



... for a brighter future



U.S. Department
of Energy

UChicago ►
Argonne_{LLC}



A U.S. Department of Energy laboratory
managed by UChicago Argonne, LLC

Task 1: Evaluation of the Causes & Mechanisms of IASCC in BWRs - Crack Growth & Fracture Toughness of Irradiated Stainless Steels

Investigators: Omesh Chopra, Gene Gruber, and Bill Shack

Experimental Effort: Ron Clark, Tom Galvin, and Loren Knoblich

September 25-26, 2007

Nuclear Engineering Division

Argonne National Laboratory, Argonne, IL 60439



Work sponsored by the US Nuclear Regulatory Commission

Objective

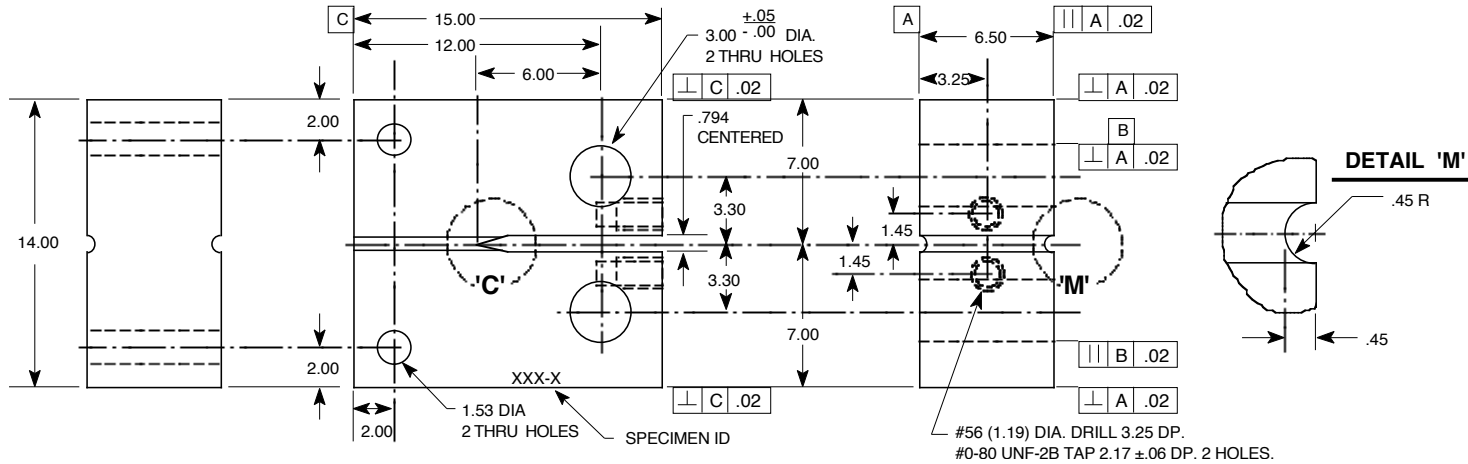
- Provide a better understanding of
 - Threshold fluence above which the effects of neutron irradiation on crack growth rates (CGRs) are significant
 - Disposition curve for cyclic & SCC growth rates of irradiated SSs
 - Fluence level above which benefit of HWC may be lost
- Significance of specimen size criteria
- Evaluate cyclic CGR data by using a superposition model
- Investigate the change in fracture toughness of austenitic SSs under LWR irradiation conditions & temperatures
 - Investigate effects of crack morphology (SCC IG vs. TG fatigue crack) and BWR environment on fracture toughness
- Review the existing fracture toughness data in order to assess potential for radiation embrittlement of reactor core internal components

Material

- CGR and/or fracture toughness J-R curve tests completed on SA Types 304L, 304, 316L, & 316 SS irradiated up to ≈ 3 dpa; sensitized 304 SS & HAZ of SAW & SMAW irradiated to ≈ 2.2 dpa; and thermally aged CF-8M cast SS irradiated to ≈ 2.5 dpa
- Materials irradiated in the Halden heavy boiling water reactor in Norway; SA SSs irradiated at $\approx 288^\circ\text{C}$ & others at $297\text{-}300^\circ\text{C}$

Heat ID	Steel	Ni	Si	P	S	Mn	C	N	Cr	Mo	O
C3	304L	9.10	0.45	0.020	0.003	1.86	0.024	0.074	18.93	0.12	0.014
C19	304	8.13	0.51	0.028	0.008	1.00	0.060	0.068	18.05	0.09	0.020
C16	316L	12.32	0.42	0.026	0.003	1.65	0.029	0.011	16.91	2.18	0.016
C21	316	10.45	0.61	0.035	0.002	1.23	0.060	0.016	16.27	2.10	0.014
10285	304	8.45	0.60	0.015	0.007	1.90	0.070	0.084	18.56	0.51	0.013
GG Top Shell	304L	9.05	0.53	0.027	0.016	1.84	0.013	0.064	18.23	0.44	0.010
GG Bottom Shell	304L	8.95	0.55	0.023	0.008	1.80	0.015	0.067	18.62	0.31	0.014
75	CF-8M	9.12	0.67	0.022	0.012	0.53	0.065	0.052	20.86	2.58	0.72

Specimen Geometry

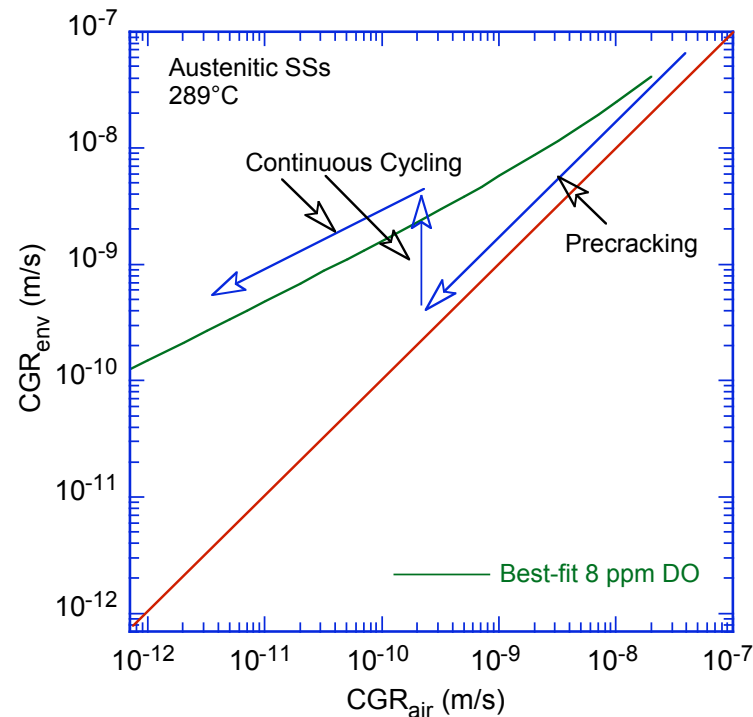


- Crack extension measured by DC potential drop method
- Current leads attached to the side of the specimen;
Potential leads attached across the notch

