

# **Irradiation-Assisted Stress Corrosion Cracking Behavior of Austenitic Stainless Steels Applicable to LWR Core Internals**

**Argonne National Laboratory**

**U.S. Nuclear Regulatory Commission  
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# **Irradiation-Assisted Stress Corrosion Cracking Behavior of Austenitic Stainless Steels Applicable to LWR Core Internals**

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H. M. Chung and W. J. Shack

## **Abstract**

This report summarizes work performed at Argonne National Laboratory on irradiation-assisted stress corrosion cracking (IASCC) of austenitic stainless steels that were irradiated in the Halden reactor in simulation of irradiation-induced degradation of boiling water reactor (BWR) core internal components. Slow-strain-rate tensile tests in BWR-like oxidizing water were conducted on 27 austenitic stainless steel alloys that were irradiated at 288°C in helium to 0.4, 1.3, and 3.0 dpa. Fractographic analysis was conducted to determine the fracture surface morphology. Microchemical analysis by Auger electron spectroscopy was performed on BWR neutron absorber tubes to characterize grain-boundary segregation of important elements under BWR conditions. At 0.4 and 1.4 dpa, transgranular fracture was mixed with intergranular fracture. At 3 dpa, transgranular cracking was negligible, and fracture surface was either dominantly intergranular, as in field-cracked core internals, or dominantly ductile or mixed. This behavior indicates that percent intergranular stress corrosion cracking determined at  $\approx 3$  dpa is a good measure of IASCC susceptibility. At  $\approx 1.4$  dpa, a beneficial effect of a high concentration of Si (0.8-1.5 wt.%) was observed. At  $\approx 3$  dpa, however, such effect was obscured by a deleterious effect of S. Excellent resistance to IASCC was observed up to  $\approx 3$  dpa for eight heats of Types 304, 316, and 348 steel that contain very low concentrations of S. Susceptibility of Types 304 and 316 steels that contain  $>0.003$  wt.% S increased drastically. This indicates that a sulfur-related critical phenomenon plays an important role in IASCC. A sulfur content of  $<0.002$  wt.% is the primary material factor necessary to ensure good resistance to IASCC. However, for Types 304L and 316L steel and their high-purity counterparts, a sulfur content of  $<0.002$  wt.% alone is not a sufficient condition to ensure good resistance to IASCC. This is in distinct contrast to the behavior of their high-C counterparts. At S concentrations  $>0.002$  wt.%, the deleterious effect of S is so dominant that a high concentration of C is not an important factor. A two-dimensional map was developed in which susceptibility or resistance to IASCC is shown as a function of bulk concentrations of S and C. Data reported in the literature are consistent with the map. The map is helpful to predict relative IASCC susceptibility of Types 304 and 316 steels. A similar but somewhat different map is helpful to predict IASCC behavior of Type 348 steels. Grain-boundary segregation of S was observed for BWR neutron absorber tubes irradiated to  $\approx 3$  dpa. On the basis of the results of the stress-corrosion-cracking tests and the microstructural characterization, a mechanistic IASCC model has been developed.

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## Foreword

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This report examines the effects of simulated light-water reactor coolants, material chemistry, and irradiation damage on the susceptibility to stress corrosion cracking of a series of commercially available and laboratory-melted stainless steels. This report is one of a series dating back about 8 years, describing such results, which are required to support analysis of the structural integrity of reactor internal components, many of which are subject to irradiation-assisted stress corrosion cracking (IASCC). Earlier publications in this series include NUREG/CR-5608, "Irradiation-Assisted Stress Corrosion Cracking of Model Austenitic Stainless Steels Irradiated in the Halden Reactor"; NUREG/CR-6826, "Fracture Toughness and Crack Growth Rates of Irradiated Austenitic Stainless Steels"; and NUREG/CR-6687, "Irradiation-Assisted Stress Corrosion Cracking of Model Austenitic Stainless Steel Alloys."

This report and its sister report, which details crack growth rates in heat-affected zones adjacent to stainless steel weldments, comprise the final publications based on specimens irradiated in Phase I (of two) in the Halden test reactor. Phase I irradiations principally comprised stainless steels of wide-ranging chemistry (including commercial steels of typical chemistry) and conventional heat treatment and product form processing, while Phase II irradiations included a plurality of innovatively fabricated and engineered alloys designed to possibly be more resistant to IASCC. Irradiation levels in both phases ranged up to about 3 dpa, and the high-temperature water environment employed in these tests contained 200 ppb to 8 ppm dissolved oxygen. The materials tested included several commonly used stainless steels, such as Types 304, 316 (and their low-carbon counterparts), and 347, and several laboratory heats with the basic 300-series stainless steel materials chemistries, with additions of specific elements that were expected to have an impact on IASCC properties. Taken together, these materials and test conditions make these results most applicable to boiling-water reactor internals. Publications concerning test results from Phase II irradiations are expected in the 2006 – 2008 time frame.

As in earlier reports, the deleterious effect of sulfur, which increases susceptibility to IASCC, was confirmed and further quantified. Reducing sulfur content to less than about 0.002 wt. % provides protection against IASCC. By contrast, susceptibility increased dramatically with heats that contain >0.003 wt. % S. A sufficiently low concentration of sulfur is the primary material factor necessary to ensure good resistance to IASCC. However, a sulfur concentration of less than 0.002 wt. % alone is not a sufficient material condition to ensure resistance to IASCC. Rather, a moderately high (>0.03 wt. %) level of carbon is also required in Type 304 and 316 steels. This is an important observation, given that many stainless steels (Type 304L or 316L) are decarburized to protect against thermal sensitization; too low a carbon level could render such steels susceptible to IASCC. Based on these results and related microstructural characteristics and data from electron microscopy studies, the authors have developed a model of grain boundary processes leading to IASCC. Such mechanistic understanding may shed light on the principles that might become the basis for a calculational model for use in predicting IASCC susceptibility.

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Carl J. Paperiello, Director  
Office of Nuclear Regulatory Research  
U.S. Nuclear Regulatory Commission

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## Acronyms

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AAEM = advanced analytical electron microscopy

AES = Auger electron spectroscopy

AISI = American Iron and Steel Institute

ASTM = American Society for Testing and Materials

BWR = boiling water reactor

CGR = crack growth rate

CP = commercial purity

CT = compact tension

dpa = displacement per atom

DO = dissolved oxygen

ECP = electrochemical potential

EDS = energy dispersive spectroscopy

FEG-AAEM = field-emission-gun advanced analytical electron microscopy

HBWR = heavy boiling water reactor

HP = high purity

IASCC = irradiation-induced stress corrosion cracking

IG = intergranular

IGC = intergranular cracking

IGSCC = intergranular stress corrosion cracking

JRR = Japan Research Reactor

LWR = light water reactor

PWR = pressurized water reactor

SAM = scanning Auger electron microscope

SEM = scanning electron microscopy

SS = stainless steel

SSRT = slow-strain-rate tensile

TE = total elongation

TEM = transmission electron microscopy

TG = transgranular

TGSCC = transgranular stress corrosion cracking

UE = uniform elongation

UHP = ultra high purity

UTS = ultimate tensile strength

YS = yield strength

## Executive Summary

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This report summarizes experimental work performed at Argonne National Laboratory on irradiation-assisted stress corrosion cracking (IASCC) of austenitic stainless steels that were irradiated in the Halden boiling heavy water reactor in order to simulate irradiation-induced degradation of boiling water reactor (BWR) core internal components. Slow-strain-rate tensile tests in BWR-like water were conducted on 27 austenitic stainless steel alloys that were irradiated at 288°C in helium to three fluence levels, i.e.,  $\approx 0.3 \times 10^{21}$  n·cm<sup>-2</sup>,  $\approx 0.9 \times 10^{21}$  n·cm<sup>-2</sup>, and  $\approx 2.0 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV) (neutron damage  $\approx 0.4$ ,  $\approx 1.3$ , and  $\approx 3.0$  displacement per atom [dpa]). Fractographic analysis by scanning electron microscopy was conducted to determine the susceptibility to IASCC as manifested by the degree of intergranular fracture surface morphology. Microchemical analysis by Auger electron spectroscopy was performed on BWR neutron absorber tubes to characterize the behavior of grain-boundary segregation of important elements under BWR operating conditions. On the basis of the results from the stress-corrosion-cracking tests and microstructural analyses, a mechanistic model for IASCC was developed.

At  $\approx 0.3 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV) ( $\approx 0.4$  dpa), IASCC susceptibility was negligible for all tested heats, with the exception of a 316L-like high-purity model alloy that contained 0.003 wt.% C and 0.004 wt.% S. At  $\approx 0.4$  and  $\approx 1.3$  dpa, significant transgranular fracture was mixed with intergranular fracture. At  $\approx 3$  dpa, susceptibility to transgranular stress corrosion cracking was negligible, and fracture surface morphology was either dominantly intergranular, as in field-cracked core internal components, or dominantly ductile or mixed. This behavior indicates that percent intergranular stress corrosion cracking determined at  $\approx 3$  dpa is a good measure of IASCC susceptibility.

At  $\approx 1.4$  dpa, a beneficial effect of a high concentration of Si (0.8-1.5 wt.%) was observed. At  $\approx 3$  dpa, however, such effect was obscured by a strongly deleterious effect of S. From the standpoint of BWR IASCC, which usually occurs at  $>2.5$  dpa, typical variation in Si concentration in the range of 0.4-1.1 wt.% in Types 304, 316, and 348 steel is, therefore, not an important factor.

Excellent resistance to IASCC was observed up to  $\approx 3$  dpa for seven heats of Types 304 and 316 steel that contain very low S concentrations of  $\leq 0.002$  wt.% and for one heat of Type 348 steel that contains 0.005 wt.% S. Susceptibility of Types 304 and 316 steel that contain  $>0.003$  wt.% S increased drastically. These observations indicate that a sulfur-related critical phenomenon plays an important role in IASCC. A sufficiently low concentration of S (i.e.,  $<0.002$  wt.%) is the primary material factor necessary to ensure good resistance to IASCC. For Types 304L and 316L steels and their high-purity counterparts that contain  $<0.025$  wt.% C, a sulfur concentration of  $<0.002$  wt.% alone is not a sufficient material condition to ensure good resistance to IASCC. This is in distinct contrast to the behavior of the high-C counterparts (i.e., Types 304 and 316 steel that contain  $<0.002$  wt.% S and  $>0.03$  wt.% C), which exhibit good resistance to IASCC. The contrasting behavior indicates that, at sufficiently low concentrations of S, a high concentration of C is beneficial. However, at S concentrations  $>0.003$  wt.%, the deleterious effect of S is so dominant that a high concentration of C is not an important factor.

A two-dimensional map was developed in which susceptibility or resistance to IASCC is shown as a function of bulk concentrations of S and C. Data reported in the literature are consistent with the map. The two-dimensional map is helpful to predict relative IASCC susceptibility of Types 304 and 316 steels. A similar map is helpful to predict IASCC behavior of Type 348 steels.

At S concentrations  $>0.003$  wt.%, IASCC susceptibility is thought to be influenced significantly by various material-related factors, such as the degree of S segregation to grain boundaries, annealing and cooling during fabrication, component thickness, presence of a small amount of delta ferrite, Mn content, precipitation of sulfides, solubility of sulfides in the crack-tip water, and the thickness and morphology of the Cr oxide on the crack-tip grain boundary, which controls oxygen permeation and diffusion along the grain boundary. A model alloy that contains  $\approx 3$  vol.% globular-shaped delta ferrite and  $\approx 21$  wt.% Cr exhibited excellent resistance to IASCC, despite the fact that it contained a very high S concentration of 0.028 wt.%. The excellent resistance is attributed to the effect of delta ferrite, which traps S atoms, and the effect of a high concentration of Cr, which forms Cr oxide and helps to block oxygen permeation and diffusion along the austenite grain boundary.

Grain-boundary segregation of S was observed for BWR neutron absorber tubes irradiated to  $\approx 3$  dpa. It is not clear, however, whether the S segregation occurred via a thermal process, an irradiation-induced process, or both. The S segregation behavior is consistent with the results of stress-corrosion-cracking tests that showed a predominantly deleterious effect of S on IASCC at high fluence. Based on the results of stress-corrosion-cracking tests and microstructural characterization, an IASCC model has been developed. The model is based on several key steps postulated to occur at the crack-tip grain boundary, i.e., grain-boundary segregation of Ni and S, preferential oxidation of Cr and Fe atoms over Ni atoms by oxygen that diffuses along the grain boundary, formation of a Cr-Fe spinel oxide ahead of the crack tip, formation of Ni- and S-rich thin films and islands at the metal/spinel boundary, and melting or amorphization of the Ni- and S-rich thin films and islands. The model also predicts that the thickness and morphology of Cr oxide that covers the crack-tip grain boundary strongly influence the rate of oxygen permeation and diffusion along the grain boundary.

## **Acknowledgments**

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# 1 Introduction

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In recent years, failures of reactor core internal components have increased after accumulating a fluence  $>0.5 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ), or  $\approx 0.7 \text{ dpa}$ , in boiling water reactors (BWRs),<sup>1</sup> and at approximately one order of magnitude higher fluences in some pressurized water reactors (PWRs).<sup>2</sup> The general pattern of the observed failures indicates that, as nuclear plants age and neutron fluence increases, various nonsensitized austenitic stainless steels (SSs) become susceptible to intergranular (IG) failure. Some components are known to have cracked under minimal applied stress. Although most failed components can be replaced, some safety-significant structural components (e.g., the BWR top guide, shroud, and core plate) would be very costly or impractical to replace. Therefore, the structural integrity of these components at high fluence or near the end of life (EOL) has been a subject of concern, and extensive research has been conducted to provide an understanding of this type of degradation, commonly known as irradiation-assisted stress corrosion cracking (IASCC).

Irradiation affects profoundly both local coolant-water chemistry and component microstructure. The primary effects of irradiation on materials include alteration of local microchemistry, microstructure, and mechanical properties of the core internal components, which are usually fabricated from American Iron and Steel Institute (AISI) Types 304, 316, or 348 SS. Irradiation produces defects and defect clusters in grain matrices, alters the dislocation and dislocation loop structures, and produces defect-impurity and defect-cluster-impurity complexes, leading to radiation-induced hardening and, in many cases, flow localization via dislocation channeling. Irradiation also leads to changes in the stability of second-phase precipitates and the local alloy chemistry near grain boundaries, precipitates, and defect clusters. Grain-boundary microchemistry that significantly differs from bulk composition can be produced in association with not only radiation-induced segregation but also thermally driven segregation of alloying and impurity elements.<sup>1-3</sup>

For many years, irradiation-induced grain-boundary depletion of Cr has been considered the primary metallurgical process that causes IASCC. One of the most important factors that has been considered by many investigators to be a support for the Cr-depletion mechanism is the similarity in dependence on water chemistry (i.e., oxidizing potential) of both intergranular stress corrosion cracking (IGSCC) of unirradiated, thermally sensitized material and IASCC of solution-annealed and quenched, BWR-irradiated material.<sup>4,5,6</sup> However, contrary to expectations based on the Cr-depletion mechanism, cracking incidents of control rod cladding and baffle bolts have been reported in numerous PWRs (i.e., under nonoxidizing potential). Also, the susceptibility of PWR-irradiated components to IASCC has been shown clearly by expanding-mandrel,<sup>2,7</sup> slow-strain-rate tensile (SSRT) tests,<sup>8,9,10</sup> and C-ring compression tests<sup>11</sup> in PWR water<sup>2,7</sup> or simulated PWR-like water,<sup>8-11</sup> although PWR water chemistry falls well within the range of protective electrochemical potential (ECP). Also, it has been reported that crack propagation by intergranular separation occurs in highly irradiated compact-tension specimens even in hydrogen-water chemistry (HWC) in a simulated BWR environment.<sup>12</sup> These observations indicate that the phenomenon of IASCC cannot be explained well by the grain-boundary Cr depletion and slip dissolution mechanism.

Another important aspect of IASCC characteristics that does not support the grain-boundary Cr-depletion mechanism is the effect of postirradiation annealing. Several investigators have reported that postirradiation annealing at 450-550°C for relatively short periods of time is sufficient to partially or fully mitigate the susceptibility to IASCC of austenitic SSs irradiated under BWR<sup>13-15</sup> and PWR<sup>10,16</sup> conditions. During short annealing at such low temperatures (e.g., 5-24 h at 475°C<sup>14</sup> and 1 h at 550°C<sup>16</sup>), it is not possible for significant back diffusion of metallic elements (Fe, Cr, and Ni) to occur from the grain boundary.



It has been shown that, during postirradiation annealing, IASCC susceptibility becomes negligible long before grain-boundary Cr concentration begins to change from its level in as-irradiated state.<sup>16,17</sup> These observations do not support the premise that grain-boundary Cr depletion is the direct cause of IASCC. The observations, however, suggest a possibility that a rapidly diffusing impurity element, segregated on the grain boundary during irradiation, plays an important role in annealing-induced restoration of the resistance to IASCC.

In general, IASCC is characterized by strong heat-to-heat variation in susceptibility, in addition to strong dependence on irradiation condition, material type, and grade, even among materials of virtually identical chemical composition. This behavior also indicates that the traditional interpretation based on the role of grain-boundary Cr depletion cannot explain well the mechanism of IASCC, because, for a given level of neutron fluence, heat-to-heat variation of Cr depletion is insignificant among a given type of solution-annealed and quenched steel.

Many investigators have implicated radiation-induced segregation of Si or P as the primary process that causes IASCC.<sup>1-3,7</sup> The superior resistance of one heat of Type 348 SS that is especially low in C, Si, and P seemed to provide support for this process,<sup>2</sup> and the same rationale was extended to Type 304 SS. However, in direct contradiction, several investigators reported that IASCC resistance of high-purity (HP) heats (low in C, Si, and P) of Type 304 SS is no better than that of commercial-purity (CP) counterparts.<sup>7-9,12,16,23</sup> Therefore, the role of grain-boundary segregation of C, Si, and P appeared poorly established.

Although C significantly increases the yield strength of irradiated stainless steels, higher C content (>0.06 wt.%) seems to be either benign or even conducive to lower susceptibility of irradiated austenitic SSs to IASCC.<sup>23,24</sup>

Deleterious effects of O on IGSCC of proton-irradiated steels have been reported by Cookson et al., who showed that oxides near the surface facilitate crack nucleation.<sup>25</sup> High concentrations of O and internal oxide precipitates were also reported for grain boundaries in the heat-affected zones of unirradiated Types 304 and 304L SSs and field-cracked BWR core shroud welds that were fabricated by the shielded-metal-arc procedure.<sup>26,27</sup> Yet, the exact role and the source of O on grain boundaries in IASCC of a solution-annealed material or intergranular failure of a BWR core internal weld remain poorly understood.

Indications of the deleterious effect of grain-boundary segregation of N have been reported for BWR neutron absorber tubes fabricated from high- and commercial-purity heats of Type 304 SS.<sup>23</sup> Similar reports suggested that a high concentration of N (e.g., >0.06 wt.%) is deleterious, at least under BWR conditions.<sup>1,28-30</sup> Indications of the deleterious role of N have also been reported for Types 304L and 316L SS that contain <0.024 wt.% C and have been irradiated in BWRs or test reactors at 240–300°C.<sup>22</sup> Kasahara et al.<sup>28</sup> also reported that higher N in Type 316L increased the susceptibility to IASCC, indicating that Type 316LN is a susceptible material. This observation appeared to be consistent with the behavior of 316NG SSs reported by Jacobs et al.<sup>1</sup> and Jenssen and Ljungberg.<sup>30</sup> In contrast to these observations, 316NG SSs irradiated at ≈50°C have been reported to be resistant to intergranular failure at ≈288°C in high-purity water that contained ≈32 ppm dissolved oxygen (DO).<sup>22</sup> Therefore, the role of N appears to be neither clear nor convincing.

The strong heat-to-heat variation in IASCC susceptibility has been customarily correlated with variations in irradiation-induced evolution of the microstructure and microchemistry of ordinary grain boundaries, i.e., grain boundaries not directly involved in crack-tip formation or crack propagation. This approach implicitly assumes that the crack tip, stress intensity, or the event of crack propagation itself does

not influence the microstructure and microchemistry of the grain boundary in front of the advancing crack tip. However, contrary to this assumption, results from several crack-tip microstructural studies indicate that the microstructure and microchemistry of a crack-tip grain boundary differ significantly from those of an ordinary grain boundary.<sup>31-34</sup>

Although intergranular failure in irradiated fuel cladding fabricated from austenitic SSs has been observed since the 1960s, extensive research and investigations on core internals were not conducted until the late 1980s. In spite of the extensive investigations, IASCC is still only poorly understood, and its mechanism remains elusive, and in some respects, more perplexing. In the context of this somewhat unusual background, we have initiated a comprehensive program to systematically investigate the effects of alloying and impurity elements (Cr, Ni, Si, P, S, Mn, C, O, and N) on the susceptibility of austenitic SSs to IASCC at several fluence levels. Construction of the test matrix was based on the method of Taguchi,<sup>35</sup> and susceptibility to IASCC was March 17, 2005 determined by well-controlled SSRT testing of the irradiated specimens in BWR-like water. Posttest fractographic examination was performed by scanning electron microscopy (SEM) and Auger electron spectroscopy (AES).

This report is the final topical report of the program; it summarizes the results obtained from 27 austenitic SS alloys that were irradiated in the Halden reactor at 288°C in helium to three fluence levels, i.e.,  $\approx 0.3 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ )( $\approx 0.4 \text{ dpa}$ ),  $\approx 0.9 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $\approx 1.3 \text{ dpa}$ ), and  $\approx 2.0 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $\approx 3.0 \text{ dpa}$ ). Several reports have been issued previously to document the results obtained from specimens irradiated to the low,<sup>36,37</sup> medium,<sup>38,39</sup> and high<sup>40</sup> fluence levels.

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## 2 Test Matrix and Specimen Fabrication

### 2.1 Construction of Test Matrix

The test matrix for irradiation and stress corrosion testing for this project was constructed according to the method of Taguchi, which is described in detail in Ref. 39. The base matrix followed Taguchi's standard orthogonal array  $L_{18} (2^1 \times 3^7)$ , which is an optimized matrix designed to systematically determine the effects of seven variables (i.e., bulk material concentrations of Cr, Si, P, S, Mn, C, and N) at three concentration levels, and one variable (bulk Ni concentration) at two levels. A possible synergistic interaction was assumed only between Ni and Si. Compositions assigned to the three levels of the seven alloying and impurity elements and two levels of Ni are given in Table 1. The basic test matrix, optimized on the basis of the orthogonal array  $L_{18} (2^1 \times 3^7)$ , is given in Table 2.

Table 1. Compositions assigned to medium, high, and low levels of alloying and impurity elements used to construct Taguchi's  $L_{18} (2^1 \times 3^7)$  array.

| FACTOR NO. | ELEMENT ASSIGNED | COMPOSITION (IN WT.%) ASSIGNED TO LEVEL |        |        |
|------------|------------------|---|--------|--------|
|            |                  | 1                                       | 2      | 3      |
| 1          | Ni               | 8.000                                   | 10.000 | -      |
| 2          | Si               | 0.700                                   | 1.400  | 0.150  |
| 3          | P                | 0.040                                   | 0.090  | 0.020  |
| 4          | S                | 0.006                                   | 0.020  | 0.002  |
| 5          | Mn               | 1.200                                   | 2.050  | 0.500  |
| 6          | C                | 0.040                                   | 0.090  | 0.010  |
| 7          | N                | 0.040                                   | 0.090  | 0.010  |
| 8          | Cr               | 17.500                                  | 18.500 | 16.000 |

In addition to the 18 statistically optimized heats given in Table 2, six supplementary heats of commercial- and high-purity grade Types 304, 316, and 348 SS were included in the test matrix. Compositions of the six supplementary heats are given in Table 3. Of the 24 heats given in Tables 2 and 3, eight (i.e., Heats 1, 3, 9, 10, 12, 16, 19, and 21) were replaced by commercially purchased heats. Compositions of major impurities (i.e., Si, P, C, and N) of each of the eight commercial heats matched closely those of each corresponding heat given in Tables 2 and 3. The prefix "C" was added to the identification number of these eight commercial heats (see Table 4).

The remaining 16 heats in Tables 2 and 3 were fabricated in the laboratory (USDOE Materials Preparation Center, Ames Laboratory, Ames, Iowa). All of the laboratory-fabricated heats were designated with identification numbers that began with "L". To this matrix of 24 heats, three laboratory heats were added to test the effects of the fabrication procedure. Compositions of these three laboratory heats (i.e., Heats L25, L26, and L27 in Table 4) closely match those of the counterpart commercial heats (i.e., Heats C3, C19,

and C21), respectively. Elemental composition of the complete test matrix, comprising 27 austenitic SSs, is given in Table 4.

Oxygen content, which is not specified in AISI Specifications for Types 304, 316, and 348 SS and their low-C counterparts, was not systematically controlled. However, oxygen content in the 27 heats ranged from 0.0037 wt.% (L4) to 0.0274 wt.% (L7) except for one heat: Heat L20 contained an unusually high concentration of O (0.0940 wt.%) and exhibited an anomalous dendritic structure that is not characteristic of an austenitic SS.

