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THE ROLE OF MICROSTRUCTURAL VARIABLES IN PRIMARY WATER STRESS CORROSION CRACKING OF INCONEL 600

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Primary water stress corrosion cracking (PWSCC) of Inconel 600 has been a wide - spread and continuous problem in steam generators. This paper focuses on the microstructural variables for the resistance to PWSCC. These include carbide distribution, grain size, impurity segregation, chromium depleted zone, and carbon-in-matrix.

On the basis of operating experiences and experimental reports, an alternative interpretation for microstructural effects on PWSCC has been newly proposed. The role of microstructural variables appear to play differently in Inconel 600SA and 600TT : carbon-in-matrix plays a dominant role in Inconel 600SA, and intergranular carbide in Inconel 600TT.

DEFINITION

Inconel 600MA : Low temperature Mill Annealed Inconel 600
Inconel 600TT : Thermally Treated Inconel 600 after Low Temperature Annealing
Inconel 600SA : High Temperature Solution Annealed Inconel 600

1. INTRODUCTION

Steam generator is one of the most important components in nuclear power plants. In general, steam generators have been operated at high temperature(about 300℃) and high pressure (about 1000psig), and the interface between primary side and secondary side, i.e., one side of steam generator tubing is exposed to the primary water and the other side to the secondary water.

Several corrosion problems have occurred in steam generators. Corrosion problems in recirculating steam generators can be classified as follows[1] : 1) primary water stress corrosion cracking, 2) secondary side stress corrosion cracking, 3) pitting, 4) intergranular attack, 5) denting, 6) wastage, etc. These problems are very critical because of its effects on integrity and safety of PWRs(Pressurized Water Reactors). In particular, PWSCC is a wide-spread and continuous problem in nuclear power plants. The potential consequences of PWSCC include : secondary side contamination, possibly having to derate the plant electrical output in the event that the number of plugged tubes exceeds the available tube margin, exceeding the plant Technical Specification allowable leakage limits, and a small, but nevertheless increased, risk of sudden rupture.

In order to mitigate PWSCC, many plants have used Inconel 600TT or Inconel 600SA as a steam generator tubing. However, no single mechanism exists for PWSCC resistance by heat treatment or high temperature solution annealing. Instead a complex process involving many of the proposed mechanisms as contributing factors has been shown to account for PWSCC phenomena.

The objective of this paper is to examine selectively the literature records on PWSCC, and then to discuss on the primary factor which may play the most important role in PWSCC of Inconel 600. A second purpose is to draw out an alternative interpretation for microstructural effects on PWSCC.

2. PHENOMENOLOGY OF PWSCC

PWSCC in Inconel 600 was first reported by Coriou in the laboratory demonstration[2]. Thereafter, PWSCC of steam generators, a so-called "Coriou Syndrome", has occurred at a substantial number of plants, and becomes a significant industry problem.

2-1 PWSCC PROCESS

As is the case with other stress corrosion cracking, PWSCC requires the coincidence of three factors : i) agressive environment, ii) tensile stress, iii) susceptible material.

i) Aggressive environment : operating experience and laboratory test[3,4] have documented that Inconel 600 tubing in some heat treatment conditions will crack in a normal PWR primary water enviroment.

ii) Tensile stress : the cracking is closely related to residual stress and/or operating stress. The main locations where it has occurred[5] are at U-bends, expansion transitions, and in roll expanded areas, as shown in Figure 1. The tensile