Experimental Conditions

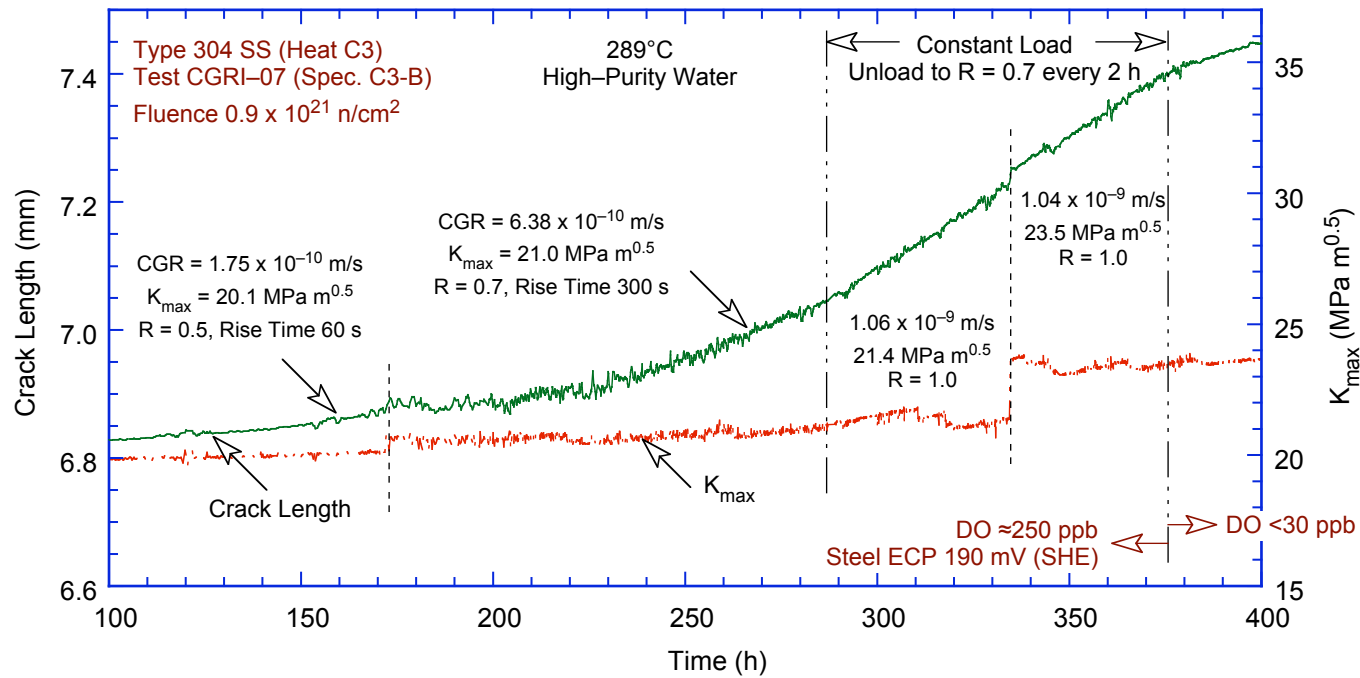
Temp:	289°C
DO:	≈350 ppb with N ₂ + 1% O ₂ cover gas <30 ppb with 5% H ₂ cover gas
Flow:	15–25 mL/min
Conductivity:	effluent 0.08 - 0.12 μS/cm
Cyclic Loading:	load ratio 0.3-0.7 sawtooth waveform with 12 to 1000 s rise time
SCC:	constant load with or w/o periodic partial unloading 1 or 2 h
K _{max} :	approximately constant by load shedding
K/size criterion:	$(W-a) \geq (2.5) (K/\sigma_{\text{effys}})^2$ with effective yield stress defined as $\sigma_{\text{effys}} = (\sigma_{\text{y nonirr}} + \sigma_{\text{y irr}})/2$
J-R curve tests:	constant extension rate of 0.026 mm/min blunting line given by $\Delta a = J/(4\sigma_{\text{feff}})$

Environmental Enhancement of Growth Rates



- Under more rapid cycling loading typically used for precracking, crack growth is dominated by mechanical fatigue
- For K_{max} 15-18 MPa $m^{1/2}$, environmental enhancement typically occurs at $R \geq 0.5$ & rise time ≥ 30 s; also fracture morphology changes from transgranular to intergranular

Enhanced Growth Rate for Irradiated Heat C3 of Type 304 SS



- Environmental enhancement observed after 170 h when load ratio & rise time changed from 0.5 & 60 s to 0.7 & 300s

Data Analysis

Cyclic CGR data analyzed using the superposition model

$$\text{CGR}_{\text{env}} = \text{CGR}_{\text{air}} + \text{CGR}_{\text{cf}} + \text{CGR}_{\text{scc}}$$

CGR_{air} determined from correlation by James & Jones

$$\text{CGR}_{\text{air}} = C_{\text{ss}} S(R) \Delta K^{3.3}/t_r$$

$$S(R) = 1.0 \quad R < 0$$

$$S(R) = 1.0 + 1.8R \quad 0 < R < 0.79$$

$$S(R) = -43.35 + 587.97R \quad 0.79 < R < 1.0$$

$$C_{\text{ss}} = \text{fn}(T) \text{ and } t_r \text{ is the rise time}$$

CGR_{cf} based on expressions proposed by Shack & Kassner

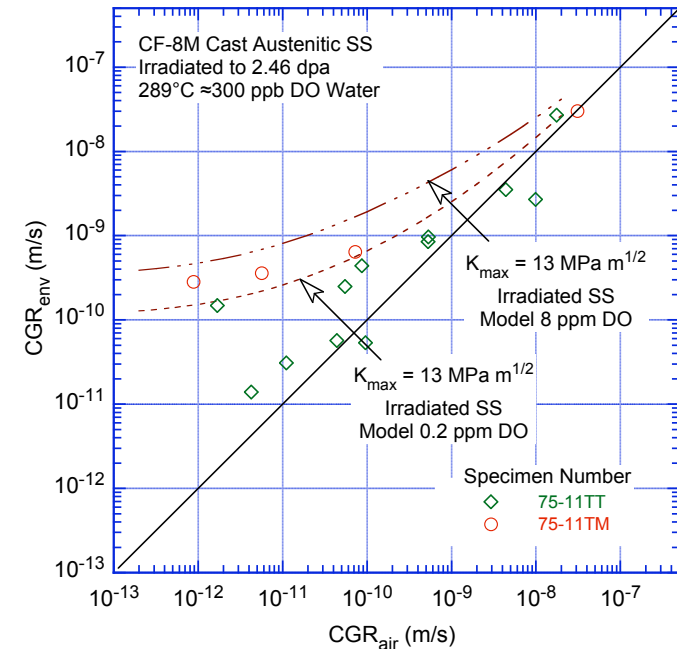
$$\text{CGR}_{\text{env}} = \text{CGR}_{\text{air}} + 4.5 \times 10^{-5} (\text{CGR}_{\text{air}})^{0.5} \approx 0.2 \text{ ppm DO}$$

$$\text{CGR}_{\text{env}} = \text{CGR}_{\text{air}} + 1.5 \times 10^{-4} (\text{CGR}_{\text{air}})^{0.5} \approx 8.0 \text{ ppm DO}$$