Table 2. Basic test matrix optimized according to Taguchi's orthogonal array L<sub>18</sub> (2<sup>1</sup> × 3<sup>7</sup>).

| <b>HEAT</b> |           | <b>COMPOSITION (IN WT.%)</b> |          |          |           |          |          |           |
|-------------|-----------|------------------------------|----------|----------|-----------|----------|----------|-----------|
| <b>No.</b>  | <b>Ni</b> | <b>Si</b>                    | <b>P</b> | <b>S</b> | <b>Mn</b> | <b>C</b> | <b>N</b> | <b>Cr</b> |
| 1           | 10.0      | 0.70                         | 0.04     | 0.006    | 1.20      | 0.04     | 0.04     | 18.5      |
| 2           | 10.0      | 0.70                         | 0.09     | 0.020    | 2.05      | 0.09     | 0.09     | 17.5      |
| 3           | 10.0      | 0.70                         | 0.02     | 0.002    | 0.50      | 0.01     | 0.01     | 16.0      |
| 4           | 10.0      | 1.40                         | 0.04     | 0.006    | 2.05      | 0.09     | 0.01     | 16.0      |
| 5           | 10.0      | 1.40                         | 0.09     | 0.020    | 0.50      | 0.01     | 0.04     | 18.5      |
| 6           | 10.0      | 1.40                         | 0.02     | 0.002    | 1.20      | 0.04     | 0.09     | 17.5      |
| 7           | 10.0      | 0.15                         | 0.04     | 0.020    | 1.20      | 0.01     | 0.09     | 16.0      |
| 8           | 10.0      | 0.15                         | 0.09     | 0.006    | 2.05      | 0.04     | 0.01     | 18.5      |
| 9           | 10.0      | 0.15                         | 0.02     | 0.002    | 0.50      | 0.09     | 0.04     | 17.5      |
| 10          | 8.0       | 0.70                         | 0.04     | 0.002    | 0.50      | 0.09     | 0.09     | 18.5      |
| 11          | 8.0       | 0.70                         | 0.09     | 0.006    | 1.20      | 0.01     | 0.01     | 17.5      |
| 12          | 8.0       | 0.70                         | 0.02     | 0.020    | 2.05      | 0.04     | 0.04     | 16.0      |
| 13          | 8.0       | 1.40                         | 0.04     | 0.020    | 0.50      | 0.04     | 0.01     | 17.5      |
| 14          | 8.0       | 1.40                         | 0.09     | 0.002    | 2.05      | 0.09     | 0.04     | 16.0      |
| 15          | 8.0       | 1.40                         | 0.02     | 0.006    | 1.20      | 0.01     | 0.09     | 18.5      |
| 16          | 8.0       | 0.15                         | 0.04     | 0.002    | 2.05      | 0.01     | 0.04     | 17.5      |
| 17          | 8.0       | 0.15                         | 0.09     | 0.006    | 0.50      | 0.04     | 0.09     | 16.0      |
| 18          | 8.0       | 0.15                         | 0.02     | 0.020    | 1.20      | 0.09     | 0.01     | 18.5      |

Table 3. Composition (wt.%) of six supplementary commercial- and high-purity heats of Types 304, 316, and 348 stainless steel.

| HEAT | SS TYPE | C    | N    | SI   | P    | S    | MN   | CR   | Ni   | NB   | MO   |
|------|---------|------|------|------|------|------|------|------|------|------|------|
| 19   | CP 304  | 0.04 | 0.04 | 0.70 | 0.04 | 0.06 | 1.20 | 18.5 | 10.0 | -    | -    |
| 20   | HP 304  | 0.01 | 0.01 | 0.15 | 0.02 | 0.02 | 0.50 | 18.5 | 10.0 | -    | -    |
| 21   | CP 316  | 0.04 | 0.04 | 0.70 | 0.04 | 0.06 | 1.20 | 16.5 | 13.5 | -    | 2.20 |
| 22   | HP 316  | 0.01 | 0.01 | 0.15 | 0.02 | 0.02 | 0.50 | 16.5 | 13.5 | -    | 2.20 |
| 23   | CP 348  | 0.04 | 0.04 | 0.70 | 0.04 | 0.06 | 1.20 | 17.5 | 11.5 | 2.20 | -    |
| 24   | HP 348  | 0.01 | 0.01 | 0.15 | 0.02 | 0.02 | 0.50 | 17.5 | 11.5 | 2.20 | -    |

Table 4. Elemental composition of 27 commercial and laboratory model austenitic stainless steel alloys irradiated in Halden Reactor.

| ANL ID <sup>a</sup> | Source Heat ID | Elemental Composition (wt.%) |       |       |        |      |       |       |       |        |        |         |
|---------------------|----------------|------------------------------|-------|-------|--------|------|-------|-------|-------|--------|--------|---------|
|                     |                | Ni                           | Si    | P     | S      | Mn   | C     | N     | Cr    | O      | B      | Mo, Nb  |
| C1                  | ASW-70378      | 8.12                         | 0.50  | 0.038 | 0.0020 | 1.00 | 0.060 | 0.060 | 18.11 | 0.0102 | <0.001 | -       |
| L2                  | BPC-4111       | 10.50                        | 0.82  | 0.080 | 0.0345 | 1.58 | 0.074 | 0.102 | 17.02 | 0.0065 | <0.001 | -       |
| C3                  | PNL-C1         | 8.91                         | 0.46  | 0.019 | 0.0040 | 1.81 | 0.016 | 0.083 | 18.55 | -      | <0.001 | -       |
| L4                  | BPC-488        | 10.20                        | 0.94  | 0.031 | 0.0100 | 1.75 | 0.110 | 0.002 | 15.80 | 0.0037 | <0.001 | -       |
| L5                  | BPC-4104       | 9.66                         | 0.90  | 0.113 | 0.0283 | 0.47 | 0.006 | 0.033 | 21.00 | 0.0068 | <0.001 | -       |
| L6                  | BPC-4127       | 10.00                        | 1.90  | 0.020 | 0.0051 | 1.13 | 0.096 | 0.087 | 17.10 | 0.0058 | <0.001 | -       |
| L7                  | BPC-4112       | 10.60                        | 0.18  | 0.040 | 0.0384 | 1.02 | 0.007 | 0.111 | 15.40 | 0.0274 | <0.001 | -       |
| L8                  | BPC-491        | 10.20                        | 0.15  | 0.093 | 0.0100 | 1.85 | 0.041 | 0.001 | 18.30 | 0.0059 | <0.001 | -       |
| C9                  | PNL-C6         | 8.75                         | 0.39  | 0.013 | 0.0130 | 1.72 | 0.062 | 0.065 | 18.48 | 0.0102 | <0.001 | -       |
| C10                 | ASW-23381      | 8.13                         | 0.55  | 0.033 | 0.0020 | 1.00 | 0.060 | 0.086 | 18.19 | 0.0074 | <0.001 | -       |
| L11                 | BPC-493        | 8.15                         | 0.47  | 0.097 | 0.0094 | 1.02 | 0.014 | 0.004 | 17.40 | 0.0071 | <0.001 | -       |
| C12                 | ASW-23805      | 8.23                         | 0.47  | 0.018 | 0.0020 | 1.00 | 0.060 | 0.070 | 18.43 | -      | <0.001 | -       |
| L13                 | BPC-496        | 8.18                         | 1.18  | 0.027 | 0.0222 | 0.36 | 0.026 | 0.001 | 17.40 | 0.0042 | <0.001 | -       |
| L14                 | BPC-4129       | 7.93                         | 1.49  | 0.080 | 0.0023 | 1.76 | 0.107 | 0.028 | 15.00 | 0.0045 | <0.001 | -       |
| L15                 | BPC-4126       | 8.00                         | 1.82  | 0.010 | 0.0129 | 1.07 | 0.020 | 0.085 | 17.80 | 0.0110 | <0.001 | -       |
| C16                 | PNL-SS14       | 12.90                        | 0.38  | 0.014 | 0.0020 | 1.66 | 0.020 | 0.011 | 16.92 | 0.0157 | <0.001 | Mo 2.30 |
| L17                 | BPC-4128       | 8.00                         | 0.66  | 0.090 | 0.0089 | 0.48 | 0.061 | 0.078 | 15.30 | 0.0090 | <0.001 | -       |
| L18                 | BPC-498        | 8.13                         | 0.14  | 0.016 | 0.0326 | 1.13 | 0.080 | 0.001 | 18.00 | 0.0055 | <0.001 | -       |
| C19                 | ASW-74827      | 8.08                         | 0.45  | 0.031 | 0.0030 | 0.99 | 0.060 | 0.070 | 18.21 | 0.0200 | <0.001 | -       |
| L20                 | BPC-4101       | 8.91                         | 0.017 | 0.010 | 0.0039 | 0.41 | 0.002 | 0.002 | 18.10 | 0.0940 | <0.001 | -       |
| C21                 | ASW-12455      | 10.24                        | 0.51  | 0.034 | 0.0010 | 1.19 | 0.060 | 0.020 | 16.28 | 0.0112 | <0.001 | Mo 2.08 |
| L22                 | BPC-4100       | 13.30                        | 0.024 | 0.015 | 0.0036 | 0.40 | 0.003 | 0.001 | 16.10 | 0.0103 | <0.001 | Mo 2.04 |
| L23                 | BPC-4114       | 12.04                        | 0.68  | 0.030 | 0.0475 | 0.96 | 0.043 | 0.092 | 17.30 | 0.0093 | <0.001 | Nb 1.06 |
| L24                 | BPC-4105       | 12.30                        | 0.03  | 0.007 | 0.0055 | 0.48 | 0.031 | 0.002 | 16.90 | 0.0129 | <0.001 | Nb 1.72 |
| L25                 | BPC-4133       | 8.93                         | 0.92  | 0.020 | 0.0082 | 1.54 | 0.019 | 0.095 | 17.20 | 0.0085 | 0.010  | -       |
| L26                 | BPC-4131       | 8.09                         | 0.79  | 0.004 | 0.0022 | 0.91 | 0.070 | 0.089 | 17.20 | 0.0080 | <0.001 | -       |
| L27                 | BPC-4132       | 10.30                        | 0.96  | 0.040 | 0.0018 | 0.97 | 0.057 | 0.019 | 15.30 | 0.0058 | 0.030  | Mo 2.01 |

<sup>a</sup>The first letter "C" or "L" denotes, respectively, a commercial or a laboratory-fabricated heat.

## 2.2 Fabrication of Specimen

Slow-strain-rate tensile (SSRT) specimens were machined from solution-annealed and water-quenched plates, pipes, or sheets that were fabricated from the 27 model austenitic SS alloys. Thick plates or pipes of various thickness and diameter were obtained only for commercial heats, and the laboratory heats were in sheet form. The geometry of the SSRT specimens, 0.76 mm thick, 57.2 mm long, and 12.7 mm wide (gauge section 19.1 mm long and 3.1 mm wide), is shown in Fig. 1. Gauge length and plane of the specimen were parallel to the rolling direction and the plane of the sheet, respectively. Subsize compact-tension (1/4TCT) specimens were also fabricated and irradiated in tandem in the same capsules with the SSRT specimens to determine J-R fracture toughness properties and crack growth rate (CGR). The geometry of the CT specimen is shown in Fig. 2. Together, 96 SSRT and 24 CT specimens were prepared and irradiated in this study.

Before machining the specimens, the surface oxide layer was removed to minimize specimen-to-specimen variation in surface roughness and uncertainty in surface chemical composition. After the specimens were mechanically machined and polished, no additional heat treatment was applied to them. Electric discharge machining was not employed because of concern about potential contamination of the surface by Cu impurity. The machined specimens were degreased in acetone and cleaned ultrasonically in alcohol before they were encapsulated in clean Type 304 SS capsules. The capsules were filled with research-grade He for irradiation in the Halden HBWR located in Halden, Norway.

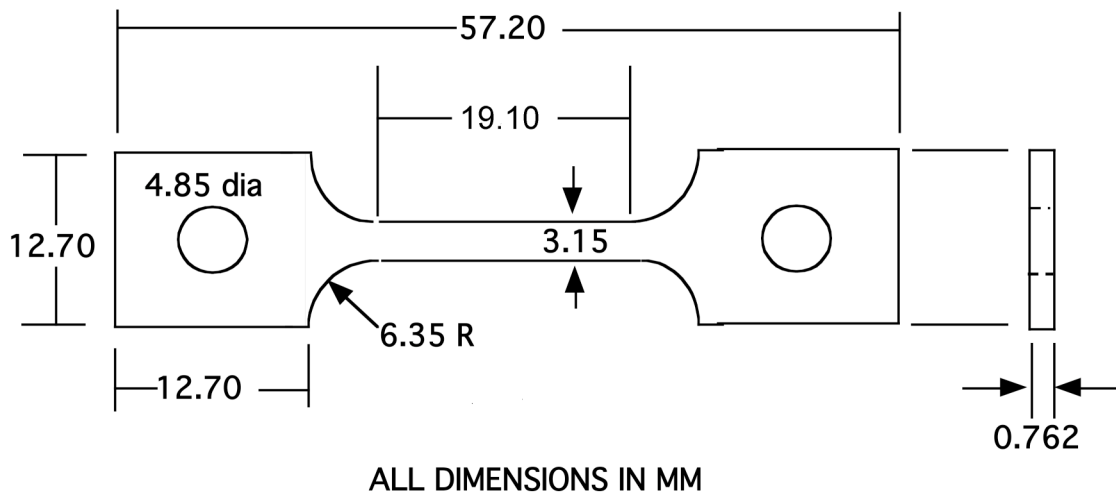


Figure 1. Geometry of slow-strain-rate tensile test specimen; all dimensions in mm.

The width of the SSRT specimens (12.7 mm, Fig. 1) was chosen similar to the width of the compact-tension specimens (14.0 mm, Fig. 2) so that an optimum combination of the two types of specimens could be stacked in the irradiation capsule of the Halden reactor. In each capsule, four CT specimens were positioned at the bottom, and 16 SSRT specimens could be stacked vertically on top of the CT specimens. In this way, it was possible to make the best use of the irradiation space. For reference, the dimension of the compact tension specimens is given in Fig. 2, although the specimens were tested in a separate investigation.

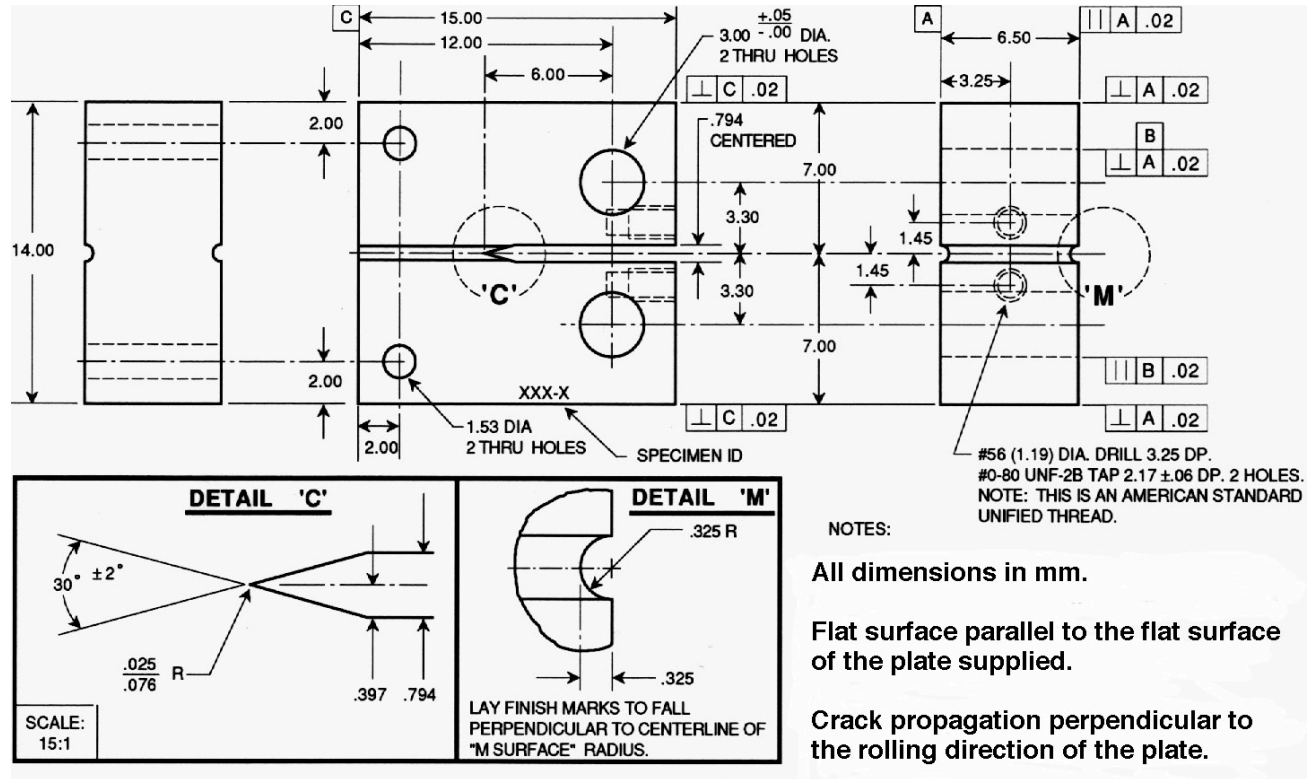


Figure 2. Geometry of compact-tension specimen.



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### 3 Specimen Irradiation

Ninety-six SSRT and 24 CT specimens were encapsulated in six capsules, each capsule containing 16 SSRT and 4 CT specimens. A fixed 0.5-mm gap, filled with He, was allowed between the inner wall of the Type 304 SS capsule and the specimen surface. The size of the gap was selected to maintain specimen temperature at 289°C during irradiation in He. To prevent capsule wall creepdown and possible changes in gap size, spacers in the form of Type 304 SS wires (0.5-mm diameter) were placed between the specimens and the capsule inner wall. Type 304 SS filler bodies were inserted on both sides of the SSRT specimen stack to avoid overheating the thin gauge section. Table 5 shows the specimens categorized according to alloy, fluence level, and postirradiation test type. The six capsules were irradiated in the Halden HBWR starting on April 8, 1992. Fast neutron ( $E > 1$  MeV) flux during the various irradiation cycles ranged from  $1.80 \times 10^{13}$  n-cm<sup>-2</sup> s<sup>-1</sup> to  $3.31 \times 10^{13}$  n-cm<sup>-2</sup> s<sup>-1</sup>. The irradiation history of the six capsules is summarized in Table 6. Specimen temperature during the irradiation, determined on the basis of the melting behavior of five to six types of low-melting alloys, was 288±2°C.

Table 5. Specimen number per alloy, irradiation fluence level, and postirradiation test type.

| Alloy ID | SSRT Test         |                     |                  | Constant Load Test |        |     | J-R, Crack Growth Rate Test |        |     |
|----------|-------------------|---------------------|------------------|--------------------|--------|-----|-----------------------------|--------|-----|
|          | high <sup>a</sup> | medium <sup>a</sup> | low <sup>a</sup> | high               | medium | low | high                        | medium | low |
| C1       | 1                 | 1                   | 1                |                    |        |     |                             |        |     |
| L2       | 1                 | 1                   |                  |                    |        |     | 1                           | 1      |     |
| C3       | 1                 | 1                   | 1                |                    |        |     | 1                           | 1      | 1   |
| L4       | 1                 | 1                   | 1                |                    |        |     |                             |        |     |
| L5       | 1                 | 1                   | 1                |                    |        |     | 1                           |        |     |
| L6       | 1                 | 1                   |                  |                    |        |     |                             |        |     |
| L7       | 1                 | 1                   |                  |                    |        |     |                             |        |     |
| L8       | 1                 | 1                   | 1                |                    |        |     |                             |        |     |
| C9       | 1                 | 1                   | 1                |                    |        |     |                             |        |     |
| C10      | 1                 | 1                   | 1                |                    |        |     |                             |        |     |
| L11      | 1                 | 1                   | 1                |                    |        |     |                             |        |     |
| C12      | 1                 | 1                   | 1                |                    |        |     |                             |        |     |
| L13      | 1                 | 1                   | 1                |                    |        |     |                             |        |     |
| L14      | 1                 | 1                   |                  |                    |        |     | 1                           |        |     |
| L15      | 1                 | 1                   |                  |                    |        |     |                             |        |     |
| C16      | 1                 | 1                   | 1                |                    |        |     | 1                           | 1      |     |
| L17      | 1                 | 1                   |                  |                    |        |     |                             |        |     |
| L18      | 1                 | 1                   | 1                |                    |        |     | 1                           | 1      |     |
| C19      | 5                 | 1                   | 1                | 4                  | 4      |     | 1                           | 1      | 1   |
| L20      | 5                 | 1                   | 1                | 4                  | 4      |     | 1                           | 1      | 1   |
| C21      | 1                 | 1                   | 1                |                    |        |     | 1                           | 1      | 1   |
| L22      | 1                 | 1                   | 1                |                    |        |     | 1                           | 1      |     |
| L23      | 1                 | 1                   |                  |                    |        |     | 1                           |        |     |
| L24      | 1                 | 1                   |                  |                    |        |     | 1                           |        |     |
| L25      | 3                 |                     |                  |                    |        |     |                             |        |     |
| L26      | 3                 |                     |                  |                    |        |     |                             |        |     |
| L27      | 2                 |                     |                  |                    |        |     |                             |        |     |

<sup>a</sup>Target fluence level in units of  $10^{21}$  n-cm<sup>-2</sup>, 0.3 (low), 0.9 (medium), and 2.5 (high).

Table 6. Summary of discharge date and fluence of model austenitic stainless steel alloys irradiated in Halden Heavy Boiling Water Reactor.

| Capsule ID | Fluence Level | Target Fluence <sup>a</sup> | Irradiation Cycles               | Discharge Date | Discharge Fluence <sup>a</sup> |
|------------|---------------|-----------------------------|----------------------------------|----------------|--------------------------------|
| 1          | Medium        | 1.0                         | IFA 530-3 to -6, D07-004-2       | Nov. 1996      | 0.9                            |
| 4          | Low           | 0.4                         | IFA 530-3                        | Oct. 1992      | 0.3                            |
| 5          | High          | 2.5                         | IFA 530-4 to -6, D07-004-1 to -3 | Nov. 1999      | 2.0                            |
| 6          | High          | 2.5                         | IFA 530-4 to -6, D07-004-1 to -3 | Nov. 1999      | 2.0                            |
| 7          | Medium        | 1.0                         | IFA 530-4 and -6, D07-004-1      | May 1996       | 0.9                            |
| 8          | high          | 2.5                         | IFA 530-5 to -6, D07-004-1 to -3 | Nov. 1999      | 2.0                            |

<sup>a</sup>In units of  $10^{21}$  n·cm<sup>-2</sup> (E >1 MeV).

## 4 Postirradiation Testing and Posttest Characterization

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After retrieval of the irradiated specimens from the capsules, slow-strain-rate tensile tests and fractographic characterization by scanning electron microscopy were performed for each specimen listed in Table 5. In addition to the irradiated specimens, unirradiated control specimens were also tested under the same conditions.

All SSRT tests were conducted at 289°C in simulated BWR-like water that contained  $\approx 8$  ppm dissolved oxygen (DO). Conductivity and pH of the water at 23°C were kept at  $\approx 0.07$ - $0.10 \mu\text{S cm}^{-1}$  and  $\approx 6.5$ , respectively. Strain rate was held constant at  $1.65 \times 10^{-7} \text{ s}^{-1}$ . Electrochemical potential (ECP) was measured at the effluent side at regular intervals.

A limited number of specimens were selected and tested in air either at 23°C or 289-325°C in as-irradiated state by the SSRT technique at a strain rate of  $1.65 \times 10^{-7} \text{ s}^{-1}$ . The objective of such testing was to determine the susceptibility of the selected heats to irradiation-assisted intergranular cracking in an inert environment at high fluence, and to compare the result with the susceptibility of the same heats to IASCC in 289°C water.

Some high-fluence specimens, irradiated to  $2.0 \times 10^{21} \text{ n cm}^{-2}$  ( $\approx 3$  dpa) and SSRT-tested in BWR-like oxidizing water at 289°C, were selected for rapid bending tests in 23°C air. Fracture in such a test was produced by simple bending of the gauge section near the specimen fracture tip that was produced during SSRT testing in 289°C water. Surprisingly, many high-fluence specimens were susceptible to intergranular cracking in 23°C air after 1-3 times of repeated bending to  $\approx 90^\circ$ .

After SSRT testing in water or in air, fracture surface morphology was imaged in a shield scanning electron microscope (SEM). From the medium-magnification (100-150X) images, a composite fracture surface was constructed for each tested specimen. The type of fracture surface morphology (i.e., ductile, intergranular, and transgranular) was identified from the high-magnification fractographs, which varied from region to region on the fracture surface. From the composite of the whole fracture surface, fractions of intergranular (IG) and transgranular (TG) fracture morphology were measured.

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## 5 SSRT SCC Test and Fractography on Unirradiated Specimens

Yield strength and total elongation, measured on the non-irradiated control specimens in the 8-ppm-DO water at 288°C, are plotted in Figs. 3 and 4, respectively. Yield strengths of the non-irradiated solution-annealed Type 304 SS heats were 170-240 MPa, whereas yield strengths of Type 316 and 348 SS heats were significantly higher, i.e., 280-300 MPa and 330-350 MPa, respectively. Total elongation measured in water on most non-irradiated specimens was 17-59% (see Fig. 4). However, heat-to-heat variation in ductility in BWR-like water was significant, even among the same type of steel. Of the 17 heats tested in 289°C water, five heats exhibited relatively low ductility. Compositional characteristics of these five heats are denoted in Fig. 4. Two Type 304 SS heats that contained unusually high concentrations of S exhibited unusually low ductility (5-8%), i.e., Heat L7 (S 0.038 wt.%) and Heat L2 (S 0.034 wt.%). Tables 7 and 8 summarize the results obtained from non-irradiated specimens.

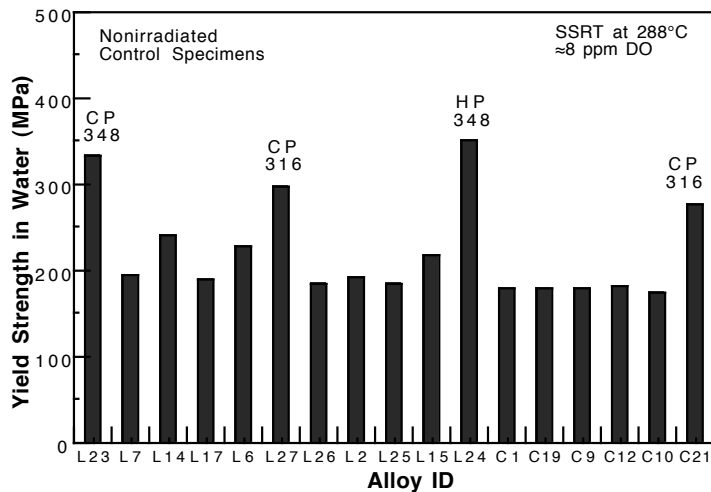


Figure 3.  
Yield strength of non-irradiated control specimens of model stainless steel alloys tested at 288°C in BWR-simulated water containing ≈8 ppm DO.

