IRRADIATION-ASSISTED STRESS CORROSION CRACKING OF MODEL AUSTENITIC STAINLESS STEELS*

by

H. M. Chung, W. E. Ruther, R. V. Strain, and W. J. Shack
Argonne National Laboratory
Argonne, IL 60439, USA

and

T. M. Karlsen
OECD Halden Reactor Project
Halden, Norway

September 1999

To be published in the Proceedings of 9th International Conference on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, August 1-5, 1999, Newport Beach, CA.

*Work supported by the U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research.

DISCLAIMER

IRRADIATION-ASSISTED STRESS CORROSION CRACKING OF MODEL AUSTENITIC STAINLESS STEELS*

H. M. Chung, W. E. Ruther, R. V. Strain, and W. J. Shack
Argonne National Laboratory
Argonne, IL 60439, USA

T. M. Karlsen
OECD Halden Reactor Project
Halden, Norway

Abstract

Slow-strain-rate tensile (SSRT) tests were conducted on model austenitic stainless steel (SS) alloys that were irradiated at 289°C in He. After irradiation to ≈0.3 x 10^{21} n·cm^{-2} and ≈0.9 x 10^{21} n·cm^{-2} (E > 1 MeV), significant heat-to-heat variations in the degree of intergranular and transgranular stress corrosion cracking (IGSCC and TGSCC) were observed. At ≈0.3 x 10^{21} n·cm^{-2}, a high-purity heat of Type 316L SS that contains a very low concentration of Si exhibited the highest susceptibility to IGSCC. In unirradiated state, Types 304 and 304L SS did not exhibit a systematic effect of Si content on alloy strength. However, at ≈0.3 x 10^{-21}n cm^{-2}, yield and maximum strengths decreased significantly as Si content was increased to >0.9 wt.%. Among alloys that contain low concentrations of C and N, ductility and resistance to TGSCC and IGSCC were significantly greater for alloys with >0.9 wt.% Si than for alloys with <0.47 wt.% Si. Initial data at ≈0.9 x 10^{-21}n cm^{-2} were also consistent with the beneficial effect of high Si content. This indicates that to delay onset of and reduce susceptibility to irradiation-assisted stress corrosion cracking (IASCC), at least at low fluence levels, it is helpful to ensure a certain minimum concentration of Si. High concentrations of Cr were also beneficial; alloys that contain <15.5 wt.% Cr exhibited greater susceptibility to IASCC than alloys with ≈18 wt.% Cr, whereas an alloy that contains >21 wt.% Cr exhibited less susceptibility than the lower-Cr alloys under similar conditions.

Key words: austenitic stainless steels, irradiation, transgranular and intergranular stress corrosion cracking, ductility, yield strength, silicon, chromium.

*Work supported by the U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research.

Introduction

As neutron fluence increases, austenitic stainless steel core internals of boiling and pressurized water reactors (BWRs and PWRs) become susceptible to IASCC. Although most failed components can be repaired or replaced, such operations are difficult and expensive. Therefore, extensive research has been conducted to develop understanding of this form of degradation.[1-18] Irradiation–induced grain–boundary depletion of Cr has been considered by most investigators to be the primary metallurgical process that causes IASCC. Chromium–depleted zones at grain boundaries have been observed, and the effects of electrochemical potential (ECP) on susceptibility to SCC of nonirradiated, thermally sensitized material (where Cr–depletion is widely recognized as the primary factor) and on susceptibility to IASCC of BWR–irradiated solution–annealed material have been reported to be similar.[1-3] However, contrary to expectations based on the strong effect of ECP on SCC associated with the Cr–depletion mechanism, cracking of highly stressed components has been reported in PWRs, which operate at low ECPs, and the susceptibility of PWR–irradiated components to cracking at low ECP has been demonstrated in several experiments.[4,5] Other investigators have suggested radiation–induced segregation of impurities, such as Si, P, and S, as a process that causes IASCC.[4,6,7] The superior resistance to IASCC of a heat of Type 348 SS that contained very low levels of C, Si, P, and S seemed to provide evidence for this.[4] However, later investigations indicated that the resistance of high–purity (HP) heats (low in C, Si, S, and P) of Type 304 SS is no better than that of commercial–purity (CP) Type 304 SSs.[8-14]

Although it has been known for many years that IASCC is generally characterized by strong heat-to-heat variations even among the same grade of SSs, the origin of the variations is not well understood. Therefore, a joint irradiation testing program was initiated to systematically investigate the effects of alloying and impurity elements (Cr, Ni, Si, P, S, Mn, C, and N) on the susceptibility of austenitic SSs to IASCC. In the joint program, many austenitic SSs were irradiated in the Halden reactor, and postirradiation tests and examinations are being conducted at Argonne National Laboratory. This paper describes the results obtained to date from slow-strain-rate-tensile (SSRT) tests on specimens irradiated to $\approx 0.3 \times 10^{21}$ n·cm^{-2} and $\approx 0.9 \times 10^{21}$ n·cm^{-2} (E > 1 MeV).

Experimental Procedure

An optimized test matrix was constructed according to the method of Taguchi.[19,20] Based on the optimized matrix, 8 commercial and 19 laboratory heats of model austenitic SS alloys were procured.[20] High- and commercial-purity (HP and CP) heats of Types 304, 316 and 348 SS were included in the test matrix. The compositions of the 27 model alloys are given in Table 1. Slow-strain-rate-tensile and 1/4T compact-tension (CT) specimens were prepared from the alloys. The specimens were irradiated in the Halden reactor at 289°C in He to three fluence levels, 0.3, 0.9, and 2.5×10^{21} n·cm^{-2} (E > 1 MeV).

Susceptibility to IASCC was determined by SSRT tests of the irradiated specimens in simulated BWR water, and posttest fractographic analysis was conducted by scanning electron microscopy (SEM) to measure the degree of transgranular and intergranular fracture. All SSRT tests were conducted at 289°C in deionized HP water that contained ≈8 ppm dissolved oxygen (DO). Conductivity and pH of the water were kept at ≈0.07-0.10

Table I. Composition of twenty-seven commercial and laboratory model austenitic stainless steel alloys irradiated in the Halden Reactor