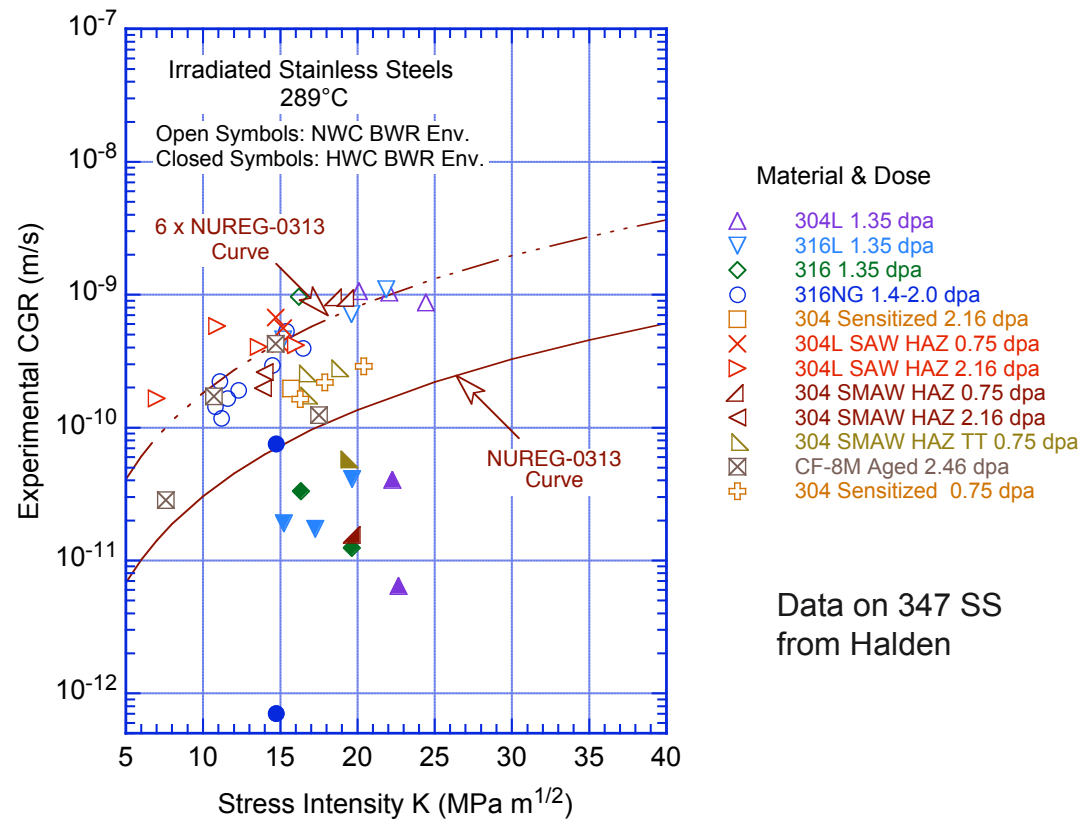
CGR_{scc} represented by correlation given in NUREG-0313

$$\text{CGR}_{\text{scc}} = A (K)^{2.161}$$

$$A = 2.1 \times 10^{-13} \text{ for sensitized SS \& } \approx 8 \text{ ppm DO}$$

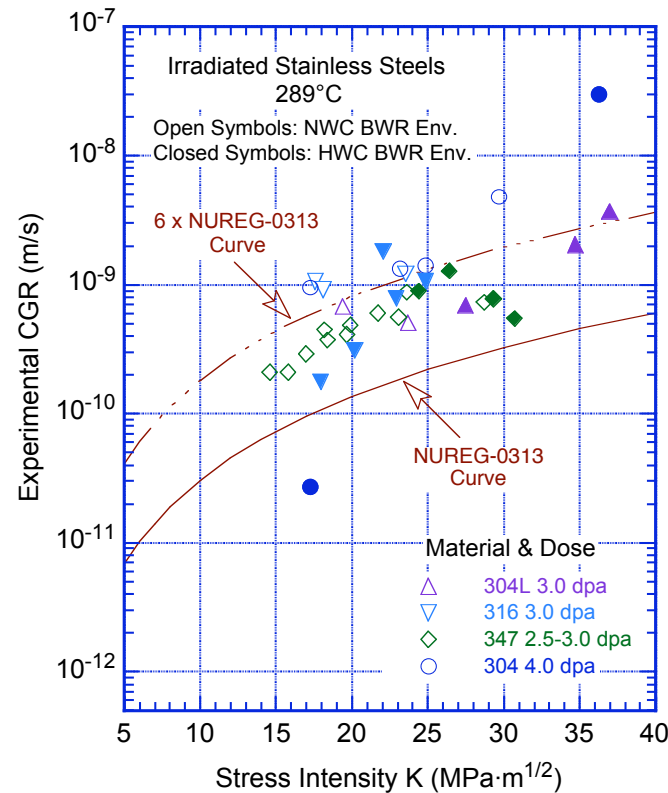
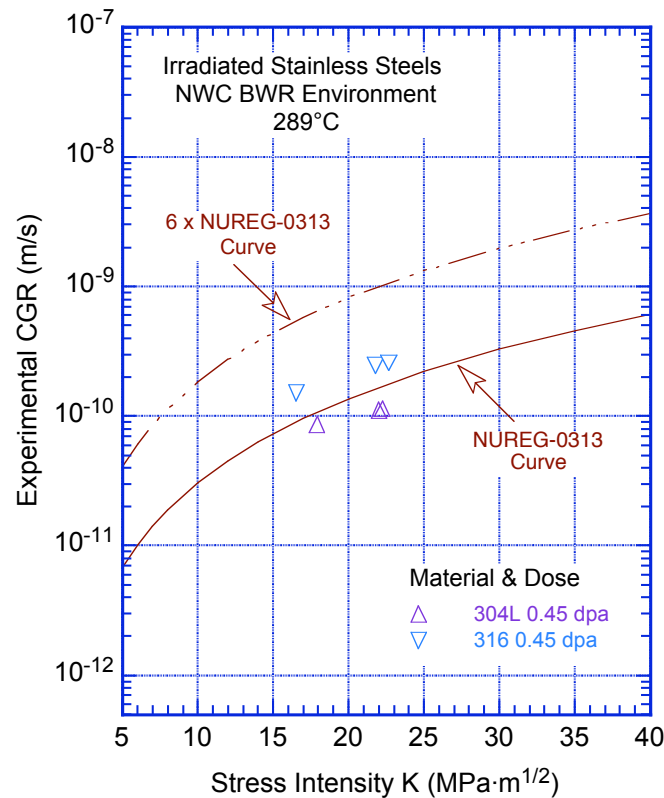


SCC Data for SSs Irradiated to 0.75-2.20 dpa



- Threshold fluence of $5 \times 10^{20} \text{ n/cm}^2$ (0.75 dpa) is inconsistent with experimental data
- At 0.75-2.20 dpa, CGRs are factor of 3-10 greater than those predicted by NUREG-0313
- CGRs of HAZ materials are generally greater than those of SA or sensitized SSs
- Benefit of HWC is observed at these fluence levels