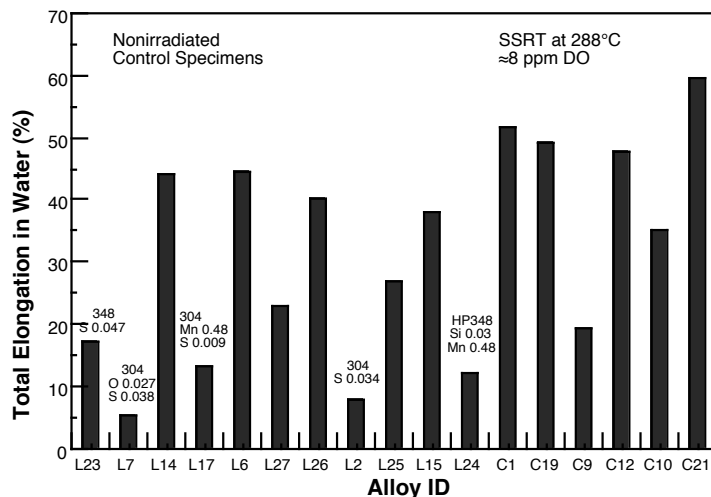


Figure 4.  
Total elongation of non-irradiated control specimens of model stainless steel alloys tested at 288°C in BWR-simulated water containing ≈8 ppm DO.

Table 7. Results of SSRT test and SEM fractography for non-irradiated control specimens of model austenitic stainless steel alloys.

| ALLOY AND SPECIMEN |          | FEEDWATER CHEMISTRY |                      |                                      |            | SSRT PARAMETERS    |                   |                    |                  | FRACTURE BEHAVIOR |           |                  |
|--------------------|----------|---------------------|----------------------|--------------------------------------|------------|--------------------|-------------------|--------------------|------------------|-------------------|-----------|------------------|
| IDENT. No.         | SSRT No. | OXYGEN CONC. (PPM)  | AVERAGE ECP (MV SHE) | COND. AT 25°C (μS·CM <sup>-1</sup> ) | PH AT 25°C | YIELD STRESS (MPA) | MAX. STRESS (MPA) | UNIFORM ELONG. (%) | TOTAL ELONG. (%) | TGSCC (%)         | IGSCC (%) | TGSCC+ IGSCC (%) |
| L23-4              | CHR-1    | 8.6                 | +228                 | 0.07                                 | 6.65       | 332                | 480               | 15.6               | 17.0             | 15                | 0         | 15               |
| L7-4               | CHR-2    | 8.0                 | +217                 | 0.07                                 | 7.37       | 195                | 370               | 2.5                | 5.2              | 20                | 0         | 20               |
| L7-B1              | CHR-7    | TESTED IN AIR       |                      |                                      |            | 282                | 676               | 42.3               | 43.9             | 0                 | 0         | 0                |
| L14-4              | CHR-3    | 8.6                 | +208                 | 0.07                                 | 7.37       | 240                | 474               | 41.8               | 44.2             | 0                 | 0         | 0                |
| L17-4              | CHR-4    | 7.5                 | +262                 | 0.06                                 | 7.09       | 189                | 412               | 11.6               | 13.3             | 60                | 0         | 60               |
| L17-B1             | CHR-19   | 7.8                 | +166                 | 0.08                                 | 6.71       | 184                | 447               | 30.1               | 31.2             | 8                 | 0         | 8                |
| L6-4               | CHR-5    | 7.9                 | +256                 | 0.08                                 | 6.85       | 227                | 545               | 43.0               | 44.5             | 0                 | 0         | 0                |
| L27-4              | CHR-6    | 9.3                 | +247                 | 0.08                                 | 6.96       | 298                | 483               | 20.6               | 22.9             | 0                 | 0         | 0                |
| L26-4              | CHR-8    | 9.4                 | +223                 | 0.07                                 | 6.65       | 184                | 596               | 38.2               | 40.2             | 0                 | 0         | 0                |
| L2-4               | CHR-9    | 8.6                 | +292                 | 0.06                                 | 6.55       | 193                | 348               | 6.6                | 7.8              | 57                | 0         | 57               |
| L25-4              | CHR-10   | 8.2                 | +239                 | 0.06                                 | 6.42       | 184                | 458               | 25.5               | 27.0             | 0                 | 0         | 0                |
| L15-4              | CHR-11   | 8.2                 | +195                 | 0.06                                 | 6.32       | 218                | 512               | 36.7               | 37.9             | 0                 | 0         | 0                |
| L24-4              | CHR-12   | 8.4                 | +200                 | 0.07                                 | 6.20       | 352                | 461               | 10.4               | 12.3             | 10                | 0         | 10               |
| C1-15              | CHR-13   | 8.1                 | +187                 | 0.07                                 | 6.33       | 179                | 498               | 49.4               | 51.7             | 0                 | 0         | 0                |
| C19-B1             | CHR-14   | 8.8                 | +179                 | 0.08                                 | 6.29       | 178                | 501               | 47.4               | 49.2             | 0                 | 0         | 0                |
| C9-B1              | CHR-15   | 8.5                 | +166                 | 0.07                                 | 6.83       | 178                | 408               | 17.4               | 19.4             | 32                | 0         | 32               |
| C12-B1             | CHR-16   | 8.5                 | +124                 | 0.07                                 | 6.18       | 182                | 511               | 46.0               | 47.6             | 0                 | 0         | 0                |
| C10-B1             | CHR-17   | 9.2                 | +145                 | 0.07                                 | 6.26       | 174                | 478               | 30.6               | 35.1             | 0                 | 0         | 0                |
| C21-9              | CHR-18   | 9.2                 | +187                 | 0.07                                 | 6.41       | 277                | 455               | 48.9               | 59.5             | 0                 | 0         | 0                |

Table 8. Composition (wt.%) of non-irradiated control specimens of model austenitic stainless steels correlated with results of SSRT test and SEM fractography.

| Alloy ID | Ni    | Si   | P     | S     | Mn   | C     | N     | Cr    | Mo or Nb | O wppm | YS (MPa) | UTS (MPa) | UE (%) | TE (%) | TG (%) | IG (%) |
|----------|-------|------|-------|-------|------|-------|-------|-------|----------|--------|----------|-----------|--------|--------|--------|--------|
| L23      | 12.04 | 0.68 | 0.030 | 0.047 | 0.96 | 0.043 | 0.092 | 17.30 | Nb 1.06  | 93     | 332      | 480       | 15.6   | 17.0   | 15     | 0      |
| L7       | 10.60 | 0.18 | 0.040 | 0.038 | 1.02 | 0.007 | 0.111 | 15.40 | -        | 274    | 195      | 370       | 2.5    | 5.2    | 20     | 0      |
| L14      | 7.93  | 1.49 | 0.080 | 0.002 | 1.76 | 0.107 | 0.028 | 15.00 | -        | 45     | 240      | 474       | 41.8   | 44.2   | 0      | 0      |
| L17      | 8.00  | 0.66 | 0.090 | 0.009 | 0.48 | 0.061 | 0.078 | 15.30 | -        | 90     | 189      | 412       | 11.6   | 13.3   | 60     | 0      |
| L6       | 10.00 | 1.90 | 0.020 | 0.005 | 1.13 | 0.096 | 0.087 | 17.10 | -        | 58     | 227      | 515       | 43.0   | 44.5   | 0      | 0      |
| L27      | 10.30 | 0.96 | 0.040 | 0.002 | 0.97 | 0.057 | 0.019 | 15.30 | Mo 2.01  | 58     | 298      | 483       | 20.6   | 22.9   | 0      | 0      |
| L26      | 8.09  | 0.79 | 0.004 | 0.002 | 0.91 | 0.070 | 0.089 | 17.20 | -        | 80     | 184      | 506       | 38.2   | 40.2   | 0      | 0      |
| L2       | 10.50 | 0.82 | 0.080 | 0.034 | 1.58 | 0.074 | 0.102 | 17.02 | -        | 66     | 193      | 348       | 6.6    | 7.8    | 57     | 0      |
| L25      | 8.93  | 0.92 | 0.020 | 0.008 | 1.54 | 0.019 | 0.095 | 17.20 | -        | 85     | 184      | 458       | 25.5   | 27.0   | 0      | 0      |
| L15      | 8.00  | 1.82 | 0.010 | 0.013 | 1.07 | 0.020 | 0.085 | 17.80 | -        | 110    | 218      | 512       | 36.7   | 37.9   | 10     | 0      |
| L24      | 12.30 | 0.03 | 0.007 | 0.005 | 0.48 | 0.031 | 0.002 | 16.90 | Nb 1.72  | 129    | 352      | 461       | 10.4   | 12.3   | 0      | 0      |
| C1       | 8.12  | 0.50 | 0.038 | 0.002 | 1.00 | 0.060 | 0.060 | 18.11 | -        | 102    | 179      | 498       | 49.4   | 51.7   | 0      | 0      |
| C19      | 8.08  | 0.45 | 0.031 | 0.003 | 0.99 | 0.060 | 0.070 | 18.21 | -        | 200    | 178      | 501       | 47.4   | 49.2   | 0      | 0      |
| C9       | 8.75  | 0.39 | 0.013 | 0.013 | 1.72 | 0.062 | 0.065 | 18.48 | -        | 102    | 178      | 408       | 17.4   | 19.4   | 32     | 0      |
| C12      | 8.23  | 0.47 | 0.018 | 0.002 | 1.00 | 0.060 | 0.070 | 18.43 | -        | -      | 182      | 511       | 46.0   | 47.6   | 0      | 0      |
| C10      | 8.13  | 0.55 | 0.033 | 0.002 | 1.00 | 0.060 | 0.086 | 18.19 | -        | 74     | 174      | 478       | 30.6   | 35.1   | 0      | 0      |
| C21      | 10.24 | 0.51 | 0.034 | 0.001 | 1.19 | 0.060 | 0.020 | 16.28 | Mo 2.08  | 112    | 277      | 455       | 48.9   | 59.5   | 0      | 0      |

Load-elongation behavior of Type 304 SS Heat L7 exhibited significantly deleterious effects of water even in the nonirradiated state, as shown in Fig. 5. Approximately 20% of the fracture surface of the specimen was characterized by transgranular fracture morphology that was mostly covered with unidentified corrosion debris (Fig. 6). Although it was suspected that such corrosion debris contained a high concentration of S, positive identification of the debris could not be determined.

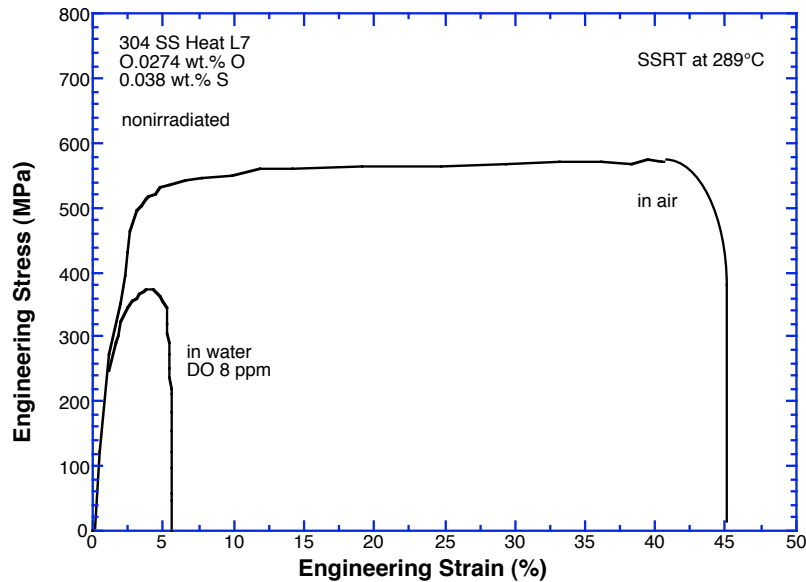


Figure 5. Load vs. elongation of non-irradiated Type 304 SS that contains unusually high concentrations of O (0.0274 wt.%) and S (0.038 wt.%); tests at 289°C in air and BWR-simulated water containing ≈8 ppm DO.

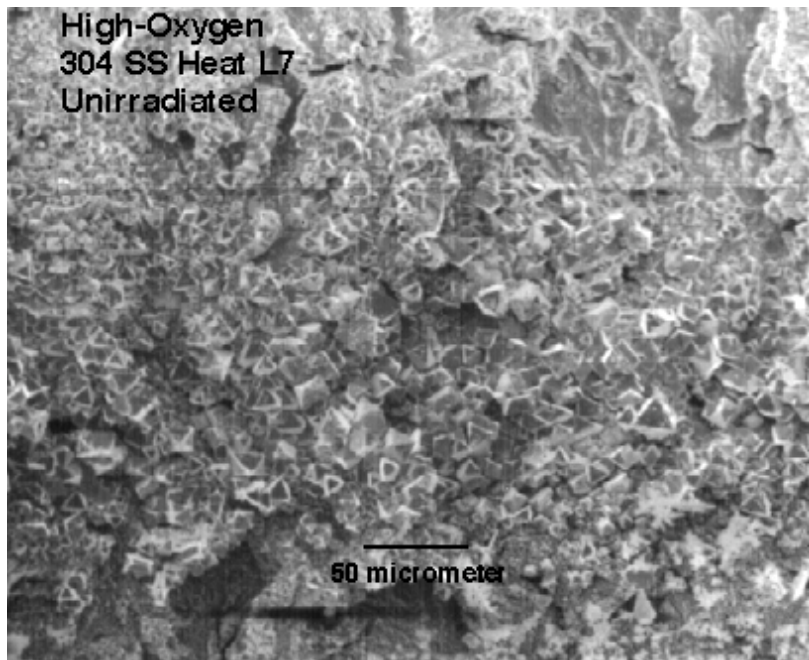


Figure 6. Corrosion debris formed on non-ductile fracture surface of non-irradiated Type 304 SS Heat L7 that contains unusually high concentrations of O and S; SSRT test at 288°C in BWR-like water containing ≈8 ppm DO.

The five heats that exhibited relatively low ductility were characterized by relatively high susceptibility to TGSCC. Other heats were very ductile and exhibited predominantly ductile dimple fracture surface morphology. However, no evidence of susceptibility to IGSCC was observed in any of the 17 heats that were tested in the BWR-like water at 289°C in the nonirradiated state.



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## 6 SSRT SCC Test and Fractography on Low-Fluence Specimens ( $\approx 0.4$ dpa)

### 6.1 Tabulation of Test Results

Results of SSRT tests on specimens irradiated to  $\approx 0.3 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV) ( $\approx 0.4$  dpa) are summarized in Tables 9 and 10. Feedwater chemistry (i.e., DO, ECP, conductivity, and pH) and SSRT behavior in water (i.e., 0.2%-offset yield strength, maximum strength, uniform strain, and total strain) are given in Table 9. Also shown in these tables are results of SEM fractographic analysis of the failure mode (i.e., ductile, intergranular, and transgranular fracture surface morphology) of the specimens. In Table 10, the results of SSRT and SEM fractographic analysis (percent TGSCC and percent IGSCC) are correlated with the compositions of the low-fluence specimens.

Table 9. Results of SSRT tests in 289°C water (DO  $\approx 8$  ppm) and SEM fractography of steels irradiated to  $\approx 0.3 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV).

| Specimen | Oxygen | Average | Cond.    | pH                        | Yield | Max.   | Uniform | Total  | Fracture |       |          |
|----------|--------|---------|----------|---------------------------|-------|--------|---------|--------|----------|-------|----------|
| Ident.   | SSRT   | Conc.   | ECP      | at 25°C                   | at    | Stress | Stress  | Elong. | Elong.   | TGSCC | IGSCC    |
| No.      | No.    | (ppm)   | (mV SHE) | ( $\mu\text{S cm}^{-1}$ ) | 25°C  | (MPa)  | (MPa)   | (%)    | (%)      | (%)   | (%)      |
| C1-1     | HR-1   | 8.3     | +184     | 0.07                      | 7.03  | 490    | 680     | 13.4   | 16.6     | 4     | 0        |
| L5-1     | HR-2   | 9.7     | +208     | 0.07                      | 6.89  | 413    | 539     | 29.5   | 32.7     | 2     | 2        |
| L22-1    | HR-3   | 8.0     | +236     | 0.07                      | 6.80  | 360    | 596     | 6.6    | 9.4      | 50    | 15       |
| C3-1     | HR-4   | 8.7     | +161     | 0.07                      | 6.68  | 338    | 491     | 27.7   | 31.6     | 5     | 0        |
| C16-1    | HR-5   | 8.3     | +204     | 0.08                      | 6.74  | 370    | 527     | 17.6   | 20.6     | 2     | 0        |
| L4-1     | HR-6   | 9.0     | +202     | 0.08                      | 6.70  | 367    | 542     | 19.7   | 22.3     | 46    | 0        |
| L18-1    | HR-7   | 9.0     | +203     | 0.08                      | 6.33  | 503    | 572     | 6.3    | 8.8      | 54    | 0        |
| C10-1    | HR-8   | 8.2     | +174     | 0.07                      | 6.35  | 523    | 640     | 17.4   | 18.9     | 6     | 0        |
| C21-1    | HR-9   | 8.1     | +149     | 0.08                      | 6.49  | 480    | 620     | 15.9   | 19.4     | 4     | 0        |
| L11-1    | HR-10  | 9.0     | +157     | 0.08                      | 6.17  | 487    | 599     | 2.3    | 3.8      | 62    | 0        |
| L13-1    | HR-11  | 8.7     | +164     | 0.08                      | 6.17  | 248    | 461     | 22.1   | 24.8     | 18    | 0        |
| L20-1    | HR-12  | 8.4     | +174     | 0.07                      | 6.20  | 454    | 552     | 2.9    | 5.1      |       | dendrite |
| C19-1    | HR-13  | 9.5     | +132     | 0.12                      | 6.36  | 554    | 682     | 10.5   | 14.7     | 7     | 0        |
| C9-1     | HR-14  | 8.0     | +192     | 0.11                      | 6.30  | 522    | 607     | 13.4   | 14.6     | 24    | 0        |
| C12-1    | HR-15  | 9.0     | +195     | 0.08                      | 6.40  | 404    | 589     | 20.4   | 24.2     | 5     | 0        |
| L8-1     | HR-16  | 9.0     | +215     | 0.08                      | 6.60  | 411    | 571     | 15.6   | 17.9     | 64    | 0        |

### 6.2 Susceptibility to IGSCC

Even at the low damage level of  $\approx 0.4$  dpa, heat-to-heat variations in susceptibility to IGSCC and TGSCC were significant among the steels tested and listed in Table 10. A high-purity Heat L22 of Type 316L SS, which contains very low concentrations of Si (0.024 wt.%) and C (0.003 wt.%), exhibited relatively low ductility and the highest susceptibility to IASCC (i.e., the highest percent IGSCC) among the 16 tested heats. The high-purity Heat L20 of Type 304L SS, which contains very low concentrations of Si (0.017 wt.%) and C (0.002 wt.%), also exhibited an indication of slight susceptibility to IGSCC. Susceptibility of all other heats to IGSCC was insignificant (see Fig. 7).

At this low fluence, the limited observation in Fig. 7 seems to indicate that very low concentrations of Si and C are deleterious from the standpoint of IASCC. The susceptible behavior of the two high-purity heats in the figure appears to be consistent with similar behavior of BWR neutron absorber tubes fabricated from

high-purity 304L SSs which exhibited relatively higher susceptibility during SSRT tests performed after irradiation to  $0.2 \times 10^{21}$  n·cm<sup>-2</sup> to  $1.4 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV).<sup>19</sup> However, field failure by IASCC in a BWR usually occurs at higher fluence levels, e.g.,  $>1.5 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV). Therefore, a comprehensive database at higher fluence levels is necessary to be able to evaluate the roles of Si, C, and other impurity elements.

Figure 8 shows that limited intergranular fracture surface morphology is surrounded by transgranular morphology in Heat L22, indicating that transgranular fracture made a transition to intergranular fracture. Heat L22 also exhibited relatively high susceptibility to IGSCC after irradiation to higher fluence (Section 7).

Table 10. Composition (wt.%) of steels irradiated to  $\approx 0.3 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV) correlated with results of SSRT tests and SEM fractography.

| Spec. ID          | Ni    | Si    | P     | S     | Mn   | C     | N     | Cr    | Others                             | YS (MPa) | UTS (MPa) | UE (%) | TE (%) | TG (%)   | IG (%) |
|-------------------|-------|-------|-------|-------|------|-------|-------|-------|------------------------------------|----------|-----------|--------|--------|----------|--------|
| C1-1              | 8.12  | 0.50  | 0.038 | 0.002 | 1.00 | 0.060 | 0.060 | 18.11 | O 0.0102                           | 490      | 680       | 13.4   | 16.6   | 4        | 0      |
| L5-1 <sup>a</sup> | 9.66  | 0.90  | 0.113 | 0.028 | 0.47 | 0.006 | 0.033 | 21.00 | O 0.0068<br>$\approx 3\% \delta^a$ | 413      | 539       | 29.5   | 32.7   | 2        | 2      |
| L22-1             | 13.30 | 0.024 | 0.015 | 0.004 | 0.40 | 0.003 | 0.001 | 16.10 | O 0.0103<br>Mo 2.04                | 360      | 596       | 6.6    | 9.4    | 50       | 15     |
| C3-1              | 8.91  | 0.46  | 0.019 | 0.004 | 1.81 | 0.016 | 0.083 | 18.55 | -                                  | 338      | 491       | 27.7   | 31.6   | 5        | 0      |
| C16-1             | 12.90 | 0.38  | 0.014 | 0.002 | 1.66 | 0.020 | 0.011 | 16.92 | O 0.0157<br>Mo 2.30                | 370      | 527       | 17.6   | 20.6   | 2        | 0      |
| L4-1              | 10.20 | 0.94  | 0.031 | 0.010 | 1.75 | 0.110 | 0.002 | 15.80 | O 0.0037                           | 367      | 542       | 19.7   | 22.3   | 46       | 0      |
| L18-1             | 8.13  | 0.14  | 0.016 | 0.033 | 1.13 | 0.080 | 0.001 | 18.00 | O 0.0055                           | 503      | 572       | 6.3    | 8.8    | 54       | 0      |
| C10-1             | 8.13  | 0.55  | 0.033 | 0.002 | 1.00 | 0.060 | 0.086 | 18.19 | O 0.0074                           | 523      | 640       | 17.4   | 18.9   | 6        | 0      |
| C21-1             | 10.24 | 0.51  | 0.034 | 0.001 | 1.19 | 0.060 | 0.020 | 16.28 | O 0.0112<br>Mo 2.08                | 480      | 620       | 15.9   | 19.4   | 4        | 0      |
| L11-1             | 8.15  | 0.47  | 0.097 | 0.009 | 1.02 | 0.014 | 0.004 | 17.40 | O 0.0071                           | 487      | 599       | 2.3    | 3.8    | 62       | 0      |
| L13-1             | 8.18  | 1.18  | 0.027 | 0.022 | 0.36 | 0.026 | 0.001 | 17.40 | O 0.0042                           | 248      | 461       | 22.1   | 24.8   | 18       | 0      |
| L20-1             | 8.91  | 0.017 | 0.010 | 0.004 | 0.41 | 0.002 | 0.002 | 18.10 | O 0.0940                           | 454      | 552       | 2.9    | 5.1    | dendrite |        |
| C19-1             | 8.08  | 0.45  | 0.031 | 0.003 | 0.99 | 0.060 | 0.070 | 18.21 | O 0.0200                           | 554      | 682       | 10.5   | 14.7   | 7        | 0      |
| C9-1              | 8.75  | 0.39  | 0.013 | 0.013 | 1.72 | 0.062 | 0.065 | 18.48 | O 0.0102                           | 522      | 607       | 13.4   | 14.6   | 24       | 0      |
| C12-1             | 8.23  | 0.47  | 0.018 | 0.002 | 1.00 | 0.060 | 0.070 | 18.43 | -                                  | 404      | 589       | 20.4   | 24.2   | 5        | 0      |
| L8-1              | 10.20 | 0.15  | 0.093 | 0.010 | 1.85 | 0.041 | 0.001 | 18.30 | O 0.0059                           | 411      | 571       | 15.6   | 17.8   | 64       | 0      |

<sup>a</sup>Heat L5 contains  $\approx 3$  vol.% globular delta ferrite.

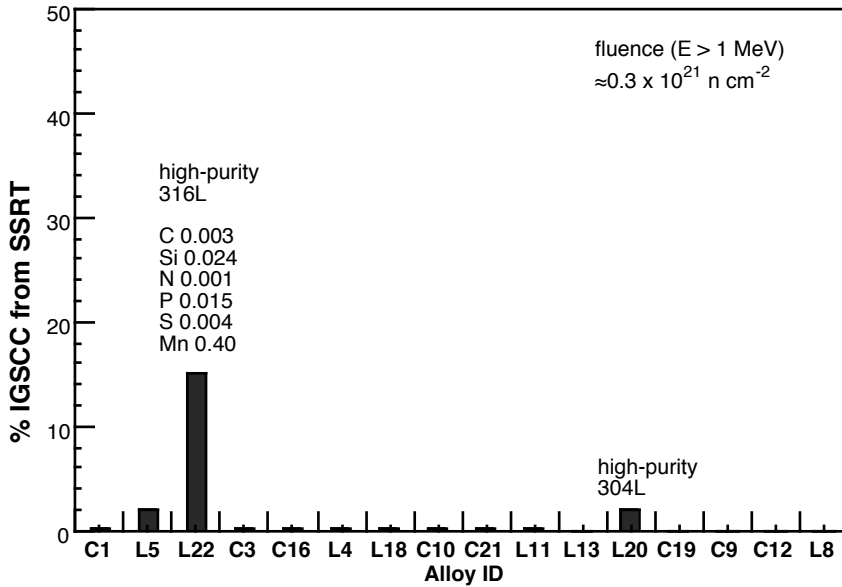


Figure 7. Percent IGSCC of model SS alloys irradiated in He in the Halden reactor to fluence of  $\approx 0.3 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ) and tested at  $289^\circ\text{C}$  in BWR-like water.