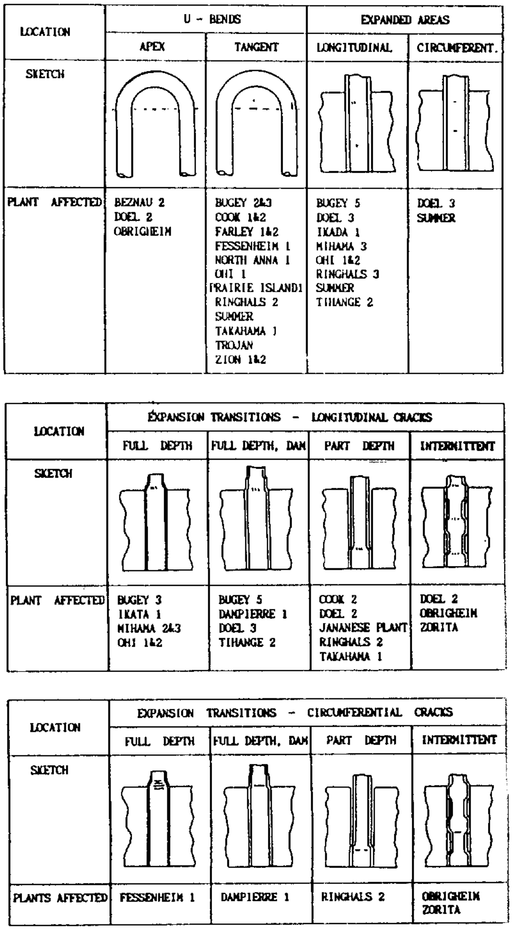

Fig. 1. PRIMARY WATER SCC CRACK LOCATIONS AND TYPES OF INCONEL 600[500]

stresses involved in primary water intergranular stress corrosion cracking could be the sum of residual plus operating stresses. Residual tensile stresses can be induced in the tubing during fabrication.

iii) Susceptible material : high Ni-alloys appear to be sensitive to primary water intergranular stress corrosion cracking[6-8]. In parti-cular, material susceptibility seems to correlate with microstructural variables such as carbide distribution, grain size, etc.

2-2 CRACKING MORPHOLOGY

Most cracking mode occurred in plants using Inconel 600MA as a steam generator tubing was axially oriented, intergranular-type[9-12]. Circum ferential cracking was also reported in several PWRs[13]. The safety implication of an axial crack is not considered a significant threat to the structural integrity of the components and most likely will result in a small leak. On the other hand, circumferential cracking poses a more serious safety concern because if it was to go undetected it could lead to a structural failure of a component rather than to a limited leak. The difference in the cracking morphology has been attributed to the different type of mechanical working(rolling vs. reaming) being performed on the components. Photo. 1 and Photo. 2 show the typical axial and the intergranular cracks occurred in steam generator tubing respectively[9,10]

2-3 IGSCC(Inter-Granular Stress Corrosion Cracking)

Although the studies of PWSCC in Inconel 600

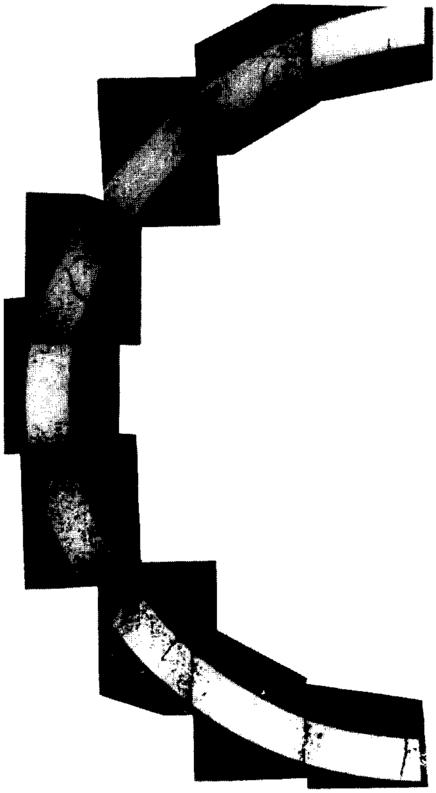

PHOTO 1. TYPICAL AXIALLY ORIENTED CRACKING IN PRIMARY SIDE OF INCONEL 600 STEAM TUBING[9]

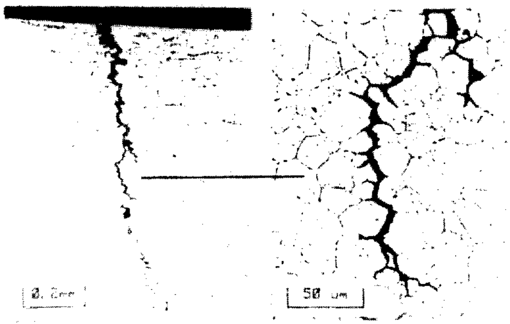

PHOTO 2. INTERGRANULAR-TYPE CRACKING IN INCONEL 600 STEAM GENERATOR TUBING[10]

have been documented in numerous published reports[14-17], the mechanism of primary water IGSCC in Inconel 600 has not been well established. Since primary water stress corrosion crack morphology was mostly intergranular type, it is useful to present a general view of the IGSCC of Ni-base alloys to clarify the comments on PWSCC.