SCC Data for SSs Irradiated to <0.5 & 3.0-4.0 dpa



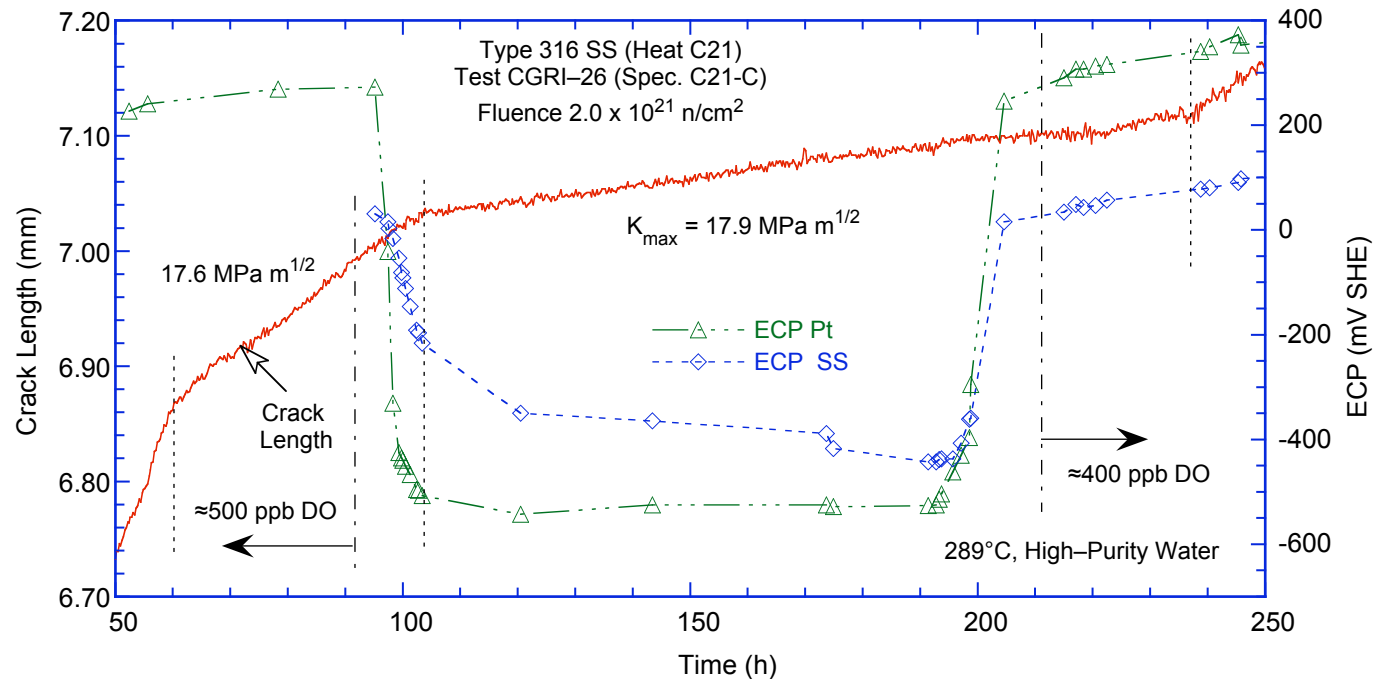
347 SS data
from Halden &
304 SS data
from GE

- At <0.5 dpa, CGRs comparable with values predicted by NUREG-0313
- At 3-4 dpa, benefit of HWC not observed for some heats at high K values
 - tests considered invalid according to size criterion proposed by Andresen

Proposed K/Size Criteria for Irradiated Materials

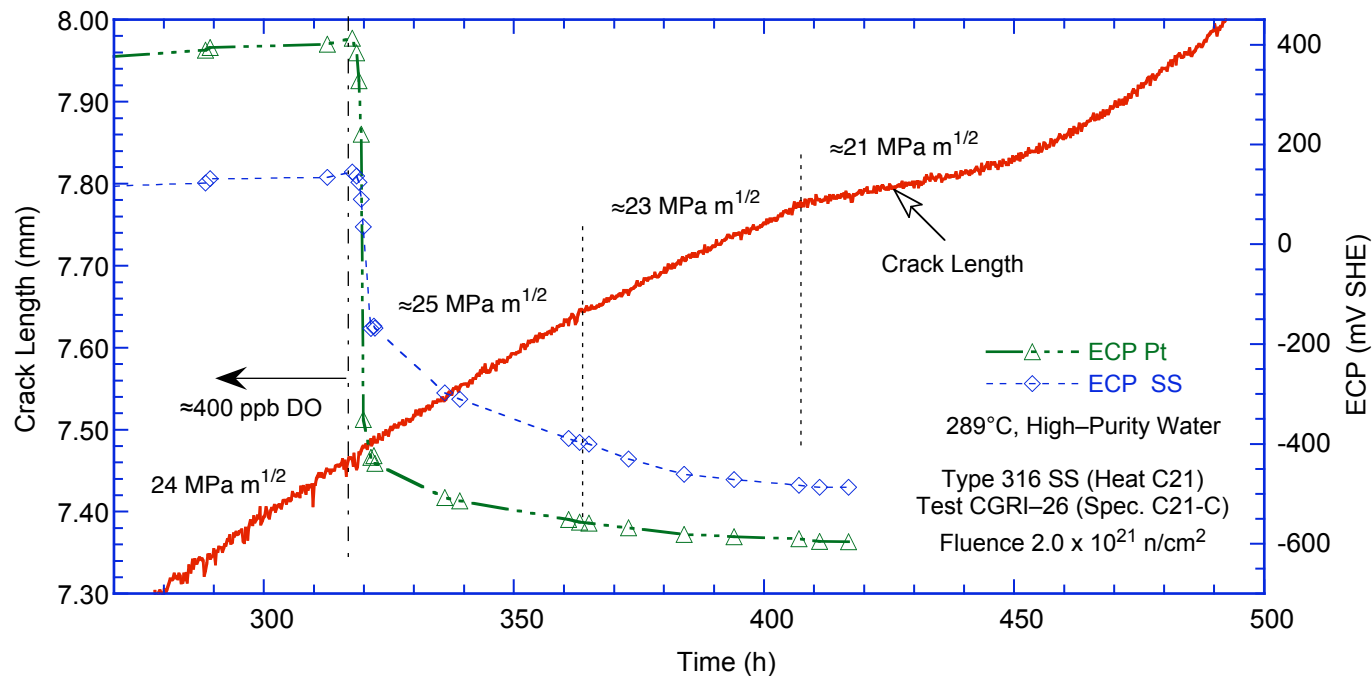
- Two K/Size criteria have been proposed for irradiated materials which generally show no strain hardening or actually show strain softening (i.e., materials that deform by dislocation channeling)
 - for moderate to highly irradiated materials (by Andresen)
$$\sigma_{\text{yeff}} = (\sigma_{\text{yirr}} + \sigma_{\text{ynonirr}})/2$$
 - for materials irradiated to very high fluences (by Anders)
$$\sigma_{\text{yeff}} = (\sigma_{\text{yirr}} + \sigma_{\text{ynonirr}})/3$$
- However, basis for these criteria is not clear
- ANL tests have tried to evaluate the K/size criteria by
 - consistency of results, &
 - evidence of loss constraint in fractography