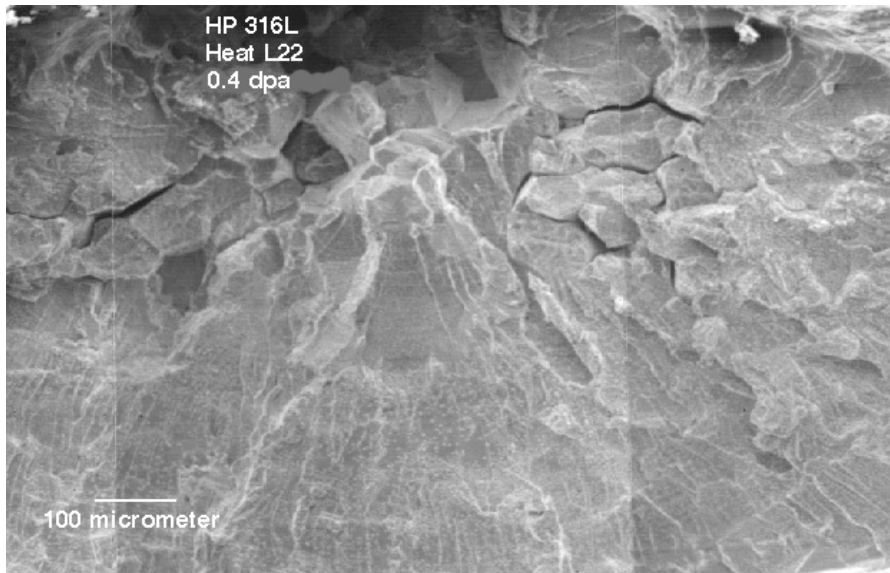


Figure 8. Limited intergranular fracture surface surrounded by transgranular surface in 316L-like SS Heat L22 irradiated to  $\approx 0.3 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ,  $\approx 0.4 \text{ dpa}$ ) and tested at  $289^\circ\text{C}$  in BWR-like water.

### 6.3 Effect of Si on Strength

The relatively higher susceptibility of the high-purity heat of Type 316L SS (i.e., Heat L22) when compared with the commercial counterpart (Heat C21) is of particular interest. This behavior is similar to that observed for BWR neutron absorber tubes fabricated from high- and commercial-purity heats of Type 304 SS.<sup>23</sup> In another investigation similar to that of the present study, Jensen and Ljunberg irradiated U-notched rod specimens that had been fabricated from two heats of Type 316 SS, then conducted postirradiation SSRT tests in a BWR loop under conditions of normal oxidizing water chemistry.<sup>30</sup> In their study, a commercial-purity heat of Type 316 SS (Heat K) was resistant to IASCC, whereas a high-purity heat of Type 316L SS (Heat F) was susceptible. The two relatively more susceptible heats (i.e., Heats L22 and F) are characterized by an unusually low Si concentration of  $<0.26 \text{ wt.}\%$ , whereas the two relatively more resistant heats (i.e., Heats C21 and K) contain a higher Si concentration of  $>0.5 \text{ wt.}\%$ .

An interesting observation from Table 10 is the effect of Si content on the alloy strength that was measured in water before and after irradiation. Yield strength of unirradiated commercial heats of Types 304 and 304L SS, measured in water, was similar to that measured in air,<sup>1</sup> i.e., 174-182 MPa vs. 170-175 MPa, indicating that the effect of water is insignificant. The yield and maximum strengths (in water) are plotted in Fig. 9 as function of Si content for the unirradiated and low-fluence specimens. Before irradiation, 304- and 304L-type steels did not exhibit a systematic effect of Si content. However, after irradiation to  $\approx 0.3 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ), yield and maximum strengths decreased as Si content increased to 0.8-1.2 wt.%.

The influence of Si content on the material strength indicates that, at high concentrations, Si atoms suppress formation of some types of irradiation-induced hardening centers and contribute to greater ductility, at least at low fluence levels. Tsukada et al.<sup>29</sup> irradiated two SSRT specimens of high-purity Type 304L SS (C 0.003, Si 0.01, Mn 1.36, P 0.001, S 0.0014, and N 0.0014 wt.%) in He at  $\approx 240^\circ\text{C}$  in the JRR-3 reactor to a fluence of  $\approx 0.67 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ). One specimen had been doped with  $\approx 0.69 \text{ wt.}\%$  Si and the other had not been doped. After SSRT testing of the irradiated specimens at  $\approx 300^\circ\text{C}$  in high-purity water (DO  $\approx 32 \text{ ppm}$ ), they observed that the Si-doped specimen exhibited significantly greater ductility than the undoped specimen. Silicon effect at 1.3 dpa is discussed in Section 7.4.

#### **6.4 Correlation of S Content and Susceptibility to TGSCC of 304 and 304L SSs**

For the fluence level of  $\approx 0.3 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ), SSRT tests in  $289^\circ\text{C}$  water and fractographic analysis were performed for 11 heats of Type 304 or 304L SS, three heats of Type 316 or 316L SS, and one heat of austenitic-ferritic duplex SS. Susceptibilities to TGSCC of the 11 heats of Type 304 or 304L SS varied strongly from heat to heat. However, it was found that bulk S content correlates reasonably well with percent TGSCC, as is shown in Fig. 10. Heats that contain  $\geq 0.009 \text{ wt.}\%$  S exhibited high susceptibility to TGSCC, whereas heats that contain  $\leq 0.004 \text{ wt.}\%$  S exhibited low susceptibility to TGSCC.

For a given heat of all types of austenitic SSs at low fluence, significant susceptibility to TGSCC is often observed after an SSRT test, even though susceptibility to IGSCC is insignificant. As fluence level increases, the susceptibility to TGSCC peaks at a certain fluence specific to the heat, and at the same time, the susceptibility to IGSCC increases at the expense of TGSCC. Considering this trend, the good correlation observed between S content and percent TGSCC in Fig. 10 is considered an important indication that may provide useful insight into the susceptibility to IGSCC at higher fluence levels.

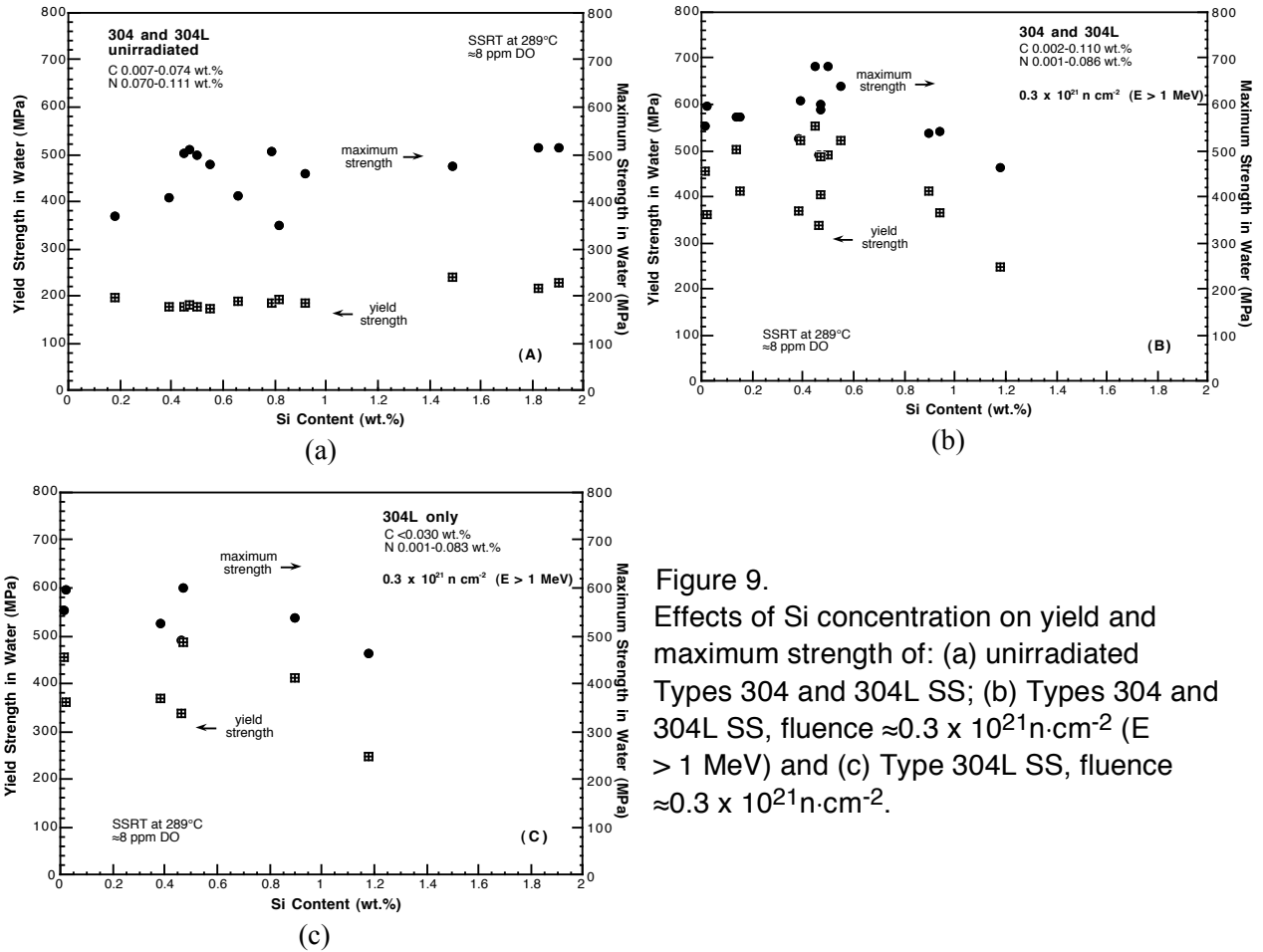


Figure 9. Effects of Si concentration on yield and maximum strength of: (a) unirradiated Types 304 and 304L SS; (b) Types 304 and 304L SS, fluence  $\approx 0.3 \times 10^{21} \text{ n cm}^{-2}$  ( $E > 1 \text{ MeV}$ ) and (c) Type 304L SS, fluence  $\approx 0.3 \times 10^{21} \text{ n cm}^{-2}$ .

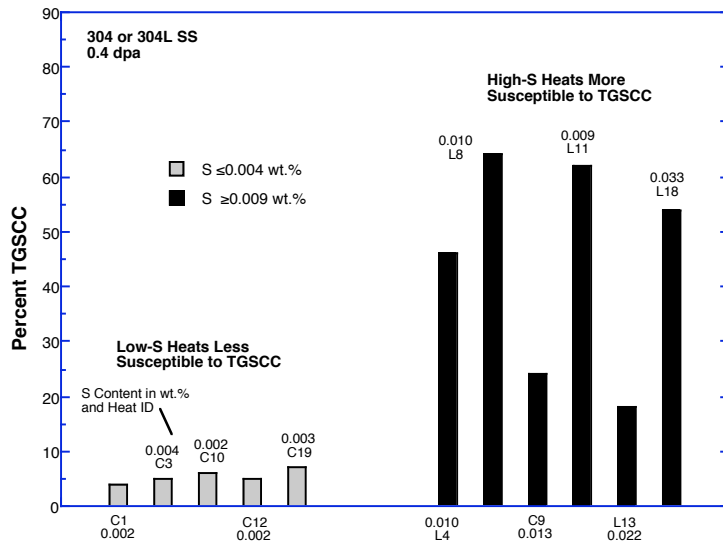


Figure 10. Effect of S content on the susceptibility to TGSCC of Type 304 or 304L heats, irradiated to fluence of  $\approx 0.3 \times 10^{21} \text{ n cm}^{-2}$  ( $E > 1 \text{ MeV}$ ) ( $\approx 0.4 \text{ dpa}$ ) and tested at 289°C in BWR-like water.

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## 7. SSRT Test and SEM Fractography on Medium-Fluence Specimens ( $\approx 1.3$ dpa)

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### 7.1 Tabulation of Test Results

Slow-strain-rate tensile tests in 289°C BWR-like water and SEM fractographic analysis were performed on 26 medium-fluence specimens irradiated to  $\approx 0.9 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV) ( $\approx 1.3$  dpa). Feedwater chemistry (i.e., DO, ECP, conductivity, and pH) and results from SSRT testing (i.e., 0.2%-offset yield strength, maximum strength, uniform plastic strain, and total plastic strain) are summarized in Table 11. Also shown in the table are results of SEM fractographic analysis of the failure mode (i.e., intergranular and transgranular fracture surface morphology) of the specimens. In Table 12, the results of SSRT and SEM fractographic analysis are correlated with compositional characteristics of the specimens.

For Heat L20, which contains an unusually high concentration of O (0.094 wt.) and a dendritic grain structure, fractographic analysis was not performed, although SSRT testing was conducted. This heat, which is not a prototypic austenitic SS, exhibited poor ductility when compared with other steels, and its behavior was excluded in the analyses of SCC test results.

### 7.2 Heat-to-Heat Variations in Susceptibility to TGSCC and IGSCC

As an example, effects of fluence and test environment on load-elongation behavior are shown for Type 304 SS commercial Heat C19 in Fig. 11. As fluence increased from  $\approx 0.3 \times 10^{21}$  n·cm<sup>-2</sup> to  $\approx 0.9 \times 10^{21}$  n·cm<sup>-2</sup>, maximum strength increased slightly (from 690 MPa to 800 MPa). In contrast, total elongation decreased significantly (from 1.45% to 0.7%).

Figures 12 and 13 show, respectively, heat-to-heat variations in percent TGSCC and IGSCC for the two tested fluence levels, i.e.,  $\approx 0.3 \times 10^{21}$  n·cm<sup>-2</sup> and  $\approx 0.9 \times 10^{21}$  n·cm<sup>-2</sup>. As fluence increased, percent TGSCC decreased significantly, and, at the same time, the percent IGSCC increased significantly for many heats, e.g., L22, L18, L11, and L8. The fact that the IGSCC fracture surfaces in these alloys were surrounded by TGSCC fracture surfaces indicates that an IGSCC fracture surface emerges from a portion of a TGSCC fracture surface. In addition, for many alloys, high susceptibility to TGSCC is a precursor to susceptibility to IGSCC at a higher fluence. However, the threshold fluence for the transition from TGSCC to IGSCC differed significantly from alloy to alloy.



Table 11. Results of SSRT test and fractography for model austenitic stainless steels irradiated to  $\approx 0.9 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ) ( $\approx 1.3 \text{ dpa}$ ).

| Speci. ID | SSRT No. | DO (ppm) | ECP (mV SHE) | Cond. at 25°C ( $\mu\text{S cm}^{-1}$ ) | pH at 25°C | Yield Stress (MPa) | Max. Stress (MPa) | Uniform Elong. (%) | Total Elong. (%) | TGSCC (%) | IGSCC (%) | TG+IG SCC (%) |
|-----------|----------|----------|--------------|---|------------|--------------------|-------------------|--------------------|------------------|-----------|-----------|---------------|
| L22-02    | HR-17    | 8.0      | +181         | 0.08                                    | 6.77       | 475                | 549               | 4.20               | 5.82             | 30        | 35        | 65            |
| L11-02    | HR-18    | 8.0      | +191         | 0.08                                    | 6.55       | 820                | 856               | 0.43               | 1.65             | 50        | 14        | 64            |
| L18-02    | HR-19    | 8.0      | +193         | 0.10                                    | 6.07       | 710                | 755               | 3.98               | 5.05             | 38        | 14        | 52            |
| L20-02    | HR-28    |          | test in      | 289°C air                               |            | 826                | 845               | 0.31               | 2.09             | dendritic |           | structure     |
| L20-05    | HR-26    | 9.0      | +182         | 0.09                                    | 6.32       | 670                | 743               | 0.37               | 1.03             | dendritic |           | structure     |
| L20-06    | HR-27    | 8.0      | +274         | 0.07                                    | 6.05       | 632                | 697               | 0.85               | 2.72             | 0         | 0         | 0             |
| C9-02     | HR-21    | 8.0      | +240         | 0.07                                    | 6.47       | 651                | 679               | 1.42               | 2.50             | 62        | 22        | 84            |
| L17-02    | HR-22    | 8.0      | +198         | 0.07                                    | 6.42       | 574                | 654               | 2.02               | 3.08             | 44        | 41        | 85            |
| L7-02     | HR-23    | 8.0      | +215         | 0.07                                    | 6.03       | 553                | 561               | 0.24               | 2.44             | 38        | 54        | 92            |
| C10-02    | HR-24    | 7.0      | +221         | 0.07                                    | 5.26       | 651                | 706               | 6.35               | 9.25             | 14        | 0         | 14            |
| C3-02     | HR-25    | 8.0      | +240         | 0.07                                    | 6.34       | 632                | 668               | 16.72              | 19.74            | 9         | 4         | 13            |
| C19-05    | HR-58    |          | test in      | 23°C air                                |            | 1130               | 1260              | 15.50              | 15.70            | -         | -         | -             |
| C19-02    | HR-30    |          | test in      | 289°C air                               |            | 888                | 894               | 6.41               | 10.21            | 1         | 0         | 1             |
| C19-04    | HR-31    | 8.0      | +252         | 0.07                                    | 6.18       | 750                | 769               | 6.06               | 8.79             | 1         | 0         | 1             |
| L6-02     | HR-32    | 8.0      | +250         | 0.07                                    | 6.40       | 493                | 546               | 2.45               | 3.77             | 8         | 27        | 35            |
| L14-02    | HR-33    | 8.0      | +246         | 0.08                                    | 6.07       | 649                | 684               | 1.90               | 4.67             | 84        | 2         | 86            |
| L13-02    | HR-34    | 7.0      | +222         | 0.09                                    | 6.85       | 602                | 624               | 1.67               | 4.95             | 55        | 12        | 67            |
| L4-02     | HR-35    | 7.0      | +259         | 0.08                                    | 6.54       | 634                | 680               | 1.07               | 2.02             | 58        | 12        | 70            |
| L5-02     | HR-36    | 7.0      | +243         | 0.07                                    | 6.85       | 665                | 725               | 3.07               | 4.57             | 3         | 5         | 8             |
| C16-02    | HR-37    | 7.0      | +230         | 0.07                                    | 6.62       | 562                | 618               | 11.99              | 15.80            | 7         | 1         | 8             |
| L8-02     | HR-38    | 8.0      | +242         | 0.07                                    | 6.57       | 838                | 838               | 0.12               | 3.12             | 15        | 22        | 37            |
| C21-02    | HR-39    | 8.0      | +231         | 0.08                                    | 6.21       | 643                | 716               | 15.38              | 18.30            | 1         | 2         | 3             |
| L2-02     | HR-40    | 7.0      | +239         | 0.07                                    | 7.11       | 839                | 849               | 0.88               | 1.56             | 31        | 11        | 42            |
| L24-02    | HR-41    | 8.0      | +239         | 0.06                                    | 6.40       | 725                | 725               | 0.15               | 2.45             | 2         | 1         | 3             |
| L23-02    | HR-42    | 7.0      | +237         | 0.08                                    | 6.60       | 787                | 818               | 0.38               | 1.24             | 3         | 24        | 27            |
| C12-02    | HR-43    | 7.0      | +227         | 0.07                                    | 6.19       | 747                | 756               | 14.96              | 18.57            | 4         | 0         | 4             |
| C1-02     | HR-44    | 8.0      | +229         | 0.07                                    | 6.30       | 707                | 763               | 13.36              | 17.04            | 2         | 0         | 2             |

### 7.3 Effect of Sulfur

The information given in Tables 11 and 12 led us to carefully examine the relationship between fracture properties and compositional characteristics of the alloys in an effort to identify the alloying and impurity elements that strongly influence stress corrosion cracking behavior. Of the steel heats tested and listed in Table 12, 17 heats are Type 304 or 304L, three heats are Type 316 or 316L, two heats are Type 348 or 348L, and one heat is duplex steel that contains  $\approx 3\%$  delta ferrite.

Of the 17 heats of Types 304 or 316 SS, steels that contain low concentrations of S ( $< 0.004 \text{ wt.}\%$  S) exhibited high resistance to IGSCC (see Fig. 14) and high uniform and total elongation (see Fig. 15), whereas steels that contain relatively higher concentrations of S ( $> 0.005 \text{ wt.}\%$  S) exhibited poor resistance to IGSCC and low uniform and total elongation.

The observations summarized in Figs. 14 and 15 indicate strongly that, for Types 304 and 304L SS, a high concentration of S ( $> 0.005 \text{ wt.}\%$ ) is significantly detrimental, and that a sufficiently low concentration of S (e.g.,  $< 0.004 \text{ wt.}\%$ ) is beneficial to render good resistance to IASCC at  $\approx 1.3 \text{ dpa}$ .

Table 12. Composition (wt.%) of stainless steels irradiated to  $\approx 0.9 \times 10^{21}$  n·cm<sup>-2</sup> (E > 1 MeV) (1.3 dpa) correlated with results of SSRT test and fractography.

| Spec. ID           | Ni    | Si    | P     | S     | Mn   | C     | N     | Cr    | O      | Other              | YS (MPa) | UTS (MPa) | UE (%) | TE (%) | TG (%)   | IG (%) |
|--------------------|-------|-------|-------|-------|------|-------|-------|-------|--------|--------------------|----------|-----------|--------|--------|----------|--------|
| L22-02             | 13.30 | 0.024 | 0.015 | 0.004 | 0.40 | 0.003 | 0.001 | 16.10 | 0.0102 | Mo 2.04            | 475      | 549       | 4.20   | 5.82   | 30       | 35     |
| L11-02             | 8.15  | 0.47  | 0.097 | 0.009 | 1.02 | 0.014 | 0.004 | 17.40 | 0.0071 | –                  | 820      | 856       | 0.43   | 1.65   | 50       | 14     |
| L18-02             | 8.13  | 0.14  | 0.016 | 0.033 | 1.13 | 0.080 | 0.001 | 18.00 | 0.0055 | –                  | 710      | 755       | 3.98   | 5.05   | 38       | 14     |
| L20-05             | 8.91  | 0.017 | 0.010 | 0.004 | 0.41 | 0.002 | 0.002 | 18.10 | 0.0940 | –                  | 670      | 743       | 0.37   | 1.03   | dendrite |        |
| L20-06             | 8.91  | 0.017 | 0.010 | 0.004 | 0.41 | 0.002 | 0.002 | 18.10 | 0.0940 | –                  | 632      | 697       | 0.85   | 2.72   | dendrite |        |
| C9-02              | 8.75  | 0.39  | 0.013 | 0.013 | 1.72 | 0.062 | 0.065 | 18.48 | 0.0102 | –                  | 651      | 679       | 1.42   | 2.50   | 62       | 22     |
| L17-02             | 8.00  | 0.66  | 0.090 | 0.009 | 0.48 | 0.061 | 0.078 | 15.30 | 0.0090 | –                  | 574      | 654       | 2.02   | 3.08   | 44       | 41     |
| L7-02              | 10.60 | 0.18  | 0.040 | 0.038 | 1.02 | 0.007 | 0.111 | 15.40 | 0.0274 | –                  | 553      | 561       | 0.24   | 2.44   | 38       | 54     |
| C10-02             | 8.13  | 0.55  | 0.033 | 0.002 | 1.00 | 0.060 | 0.086 | 18.19 | 0.0074 | –                  | 651      | 706       | 6.35   | 9.25   | 14       | 0      |
| C3-02              | 8.91  | 0.46  | 0.019 | 0.004 | 1.81 | 0.016 | 0.083 | 18.55 | –      | –                  | 632      | 668       | 16.72  | 19.74  | 9        | 4      |
| C19-04             | 8.08  | 0.45  | 0.031 | 0.003 | 0.99 | 0.060 | 0.070 | 18.21 | 0.0200 | –                  | 750      | 769       | 6.06   | 8.79   | 1        | 0      |
| L6-02              | 10.00 | 1.90  | 0.020 | 0.005 | 1.13 | 0.096 | 0.087 | 17.10 | 0.0058 | –                  | 493      | 546       | 2.45   | 3.77   | 8        | 27     |
| L14-02             | 7.93  | 1.49  | 0.080 | 0.002 | 1.76 | 0.107 | 0.028 | 15.00 | 0.0045 | –                  | 649      | 684       | 1.90   | 4.67   | 84       | 2      |
| L13-02             | 8.18  | 1.18  | 0.027 | 0.022 | 0.36 | 0.026 | 0.001 | 17.40 | 0.0042 | –                  | 602      | 624       | 1.67   | 4.95   | 55       | 12     |
| L4-02              | 10.20 | 0.94  | 0.031 | 0.010 | 1.75 | 0.110 | 0.002 | 15.80 | 0.0037 | –                  | 634      | 680       | 1.07   | 2.02   | 58       | 12     |
| L5-02 <sup>a</sup> | 9.66  | 0.90  | 0.113 | 0.028 | 0.47 | 0.006 | 0.033 | 21.00 | 0.0068 | a <sub>3</sub> % δ | 665      | 725       | 3.07   | 4.57   | 3        | 5      |
| C16-02             | 12.90 | 0.38  | 0.014 | 0.002 | 1.66 | 0.020 | 0.011 | 16.92 | 0.0157 | Mo 2.30            | 562      | 618       | 11.99  | 15.80  | 7        | 1      |
| L8-02              | 10.20 | 0.15  | 0.093 | 0.010 | 1.85 | 0.041 | 0.001 | 18.30 | 0.0059 | –                  | 838      | 838       | 0.12   | 3.12   | 15       | 22     |
| C21-02             | 10.24 | 0.51  | 0.034 | 0.001 | 1.19 | 0.060 | 0.020 | 16.28 | 0.0112 | Mo 2.08            | 643      | 716       | 15.38  | 18.30  | 1        | 2      |
| L2-02              | 10.50 | 0.82  | 0.080 | 0.034 | 1.58 | 0.074 | 0.102 | 17.02 | 0.0065 | –                  | 839      | 849       | 0.88   | 1.56   | 31       | 11     |
| L24-02             | 12.30 | 0.03  | 0.007 | 0.005 | 0.48 | 0.031 | 0.002 | 16.90 | 0.0129 | Nb 1.72            | 725      | 725       | 0.15   | 2.45   | 2        | 1      |
| L23-02             | 12.04 | 0.68  | 0.030 | 0.047 | 0.96 | 0.043 | 0.092 | 17.30 | 0.0093 | Nb 1.06            | 787      | 818       | 0.38   | 1.24   | 3        | 24     |
| C12-02             | 8.23  | 0.47  | 0.018 | 0.002 | 1.00 | 0.060 | 0.070 | 18.43 | –      | –                  | 747      | 756       | 14.96  | 18.57  | 4        | 0      |
| C1-02              | 8.12  | 0.50  | 0.038 | 0.002 | 1.00 | 0.060 | 0.060 | 18.11 | 0.0102 | –                  | 707      | 763       | 13.36  | 17.04  | 2        | 0      |

<sup>a</sup>L5-02 contains  $\approx 3$  vol.% globular-shaped delta ferrite.