ANL	Source	Composition (wt.%)										
ID[a]	Heat ID	Ni	Si	P	S	Mn	C	N	Cr	O	B	Mo or Nb
C1	DAN-70378	8.12	0.50	0.038	0.002	1.00	0.060	0.060	18.11	-	<0.001	-
L2	BPC-4-111	10.50	0.82	0.080	0.034	1.58	0.074	0.102	17.02	0.0065	<0.001	-
C3	PNL-C-1	8.91	0.46	0.019	0.004	1.81	0.016	0.083	18.55	-	<0.001	-
L4	BPC-4-88	10.20	0.94	0.031	0.010	1.75	0.110	0.002	15.80	-	<0.001	-
L5	BPC-4-104	9.66	0.90	0.113	0.028	0.47	0.006	0.033	21.00	-	<0.001	-
L6	BPC-4-127	10.00	1.90	0.020	0.005	1.13	0.096	0.087	17.10	0.0058	<0.001	-
L7	BPC-4-112	10.60	0.18	0.040	0.038	1.02	0.007	0.111	15.40	0.0274	<0.001	-
L8	BPC-4-91	10.20	0.15	0.093	0.010	1.85	0.041	0.001	18.30	-	<0.001	-
C9	PNL-C-6	8.75	0.39	0.013	0.013	1.72	0.062	0.065	18.48	-	<0.001	-
C10	DAN-23381	8.13	0.55	0.033	0.002	1.00	0.060	0.086	18.19	-	<0.001	-
L11	BPC-4-93	8.15	0.47	0.097	0.009	1.02	0.014	0.004	17.40	-	<0.001	-
C12	DAN-23805	8.23	0.47	0.018	0.002	1.00	0.060	0.070	18.43	-	<0.001	-
L13	BPC-4-96	8.18	1.18	0.027	0.022	0.36	0.026	0.001	17.40	-	<0.001	-
L14	BPC-4-129	7.93	1.49	0.080	0.002	1.76	0.107	0.028	15.00	0.0045	<0.001	-
L15	BPC-4-126	8.00	1.82	0.010	0.013	1.07	0.020	0.085	17.80	0.0110	<0.001	-
C16	PNL-SS-14	12.90	0.38	0.014	0.002	1.66	0.020	0.011	16.92	-	<0.001	-
L17	BPC-4-128	8.00	0.66	0.090	0.009	0.48	0.061	0.078	15.30	0.0092	<0.001	-
L18	BPC-4-98	8.13	0.14	0.016	0.033	1.13	0.080	0.001	18.00	-	<0.001	-
C19	DAN-74827	8.08	0.45	0.031	0.003	0.99	0.060	0.070	18.21	-	<0.001	-
L20	BPC-4-101	8.91	0.017	0.010	0.004	0.41	0.002	0.002	18.10	-	<0.001	-
C21[b]	DAN-12455	10.24	0.51	0.034	0.001	1.19	0.060	0.020	16.28	-	<0.001	Mo 2.08
L22[c]	BPC-4-100	13.30	0.024	0.015	0.004	0.40	0.003	0.001	16.10	-	<0.001	Mo 2.04
L23[d]	BPC-4-114	12.04	0.68	0.030	0.047	0.96	0.043	0.092	17.30	0.0093	<0.001	Nb 1.06
L24[e]	BPC-4-105	12.30	0.03	0.007	0.005	0.48	0.031	0.002	16.90	0.0129	<0.001	Nb 1.72
L25C3	BPC-4-133	8.93	0.92	0.020	0.008	1.54	0.019	0.095	17.20	0.0085	0.010	-
L26C19	BPC-4-131	8.09	0.79	0.004	0.002	0.91	0.070	0.089	17.20	0.0080	<0.001	-
L27C21	BPC-4-132	10.30	0.96	0.040	0.002	0.97	0.057	0.019	15.30	0.0058	0.030	Mo 2.01

[a]First letters "C" and "L" denote commercial and laboratory heats, respectively.
[b]Commercial-purity Type 316 SS.
[c]High-purity Type 316 SS.
[d]Commercial-purity Type 348 SS.
[e]High-purity Type 348 SS.

and 6.3-6.8, respectively. Strain rate was held constant at $1.65 \times 10^{-7}\ s^{-1}$. Electrochemical potential (ECP) was measured at the effluent side at regular intervals. SSRT tests and fractographic analysis have been completed for 16 alloys that were irradiated to a fluence of $\approx 0.3 \times 10^{21}$ $n \cdot cm^{-2}$ (E > 1 MeV) at ≈288°C in He. Initial tests were also conducted on 9 alloys of the total of 24 "medium-fluence" alloy specimens that were irradiated to $\approx 0.9 \times 10^{21}$ $n \cdot cm^{-2}$ (E > 1 MeV). In addition to the irradiated specimens, unirradiated control specimens were also tested under the same conditions to provide data on baseline properties (see Table 2). More detailed results on the unirradiated specimens can be found elsewhere.[21]

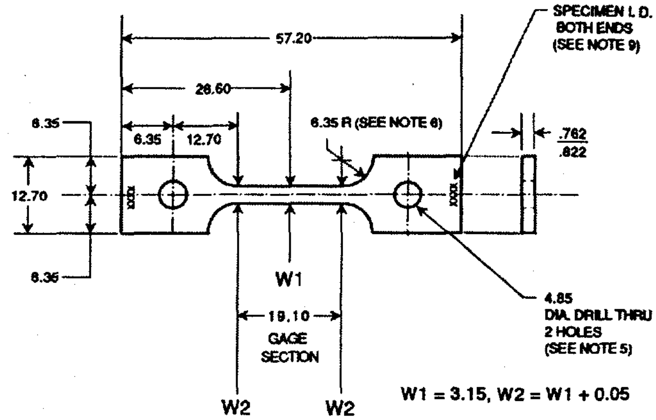


Figure 1. SSRT specimens irradiated at 289°C in He in the Halden reactor. All dimensions are in mm.

Results and Discussion - SSRT Behavior at Low Fluence

Feedwater chemistry (i.e., DO, ECP, conductivity, and pH) and results from SSRT tests (i.e., 0.2%-offset yield strength, maximum strength, uniform strain, and total strain) are summarized in Tables 3 and 4, respectively, for "low-fluence" specimens, i.e., the specimens irradiated to $\approx 0.3 \times 10^{21}$ $n \cdot cm^{-2}$ (E > 1 MeV). 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 4, the results of SSRT and SEM fractographic analysis (percent IGSCC, percent TGSCC, and combined percent IGSCC+TGSCC) are correlated with the compositions of the low-fluence specimens.

Heat-to-heat variations in susceptibility to IGSCC and TGSCC were significant, even at the low fluence of $\approx 0.3 \times 10^{-21} n\ cm^{-2}$ (E > 1 MeV). An high-purity heat L22 of Type 316L SS that contains the very low Si concentration of ≈0.024 wt.% exhibited relatively low ductility and the highest susceptibility to IGSCC (highest percent IGSCC) during the SSRT tests. At this low fluence, susceptibility of all other heats to IGSCC was insignificant. Heat L22 also exhibited relatively high susceptibility to IGSCC after irradiation to $\approx 0.9 \times 10^{-21} n\ cm^{-2}$ (E > 1 MeV) (see later).

The relatively higher susceptibility of the HP heat of Type 316L SS (i.e., Heat L22) than the CP counterpart (i.e., Heat C21), which is similar to that observed for BWR neutron absorber tubes fabricated from HP heats of Type 304 SS,[14] is of particular interest. In an SSRT experiment similar to those of the present study, Jenssen and Ljunberg irradiated U-notched rod specimens that had been fabricated from two heats of Type 316 SS, then postirradiation SSRT tests were conducted in a BWR loop under conditions of normal oxidizing water chemistry.[18] A CP heat of Type 316 SS (Heat K) was resistant to IASCC, whereas an HP heat of Type 316L SS (Heat F) was susceptible. Table 5 presents the composition, irradiation conditions, and test results of the four heats of Type 316 SS. The two relatively more susceptible heats in the table (i.e., Heats L22 and F) are characterized by an unusually low Si concentration of <0.26 wt.%, whereas the two relatively more resistant heats (i.e., Heats C21 and K) contain a higher Si concentration of >0.5 wt.%.