Benefit of Reduced DO Level (or ECP) on Growth Rates



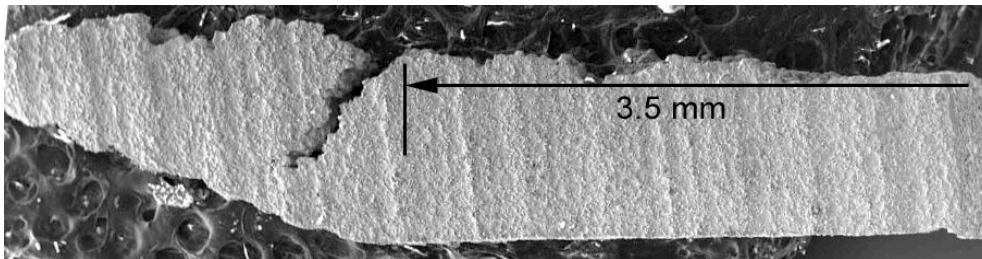
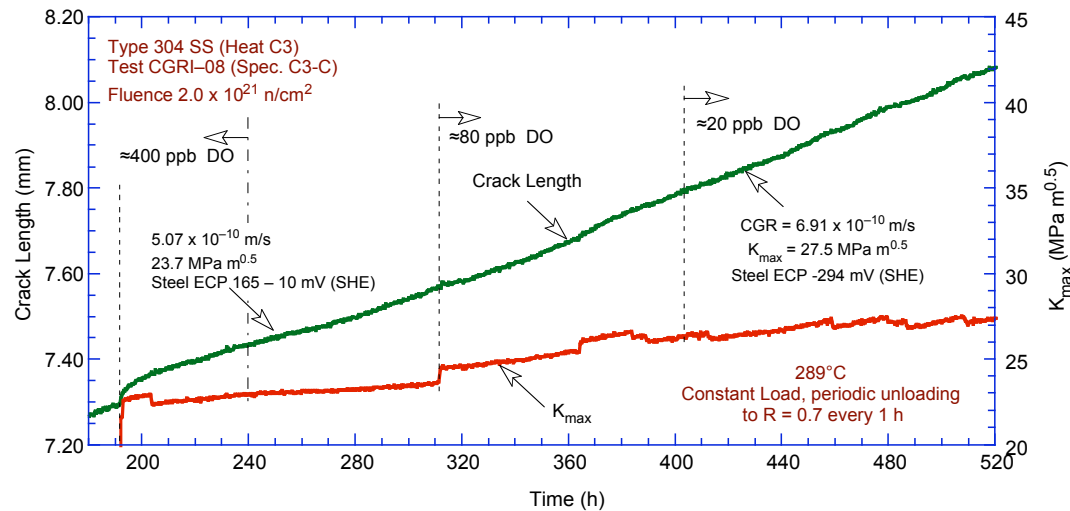
- At $K_{\max} \approx 17.8 \text{ MPa m}^{1/2}$, CGRs decreased a factor of ≈ 8 when ECP decreased below -200 mV (DO from ≈ 500 ppb to < 30 ppb)
- Rates increased back to old value when ECP increased above ≈ 100 mV

Effect of Reduced DO Level on Growth Rates



- At the value allowed by $\sigma_{\text{yeff}} = (\sigma_{\text{yirr}} + \sigma_{\text{y nonirr}})/2$, $K_{\text{max}} \approx 24 \text{ MPa m}^{1/2}$, no benefit of reduced DO on CGRs even after ECP decreased below -200 mV
- In low-DO water, rates did not change significantly even when K_{max} decreased to $\approx 21 \text{ MPa m}^{1/2}$

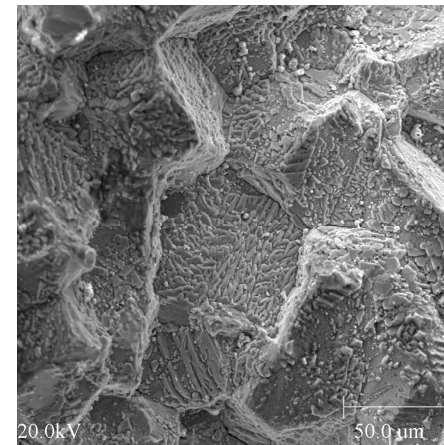
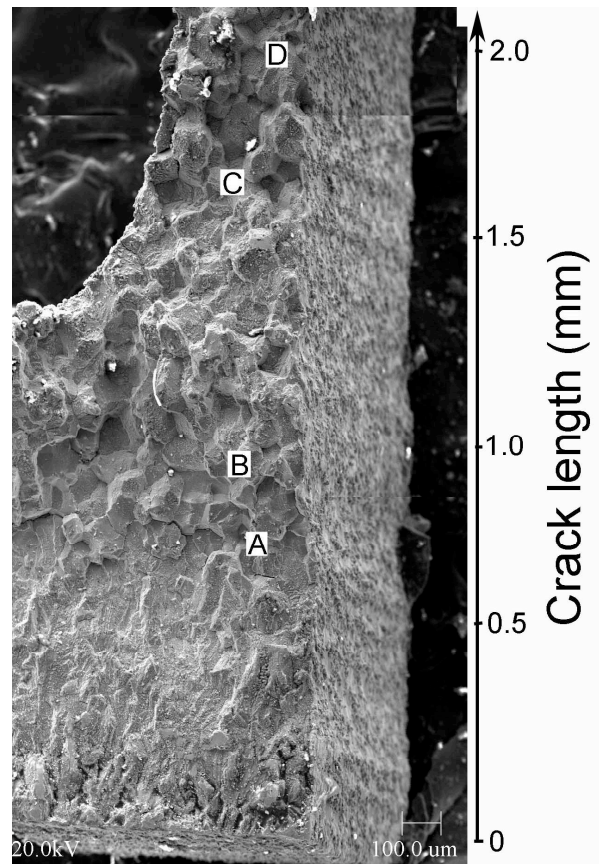
Specimen K/size Criterion



- Expected decrease in CGR not observed when DO decreased from 400 to 20 ppb.
- If applied load had exceeded the value allowed by K/size criterion, then CGRs should have increased in high-DO water
- Loading conditions seem to have had no effect until the DO level was decreased

- There is no change in fracture plane, DO level was changed at 1.7 mm crack length; fracture plane is straight & normal to stress axis
- If thickness or ligament criterion is exceeded, crack propagates away from the normal plane at an angle of 45°

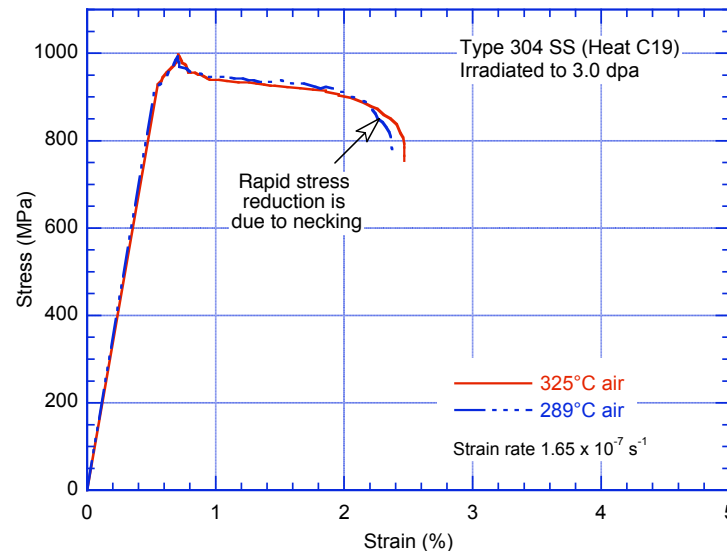
Specimen K/size Criterion (Contd.)