It is well known that high nickel alloys are susceptible to IGSCC[6]. As the nickel content increases, in general, the rate of intergranular corrosion increases. The IGSCC sensitivity appears to be primarily related to i) the precipitation of chromium carbides, and ii) the segregation of impurities.

2-3-1 PRECIPITATION OF CHROMIUM CARBIDES

It has been suggested that the chromium depleted zone produced around carbides formed along the grain boundaries cause intergranular stress corrosion cracking of the active path type[18-20]. Since the chromium depleted zone may become anodic to both the solid solution within the grains and the precipitates, the depleted zone would be rapidly dissolved by electrolytic attack, thus producing a weakened network on the surface in a corrosive environment. A high tensile stress would then tend to cause the material to fracture through this depleted zone.

2-3-2 SEGREGATION OF IMPURITIES

It has long been known that segregated impurities can cause intergranular fracture in which segregated impurities in the grain boundaries can affect the chemical-mechanical stability. These species can dissolve, forming high localized concentrations of corrosive ions which attack the crack-tip region. Thus, the repassivation rate of the ruptured passive film can be reduced causing localized rapid dissolution of the grain boundary which can increase the anodic dissolution kinetics. Hence, impurities acting as depassivator segregates along grain boundaries to make it easy for the IGSCC of the active path[21]. Other results[22] suggest that the presence of segregated impurities in the boundary which are hydrogen recombination poisons are responsible for intergranular cracking in the presence of hydrogen.

3. MICROSTRUCTURAL VARIABLES

Microstructural variables such as grain size, carbide distribution and morphology, which are primarily affected by thermal history, deserve particular consideration. The changes of microstructures due to annealing or thermal treatment can be classified as follows[23-26] :

i) Grain size
ii) Carbide precipitation and distribution
iii) Grain boundary chromium depletion
iv) Grain boundary segregation of phosphorus, etc.
v) Carbon in matrix
vi) Other(reduction of residual stress)

These variables are discussed seperately below.

3-1 GRAN SIZE

Conflicting results have appeared on the issue of the role of grain size in PWSCC. Table 1 shows the experimental results of the correlation between microstructure and SCC resistance of Inconel

600MA[27]. Several points in Table 1 are noteworthy. As the mill annealing temperatures are elevated, the grain size is enlarged, and carbides appear to precipitate along grain boundaries.

Table 1. Microstructures of the mill annealed Inconel 600 and SCC resistance in pure water at 360[27]

CODE	MILL ANNEALING TEMPERATURE ℃	GRAIN SIZE (ASTM)	GRAIN BOUNDARY PRECIPITATION YES	GRAIN BOUNDARY PRECIPITATION NO	CRACKING TIME (HRS)
6A	1020	10		0	5500
6D	1070	8	0		12000, no cracks
6E	1070	8	0		12000, no cracks
6F	1020	8		0	7000
6J1	980	10		0	3500
6L	980	10 11		0	1000
6P	980	9		0	1000
6R	980	9		0	7000

Table. 2 Grani size and primary crack locations of PWR plants with non thermally treated Inconel 600[5]

PLANT	GRAIN SIZE(ASTM)	PRIMARY CRACKS
BURGEY 3	9-10	UT
BURGEY 5	11	ET
DOEL 2	6-10	UT
DOEL 3	7-11	ET
FESSENFEIM 1	7-9	UT
IKATA 1	9	ET
OHI 1	9	UET
OBRIGHEIM	9-10	UT
RINGHALS 2	8-10	UT
RINGHALS 3	8-9	E
TAKAHMA 1	10	UT
TIHANGE 2	7-11	ET
TROJAN	8	U
SURRY 1	8-12	D
ZION 1	8	U