## 7.4 Effect of Silicon

The yield strength of the model steel alloys, measured in BWR-like water at 289°C (DO  $\approx 8$  ppm), was nearly constant at  $\approx 200$  MPa in the unirradiated state and was more or less independent of Si concentration (see Fig. 16). However, as fluence was increased to  $\approx 0.3 \times 10^{21}$  n·cm<sup>-2</sup> and  $\approx 0.9 \times 10^{21}$  n·cm<sup>-2</sup>, the degree of increase in yield strength was significantly lower for alloys that contain  $>0.9$  wt.% Si than for alloys that contain  $<0.8$  wt.% Si. This behavior indicates that the nature of irradiation-induced hardening centers and the degree of irradiation hardening are influenced by Si. Silicon atoms in austenitic stainless steels occupy substitutional sites; therefore, they are likely to interact preferentially with irradiation-induced vacancy sites. This effect is likely to inhibit the formation of vacancy clusters or vacancy-impurity complexes, and is, therefore, conducive to a less significant degree of irradiation-induced hardening. An effect similar to that of Si was, however, not observed for C and N.

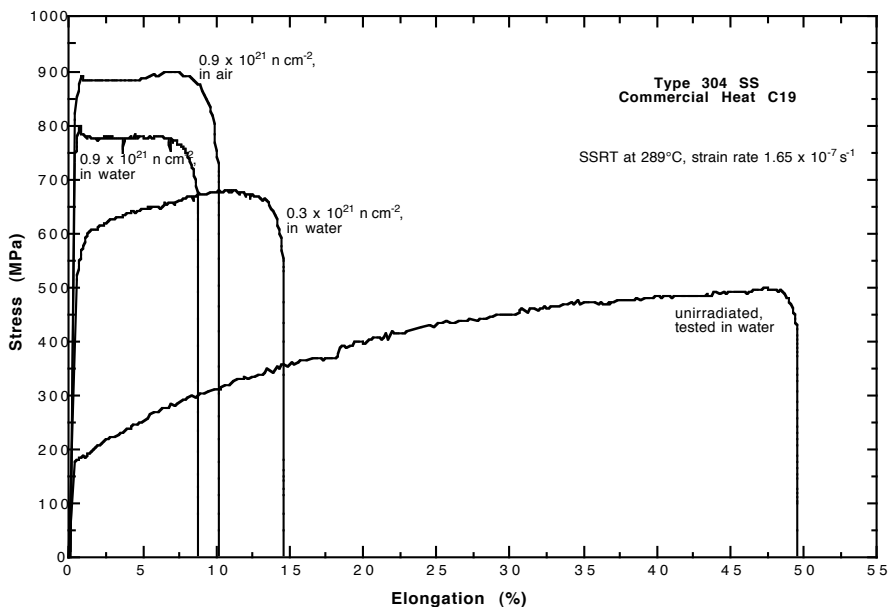


Figure 11. Effects of fluence and test environment on load elongation behavior of Type 304 SS commercial heat C19.

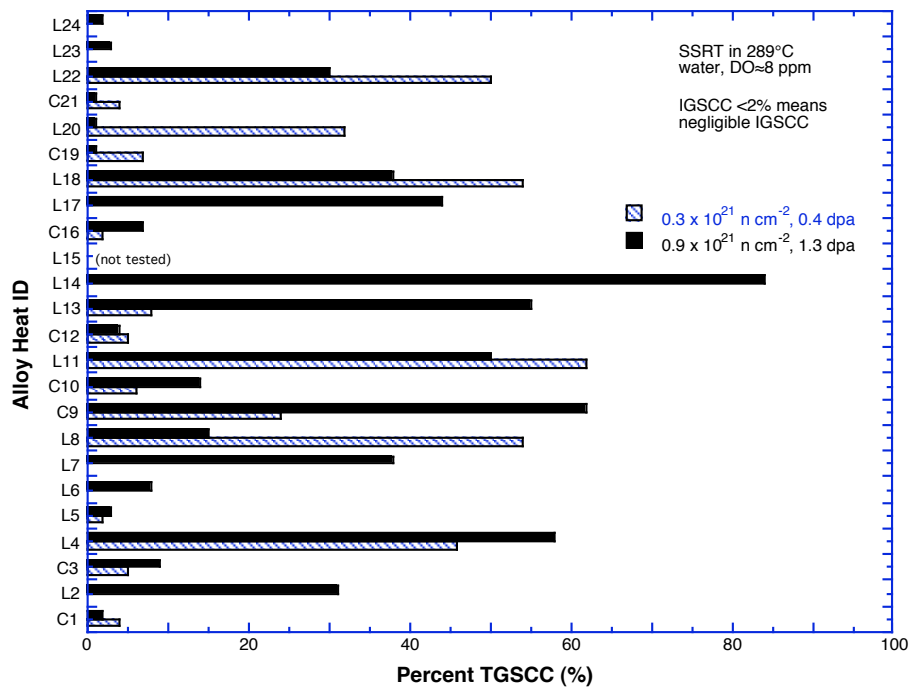


Figure 12. Effects of fluence on percent TGSCC measured in 289°C water containing ≈8 ppm DO, 0.4 to 1.3 dpa.

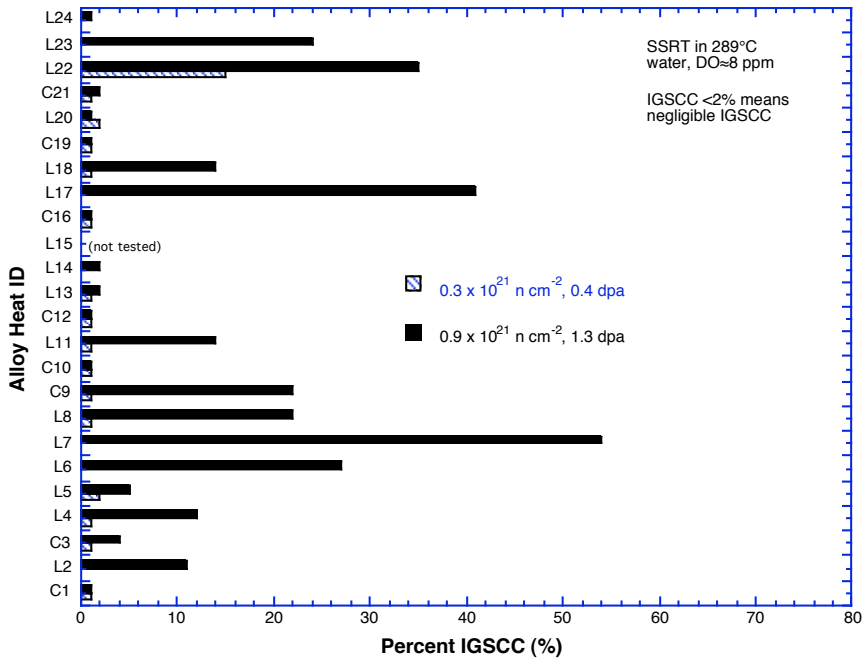


Figure 13. Effects of fluence on percent IGSC measured in 289°C water containing ≈ 8 ppm DO, 0.4 and 1.3 dpa.

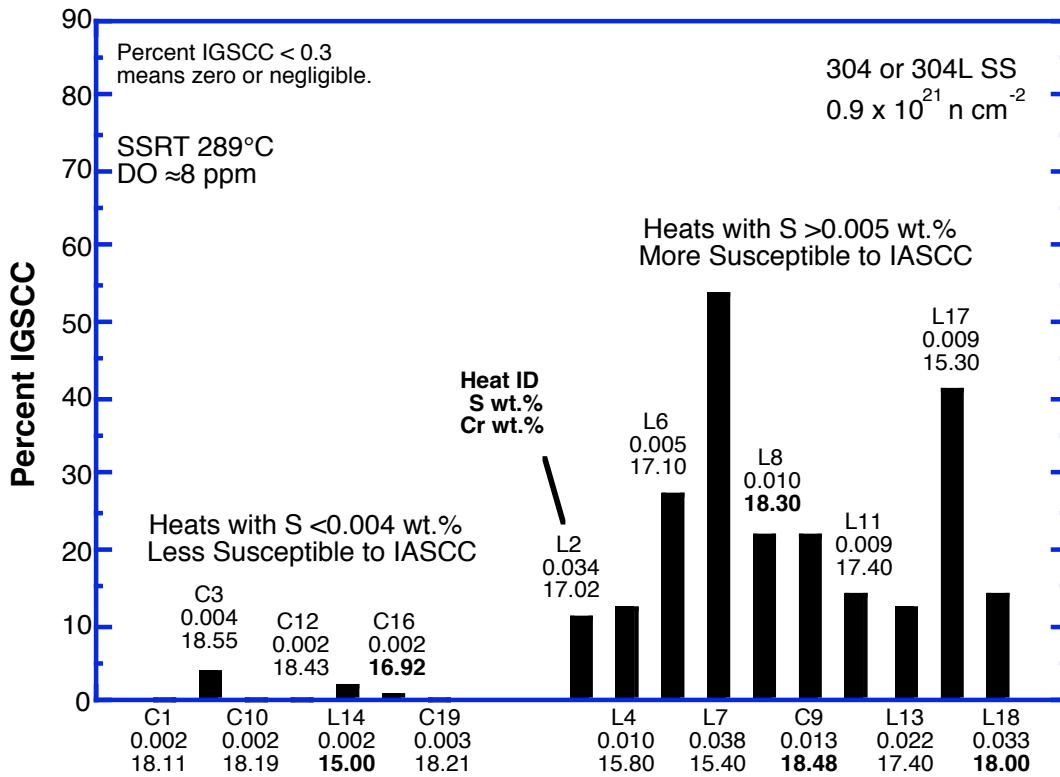


Figure 14. Effect of S on susceptibility to IGSC of Type 304 and 304L steels irradiated to  $\approx 0.9 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ); one group of steels contains < 0.004 wt.% S and the other group contains > 0.005 wt.% S.

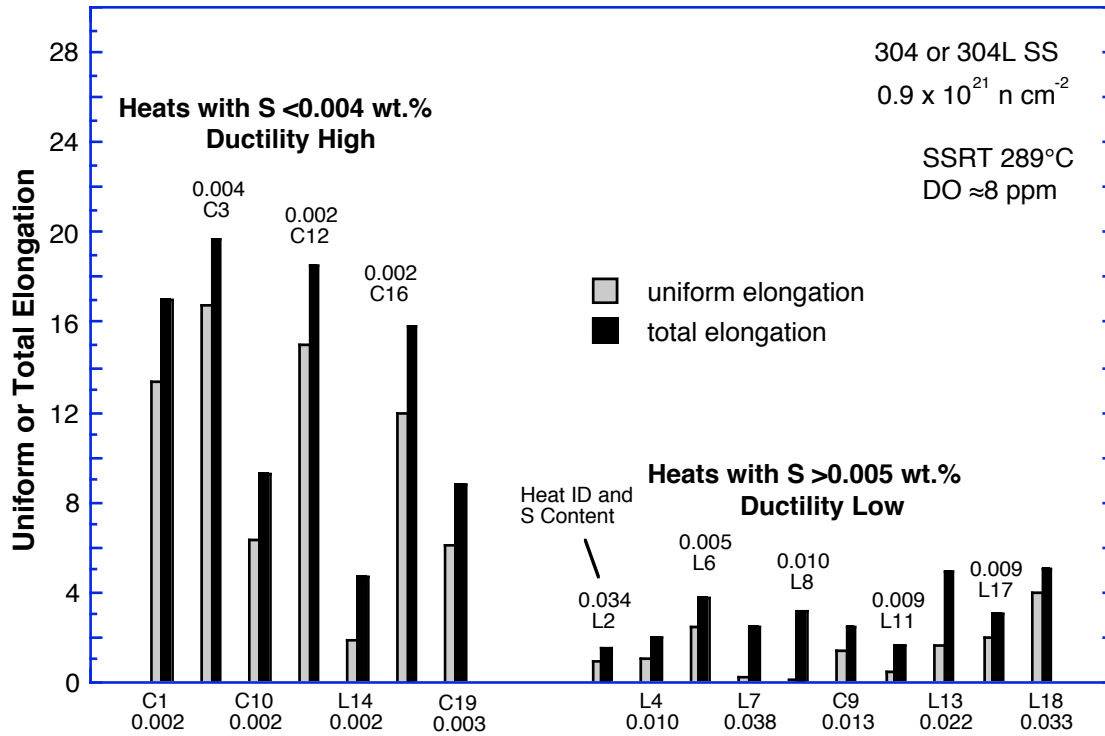


Figure 15. Effect of S on uniform and total elongation of Type 304 and 304L steels irradiated to  $\approx 0.9 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ); one group of steels contains  $< 0.004 \text{ wt.}\%$  S and the other group contains  $> 0.005 \text{ wt.}\%$  S.

Among the laboratory-fabricated heats of Types 304 and 304L SS that contain relatively high concentrations of S (0.009-0.038 wt.%), alloys that contain  $\leq 0.66 \text{ wt.}\%$  Si exhibited significant susceptibility to IGSCC, whereas alloys with  $\geq 0.82 \text{ wt.}\%$  Si exhibited relatively lower susceptibility to IGSCC (see Fig. 17). However, Heat L6, which contains  $\approx 1.9 \text{ wt.}\%$  Si, exhibited susceptibility to IGSCC (percent IGSCC  $\approx 27$ ). These observations indicate that Si concentration in the range of  $\approx 0.8$  to  $\approx 1.5 \text{ wt.}\%$  is beneficial in delaying the onset of or suppressing the susceptibility to IASCC, at least up to 1.3 dpa.

However, it is not clear if such a beneficial effect of Si (in the range of  $\approx 0.8$  to  $\approx 1.5 \text{ wt.}\%$ ) is also significant at higher fluence levels. Considering the strongly deleterious effect of S shown in Figs. 14 and 15, it is possible that, as fluence increases to higher levels, the deleterious effect of S more than offsets the beneficial effect of Si. As such, the effect of Si could be secondary, that is, manifested only at low fluence levels.

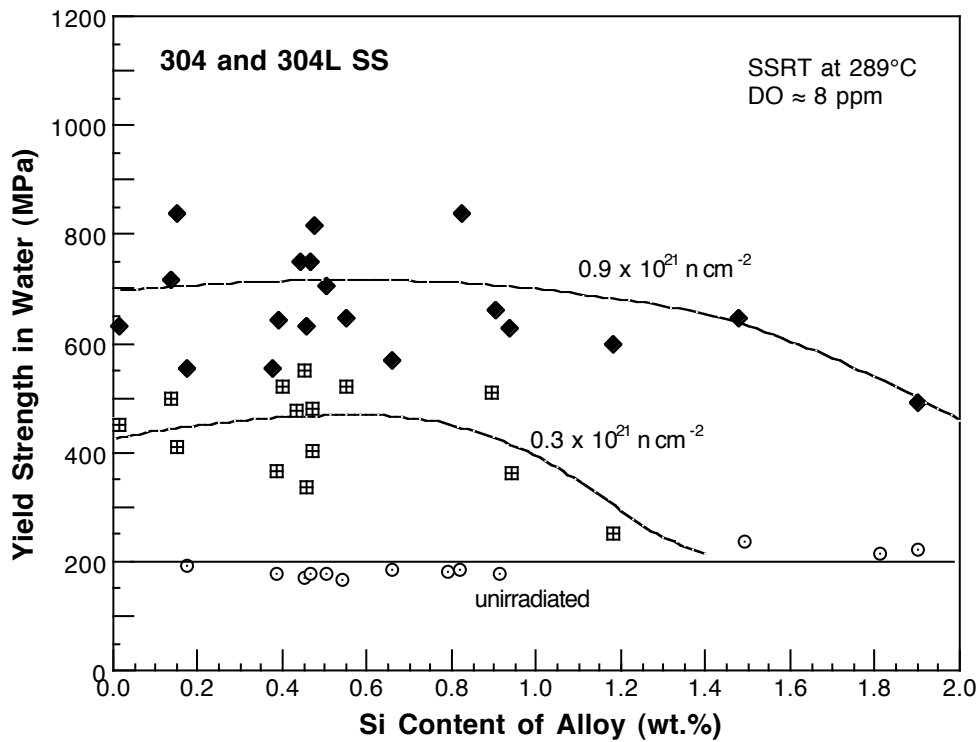


Figure 16. Effect of Si concentration on yield strength of Types 304 and 304L steel measured in 289°C water before and after irradiation.

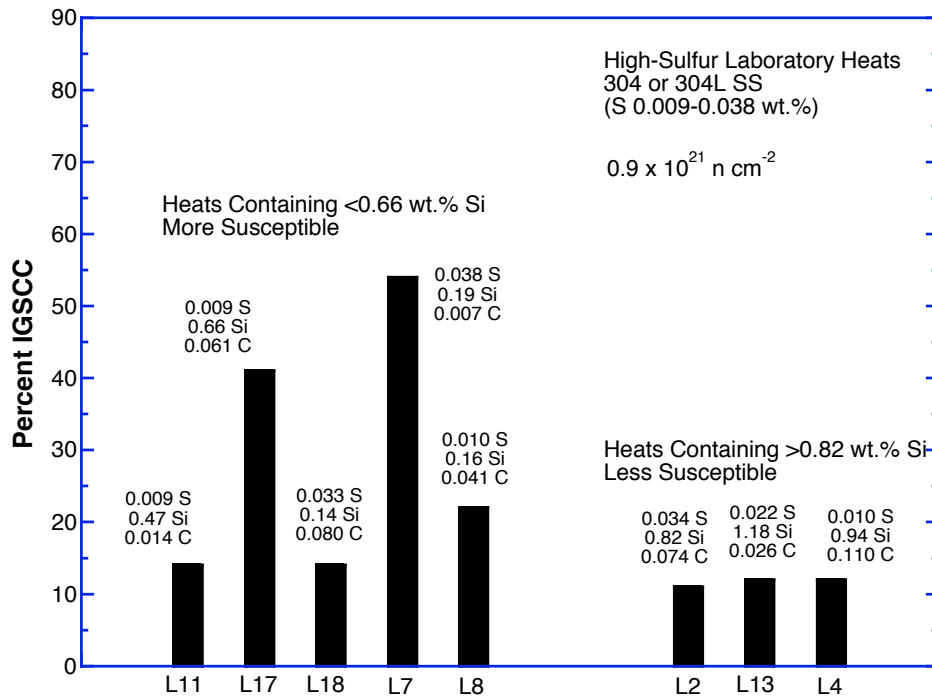


Figure 17. Effect of Si on IGSCC of lab heats of Types 304 and 304L SS containing high concentrations of S and irradiated to ≈1.3 dpa; susceptible heats contain lower concentrations of Si; resistant heats contain higher concentrations of Si.

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## 8. SSRT SCC Test and Fractography on High-Fluence Specimens ( $\approx 3.0$ dpa)

### 8.1 Tabulation of Test Results

Twenty-four steel alloys were tested by the SSRT technique after irradiation to  $\approx 2.0 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV) ( $\approx 3.0$  dpa) in BWR-like oxidizing water at 289°C (DO  $\approx 8$  ppm). Of the 24 alloys, 18 were 304- or 304L-type steels, 3 were 316- or 316L-type steels, 2 were 348- or 348L-type steels, and one alloy was austenitic-ferritic duplex steel (delta ferrite  $\approx 3$  vol.%). At this fluence, some of the specimens (Heats L13 and L8) were so brittle that they failed in 23°C air during assembly of the test train. Table 13 summarizes the test conditions and results of the SSRT testing and fractography. Table 14 summarizes correlation of the fracture characteristics with chemical composition of the steels.

Table 13. Results of SSRT tests and SEM fractography for model austenitic stainless steels irradiated to  $\approx 2.0 \times 10^{21}$  n·cm<sup>-2</sup> ( $E > 1$  MeV) ( $\approx 3.0$  dpa)

| Test Speci. ID | SSRT No. | Oxygen Conc. (ppm) | ECP (mV SHE) | Cond. ( $\mu\text{Scm}^{-1}$ )                  | pH at 25°C | Yield Stress (MPa)           | Max. Stress (MPa) | Uniform Elong. (%) | Total Elong. (%) | TG (%) | IG (%) |
|----------------|----------|--------------------|--------------|---|------------|------------------------------|-------------------|--------------------|------------------|--------|--------|
| L4-03          | HR-45    | 9.0                | +198         | 0.09  | 6.5        | 876                          | 1068              | 2.49               | 3.27             | 5      | 95     |
| C12-03         | HR-46    | 8.0                | +208         | 0.09  | 6.5        | 922                          | 996               | 1.28               | 2.73             | 8      | 2      |
| C9-03          | HR-47    | 8.0                | +212         | 0.11  | 6.5        | early failure in 289°C water |                   |                    |                  | 6      | 94     |
| L5-03          | HR-48    | 8.0                | +204         | 0.10  | 6.5        | 953                          | 985               | 0.59               | 2.97             | 2      | 4      |
| C19-08         | HR-59    |                    | test in      | 289°C air                                       |            | 1001                         | 1001              | 0.27               | 1.95             | -      | -      |
| C19-09         | HR-60    |                    | test in      | 325°C air                                       |            | 998                          | 998               | 0.27               | 2.02             | -      | -      |
| C19-03         | HR-49    | 8.0                | +171         | 0.11  | 6.5        | 787                          | 801               | 0.89               | 3.32             | 2      | 62     |
| C16-03         | HR-50    | 8.0                | +202         | 0.10  | 6.5        | 766                          | 803               | 0.83               | 1.84             | 2      | 29     |
| C10-03         | HR-51    | 8.0                | +167         | 0.11  | 6.5        | 1062                         | 1065              | 3.15               | 4.51             | 3      | 0      |
| L18-03         | HR-52    | 8.0                | +169         | 0.11  | 6.5        | 795                          | 779               | 0.35               | 1.75             | 5      | 86     |
| L13-03         | HR-53    | -                  | -            | brittle fracture during test preparation in air |            |                              |                   | -                  | -                | -      | -      |
| C1-03          | HR-54    | 8.0                | +161         | 0.10  | 6.5        | 802                          | 833               | 3.38               | 5.27             | 2      | 0      |
| C3-03          | HR-55    | 8.0                | +160         | 0.10  | 6.5        | 796                          | 826               | 5.05               | 7.31             | 0      | 26     |
| C21-03         | HR-56    | 8.0                | +156         | 0.10  | 6.5        | 893                          | 924               | 1.89               | 4.85             | 2      | 1      |
| L8-03          | HR-57    | -                  |              | brittle fracture during test preparation in air |            |                              |                   | -                  | -                | -      | -      |
| L26-01         | HR-61    | 8.0                | +221         | 0.11  | 6.5        | 795                          | 821               | 0.58               | 1.83             | 32     | 2      |
| L25-01         | HR-62    | 8.0                | +220         | 0.09  | 6.5        | 867                          | 867               | 0.05               | 0.48             | 8      | 43     |
| L2-03          | HR-63    | 8.0                | +204         | 0.10  | 6.5        | 916                          | 916               | 0.26               | 0.50             | 58     | 32     |
| L7-03          | HR-64    | 8.0                | +196         | 0.11  | 6.5        | 889                          | 889               | 0.02               | 0.05             | 10     | 52     |
| L23-03         | HR-65    | 8.0                | +189         | 0.11  | 6.5        | 887                          | 887               | 0.06               | 0.41             | 16     | 22     |
| L17-03         | HR-66    | 8.0                | +188         | 0.11  | 6.5        | 1105                         | 1116              | 0.23               | 0.28             | 56     | 20     |
| L14-03         | HR-67    | 8.0                | +174         | 0.11  | 6.5        | 917                          | 951               | 1.64               | 2.01             | 16     | 0      |
| L27-01         | HR-68    | 8.0                | +175         | 0.11  | 6.5        | 776                          | 808               | 0.73               | 1.02             | 12     | 2      |
| L15-03         | HR-69    | 8.0                | +180         | 0.11  | 6.5        | 946                          | 946               | 0.78               | 1.17             | 8      | 32     |
| L6-03          | HR-70    | 8.0                | +171         | 0.11  | 6.5        | 794                          | 805               | 2.02               | 2.44             | 12     | 36     |
| L24-03         | HR-71    | 8.0                | +173         | 0.11  | 6.5        | 862                          | 862               | 0.11               | 0.86             | 3      | 0      |



Table 14. Composition (wt.%) of stainless steels irradiated to  $\approx 2.0 \times 10^{21}$  n-cm<sup>-2</sup> (E > 1 MeV) ( $\approx 3.0$  dpa) correlated with results of SSRT test and SEM fractography.