Location D

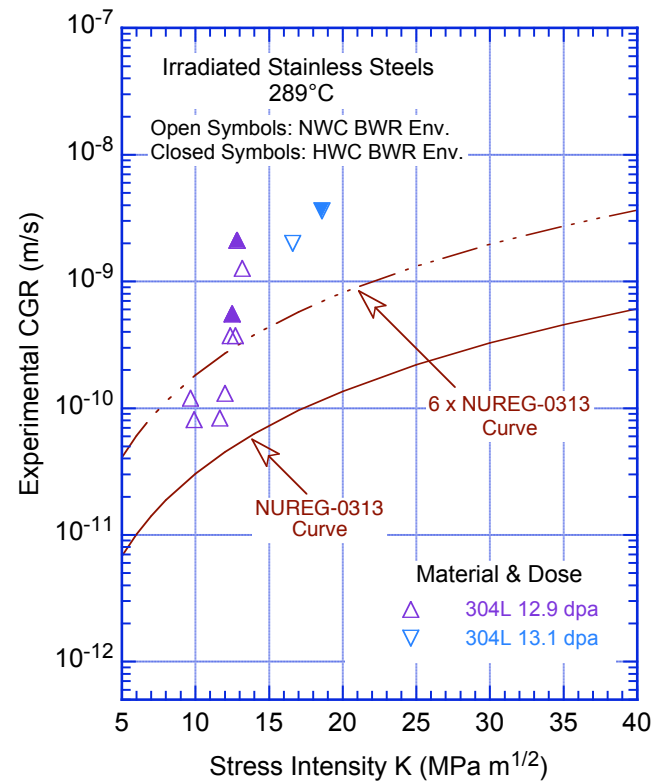
- No change in fracture morphology, complete intergranular fracture during SCC test. DO level was decreased at 1.7 mm crack length

Specimen K/size Criterion (Contd.)



- Arguments against the proposed K/size criterion:
 - strain softening in irradiated materials is rarely more than 10-15%
 - in most plastic zones, the plastic strains are so low that the material never passes the max tensile stress
 - FEA indicate difference between strain distributions ahead of a advancing crack, in a strain-hardening vs. strain-softening material, is marginal
- Adequacy of proposed K/size criterion for irradiated SSs needs to be examined

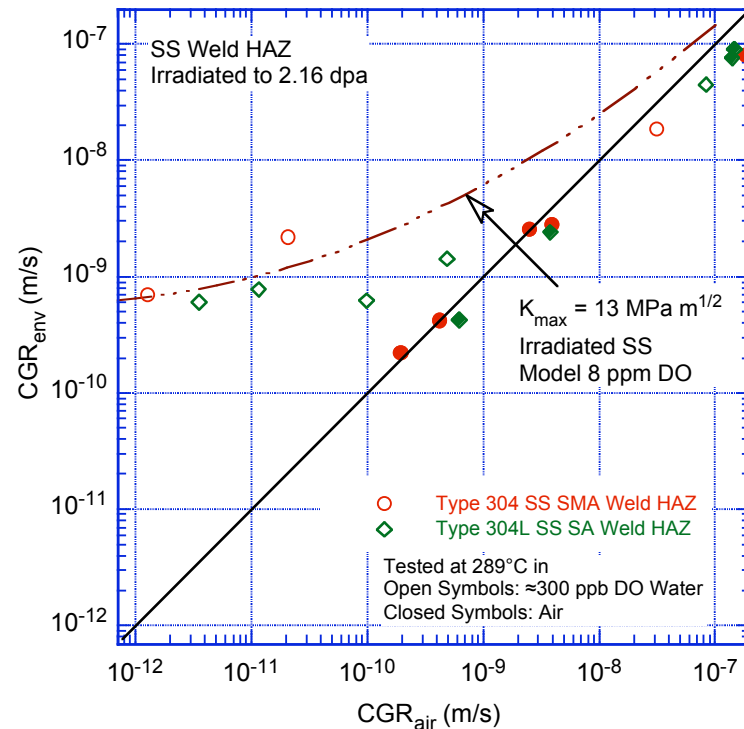
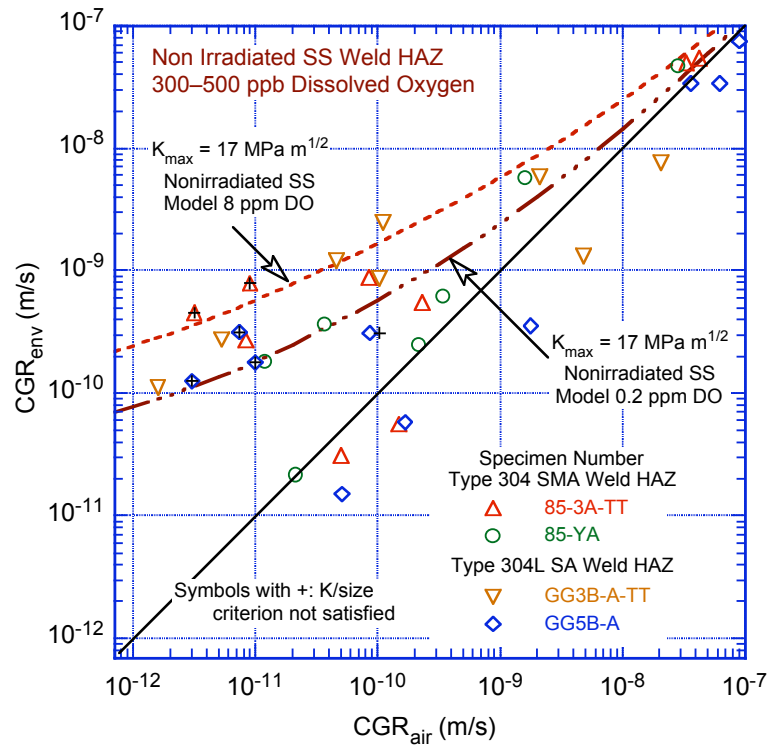
SCC Data for SSs Irradiated to ≈ 13 dpa