Table II. Composition (wt.%) of nonirradiated control specimens correlated with results of SSRT tests[a] and SEM fractography

Alloy ID	Ni	Si	P	S	Mn	C	N	Cr	Mo/Nb	O (wppm)	Remark[b]	YS (MPa)	UTS (MPa)	UE (%)	TE (%)	TGSCC (%)	IGSCC (%)	TG+IGSCC (%)
L23	12.04	0.68	0.030	0.047	0.96	0.043	0.092	17.30	Nb 1.06	93	CP 348	332	480	15.6	17.0	15	0	15
L7	10.60	0.18	0.040	0.038	1.02	0.007	0.111	15.40	-	274	High N, O; Low Si, C	195	370	2.5	5.2	20	0	20
L14	7.93	1.49	0.080	0.002	1.76	0.107	0.028	15.00	-	45	High Si, P, C; Low S	240	474	41.8	44.2	0	0	0
L17	8.00	0.66	0.090	0.009	0.48	0.061	0.078	15.30	-	90	High P; Low Cr, Mn, S	189	412	11.6	13.3	60	0	60
L17	8.00	0.66	0.090	0.009	0.48	0.061	0.078	15.30	-	90	High P; Low Cr, Mn, S	184	447	30.1	31.2	8	0	8
L6	10.00	1.90	0.020	0.005	1.13	0.096	0.087	17.10	-	58	High Si, C, Cr; Low S	227	515	43.0	44.5	0	0	0
L27	10.30	0.96	0.040	0.002	0.97	0.057	0.019	15.30	Mo 2.01	-	CP 316 ; high B (0.030)	298	483	20.6	22.9	0	0	0
L26	8.09	0.79	0.004	0.002	0.91	0.070	0.089	17.20	-	80	Low P, S	184	506	38.2	40.2	0	0	0
L2	10.50	0.82	0.080	0.034	1.58	0.074	0.102	17.02	-	66	High P, S, Mn, N	193	348	6.6	7.8	57	0	57
L25	8.93	0.92	0.020	0.008	1.54	0.019	0.095	17.20	-	85	high B (0.010)	184	458	25.5	27.0	0	0	0
L15	8.00	1.82	0.010	0.013	1.07	0.020	0.085	17.80	-	110	High N; Low C	218	512	36.7	37.9	0	0	0
L24	12.30	0.03	0.007	0.005	0.48	0.031	0.002	16.90	Nb 1.72	-	HP 348; Low Si, N	352	461	10.4	12.3	10	0	10
C1	8.12	0.50	0.038	0.002	1.00	0.060	0.060	18.11	-	-	Low S, CP 304	179	498	49.4	51.7	0	0	0
C19	8.08	0.45	0.031	0.003	0.99	0.060	0.070	18.21	-	-	Low Si, S, CP 304	178	501	47.4	49.2	0	0	0
C9	8.75	0.39	0.013	0.013	1.72	0.062	0.065	18.48	-	-	Low Si, High Mn	178	408	17.4	19.4	32	0	32
C12	8.23	0.47	0.018	0.002	1.00	0.060	0.070	18.43	-	-	Low Si, S, P	182	511	46.0	47.6	0	0	0
C10	8.13	0.55	0.033	0.002	1.00	0.060	0.086	18.19	-	-	Low S, high N	174	478	30.6	35.1	0	0	0
C21	10.24	0.51	0.034	0.001	1.19	0.060	0.020	16.28	Mo 2.08	-	CP 316; low B (0.001)	277	455	48.9	59.5	0	0	0

[a]Test at 289°C at strain rate of $1.65 \times 10^{-7}\ s^{-1}$ in BWR-like water that contained ≈8 ppm DO.
[b]HP = high purity, CP = commercial purity.

Table III. Results of SSRT tests[a] and SEM fractography of specimens irradiated to ≈0.3×10^{21} $n \cdot cm^{-2}$ (E > 1 MeV)

Alloy and Specimen Ident. No.	SSRT No.	Feedwater Chemistry: Oxygen Conc. (ppm)	Average ECP (mV SHE)	Cond. at 25°C ($\mu S \cdot cm^{-1}$)	pH at 25°C	SSRT Parameters: Yield Stress (MPa)	Max. Stress (MPa)	Uniform Elongation (%)	Total Elongation (%)	Fracture Behavior: TGSCC (%)	IGSCC (%)	TGSCC + IGSCC (%)
C1-1	HR-1	8.3	+184	0.07	7.03	490	680	13.4	16.6	4	0	4
L5-1	HR-2	9.7	+208	0.07	6.89	413	539	29.5	32.7	2	2	4
L22-1	HR-3	8.0	+236	0.07	6.80	360	596	6.6	9.4	50	15	65
C3-1	HR-4	8.7	+161	0.07	6.68	338	491	27.7	31.6	5	0	5
C16-1	HR-5	8.3	+204	0.08	6.74	370	527	17.6	20.6	2	0	2
L4-1	HR-6	9.0	+202	0.08	6.70	367	542	19.7	22.3	46	0	46
L18-1	HR-7	9.0	+203	0.08	6.33	503	572	6.3	8.8	54	0	54
C10-1	HR-8	8.2	+174	0.07	6.35	523	640	17.4	18.9	6	0	6
C21-1	HR-9	8.1	+149	0.08	6.49	480	620	15.9	19.4	4	0	4
L11-1	HR-10	9.0	+157	0.08	6.17	487	599	2.3	3.8	62	0	62
L13-1	HR-11	8.7	+164	0.08	6.17	248	461	22.1	24.8	8	0	8
L20-1	HR-12	8.4	+174	0.07	6.20	454	552	2.9	5.1	32	2	34
C19-1	HR-13	9.5	+132	0.12	6.36	554	682	10.5	14.7	7	0	7
C9-1	HR-14	8.0	+192	0.11	6.30	522	607	13.4	14.6	24	0	24
C12-1	HR-15	9.0	+195	0.08	6.40	404	589	20.4	24.2	5	0	5
L8-1	HR-16	9.0	+215	0.08	6.60	411	571	15.6	17.9	54	0	54

[a]Tested at 289°C at strain rate of 1.65×10^{-7} s^{-1} in BWR-like water that contained ≈8 ppm DO.

Table IV. Composition (wt.%) of specimens irradiated to fluence of ≈0.3×10^{21} $n \cdot cm^{-2}$ (E > 1 MeV) correlated with results of SSRT tests[a] SEM fractography

