hole crevices or under sludge should be investigated to determine the relevance of existing data for corrosion, most of which have been obtained in aqueous solutions.

10. Possible accident conditions involving resin beads, carbonate ingress, chloride ingress, lead, and other possibly aggressive and reasonably probable chemicals should be investigated.

11. The possible ingress of oxygen during start-up and shutdown should be prevented or greatly minimized. Such oxygen reacts to form Fe^{3+} and Cu^{2+} . These species can remain after start-up and be released to produce corrosion.

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ABBREVIATIONS AND ACRONYMS

AcSCC	acidic stress corrosion cracking
AES	Auger electron microscope
AkIGC	alkaline intergranular corrosion
AkSCC	alkaline stress corrosion cracking
ANL	Argonne National Laboratory
ARE	average relative error
ASME	American Society of Mechanical Engineers
ASTM	American Society for Testing and Materials
ASSCC	alumino-silicate stress corrosion cracking
ATEM	advanced transmission electron microscope
AVT	all volatile treatment
B&W	Babcock & Wilcox
BWR	boiling water reactor
BAT	boric acid treatment
CBDA	corrosion-based design approach
CE	Combustion Engineering
CERT	constant extension rate test
CF	corrosion fatigue
CGR	crack growth rate
CISCC	chloride stress corrosion cracking
CRDM	control rod drive mechanism
CuSCC	copper stress corrosion cracking
DSSCC	doped steam stress corrosion cracking
EcP	electrochemical potential
ECT	eddy-current testing
EDF	Electricite de France
EPFY	equivalent full power years
EOC	end of cycle
EPRI	Electric Power Research Institute
FAC	flow-accelerated corrosion
FDE	full-depth explosive
FDH	full-depth hydraulic
FDR	full-depth roll
FFCP	full-flow condensate polishing
GC	general corrosion
HPSCC	high-potential stress corrosion cracking
HTMA	high-temperature mill-annealed
HTSCC	high-temperature stress corrosion cracking
ID	inside diameter
IDSICC	inside diameter stress corrosion cracking
IGA	intergranular attack
IGA/SCC	intergranular attack/stress corrosion cracking
IGC	intergranular corrosion
IGSCC	intergranular stress corrosion cracking
LME	liquid metal embrittlement
LPSCC	low-potential stress corrosion cracking
LTMA	low-temperature mill-annealed
LTSCC	low-temperature stress corrosion cracking
MA	mill-annealed

MRI	molar ratio index	RUB	reverse U-bend
MULTEQ	proprietary name of software for thermodynamic calculation	SCC	stress corrosion cracking
NDE	nondestructive examination	SG	steam generator
NG	nuclear grade	SHE	standard hydrogen electrode
NSSS	nuclear steam supply system	SME	solid metal embrittlement
OD	outside diameter	SN	sensitized heat treatment
ODSCC	outside diameter stress corrosion cracking	SSRT	slow strain rate test (same as CERT)
OgSCC	organic stress corrosion cracking	S ^y SCC	low-valence stress corrosion cracking
OTSG	once-through steam generator	T _C	cold leg temperature
P*, F*	refer to alternate repair criteria at TTS	T _P	primary temperature
PbSCC	lead stress corrosion cracking	T _H	hot leg temperature
PDR	partial-depth roll	T _S	secondary temperature
PFCP	partial-flow condensate polishing	TGSCC	transgranular stress corrosion cracking
PHWR	pressurized heavy water reactor	TLT	two legs touching; applied to test specimen of C-ring or U-bend
PWR	pressurized water reactor	TSP	tube support plate
RSG	recirculating steam generator	TT	thermally treated
RT	room temperature	TTS	top of tubesheet

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