Data from
Studsvik & Halden

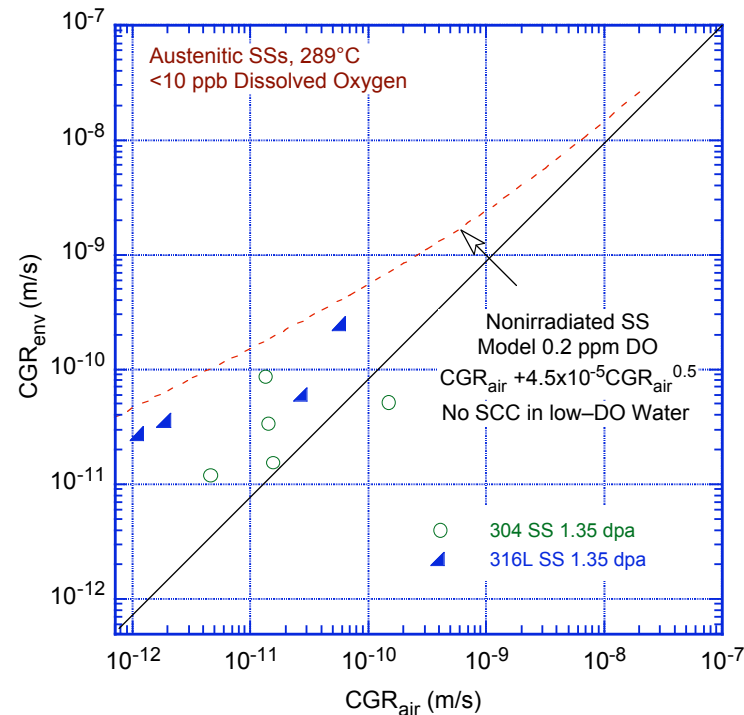
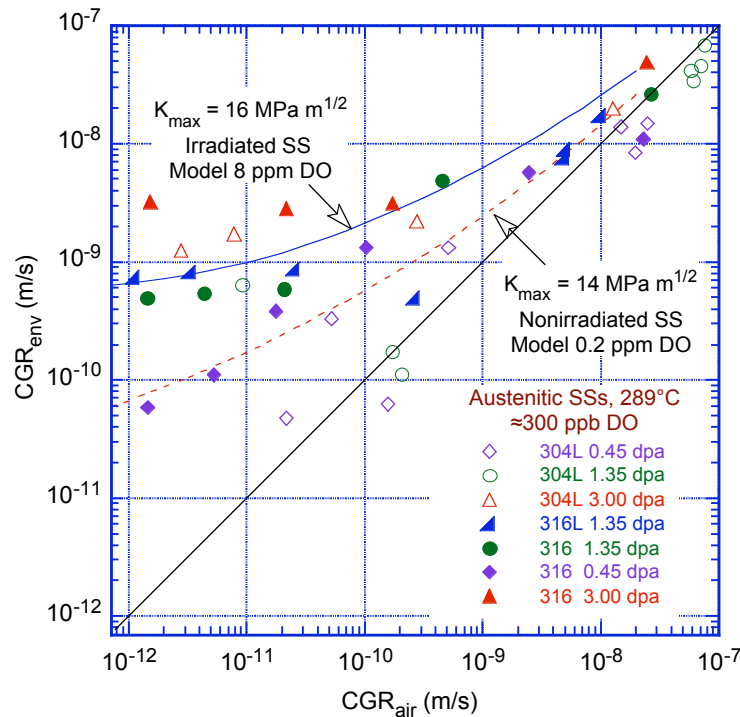
- CGRs show strong dependence on K at less than 15 MPa m^{1/2}
- At K > 15 MPa m^{1/2}, CGRs may be factor of 30 higher than NUREG-0313 curve
- Beneficial effect of low corrosion potential not observed at 13 dpa

Fatigue CGR Data for SS Weld HAZs



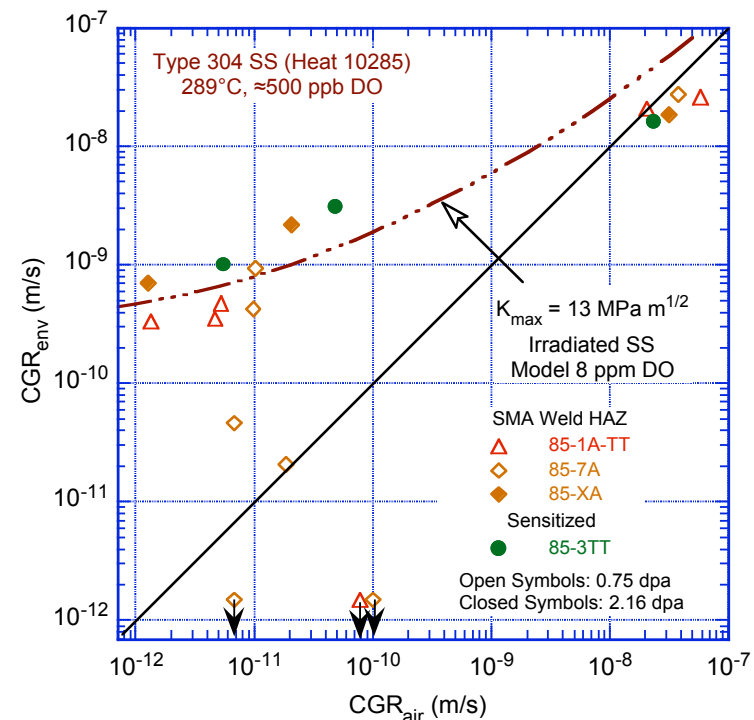
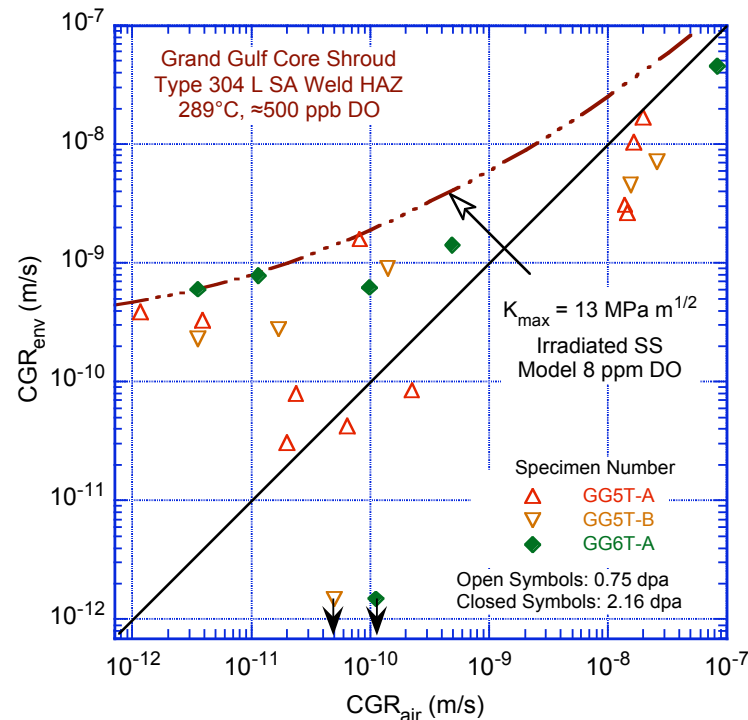
- CGRs of nonirradiated weld HAZ are consistent with Shack/Kassner model
- Irradiation up to 2.2 dpa has only marginal effect on CGRs in air