Alloy ID	Ni	Si	P	S	Mn	C	N	Cr	Mo/Nb	Remarks[b]	YS (MPa)	UTS (MPa)	UE (%)	TE (%)	TGSCC (%)	IGSCC (%)	TG+IGSCC (%)
C1	8.12	0.50	0.038	0.002	1.00	0.060	0.060	18.11	-	Low S, CP 304	490	680	13.4	16.6	4	0	4
L5	9.66	0.90	0.113	0.028	0.47	0.006	0.033	21.00	-	High P, Cr; Low C	413	539	29.5	32.7	2	2	4
L22	13.30	0.024	0.015	0.004	0.40	0.003	0.001	16.10	Mo 2.04	HP 316L, low Si, N	360	596	6.6	9.4	50	15	65
C3	8.91	0.46	0.019	0.004	1.81	0.016	0.083	18.55	-	CP 304L, Low Si	338	491	27.7	31.6	5	0	5
C16	12.90	0.38	0.014	0.002	1.66	0.020	0.011	16.92	-	High Ni; Low Si, S	370	527	17.6	20.6	2	0	2
L4	10.20	0.94	0.031	0.010	1.75	0.110	0.002	15.80	-	High Ni, Mn, C; Low N	367	542	19.7	22.3	38	0	38
L18	8.13	0.14	0.016	0.033	1.13	0.080	0.001	18.00	-	Low Si, N	503	572	6.3	8.8	54	0	54
C10	8.13	0.55	0.033	0.002	1.00	0.060	0.086	18.19	-	Low S, CP 304	523	640	17.4	18.9	6	0	6
C21	10.24	0.51	0.034	0.001	1.19	0.060	0.020	16.28	Mo 2.08	CP316	480	620	15.9	19.4	4	0	4
L11	8.15	0.47	0.097	0.009	1.02	0.014	0.004	17.40	-	High P; Low Si, C, S, N	487	599	2.3	3.8	62	0	62
L13	8.18	1.18	0.027	0.022	0.36	0.026	0.001	17.40	-	High Si; Low Mn, C, N	248	461	22.1	24.8	8	0	8
L20	8.91	0.017	0.010	0.004	0.41	0.002	0.002	18.10	-	HP 304L, Low Si, N	454	552	2.9	5.1	32	2	34
C19	8.08	0.45	0.031	0.003	0.99	0.060	0.070	18.21	-	Low Si, S	554	682	10.5	14.7	7	0	7
C9	8.75	0.39	0.013	0.013	1.72	0.062	0.065	18.48	-	Low Si; High Mn	522	607	13.4	14.6	24	0	24
C12	8.23	0.47	0.018	0.002	1.00	0.060	0.070	18.43	-	Low Si, P, S	404	589	20.4	24.2	5	0	5
L8	10.20	0.15	0.093	0.010	1.85	0.041	0.001	18.30	-	High Ni, P, Mn; Low Si, N	411	571	15.6	17.8	64	0	64

[a]Test at 289°C at strain rate of 1.65×10^{-7} s^{-1} in BWR-like water; DO ≈8 ppm
[b]HP = high purity, CP = commercial purity.

Table V. Composition (wt.%) and relative susceptibility to IASCC of Type 316 stainless steels irradiated and tested under BWR-like conditions.

Heat ID	Steel Type[a]	Source	Ni	Si	P	S	Mn	C	N	B	Cr	Mo	Irradiated in Reactor	Fluence, 10^{21} n cm^{-2}	Type of SCC Test	Relative Susceptibility
L22	HP316L	ANL	13.30	0.024	0.015	0.004	0.40	0.003	0.001	<0.001	16.10	2.04	Halden, He	0.3 and 0.9	SSRT in hot cell	high
C21	CP316	ANL	10.24	0.51	0.034	0.001	1.19	0.060	0.020	<0.001	16.28	2.08	Halden, He	0.3	SSRT in hot cell	low
F	HP316L	ABB	11.60	0.26	0.021	0.001	1.44	0.009	0.062	0.001	16.69	2.65	BWR	0.3-9.0	SSRT in BWR loop	high
K	CP316	ABB	12.40	0.64	0.016	0.006	1.73	0.055	0.029	<0.0004	16.51	2.25	BWR	0.3-9.0	SSRT in BWR loop	low

[a]HP = high purity, CP = commercial purity.

exhibit a systematic effect of Si content. However, after irradiation to ≈0.3 x 10^{-21}n cm^{-2} (E > 1 MeV), yield and maximum strength decreased significantly as Si content increased to >0.9 wt.%.

The systematic influence of Si content indicates that at high concentrations, Si atoms suppress formation of some type of irradiation-induced hardening centers and contribute to greater ductility and lower susceptibility to IASCC, at least at low fluence. To show this more directly, total strain and the combined percent TGSCC plus percent IGSCC were correlated with Si concentration for four low-fluence specimens that contain low concentrations of C (0.003-0.26 wt.%) and N (0.001-0.033 wt.%). These 4 alloys (i.e., L22, L11, L5, and L13) were considered an ideal combination that could provide more direct information on the effect of Si. The results are shown in Fig. 3. The two alloys that contained low concentrations of Si (0.02-0.47 wt.%) exhibited consistently smaller total elongation and higher percent TGSCC plus percent IGSCC than the two alloys that contain high concentrations of Si (0.90-1.18 wt.%).

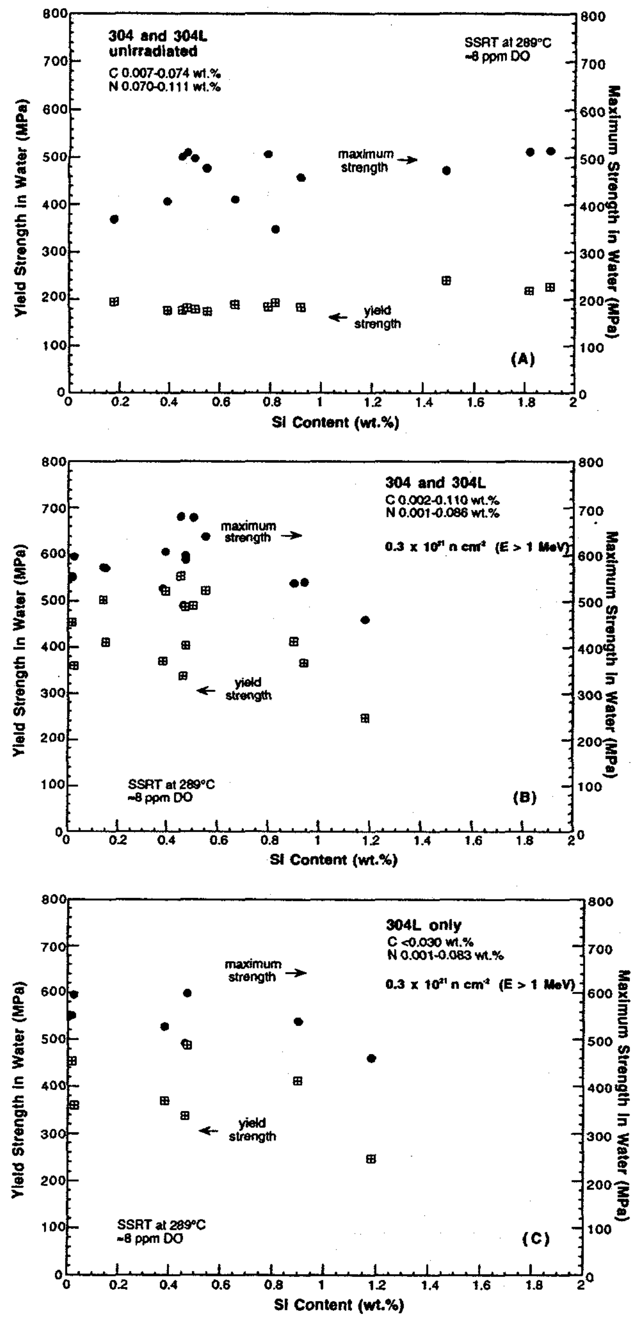


Figure 2. Effects of Si concentration on yield and maximum strength of (top) unirradiated Types 304 and 304L SS (middle) Types 304 and 304L SS irradiated to ≈0.3 x 10^{-21}n cm^{-2} (E > 1 MeV) and (bottom) Type 304L SS irradiated to ≈0.3 x 10^{-21}n cm^{-2}.