| Test Speci. ID     | Ni    | Si   | P     | S      | Mn   | C     | N     | Cr    | O      | Other                  | Unif. Elong. (%) | Total Elong. (%) | IG (%) |
|--------------------|-------|------|-------|--------|------|-------|-------|-------|--------|------------------------|------------------|------------------|--------|
| L4-03              | 10.20 | 0.94 | 0.031 | 0.0100 | 1.75 | 0.110 | 0.002 | 15.80 | 0.0037 | –                      | 2.49             | 3.27             | 95     |
| C12-03             | 8.23  | 0.47 | 0.018 | 0.0020 | 1.00 | 0.060 | 0.070 | 18.43 | –      | –                      | 1.28             | 1.73             | 2      |
| C9-03              | 8.75  | 0.39 | 0.013 | 0.0130 | 1.72 | 0.062 | 0.065 | 18.48 | 0.0102 | –                      | –                | –                | 94     |
| L5-03 <sup>a</sup> | 9.66  | 0.90 | 0.113 | 0.0283 | 0.47 | 0.006 | 0.033 | 21.00 | 0.0068 | $\approx 3\% \delta^a$ | 0.59             | 2.97             | 4      |
| C19-03             | 8.08  | 0.45 | 0.031 | 0.0030 | 0.99 | 0.060 | 0.070 | 18.21 | 0.0200 | –                      | 0.89             | 3.32             | 62     |
| C16-03             | 12.90 | 0.38 | 0.014 | 0.0020 | 1.66 | 0.020 | 0.011 | 16.92 | 0.0150 | Mo 2.30                | 0.83             | 1.84             | 29     |
| C10-03             | 8.13  | 0.55 | 0.033 | 0.0020 | 1.00 | 0.060 | 0.086 | 18.19 | 0.0074 | –                      | 3.15             | 4.51             | 0      |
| L18-03             | 8.13  | 0.14 | 0.016 | 0.0326 | 1.13 | 0.080 | 0.001 | 18.00 | 0.0055 | –                      | 0.35             | 1.75             | 86     |
| L13-03             | 8.18  | 1.18 | 0.027 | 0.0222 | 0.36 | 0.026 | 0.001 | 17.40 | 0.0042 | –                      | –                | –                | –      |
| C1-03              | 8.12  | 0.50 | 0.038 | 0.0020 | 1.00 | 0.060 | 0.060 | 18.11 | 0.0102 | –                      | 3.38             | 5.27             | 0      |
| C3-03              | 8.91  | 0.46 | 0.019 | 0.0040 | 1.81 | 0.016 | 0.083 | 18.55 | –      | –                      | 5.05             | 7.31             | 26     |
| C21-03             | 10.24 | 0.51 | 0.034 | 0.0010 | 1.19 | 0.060 | 0.020 | 16.28 | 0.0112 | Mo 2.08                | 1.89             | 4.85             | 1      |
| L8-03              | 10.20 | 0.15 | 0.093 | 0.0100 | 1.85 | 0.041 | 0.001 | 18.30 | 0.0059 | –                      | –                | –                | –      |
| L26-01             | 8.09  | 0.79 | 0.004 | 0.0022 | 0.91 | 0.070 | 0.089 | 17.20 | 0.0080 | –                      | 0.58             | 1.83             | 2      |
| L25-01             | 8.93  | 0.92 | 0.020 | 0.0082 | 1.54 | 0.019 | 0.095 | 17.20 | 0.0085 | –                      | 0.05             | 0.48             | 43     |
| L2-03              | 10.50 | 0.82 | 0.080 | 0.0345 | 1.58 | 0.074 | 0.102 | 17.02 | 0.0065 | –                      | 0.26             | 0.50             | 32     |
| L7-03              | 10.60 | 0.18 | 0.040 | 0.0384 | 1.02 | 0.007 | 0.111 | 15.40 | 0.0274 | –                      | 0.02             | 0.05             | 52     |
| L23-03             | 12.04 | 0.68 | 0.030 | 0.0475 | 0.96 | 0.043 | 0.092 | 17.30 | 0.0093 | Nb 1.06                | 0.06             | 0.41             | 22     |
| L17-03             | 8.00  | 0.66 | 0.090 | 0.0089 | 0.48 | 0.061 | 0.078 | 15.30 | 0.0090 | –                      | 0.23             | 0.28             | 20     |
| L14-03             | 7.93  | 1.49 | 0.080 | 0.0023 | 1.76 | 0.107 | 0.028 | 15.00 | 0.0045 | –                      | 1.64             | 2.01             | 0      |
| L27-01             | 10.30 | 0.96 | 0.040 | 0.0018 | 0.97 | 0.057 | 0.019 | 15.30 | 0.0058 | Mo 2.01                | 0.73             | 1.02             | 2      |
| L15-03             | 8.00  | 1.82 | 0.010 | 0.0129 | 1.07 | 0.020 | 0.085 | 17.80 | 0.0110 | –                      | 0.78             | 1.17             | 32     |
| L6-03              | 10.00 | 1.90 | 0.020 | 0.0051 | 1.13 | 0.096 | 0.087 | 17.10 | 0.0058 | –                      | 2.02             | 2.44             | 36     |
| L24-03             | 12.30 | 0.03 | 0.007 | 0.0055 | 0.48 | 0.031 | 0.002 | 16.90 | 0.0129 | Nb 1.72                | 0.11             | 0.86             | 0      |

<sup>a</sup>L5-03 contains globular-shaped  $\approx 3$  vol.% delta ferrite.

Examples of load-elongation behavior are shown in Fig. 18; they were obtained on SSRT specimens fabricated from a commercial heat of Type 304 SS (Heat C19) and tested in water at 289°C and in air at 23, 289, and 325°C. Effects of neutron damage levels of 0, 0.4, 1.3, and 3 dpa are also shown in the figure. Several observations can be inferred from the results: (a) the increase in yield strength from 1.3 dpa to 3.0 dpa is small, indicating that irradiation hardening reaches near saturation at  $\approx 2$  dpa; (b) by the time the damage level reaches  $\approx 3$  dpa, work hardening capability of the steel is insignificant in water, and strain-softening occurs to some extent in air at 289-325°C; (c) at  $\approx 3$  dpa, tensile behavior in 289°C and 325°C air is similar; (d) ductility, i.e., uniform and total elongations in 1.3-dpa specimens tested in air, is lower at 289°C than at 23°C. The mechanism that leads to the last behavior is not known. However, the degree of strain aging or localized deformation via dislocation channeling or twinning could be more pronounced at the higher temperatures than at 23°C.

## 8.2 Correlation of Percent IGSCC with Elemental Composition

From the results summarized in Table 14, percent IGSCC obtained on 304- and 316-type steels was plotted as a function of bulk concentration of Cr, Ni, Mn, Si, P, N, O, C, and S to see if a good correlation can be obtained with any of the elements. Such plots are shown in Figs. 19-27, respectively. In these plots, the results obtained on the 348-type steels (Alloys L23 and L24) and the austenitic-ferritic steel (i.e., Alloy L5 that contains  $\approx 3$  vol.% delta ferrite) were not included.

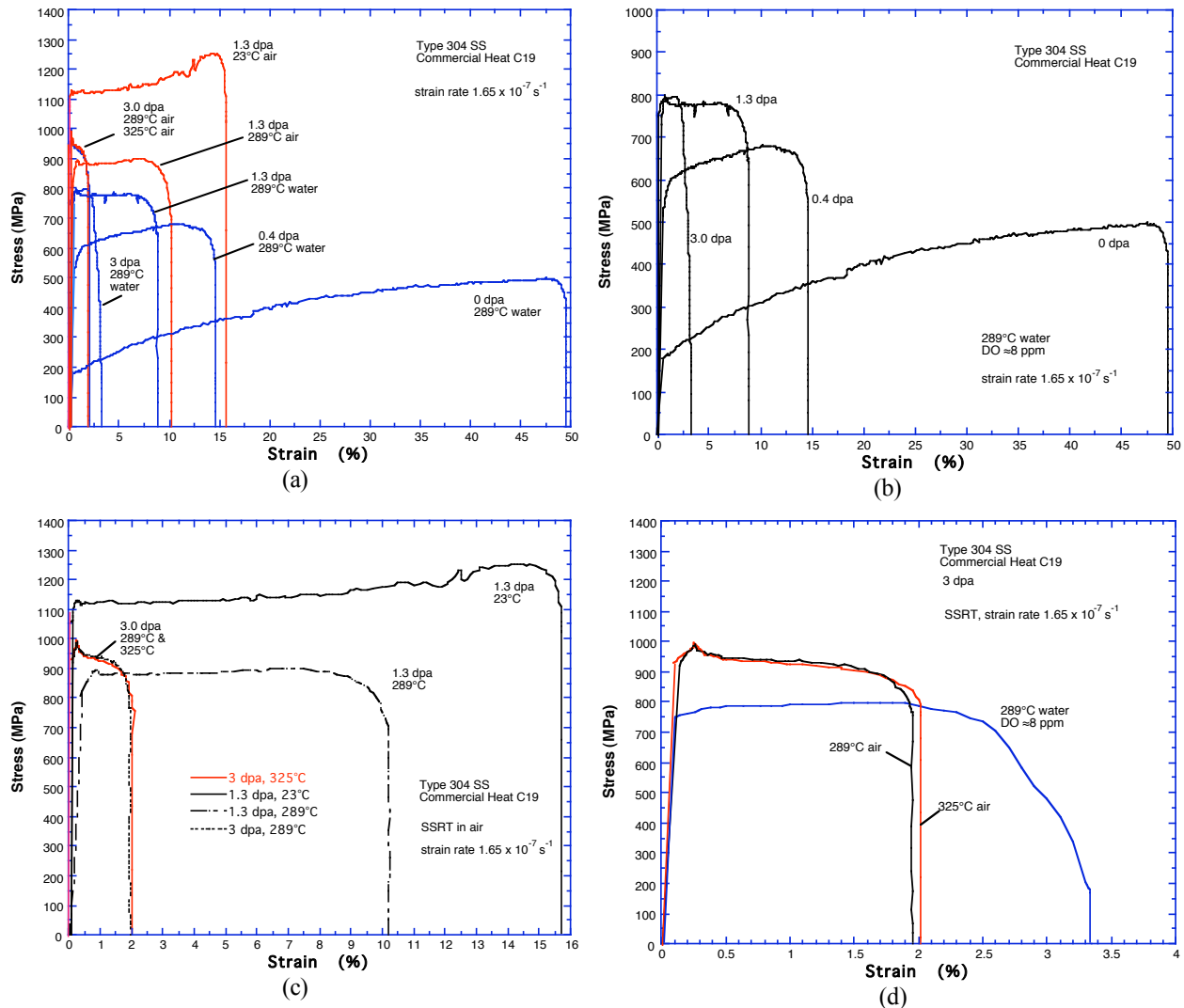


Figure 18. SSRT load-elongation behavior of Type 304 SS Heat C19: (a) overall data; (b) effect of damage level in 289°C water, DO ≈ 8 ppm; (c) effects of damage level and temperature in air; and (d) behavior at ≈ 3 dpa in 289°C water and 289-325°C air.

A careful examination of the data in Fig. 19 shows that relative IASCC susceptibility (i.e., percent IGSCC from the SSRT tests in this study) cannot be correlated well with bulk Cr concentration. That is, for a given Cr content between 15 and 19 wt.%, percent IGSCC is independent of Cr content, although by the time the damage level reaches 3 dpa, grain-boundary Cr depletion should be significant in all the tested steels and should be near the saturated levels. Therefore, the results in Fig. 19 do not support the premise that Cr atoms in bulk or on grain boundaries play a direct (primary) role in IASCC. In relation with this observation, it is interesting to note that the grain-boundary Cr-depletion mechanism is not supported by the results of postirradiation-annealing studies reported for BWR<sup>14-15</sup> and PWR<sup>10,16</sup> conditions. It has been shown in these studies that, during post-irradiation annealing at 400-550°C, IASCC susceptibility becomes negligible long before grain-boundary Cr concentration begins to change from its level in the as-irradiated state.

Similar examination of Figs. 20-26 strongly indicates that Ni, Mn, Si, P, N, O, or C do not play a primary role.

Of the 19 alloys analyzed in Figs. 19-27, of particular interest is the behavior of seven 304- or 316-type alloys that were found to be resistant to IASCC, i.e., Alloys C1, C10, C12, C21, L14, L26, and L27. Understanding why these heats are resistant is considered to hold an important clue to understanding the mechanism of IASCC. From Figs. 19-26, it is evident that IASCC susceptibilities (i.e., percent IGSCC) of the seven alloys are zero or insignificant, regardless of the concentrations of Cr, Ni, Mn, Si, P, N, O, or C. The only exception is S (see Fig. 27).

The S and C content of the seven resistant heats is given in Figs. 19-26. The S and C content of C16, which is susceptible to IASCC, is also given. Sulfur content of all other heats not denoted in the figures is 0.003 wt.% or higher. It can be deduced from the figures that, whereas the IASCC-resistant behavior of the seven heats is independent of the concentration of the element of each figure, they possess a unique and common character. That is, S concentration is very low ( $<0.002$  wt.%) and, at the same time, C concentration is relatively high ( $>0.05$  wt.%).

Furthermore, Fig. 27 shows that all 304- or 316-type steels containing  $>0.002$  wt.% S and tested in this investigation were susceptible, whereas steels containing  $\leq 0.002$  wt.% S were either susceptible or resistant. Therefore, the results in Figs. 19-27 strongly indicate that S concentration of  $\leq 0.002$  wt.% is a necessary but not a sufficient condition for a 304- or 316-type steel to be resistant to IASCC under BWR-like conditions.

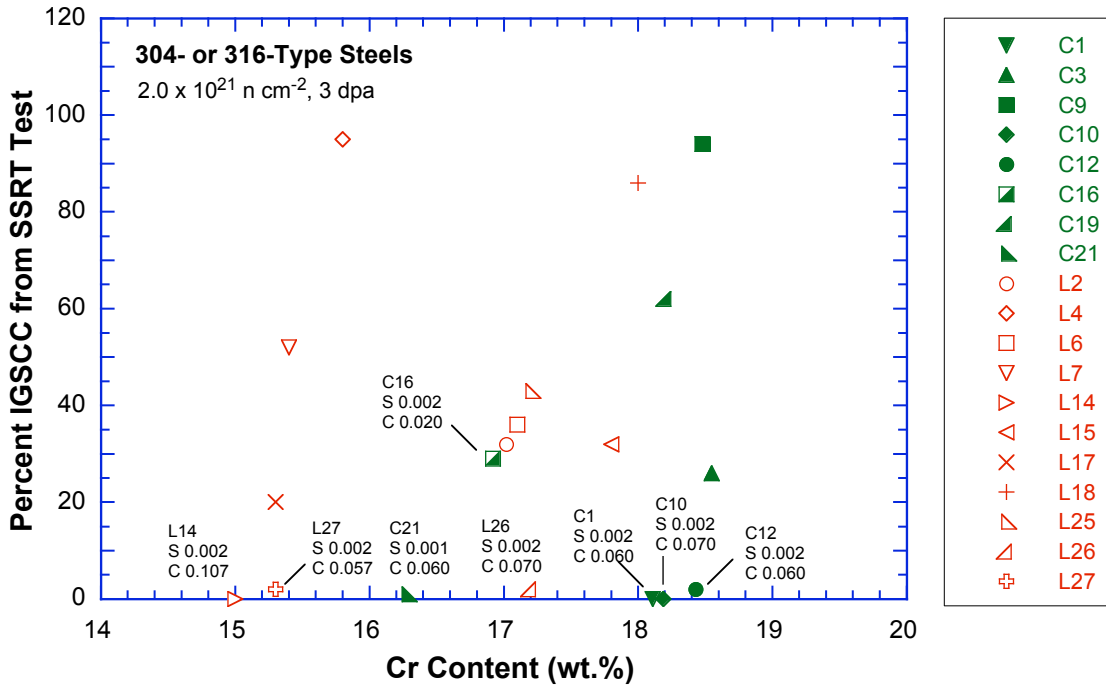


Figure 19. Percent IGSCC at  $\approx 3$  dpa correlated with bulk Cr concentration, 304- and 316-type steels.

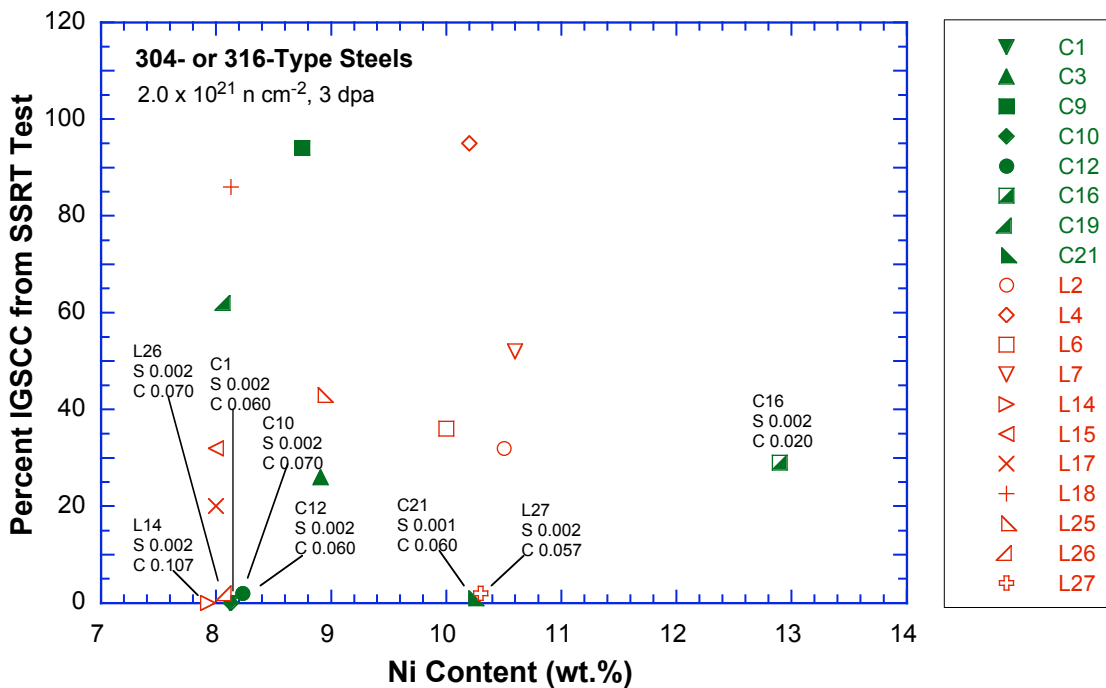


Figure 20. Percent IGSCC at  $\approx 3$  dpa correlated with bulk Ni concentration, 304- and 316-type steels.

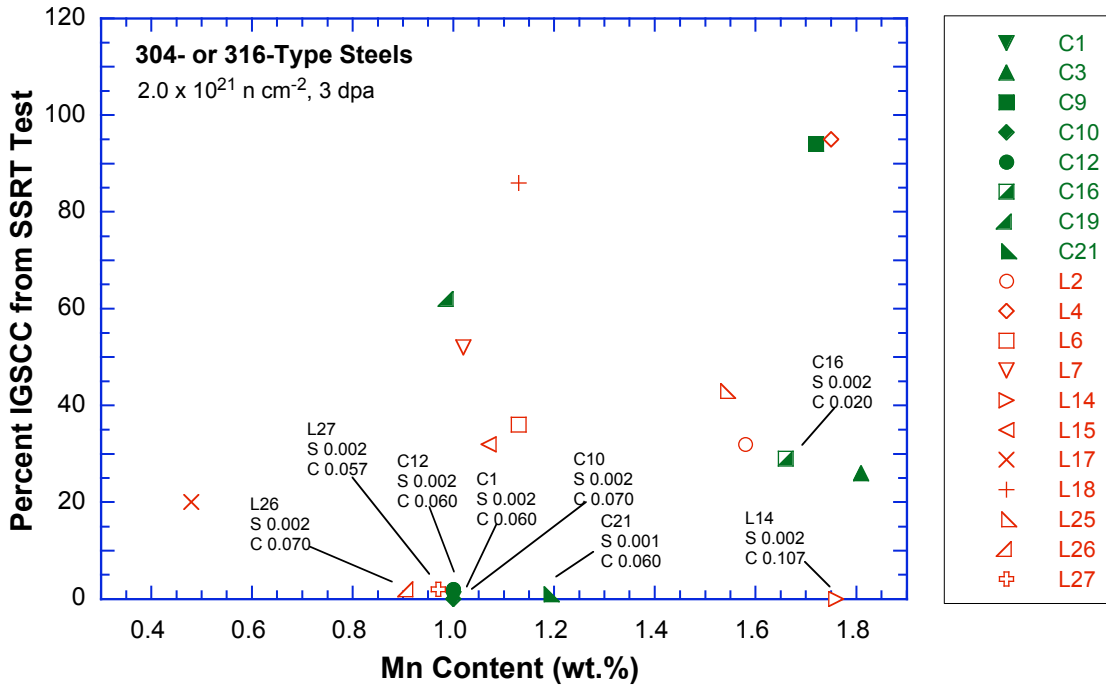


Figure 21. Percent IGSCC at  $\approx 3$  dpa correlated with bulk Mn concentration, 304- and 316-type steels.

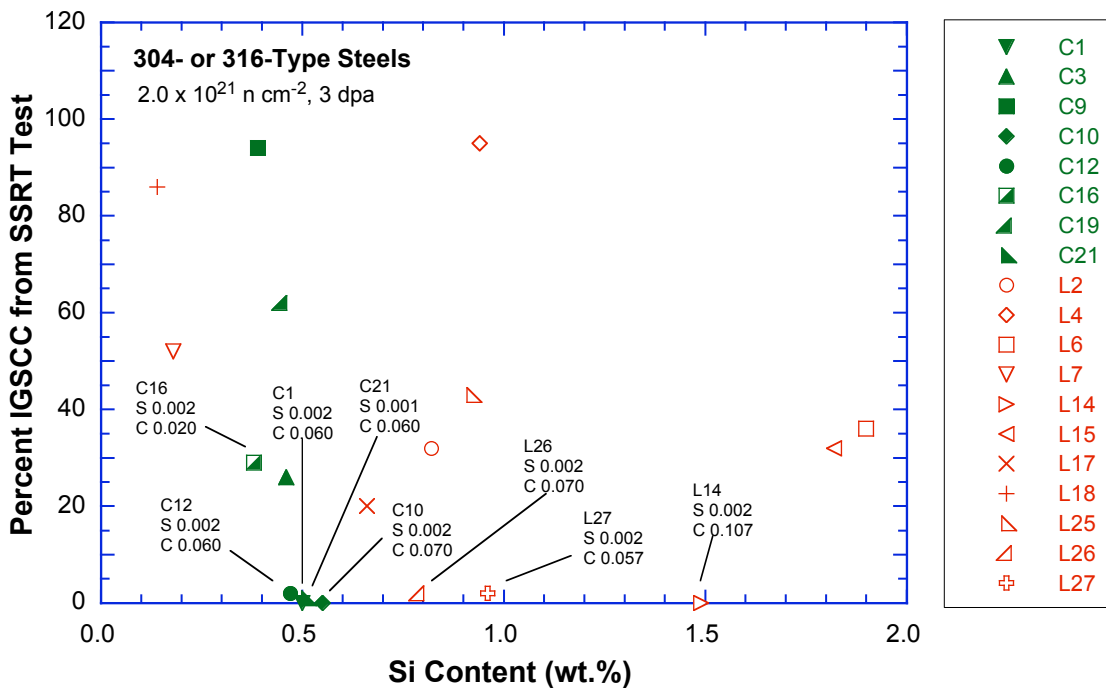


Figure 22. Percent IGSCC at  $\approx 3$  dpa correlated with bulk Si concentration, 304- and 316-type steels.

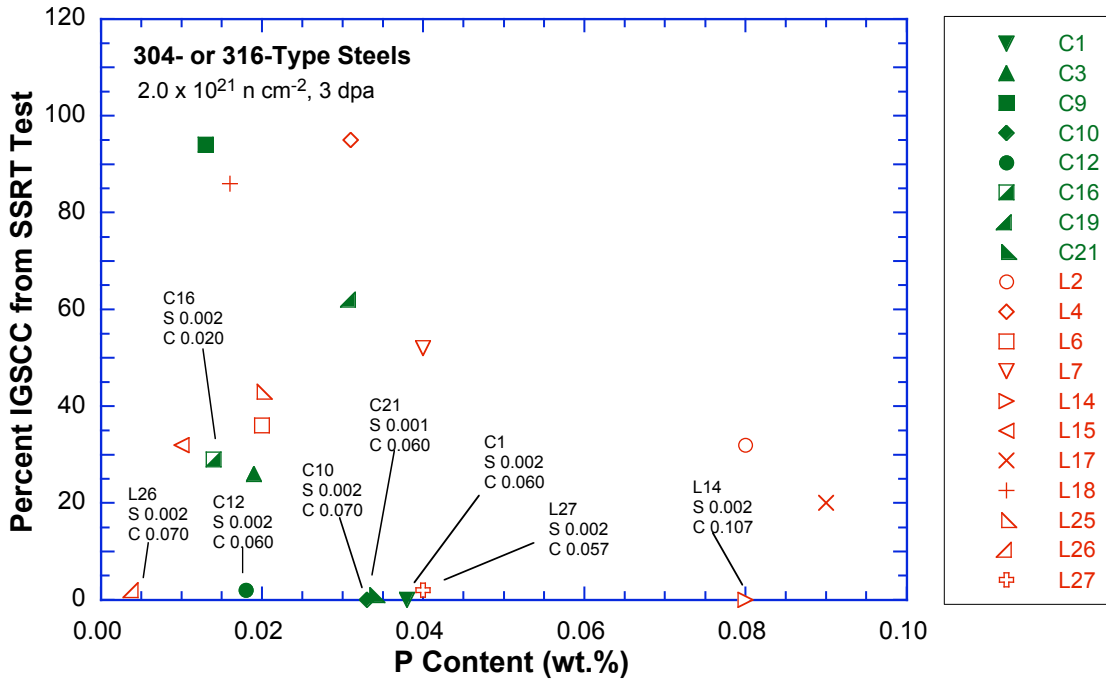


Figure 23. Percent IGSCC at  $\approx 3$  dpa correlated with bulk P concentration, 304- and 316-type steels.