E : PRIMARY SIDE IGSCC IN EXPANDED AREA
T : PRIMARY SIDE IGSCC IN EXPANSION TRANSITION REGION
U : PRIMARY SIDE IGSCC IN U-BEND REGION
D : PRIMARY SIDE IGSCC WHICH IS DENTING RELATED

Especially, the materials with large grain size and grain boundary precipitation, partly due to higher mill annealing temperature, appeared to be highly resistant to SCC in pure water at 360℃[27] Even a glance at Table 1 makes it obvious that grain size and carbide precipitation are primarily controlled by annealing temperatures, but the relation between them is not linear. That is, observed microstructural differences are determined not only by the final anneal but also by the intermediate anneals, and possibly by variations of the degree of cold work applied. Table 2 shows the grain size of cracked materials observed in steam generators with non-thermally-treated Inconel 600 of operating PWR plants[5]. Operation experiences in several plants indicated that small grain size materials were relatively more susce-ptible to PWSCC. On the other hand, steam generators using Inconel 600SA of which the grain size was larger than that of low temperature annealed materials, have not experienced PWSCC for as long as nine years[28]

PWSCC has mainly been occurred in steam generators using Inconel 600MA[5, 12]. Grain morphology for Inconel 600MA annealed at low temperature was generally bimodal(two uniform size) and small(ASTM 8−10). The main objectives of this mill anneal process as developed by Mannesman[29, 31] was a fine grained structure for high strength and virtually carbide−free grain boundaries for SCC−resistance in oxidizing environment. Mannesman argued that this kind of microstructure could be achieved by recrystallization treatments of the cold worked material during intermediate and final anneals, at low enough temperatures to avoid secondary recrystallization or grain growth. However, Inconel 600MA obtained by the above process have also experienced PWSCC[5].

Therefore, it is inferred that the effect of grain size on PWSCC interrelated with other microstructural variables.

3-2 CARBIDE PRECIPITATION AND DISTRIBUTION

Carbide precipitation in Inconel 600 seems like to be inevitable. Even cyclic sequence of cold work and annealing can produce a microstructure with predominant intragranular carbides, because grain boundaries may have moved away after carbide precipitation on them[31].

Tubes with copious intergranular carbides were reported to be less susceptible to PWSCC than tubes with intragranular carbides. The main fabrication variable which controls carbide morphology and distribution appeared to be the final mill anneal heat treatment : i.e., if annealing temperature is high, intragranular carbides will be dissolved, while intergranular carbides tend to precipitate because grain boundaries are preferential nucleation sites for precipitation. Also, if the annealing temperature is higher, the materials become less sensitive to PWSCC[30, 32, 33].

Table 1 indicates that grain boundary carbides could afford the resistance to SCC. In addition, to improve the resistance to IGSCC of Inconel 600MA, special thermal treatment was proposed [23, 24, 34]. For example, 704°C X 15hours was applied to Inconel 600MA steam generator tubing, which resulted in the microstructure with copious intergranular carbides. Inconel 600TT tubing has been commercially first used in KORI Unit 2[35].

The role of intergranular carbides in PWSCC may be explained through microdeformation view points. In general, macroscopic deformation can be accommodated by localized dislocation movement within planar arrays. Grain boundary carbides could be as the primary dislocation source, being activated at lower bulk deformation levels. As bulk deformation increased, cracks initiated and propagated along these planar arrays[36]. The differences in microdeformation characteristics between MA and TT conditions may have significant implications on the mechanism(s) controlling IGSCC of Inconel 600 steam generator tubing. Significant differences were observed during dynamic straining comparing MA and TT Inconel 600 tubing. The higher density of grain boundary carbides present in the TT microstructure would provide many more dislocation sources, shoter pile-ups and more homogeneous plastic deformation than in the MA tubing. On the other hand, the isolated grain boundary carbides in the MA microstructure would provide a limited number of sources per grain resulting in longer pile-ups, less homogeneous deformation and higher local stresses at grain boundaries, and then cracks in Inconel 600MA would tend to propagate more than in the TT materials.