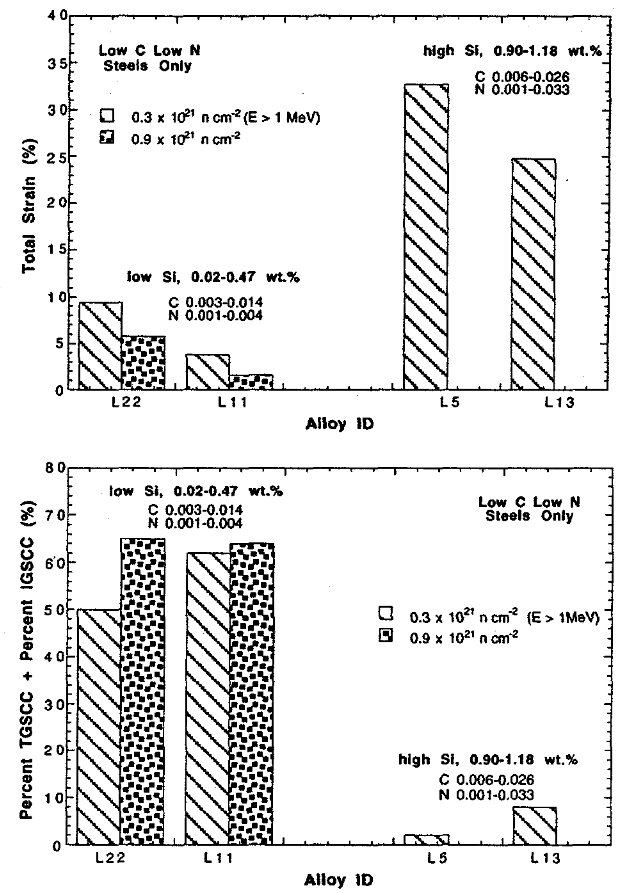


Figure 3. Effects of Si concentration on (top) total elongation and (bottom) percent TGSCC plus IGSCC of alloys that contain low C (0.003-0.026 wt.%) and low N (0.001-0.033 wt.%) and were irradiated to ≈0.3 x 10^{-21}n cm^{-2} (E > 1 MeV).

An interesting observation from Tables 2 and 4 is the effect of Si content on the alloy strength that was measured in water before and after irradiation. The yield strengths of the unirradiated commercial heats of Types 304 and 304L SS, measured in water, were similar to those measured in air,[6] 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. 2 for the unirradiated and low-fluence specimens. In the unirradiated state, Types 304 and 304L SS did not

Susceptibilities of the 16 tested alloys to TGSCC and IGSCC after irradiation to ≈0.3 x 10^{21} n·cm^{-2} (E > 1 MeV) are shown in Figs. 4 and 5, respectively. In contrast to

IGSCC, susceptibility to TGSCC was significant for seven alloys, whereas for the other nine alloys, susceptibility was insignificant. Compositional characteristics of the seven TGSCC-susceptible alloys, presented in Fig. 4 and Table 4, indicate that Si and N play a role in TGSCC. In Fig. 6, susceptibilities of all 16 alloys to TGSCC are classified and replotted in terms of N and Si concentrations. All alloys that contain <0.01 wt.% N and <1.0 wt.% Si were susceptible to TGSCC, whereas all alloys that contain >0.01 wt.% N or >1.0 wt.% Si were relatively resistant to TGSCC. This result indicates that, to delay onset of and to reduce susceptibility to IASCC, it is helpful to ensure that the alloy N concentration is >0.01 wt.% and Si concentration is >1.0 wt.%. Because practically all commercial heats of Types 304 or 304L SSs contain >0.01 wt.% N, to delay onset of and increase resistance to IASCC, it is helpful to ensure a certain minimum concentration of Si in steels. However, a Si concentration that is too high would not be desirable, because other factors, such as weldability and austenite stability, would complicate overall performance. Therefore, further investigation is needed to determine an optimal range of Si concentration, especially testing of steels irradiated to higher damage levels (e.g., >3 dpa).

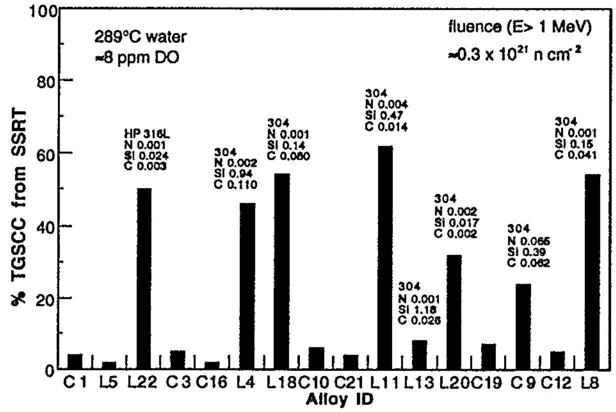


Figure 4. Percent TGSCC of model SS alloys irradiated in He in Halden reactor to fluence of ≈0.3 x 10^{21} n cm^{-2} (E > 1 MeV) and tested at 289°C in simulated BWR water.

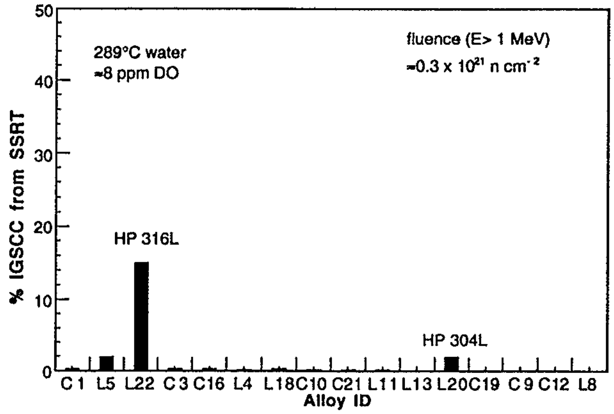


Figure 5. Percent IGSCC of model SS alloys irradiated in He in Halden reactor to fluence of ≈0.3 x 10^{21} n cm^{-2} (E > 1 MeV) and tested at 289°C in BWR-like water.

Miwa et al.[22] and Tsukada et al.[23] irradiated two SSRT specimens of HP 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 ≈240°C in the JRR-3 reactor to a fluence of ≈0.67 x 10^{-21}n cm^{-2} (E > 1 MeV). One specimen had been doped with ≈0.69 wt.% Si and the other had not been doped. After SSRT testing of the irradiated specimens at ≈300°C in HP water (DO ≈32 ppm), they observed that the Si-doped specimen exhibited significantly greater ductility than the undoped specimen, i.e., total elongation was ≈21 vs. 11 %. The results shown in Fig. 3A appear to be consistent with these reports.