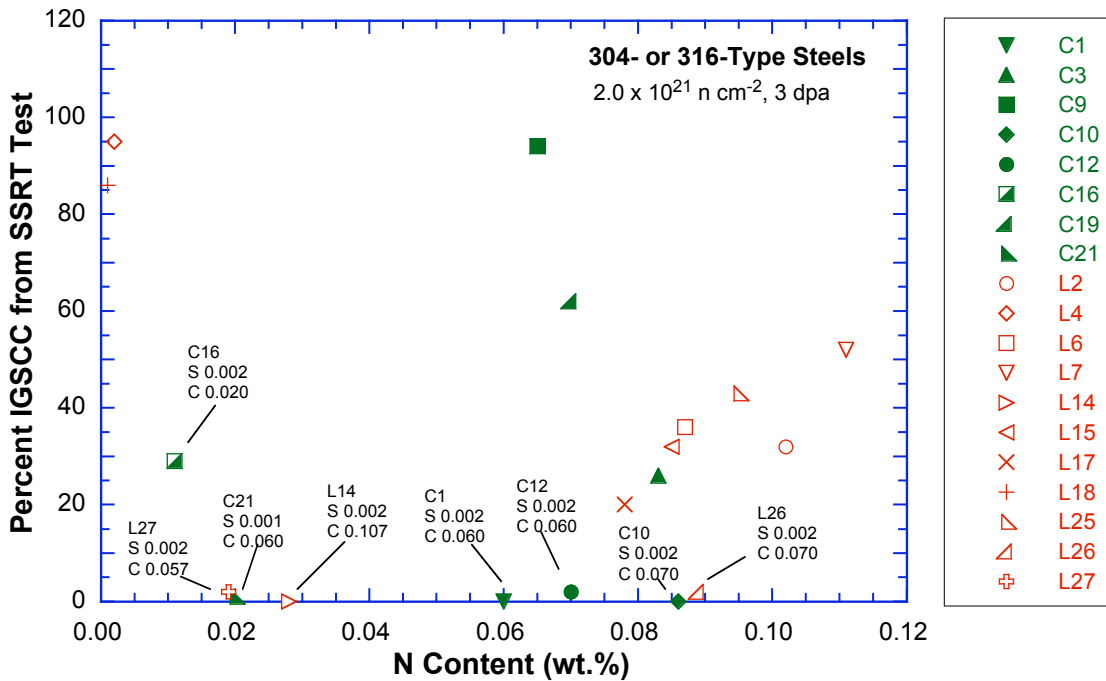


Figure 24. Percent IGSCC at  $\approx 3$  dpa correlated with bulk N concentration, 304- and 316-type steels.

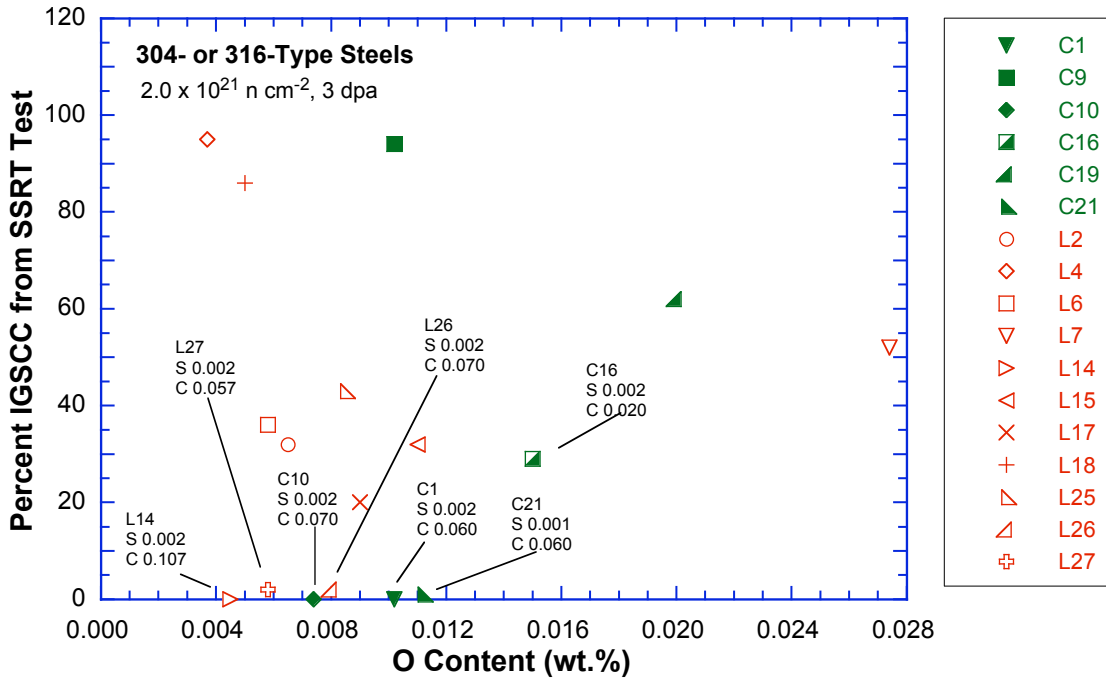


Figure 25. Percent IGSCC at  $\approx 3$  dpa correlated with bulk O concentration, 304- and 316-type steels.

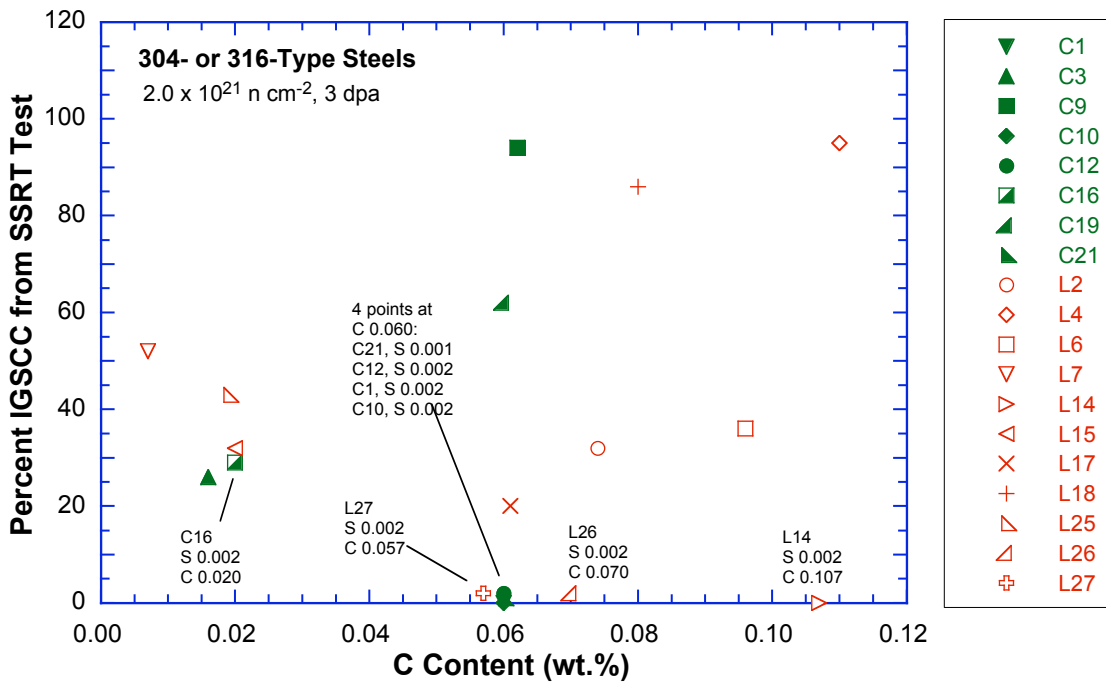
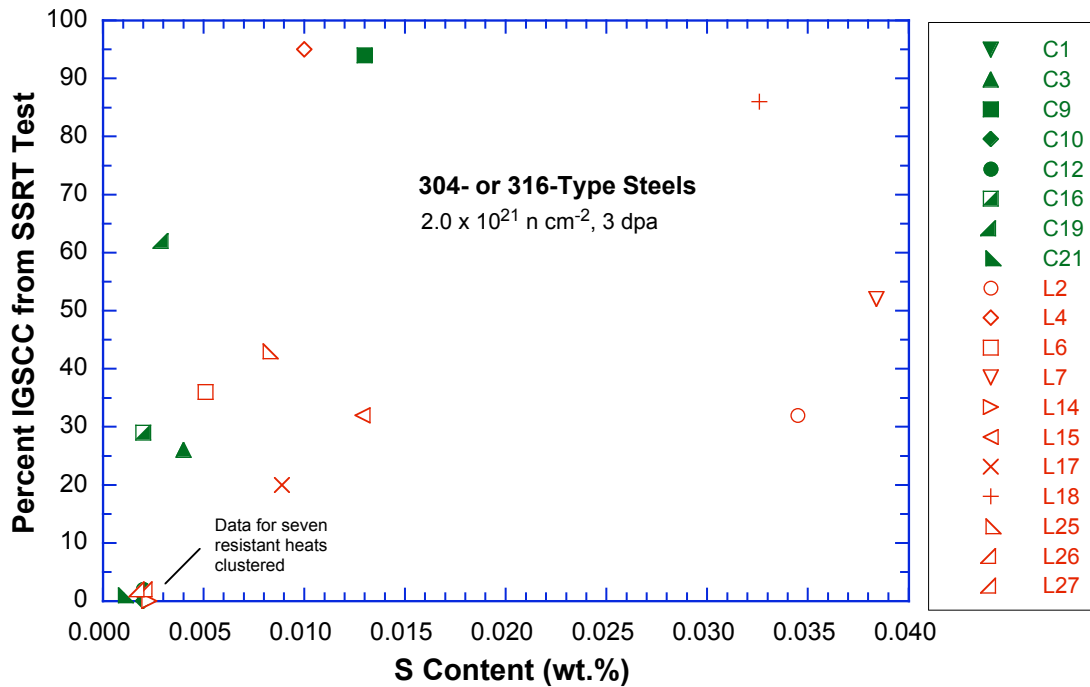
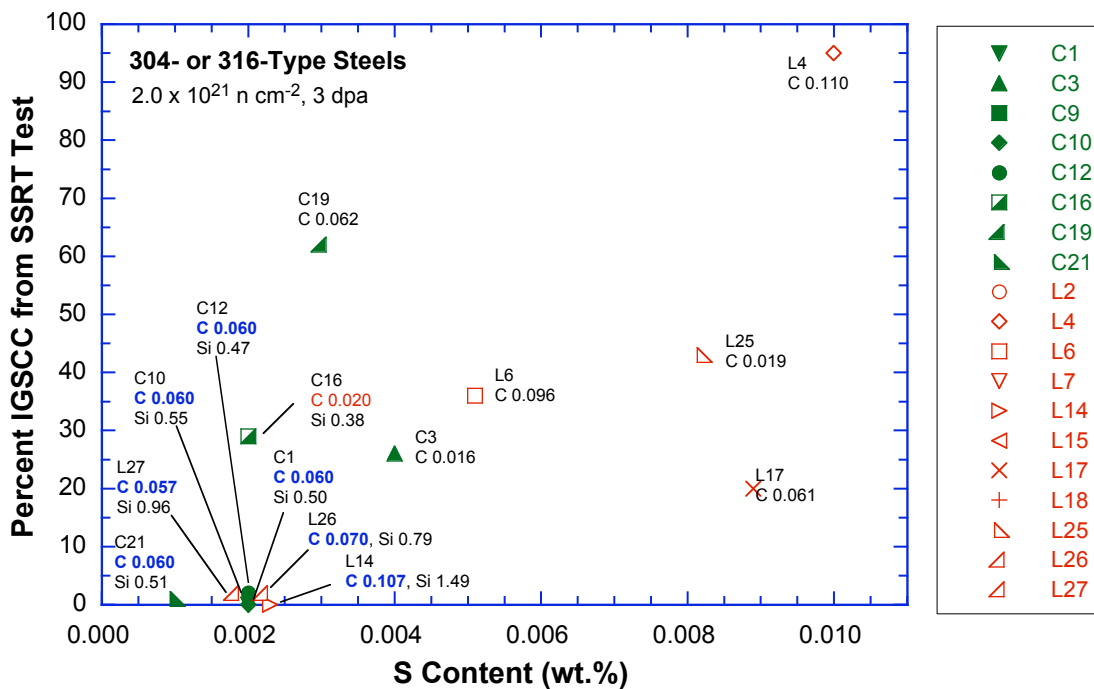


Figure 26. Percent IGSCC at  $\approx 3$  dpa correlated with bulk C concentration, 304- and 316-type steels.



(a)



(b)

Figure 27. Percent IGSCC at  $\approx 3 \text{ dpa}$  correlated with bulk S concentration, 304- and 316-type steels: (a) all data at  $S < 0.004 \text{ wt.}\%$  and (b) expanded plot for  $S < 0.01 \text{ wt.}\%$  showing details.



In Fig. 26, the behavior of Heat C16, which contains a very low concentration of S (0.002 wt.%) and a relatively low concentration of C (0.02 wt.%), is compared with the behavior of other similar heats that contain  $\leq 0.002$  wt.% S but higher concentrations of C. In spite of the very low concentration of S, the heat was susceptible. This finding indicates that C concentration higher than a threshold value (somewhere between 0.02 and 0.057 wt.%) is also a necessary (but not a sufficient) condition for a 304- or 316-type steel to be resistant to IASCC.

However, a combined condition that S concentration be less than  $\approx 0.002$  wt.% and, at the same time, that C concentration be higher than a threshold value (e.g.,  $> 0.03$  wt.%) appears to be a sufficient condition for a 304- or 316-type steel to be resistant to IASCC. This observation strongly indicates that S is the primary contributor to IASCC susceptibility; therefore, the effect of S and relevant data were analyzed more systematically.

### 8.3 Effect of Fluence

Figure 28 shows the effect of fluence on the IASCC-resistant behavior of the seven heats of 304- or 316-type SSs that contain  $\leq 0.002$  wt.% S and  $> 0.05$  wt.% C. For damage levels of 0.4, 1.3, and 3.0 dpa, no appreciable susceptibility to IASCC was observed in these heats in 289°C BWR-like high-purity water that contains  $\approx 8$  ppm DO (ECP  $\approx 160$ -220 mV SHE). The IASCC-resistant behavior of these heats is expected to be valid also in similar water that contains lower levels of DO and ECP.

In contrast, a strong effect of fluence was observed for two groups of 304- or 316-type steels that are susceptible to IASCC, i.e., heats that contain high S ( $\geq 0.003$  wt.%) and heats that contain low S ( $< 0.002$  wt.%) and low C ( $< 0.03$  wt.%). The effect of fluence for the 16 susceptible heats of 304- or 316-type SSs tested in this study is shown in Fig. 29. The effect of fluence of the susceptible heats varied significantly from heat to heat. However, the root cause of the strong variation in fluence effect is not understood. Strong variation is evident for both laboratory and commercial heats.

### 8.4 Role of S in IASCC at High Fluence

In contrast to the laboratory-fabricated model SSs that have compositions outside of the AISI specifications, for all eight commercial heats of Type 304, 304L, 316, and 316L austenitic SSs investigated in this study, bulk S content provided a good monotonic correlation with percent IGSCC (see Fig. 30). Four commercial heats of Type 304 and 316 SS that contain very low concentrations of S ( $\leq 0.002$  wt.%) exhibited only negligible susceptibility to IGSCC.

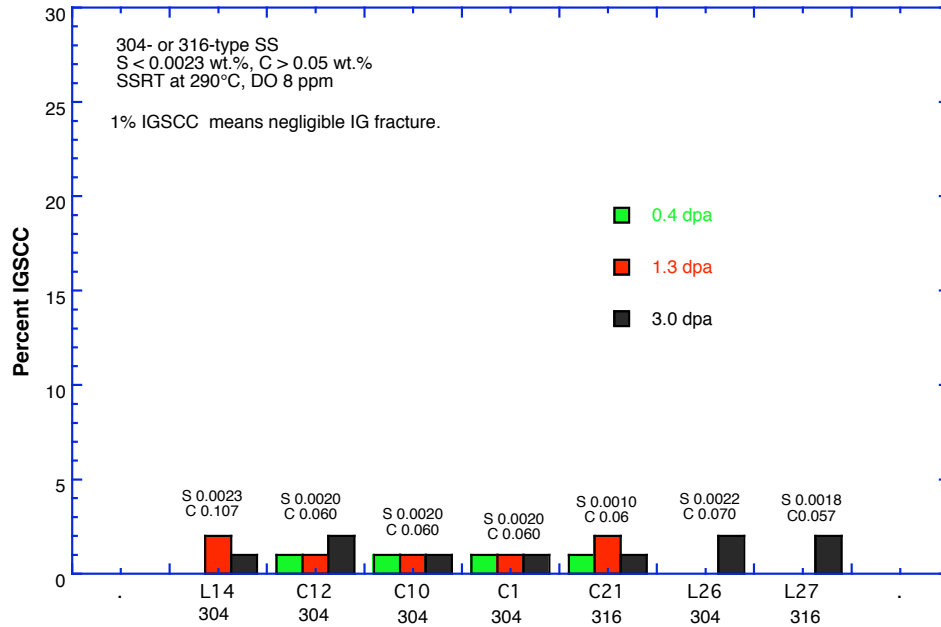


Figure 28. Effect of neutron damage on IASCC susceptibility (percent IGSCC) of Type 304 or 316 steels that contain low concentrations of S and high concentrations of C (i.e., <0.0023 wt.% S and >0.05 wt.% C).

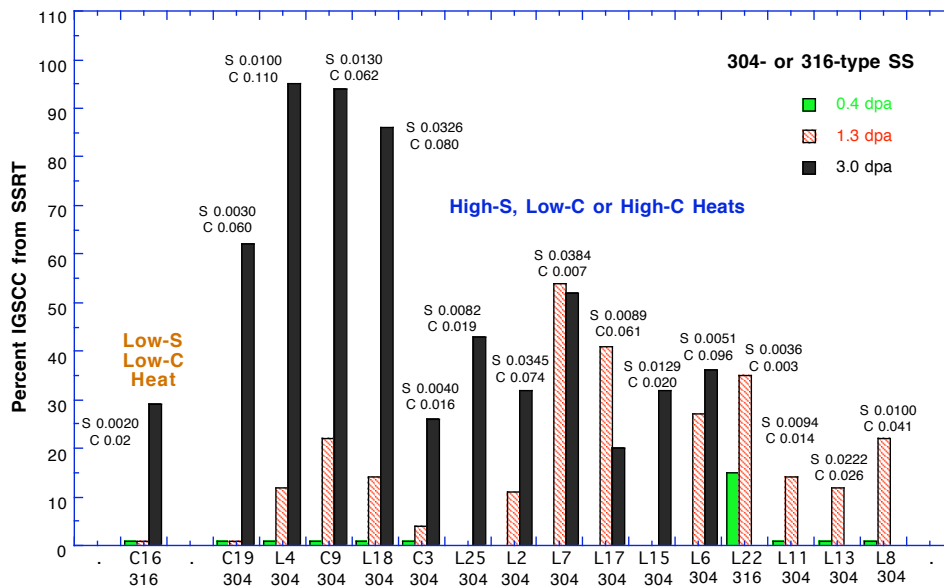


Figure 29. Effect of neutron damage on IASCC susceptibility (percent IGSCC) of Type 304 or 316 steels that contain low S and low C concentrations (i.e.,  $\leq 0.002$  wt.% S and <0.03 wt.% C), high S and high C concentrations (i.e., >0.003 wt.% S and >0.05 wt.% C), or high S and low C concentrations (i.e., >0.003 wt.% S and <0.03 wt.% C).

However, as S content increased to  $\geq 0.003$  wt.%, susceptibility increased drastically. At an S content of  $>0.005$  wt.%, the specimens from the commercial heats failed in virtually complete IG mode, just as in IASCC of field components. The steep increase of susceptibility at S levels  $>0.003$  wt.% indicates that an S-related critical phenomenon plays an important role in IASCC.

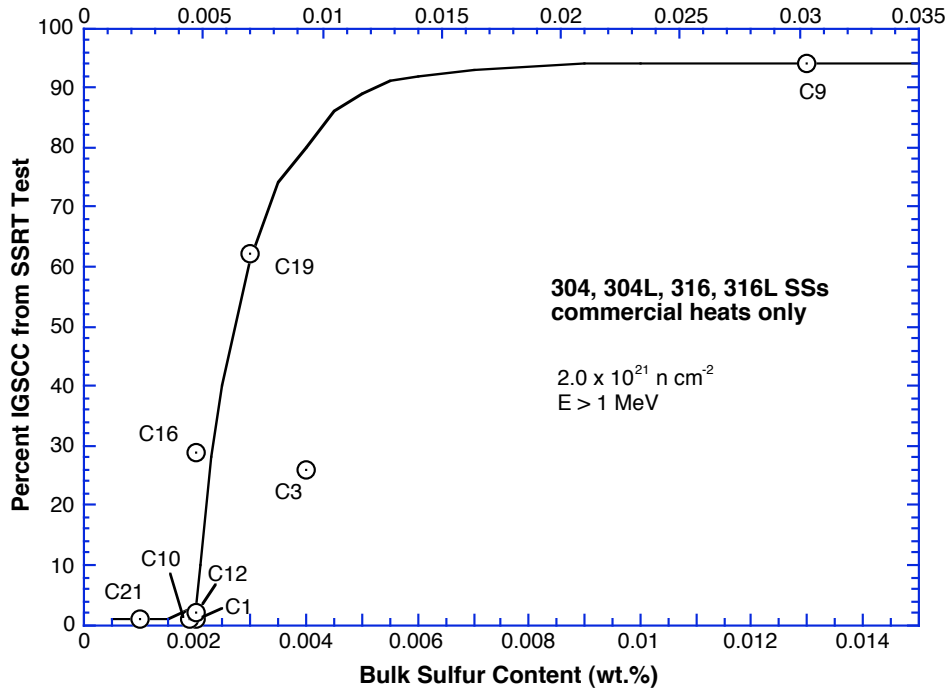


Figure 30. Effect of bulk S content on IASCC susceptibility (percent IGSCC) of commercial heats of Types 304, 304L, 316, and 316L steels.

The deleterious effect of S can be discerned more directly by comparing the IASCC susceptibilities of a pair of high- and low-S heats of the same grade that contain otherwise similar elemental compositions, e.g., Heat C12 (S 0.002 wt.%) and Heat C9 (S 0.013 wt.%). As fluence increased, the susceptibility of Heat C12 remained negligible, whereas the susceptibility of Heat C9 increased drastically (see Fig. 31).

The relative behavior of Heats C9 and C12 is consistent with the observation reported by Tsukada et al. on their HP and HP+S heats.<sup>24,29</sup> They reported that an HP heat that contained 0.0318 wt.% S and 0.002 wt.% C (Heat HP+S, see Fig. 32) was significantly more susceptible than an HP heat that contained 0.0014 wt.% S and 0.003 wt.% C (Heat HP). They also reported that the latter heat was more susceptible than a C-doped HP heat that contains 0.0020 wt.% S and 0.098 wt.% C (Heat HP+C). A deleterious effect of S is also indicated from the expanding-tube-test data reported by Kasahara et al.,<sup>28</sup> Jacobs et al.,<sup>21</sup> and Garzarolli et al.<sup>2</sup> (see Figs. 33-35, respectively).

The tests used to obtain the data in Figs. 31 and 33-35 are based on the “binary-like” approach. In this approach, S content was varied high or low while the composition of other elements was kept similar to that of commercial steels used in reactor components. In contrast, the result in Fig. 32 is based on the “single-element” approach in which high-purity heats are used.

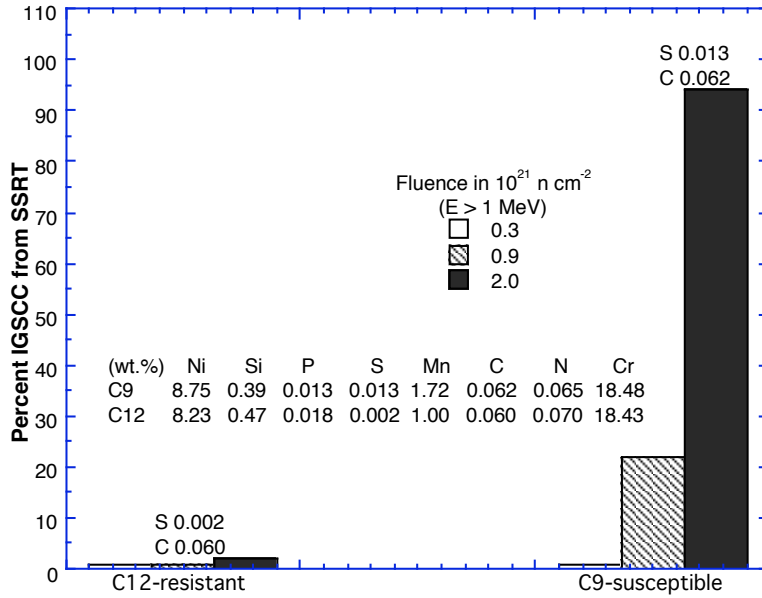


Figure 31. IASCC susceptibility of low-S (Heat C12) and high-S (Heat C19) heats of comparable chemical composition; Type 304 SS.

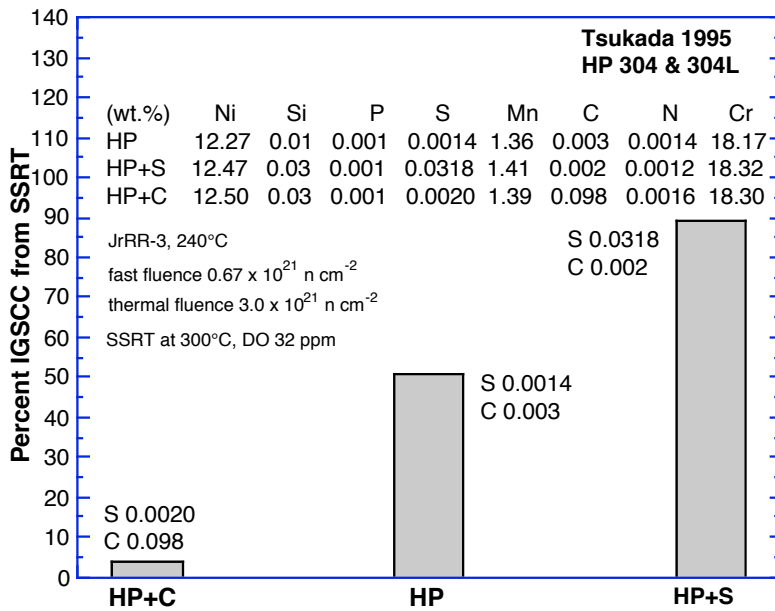


Figure 32. IASCC susceptibility of high-purity heats of Type 304L SS doped with C or S, from Tsukada et al., Ref. 29.