Therefore, from the microdeformation view points, it can be suggested that carbide distribution and morphology such as semi-continuous or possibly continous carbide precipitation along grain boundaries may play the important roles in the resistance to PWSCC.

3-3 CHROMIUM DEPLETION

Although Inconel 600TT has been reported to provide a good resistance to PWSCC, it should be noted that thermal treatment would not render the materials immune to PWSCC[27, 33, 37]. There is also a possibility that cracks would tend to propagate along chromium - depleted zone, while intergranular carbides may afford the resistance to PWSCC in microdeformation points.

3-4 IMPURITY SEGREGATION

During heat treatment, impurities such as phosporus or sulfur may segregate at grain boundaries along with carbide precipitation. However, relatively little work has been reported, and its effect on PWSCC is not well established[26, 33].

3-5 CARBN-IN-MATRIX

A recent result[33] suggested that the susceptibility to PWSCC may not depend on the grain size and the intragranular carbides. It was proposed that small grain size and intragranular carbides may not major factors in PWSCC, and interpreted PWSCC as a new concept of "Environmental-Assisted Creep Fracture" : the retardation of creep by carbon in matrix in terms of a carbon-dislocation interaction. Increased carbon-in-matrix may result in a decrease of steady state creep rate, since dislocations are always in the waiting stage and are thereby immobiled because the pinned dislocations cannot be released from the carbon atmosphere without an additional force.

As indicated above, several microstructural factors affect PWSCC of Inconel 600. Therefore, it may not be possible to discuss these factors seperately, and may be necessary to investigate only the primary factors on PWSCC.

4. DISCUSSION

In the preceding section, several kinds of micro structural variables were described. We now turn to a discussion of some of these and to provide an alternative viewpoint for PWSCC.

In general, there is a close relationship between microstructure and annealing temperature, making it difficult to seperate these interaction. When annealing temperature is sufficiently high, it is expected that impurities such as phosphorus or sulfur and intragranular carbides would be dissolved into matrix, which results in the increased amount of carbon in matrix. Also grain growth would occur. In addition, most carbide precipitation would occur along grain boundaries, since grain boundaries are favorable carbide nucleation sites.

Intergranular carbides and carbon-in-matrix seem to play a different role in PWSCC of Inconel 600SA and Inconel 600TT, because carbon contents in matrix would be decreased with carbide-formation during thermal treatment.

Therefore, an alternative interpretation for the effects of microstructure on PWSCC can be proposed as follows. : i) carbon-in-matrix plays a dominant role in high temperature solution annealed materials, and intergranular carbides and dissolution of impurities would be secondary factors in the resistance to PWSCC. As high as annealing temperatures are, carbon-in-matrix would be increased and then play a role against PWSCC by way of the carbon-dislocation interactions. While large grained microstructure appears to be favorable in Inconel 600MA, grain size variations are secondary to the dominant role that carbon-in-matrix plays. ii) Intergranular carbides plays a major role in thermally treated materials. If carbon-in-matrix is assumed to play a major role in Inconel 600TT, this material should be susceptible to PWSCC. However, Inconel 600TT has been proven to be resistant to PWSCC in many operating experiences and experimental reports, even if the amount of carbon in matrix would be decreased during thermal treatment. Therefore, it can be suggested that intergranular carbides play a dominant role in thermally treated materials. Nevertheless, it should be noted that impurities segregated at grain bundaries and chromium depleted zone due to carbide precipitation may also affect PWSCC.

5. SUMMARY

PWSCC susceptibility appears to be primarily related with carbide precipitation and carbon-in-matrix compared to the other microstructural variables. Carbon-in-matrix and intergranular carbides may play a dominant role in PWSCC in Inconel 600SA and Inconel 600TT, respectively.

Much research remains to be done on microstructural effects, and it is to be hoped that future studies will attempt to identify synergistic effects, keeping in mind that interactions do occur.

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