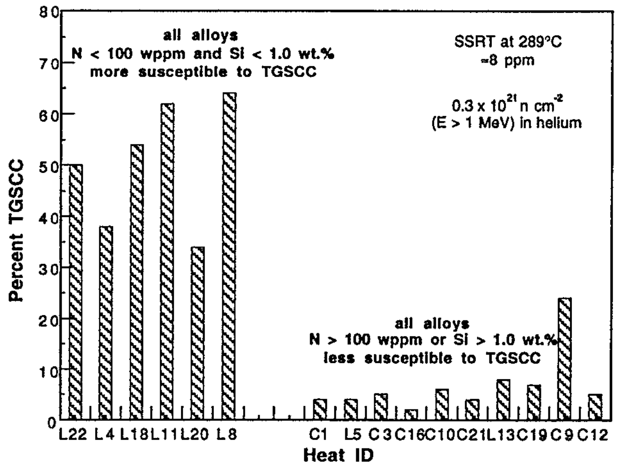


Figure 6. Susceptibility to TGSCC of model SS alloys, irradiated in He in Halden reactor to fluence of ≈0.3 x 10^{21} n cm^{-2} (E > 1 MeV) and tested at 289°C in simulated BWR water, classified as a function of N and Si content of the alloys.

Results and Discussion - SSRT Behavior at Medium Fluence

Initial tests were conducted on nine "medium-fluence" specimens irradiated to ≈0.9 x 10^{21} n cm^{-2} (E > 1 MeV); see Tables 6 and 7. For all tested medium-fluence specimens, the effects of the higher fluence on yield stress, maximum stress, uniform strain, total strains, percent IGSCC, and percent TGSCC were significant., as shown in Fig. 7. Preliminary results from the tests also indicate that when fluence was increased from ≈0.3 x 10^{21} n·cm^{-2} (E > 1 MeV) to ≈0.9 x 10^{21} n·cm^{-2} in the low-N and low-Si alloys (e.g., Heats L22, L18, and L11), susceptibility to TGSCC decreased, and at the same time, susceptibility to IGSCC increased at the expense of percent TGSCC (see Figs. 7D and E). However, the threshold fluence for the transition from TGSCC to IGSCC appears to differ from alloy to alloy. For example, susceptibility to TGSCC of a commercial Type 304 SS, Heat C9, still increased when fluence was increased from ≈0.3 to 0.9 x 10^{21} n cm^{-2} (E > 1 MeV).

Susceptibility to IGSCC of all alloys that contain <0.5 wt.% Si (i.e., L22, L18, L11, C9, and L7) increased significantly when fluence was increased from ≈0.3 x 10^{21} n·cm^{-2} (E > 1 MeV) to ≈0.9 x 10^{21} n·cm^{-2} (see Fig. 7E), indicating deleterious effect of this low concentration of Si. That is, under otherwise similar conditions, a low concentration of Si appears to promote susceptibility to TGSCC and IGSCC at relatively low fluences.

There was also strong evidence to support the premise that low concentrations of Cr (<15.5 wt.%) promote susceptibility of Type 304 SS to IASCC. For the same fluence level of ≈0.9 x 10^{21} n·cm^{-2} (E > 1 MeV), susceptibilities of L17 (Cr ≈ 15.3 wt.%) and L7 (Cr ≈ 15.4 wt.%) to IGSCC were significantly higher than that of other alloys that contain typical Cr concentrations of ≈18 wt.% (see Fig. 7E). Consistent with this observation, Alloy L5, which contains an unusually high Cr concentration of ≈ 21.0 wt.%, was resistant to both TGSCC and IGSCC at ≈0.3 x 10^{21} n·cm^{-2} (E > 1 MeV) (see Figs. 7D and E, respectively). This alloy also contains a relatively high concentration of Si (≈0.90 wt.%). The relatively good performance of Alloy L5 is also manifested by the highest ductility among all the alloys that were irradiated to ≈0.3 x 10^{21} n·cm^{-2} (Figs. 7B and C).

Alloy L7, a laboratory heat of Type 304L SS, exhibited significant susceptibility to TGSCC, even in the unirradiated state, [21] and the highest susceptibility to IGSCC after irradiation to ≈0.9 x 10^{21} n·cm^{-2} (E > 1 MeV). It appears that the high susceptibility to IASCC of this alloy is related to several deleterious compositional characteristics, i.e., unusually low concentration of Cr (≈15.3 wt.%), unusually high concentration of O (≈0.027 wt.%),[14,15,24] unusually low concentration of Si (≈0.18 wt.%), and a low concentration of C (≈0.007 wt.%).[14]

There are indications that a high concentration of Mn and low concentration of S are beneficial. For example, Alloy C3, a CP heat of Type 304L SS that contains ≈1.81 wt.% Mn and ≈0.004 wt.% S, exhibited unusually high ductility (>20%), low percent TGSCC (<9%), and low percent IGSCC (<4%) after irradiation to ≈0.3 x 10^{21} n·cm^{-2} (E > 1 MeV) and ≈0.9 x 10^{21} n·cm^{-2} (E > 1 MeV)(see Figs. 7D, E, and F, respectively). However, conclusive evidence for the effects of Mn, S, N, O, and C need to be established on the basis of more comprehensive data to be obtained on the whole test matrix, including specimens irradiated to ≈2.5 x 10^{21} n·cm^{-2} (E > 1 MeV).

Conclusions

1. Slow-strain-rate tensile tests were conducted on 16 austenitic stainless steel alloys that were irradiated at 289°C in He. After irradiation to ≈0.3 x 10^{21} n·cm^{-2} or ≈0.9 x 10^{21} n·cm^{-2} (E > 1 MeV), strong heat-to-heat variation in irradiation-induced hardening was observed. Heat-to-heat variations in susceptibility to IGSCC and TGSCC were also very significant among steels of the same grade that contain nominally similar concentrations of alloying and impurity elements. After irradiation to a fluence of ≈0.3 x 10^{21} n·cm^{-2} (E > 1 MeV), a high-purity laboratory heat of Type 316L SS that contains very low

Table VI. Results of SSRT tests[a] and SEM fractography for model austenitic stainless steels irradiated in He at 289°C to fluence of ≈0.9 x 10^{21} n·cm^{-2} (E > 1 MeV)

		Feedwater Chemistry				SSRT Parameters				Fracture Behavior		
Specimen Ident. No.	SSRT No.	Oxygen Conc. (ppm)	Average ECP (mV SHE)	Cond. at 25°C (μS·cm^{-1})	pH at 25°C	Yield Stress (MPa)	Max. Stress (MPa)	Uniform Elongation (%)	Total Elongation (%)	TGSCC (%)	IGSCC (%)	TGSCC + IGSCC (%)
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
L20-05	HR-26	9.0	+182	0.09	6.32	670	743	0.37	1.03	0	0	0
L20-06	HR-27	8.0	+274	0.07	6.05	632	697	0.85	2.72	-	-	-
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	490	531	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

[a]Tested at 289°C at a strain rate of 1.65 x 10^{-7} s^{-1} in simulated BWR water that contained ≈8 ppm DO.