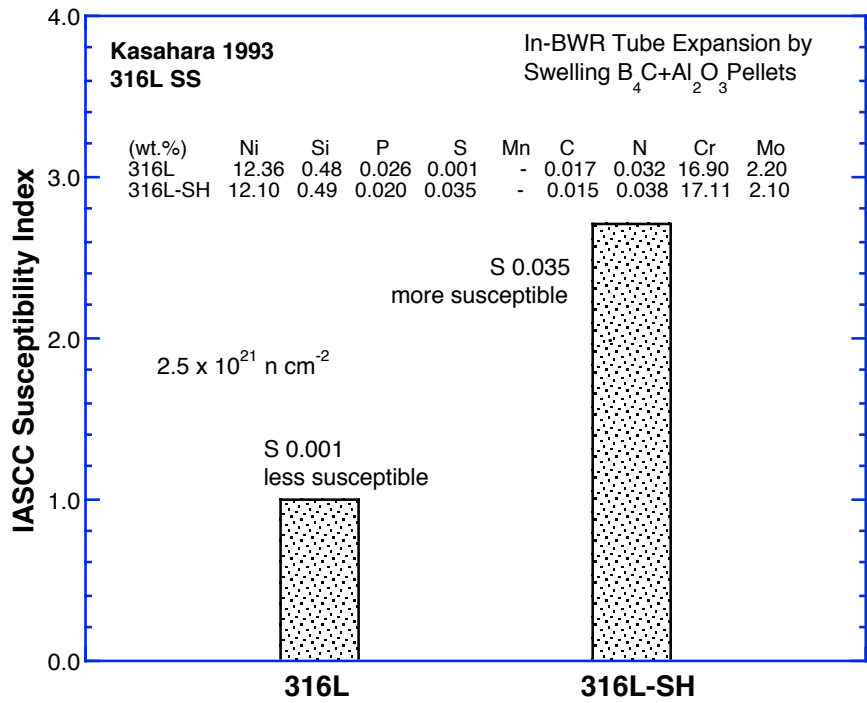


Figure 33. IASCC susceptibility of low- and high-S heats of comparable composition, from Kasahara et al., Ref. 28.

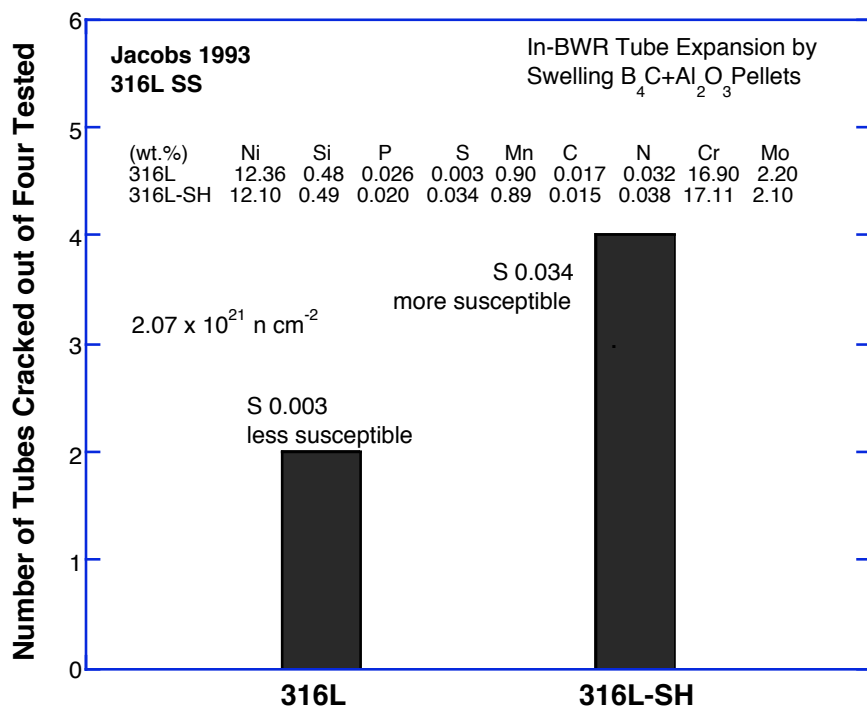


Figure 34. IASCC susceptibility of low- and high-S heats of comparable composition, from Jacobs et al., Ref. 21.

Although the latter approach is advantageous when used to isolate more clearly the effect of a single element, it is difficult to determine which element plays a dominant or secondary role and to determine if the

effect of that particular element is influenced by other elements present in a steel similar to those used in a reactor. Therefore, a finding from such an approach must be confirmed for the type of steels used to fabricate actual field components. Tests on a sufficiently large number of commercial heats, such as those in Fig. 30, serve this purpose. Also, any potential effect of uncommon parameters, such as the presence of ferrite or dendrites, should be screened carefully. The deleterious effect of S indicated from the three approaches is consistent, i.e., the binary-like single-element approach with commercial steels (Figs. 31 and 33-35), the single-element approach with HP steels (Fig. 32), and the multiple-elements approach with commercial steels (Fig. 30).

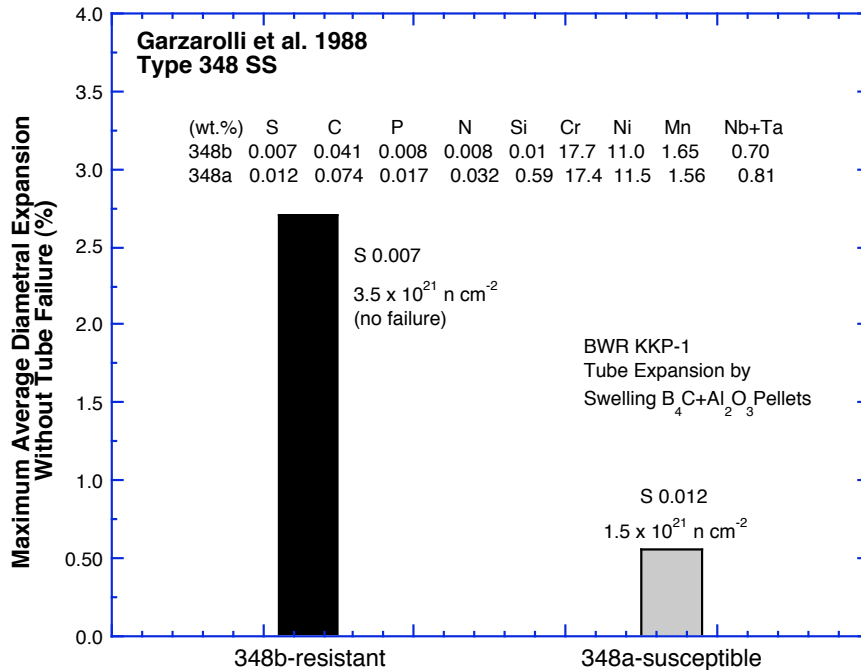


Figure 35. IASCC susceptibility of low- and high-S heats of comparable composition, from Garzarolli et al. Ref. 2.

## 8.5 Effect of Carbon on Deleterious Role of Sulfur

For high-carbon heats of Types 304 and 316 SS, it was shown in Fig. 28 that S content of  $\leq 0.002$  wt.% provides good resistance to IASCC. However, whether the same is true or not with low-carbon heats of Types 304L, 316L, or high-purity SSs needs careful examination.

In this investigation, Type 316 SS Heat C21 that contains 0.001 wt.% S and 0.060 wt.% C was resistant to IASCC. In contrast, a similar heat of Type 316L SS (Heat C16) that contains 0.002 wt.% S and 0.020 wt.% C was susceptible (Fig. 30). This behavior appears to be consistent with the data of Tsukada et al.,<sup>29</sup> i.e., the behavior of Heats HP+C vs. HP shown in Fig. 32. Tanaka and coworkers performed SSRT tests on 304L- and 316L-type SS heats that contained S in nominal concentrations between 0.000 and 0.003 wt.% after irradiation in a BWR to  $\approx 1.9$  to  $2.1 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ).<sup>41</sup> A 304-type heat that contained 0.002 wt.% S and 0.060 wt.% C was resistant to IASCC at  $1.3 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$  ( $E > 1 \text{ MeV}$ ), whereas a 304L-type heat that contained 0.002 wt.% S and 0.013 wt.% C was susceptible to IASCC at  $2.1 \times 10^{21} \text{ n}\cdot\text{cm}^{-2}$ . Of the nine 316L-type heats investigated by Tanaka et al., three exhibited negligible susceptibility to IASCC, whereas the other six exhibited susceptibility. Fukuya et al. reported similar results.<sup>20</sup>

Summarizing the results from the four investigations, Figs 36 and 37 show, respectively, relative IASCC susceptibilities (i.e., percent IGSCC from SSRT test) of low-S heats ( $S \leq 0.002$  wt.%) of high-C steels (i.e., Types 304 and 316 SS) and those of their low-C counterparts. The trend in the figures indicates that low-S heats ( $S \leq 0.002$  wt.%) of 304- and 316-type SS are resistant to IASCC. However, all low-S heats of 304L- and 316L-type SS that contain  $\leq 0.002$  wt.% S are not necessarily resistant.

A similar trend is also evident for 348- and 348L-type SSs investigated by Garzarolli and coworkers by swelling-tube tests<sup>2,7,42</sup> in a PWR (see Figs. 38 and 39). In their experiments, average diametral strain was measured for intact and cracked tubes from the same heat after irradiation to one to three fuel cycles. Sulfur concentration in the tubes ranged from 0.002 to 0.007 wt.%. Their one-cycle data in Fig. 38 show that, under the same irradiation conditions, high-C heats (C 0.027-0.043 wt.%) could tolerate more diametral expansion without incurring a failure than low-C heats (C 0.003-0.015 wt.%), in spite of the fact that the high-C heats contained somewhat higher levels of S than the latter (i.e., 0.005-0.007 wt.% vs. 0.003-0.004 wt.%). A similar trend is observed from the one-to-three cycle data shown in Fig. 39. That is, high-C heats (C 0.027-0.048 wt.%) could withstand more diametral expansion without a failure than low-C heats (C 0.003-0.007 wt.%), in spite of the fact that the former heats contained higher levels of S than the latter and that fluence was higher in the former than in the latter.

The consistent behavior from the six investigations summarized in Figs. 36-39 shows that high C content suppresses the deleterious effect of S, if the S concentration is very low. In 304- or 316-type SSs, the beneficial effect of C (in concentrations  $\geq 0.03$  wt.%) is manifested significantly if S concentration is less than  $\approx 0.004$  wt.%. In the absence of such a beneficial effect, 304L- or 316L-type steels seem to be inherently less resistant than their high-C counterparts; this seems to lead to significant susceptibility even in some low-S heats, e.g., those shown in Figs. 36 and 37.

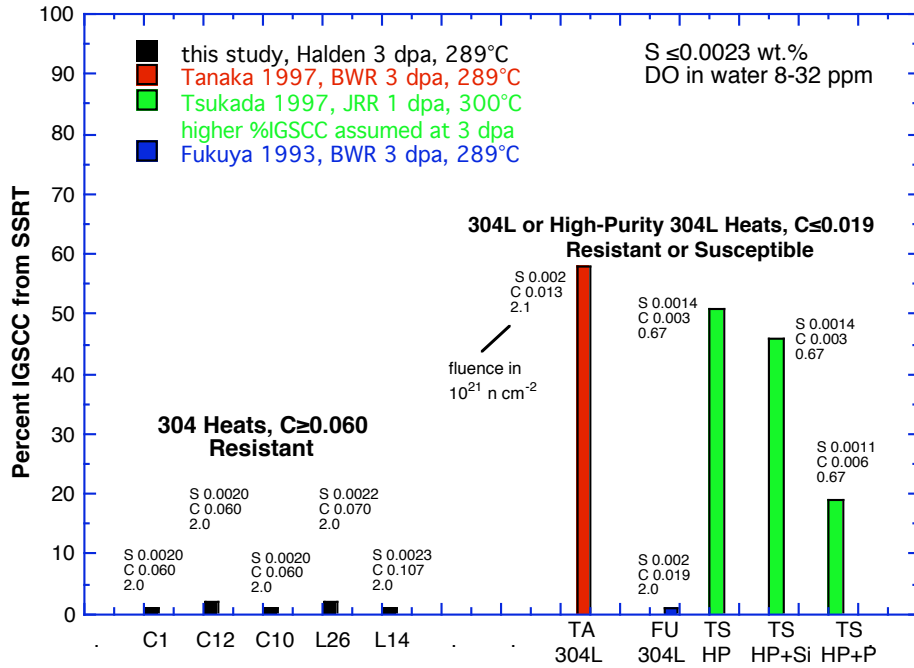


Figure 36. Percent IGSCC from SSRT tests of low-S high-C and low-S low-C heats of Type 304 or 304L SSs.

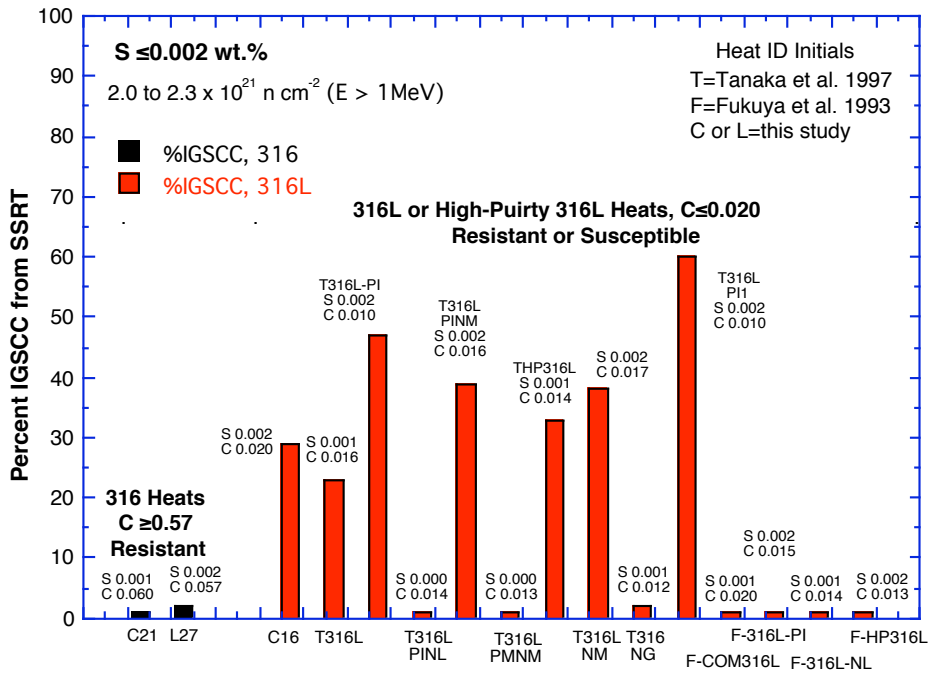


Figure 37. Percent IGSCC from SSRT test of low-S high-C and low-S low-C heats of Type 316 or 316L SSs.



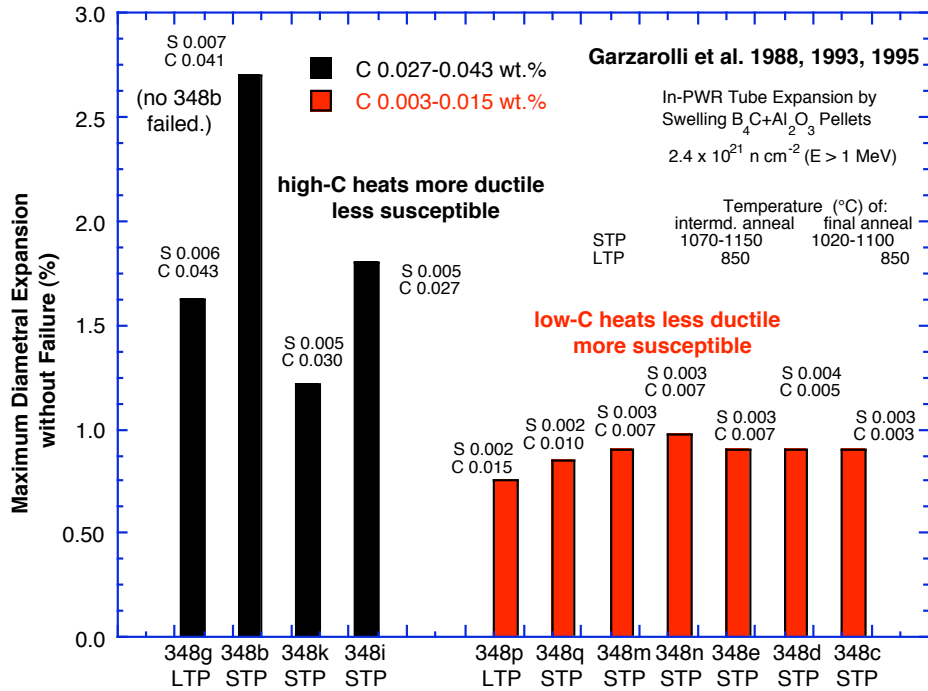


Figure 38. Diametral strain at failure for low-S high-C and low-S low-C heats of Type 348 SSs. Data from Garzarolli et al., Refs. 2, 7, and 42, one-cycle irradiation.

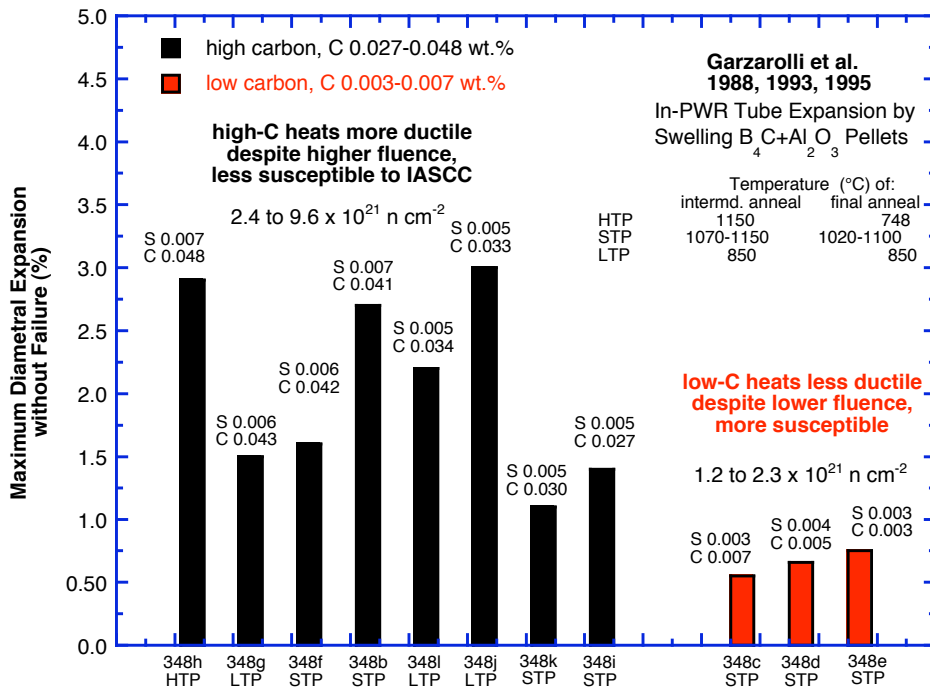


Figure 39. Diametral strain at failure for low-S high-C and low-S low-C heats of Type 348 SSs. Data from Garzarolli et al. Refs. 2, 7, and 42, one-to-three-cycle irradiation.

When S concentration is high, e.g.,  $\geq 0.004$  wt.% in 304- or 316-type SSs and  $\geq 0.01$  wt.% in 348-type SSs, it appears that the deleterious effect of S is so dominant that the effect of C is negligible or insignificant, even at high concentrations of C.

## 8.6 Effect of Si at High Fluence

As pointed out in Section 7, for a medium level of damage ( $\approx 1.3$  dpa), the data in Fig. 17 indicate that Si in high concentrations either delays the onset of IGSCC or suppresses the susceptibility to IGSCC. However, by the time the damage level reaches  $\approx 3$  dpa, it appears that the deleterious effect of S is so dominant that the effect of Si is either insignificant or secondary at best. This conclusion can be deduced from Fig. 22, in which percent IGSCC is plotted as a function of Si content.

Three 304-type laboratory heats that contained unusually high concentrations of Si were tested in 289°C water in this investigation after irradiation to  $\approx 3$  dpa, i.e., Heats L14 (Si 1.49 wt.%), L15 (Si 1.82), and L6 (Si 1.90). These Si concentrations well exceed the AISI-specified limit of 1.0 wt.%. As shown in Table 14, the three heats exhibited relatively higher level of uniform elongation compared to other laboratory heats of 304- or 316-type SSs. However, in spite of this, the susceptibility of the three heats appeared to be determined largely by the concentrations of S and C. This is shown in Fig. 40. At  $\approx 3$  dpa, there was no clear evidence that high Si content suppresses the susceptibility to IASCC of the steels investigated in this study.

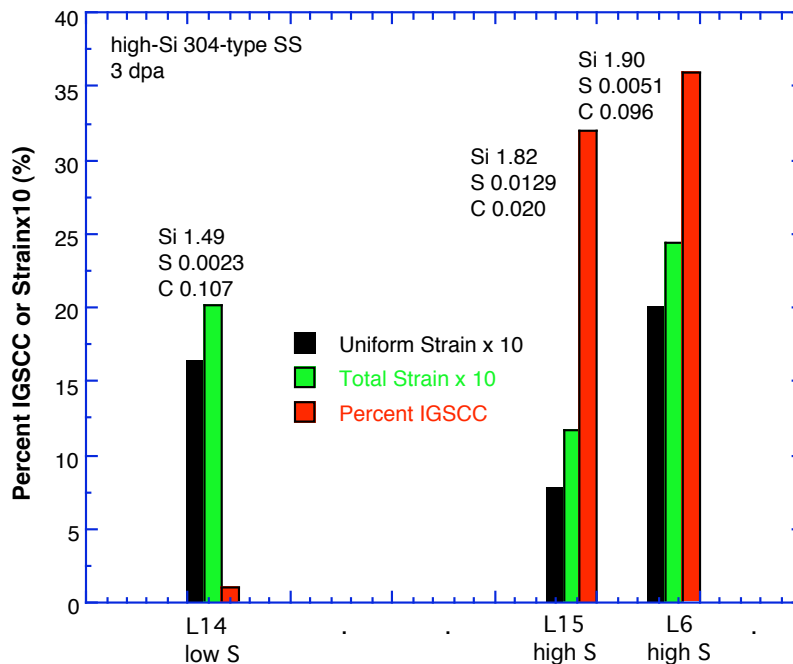


Figure 40. Ductility and percent IGSCC of low- and high-S laboratory heats of Type 304 SSs that contain unusually high Si content.

Some of the low-C, low-S heats of 316L-type SSs (damage level  $\approx 3$  dpa), shown in Fig. 37, contain an unusually low Si concentration of  $\leq 0.05$  wt.%.<sup>19,41</sup> When compared with the other heats in the figure that contain higher levels of Si (0.38-0.59 wt.%), the IASCC behavior of the high- and low-Si heats was similar; this is shown in Fig. 41.

As in the case of 304- and 316-type SSs, IASCC susceptibility of 348-type SS at high fluence ( $\approx 3$  dpa) seems to be determined largely by the concentrations of S and C, and the effect of Si appears secondary.

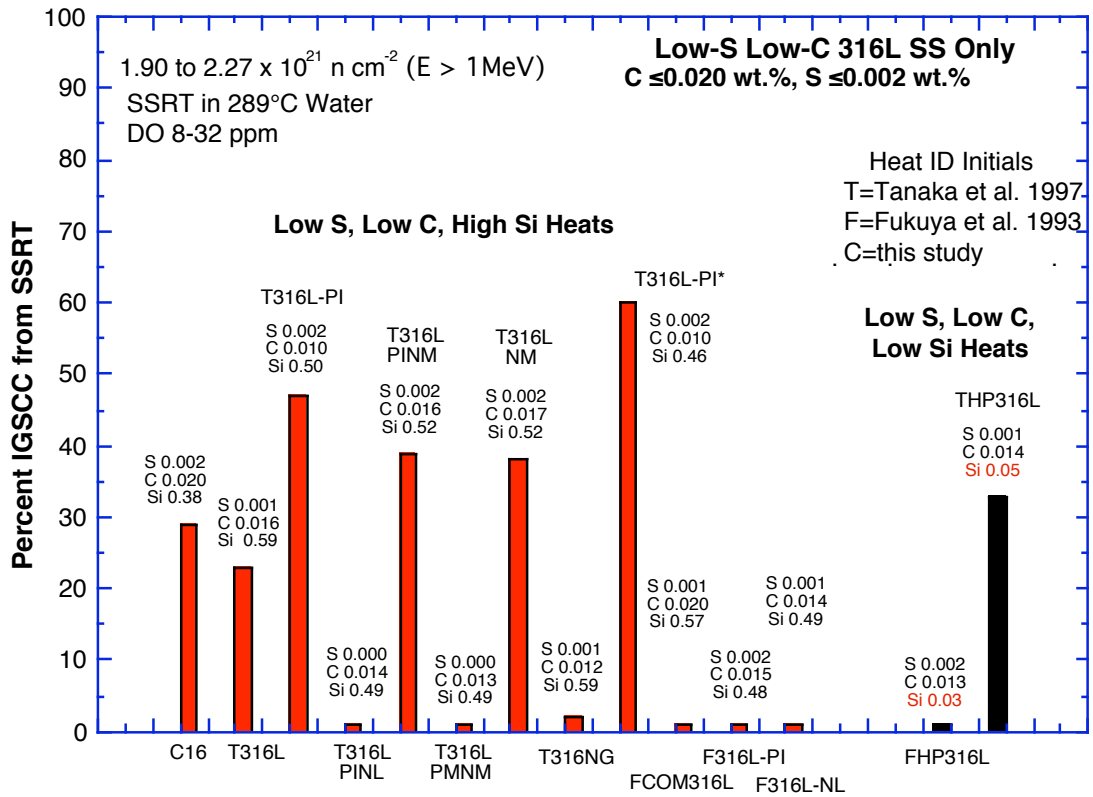


Figure 41. Comparison of failure diametral strain for high- and low-Si heats of Type 316L SSs that contain low-S and low-C concentrations. Data from Fukuya et al., Ref. 19 and Tanaka et al., Ref. 41.