Table VII. Composition (wt.%) of steels irradiated to fluence of ≈0.9 x 10^{21} n·cm^{-2} (E > 1 MeV) correlated with results of SSRT tests[a] and SEM fractography

Specimen ID	Ni	Si	P	S	Mn	C	N	Cr	O, B, Mo, Nb	Remarks[b]	YS (MPa)	UTS (MPa)	UE (%)	TE (%)	TGSCC (%)	IGSCC (%)	TG+IGSCC (%)
L22-02	13.30	0.024	0.015	0.004	0.40	0.003	0.001	16.10	Mo 2.04	HP 316L; Low Si, N	475	549	4.20	5.82	30	35	65
L11-02	8.15	0.47	0.097	0.009	1.02	0.014	0.004	17.40	-	high P; low Si, C, S, N	820	856	0.43	1.65	50	14	64
L18-02	8.13	0.14	0.016	0.033	1.13	0.080	0.001	18.00	-	low Si, N	710	755	3.98	5.05	38	14	52
L20-05	8.91	0.017	0.010	0.004	0.41	0.002	0.002	18.10	-	HP 304L; low Si, N, Mn	670	743	0.37	1.03	0	0	0
L20-06	8.91	0.017	0.010	0.004	0.41	0.002	0.002	18.10	-	HP 304L; low Si, N, Mn	632	697	0.85	2.72	-	-	-
C9-02	8.75	0.39	0.013	0.013	1.72	0.062	0.065	18.48	-	low Si; high Mn	651	679	1.42	2.50	62	22	84
L17-02	8.00	0.66	0.090	0.009	0.48	0.061	0.078	15.30	-	high P; low Cr, Mn, S	574	654	2.02	3.08	44	41	85
L7-02	10.60	0.18	0.040	0.038	1.02	0.007	0.111	15.40	O 0.0274	high N, O; low Si, C	490	531	0.24	2.44	38	54	92
C10-02	8.13	0.55	0.033	0.002	1.00	0.060	0.086	18.19	-	CP 304; low S; high N	651	706	6.35	9.25	14	0	14
C3-02	8.91	0.46	0.019	0.004	1.81	0.016	0.083	18.55	-	CP 304L; high Mn, N; low S	632	668	16.72	19.74	9	4	13

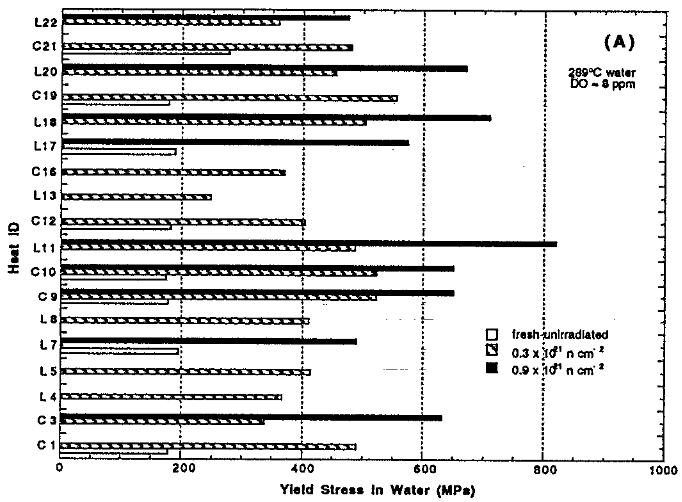
[a]Test at 289°C at a strain rate of 1.65 x 10^{-7} s^{-1} in BWR-simulated water; DO ≈8 ppm.
[b]HP = high purity, CP = commercial purity.

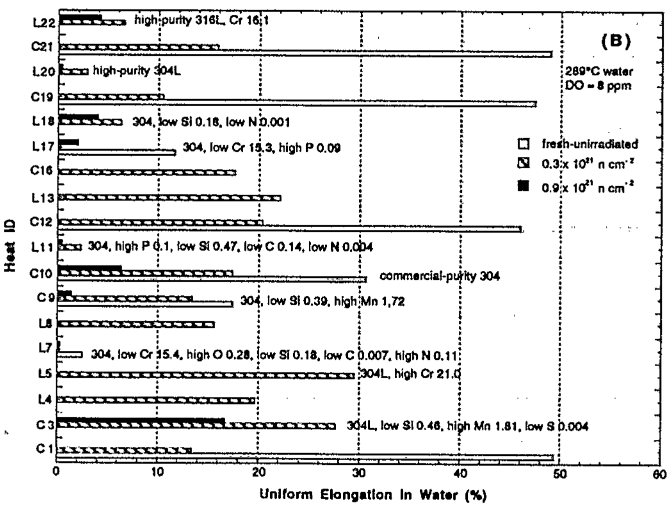
Si exhibited the highest susceptibility to intergranular stress corrosion cracking.

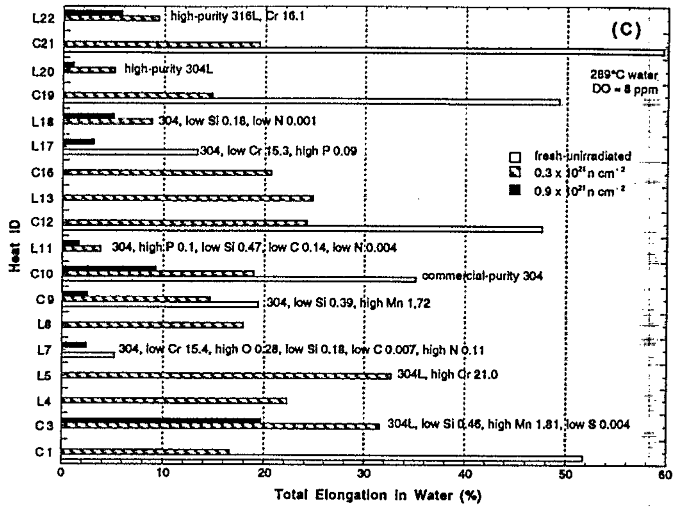
2. Yield and maximum strengths of nonirradiated Types 304 and 304L SS (measured in water) were not significantly influenced by Si concentration. However, Si concentration >0.9 wt.% was conducive to significantly lower yield and maximum strengths of the steels in water after irradiation.

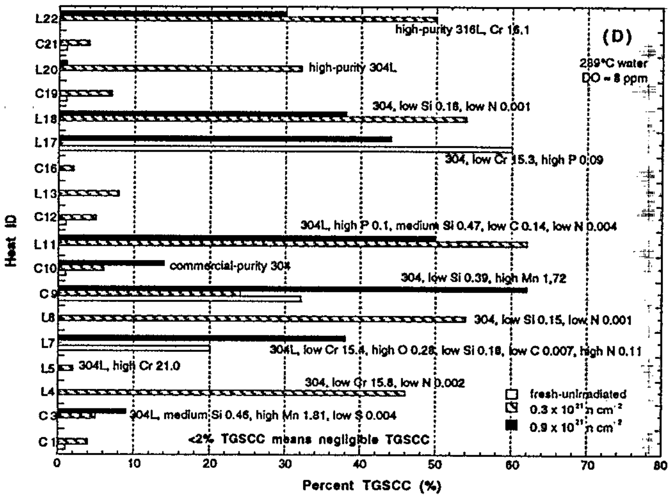
3. Susceptibilities to TGSCC of 16 alloys at ≈0.3 x 10^{21} $n \cdot cm^{-2}$ (E > 1 MeV) could be correlated with N and Si concentrations. All alloys that contained <0.01 wt.% N and <1.0 wt.% Si were susceptible, whereas all alloys that contained >0.01 wt.% N or >1.0 wt.% Si were relatively resistant. Practically all commercial heats of Types 304 or 304L SS contain >0.01 wt.% N, but Si concentration would typically be ≈0.5 wt.%. A certain minimum concentration of Si higher than this level appears to help suppress susceptibility to irradiation-assisted stress corrosion cracking, at least at low fluence.

4. High concentrations of Cr were beneficial; alloys that contain <15.5 wt.% Cr exhibited greater susceptibility to TGSCC and IGSCC than alloys that contain ≈18 wt.% Cr,









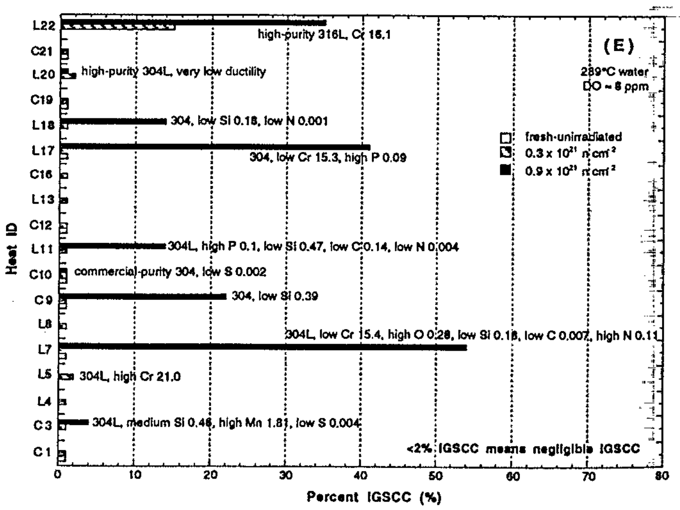


Figure 7. Effects of fluence on (A) yield strength, (B) uniform elongation, (C) total elongation, (D) percent TGSCC, and (E) percent IGSCC of various alloys measured in 289°C water that contained ≈8 ppm DO.

whereas an alloy that contains >21 wt.% Cr was less susceptible than the lower-Cr alloys under similar conditions.

Acknowledgments

The authors thank M. B. McNeil for many helpful discussions, and D. Perkins and L. J. Nowicki for contributions to the experimental efforts. This work was supported by the U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research.

References

1. M. E. Indig, J. L. Nelson, and G. P. Wozadlo, in Proc. 5th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, D. Cubicciotti, E. P. Simonen, and R. Gold, eds., American Nuclear Society, La Grange Park, IL, 1992, p. 941.
2. M. Kodama, S. Nishimura, J. Morisawa, S. Shima, S. Suzuki, and M. Yamamoto, ibid., p. 948.
3. H. M. Chung, W. E. Ruther, J. E. Sanecki, A. G. Hins, and T. F. Kassner, in Proc. 7th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, NACE International, Houston, 1995, p. 1133-1143.
4. F. Garzarolli, P. Dewes, R. Hahn, and J. L. Nelson, in Proc. 6th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, R. E. Gold and E. P. Simonen, eds., The Minerals, Metals, and Materials Society, Warrendale, PA, 1993, pp. 607-613.
5. I. Suzuki, M. Koyama, H. Kanasaki, H. Mimaki, M. Akiyama, T. Okubo, Y. Mishima, and T. R. Mager, in Proc. ASME–JSME 4th Intl. Conf. on Nuclear Engineering, March 10–14, 1996, New Orleans, American Society of Mechanical Engineers, New York, 1996, Vol. 5, pp. 205–213.
6. A. J. Jacobs, G. P. Wozadlo, K. Nakata, T. Yoshida, and I. Masaoka, in Proc. 3rd Int. Symp. Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, G. J. Theus and J. R. Weeks, eds., The Metallurgical Society, Warrendale, PA, 1988, p. 673.
7. K. Fukuya, K. Nakata, and A. Horie, in Proc. 5th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, D. Cubicciotti, E. P. Simonen, and R. Gold, eds., American Nuclear Society, La Grange Park, IL, 1992, pp. 814–820.
8. H. M. Chung, W. E. Ruther, J. E. Sanecki, A. G. Hins, and T. F. Kassner, in Effects of Radiation on Materials: 16th Int. Symp., ASTM STP 1175, A. S. Kumar, D. S. Gelles, R. K. Nanstad, and T. A. Little, eds., American Society for Testing and Materials, Philadelphia, 1993, pp. 851-869.
9. H. M. Chung, W. E. Ruther, J. E. Sanecki, and T. F. Kassner, in Proc. 6th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, R. E. Gold and E. P. Simonen, eds., The Minerals, Metals, and Materials Society, Warrendale, PA, 1993, pp. 511-519.
10. J. M. Cookson, D. L. Damcott, G. S. Was, and P. L. Anderson, ibid., pp. 573-580.
11. M. Kodama, J. Morisawa, S. Nishimura, K. Asano, S. Shima, and K. Nakata, J. Nucl. Mater., 1509 (1994) pp. 212–215.
12. T. Tsukada and Y. Miwa, in Proc. 7th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, NACE International, Houston, 1995, pp. 1009–1018.
13. F. Garzarolli, P. Dewes, R. Hahn, and J. L. Nelson, ibid., 1055–1065.
14. H. M. Chung, W. E. Ruther, J. E. Sanecki, A. G. Hins, N. J. Zaluzec, and T. F. Kassner, J. Nucl. Mater., 239 (1996) 61.
15. J. M. Cookson, G. S. Was, and P. L. Anderson, Corrosion 54 (1998) 299.
16. S. Kasahara, K. Nakata, K. Fukuya, S. Shima, A. J. Jacobs, G. P. Wozadlo, and S. Suzuki, in Proc. 6th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, R. E. Gold and E. P. Simonen, eds., The Minerals, Metals, and Materials Society, Warrendale, PA, 1993, pp. 615-623.
17. A. J. Jacobs, G. P. Wozadlo, K. Nakata, S. Kasahara, T. Okada, S. Kawano, and S. Suzuki, ibid., pp. 597-606.
18. A. Jenssen and L. G. Ljungberg, in Proc. 7th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, NACE International, Houston, 1995, 1043–1052.
19. G. Taguchi, in Quality Engineering Using Robust Design, M. S. Phadke, ed., Prentice Hall, Englewood Cliffs, NJ, 1989.
20. H. M. Chung, W. E. Ruther, R. V. Strain, and T. M. Karlsen, in Environmentally Assisted Cracking in Light Water Reactors, Semiannual Report, January-June 1998, NUREG/CR-4667, Vol. 26, ANL-98/30, pp. 10-21 (March 1999).
21. H. M. Chung, W. E. Ruther, R. V. Strain, and T. M. Karlsen, in Environmentally Assisted Cracking in Light Water Reactors, Semiannual Report, July 1997 - December 1997, NUREG/CR-4667, Vol. 25, ANL-98/18, pp. 26-38 (Sept. 1998).
22. Y. Miwa, T. Tsukada, S. Jitsukawa, S. Kita, S. Hamada, Y. Matsui, and M. Shindo, J. Nucl. Mater. 1393, (1996) pp. 233-237.
23. T. Tsukada, Y. Miwa, and H. Nakajima, in Proc. 8th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, Aug. 10-14, 1997, Amelia Island, FL, S. M. Bruemmer, ed., American Nuclear Society, La Grange Park, IL, 1997, pp. 846-856.
24. H. M. Chung, J.-H. Park, W. E. Ruther, J. E. Sanecki, R. V. Strain, and N. J. Zaluzec, in Proc. 8th Intl. Symp. on Environmental Degradation of Materials in Nuclear Power Systems - Water Reactors, August 10-14, 1997, Amelia Island, FL, S. M. Bruemmer, ed., American Nuclear Society, La Grange Park, IL, 1997, pp. 846-856.