Fatigue CGR Data for Irradiated SSs in NWC & HWC Water



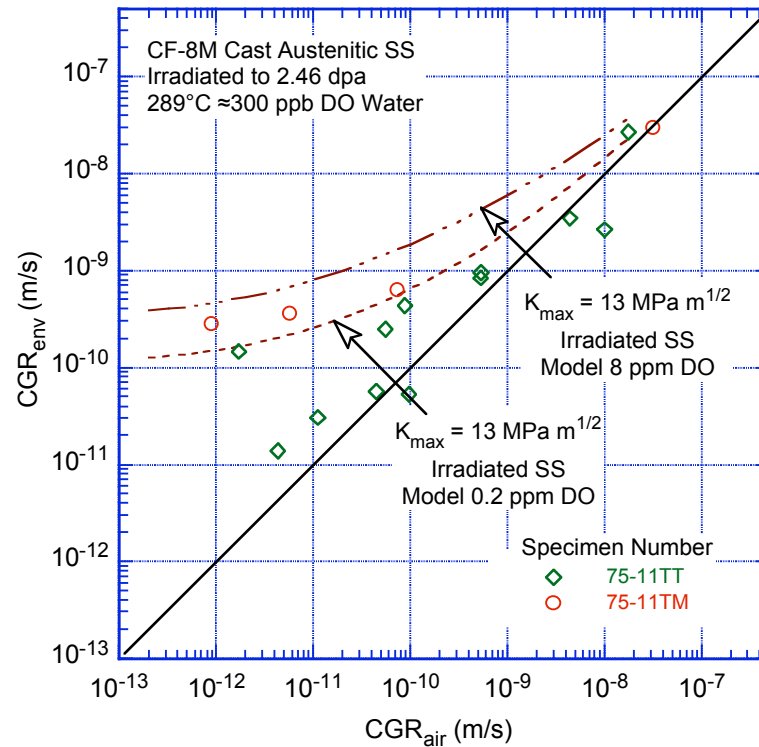
- At >0.5 dpa, cyclic CGRs in NWC represented by CGR_{SCC} for irradiated steel (i.e., 6 x NUREG-0313 rates) & Shack/Kassner model for 8 ppm DO
- At <0.5 dpa in NWC & irradiated SSs in HWC, cyclic CGRs represented by CGR_{SCC} given in NUREG-0313 & Shack/Kassner model for 0.2 ppm DO

Fatigue CGR Data for Irradiated SS HAZs in NWC Water



- Cyclic CGR data for weld HAZ materials are similar to those for SA SSs; CGR_{scc} given by 6 x NUREG-0313 growth rates & CGR_{cf} given by Shack/Kassner model for 8 ppm DO

Fatigue CGR Data for Irradiated Cast SS in NWC Water



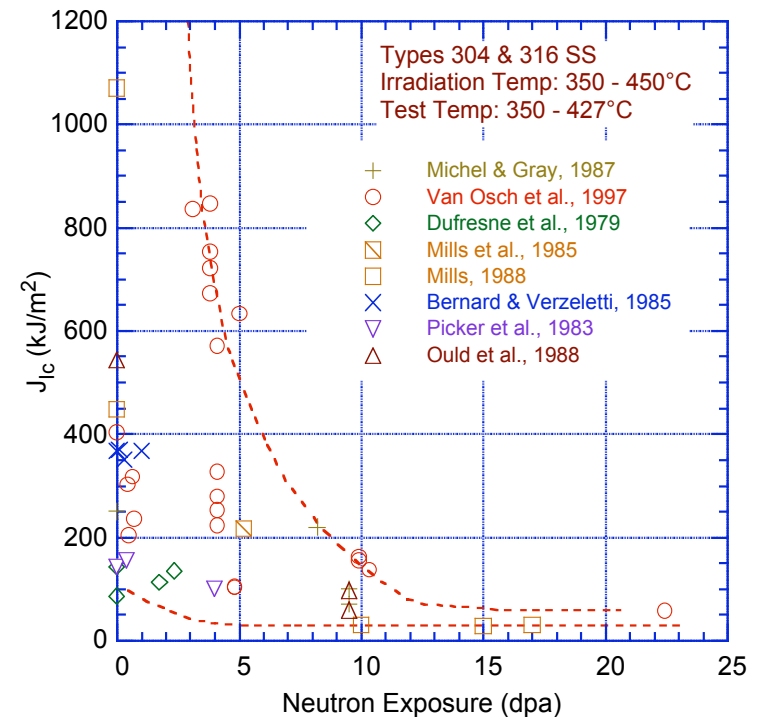
- Under similar loading & environmental conditions, cyclic CGRs for CF-8M cast SS appear to be lower than those for wrought SSs & HAZ materials

Summary

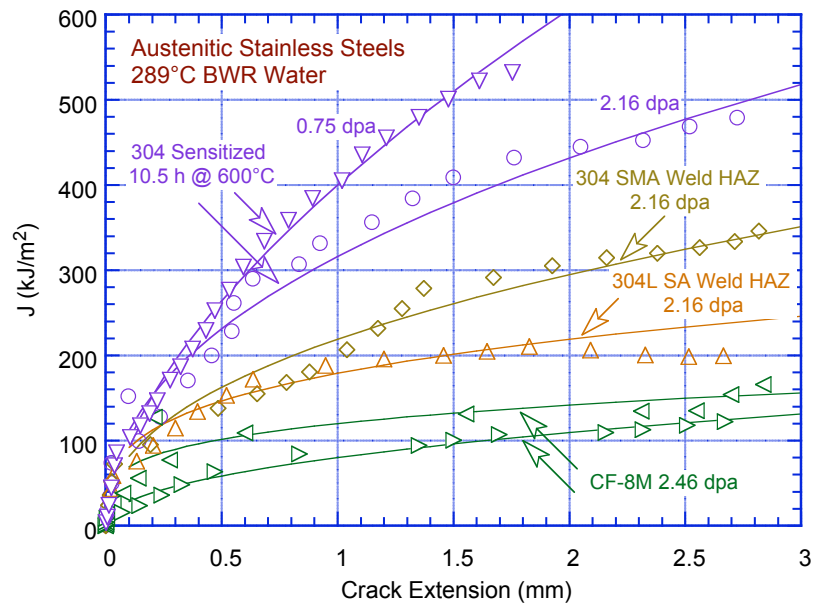
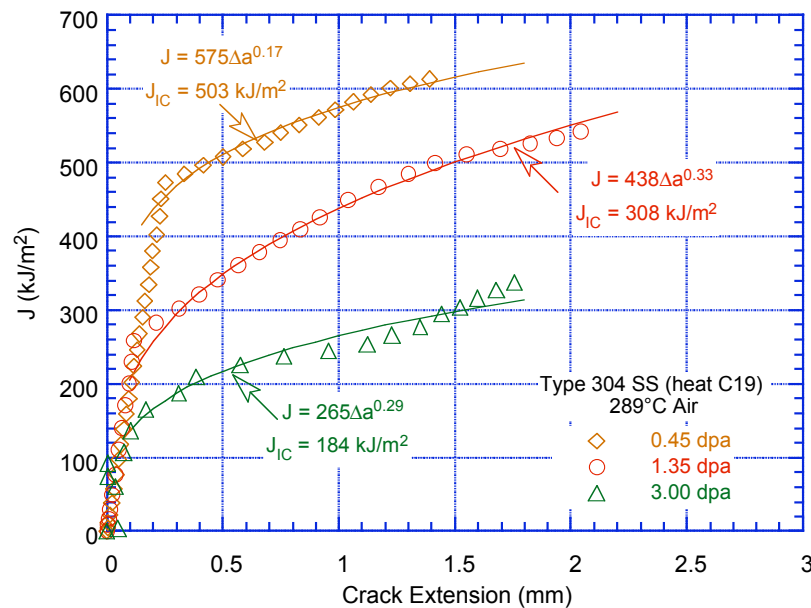
- Threshold fluence for irradiation effects to be significant: $\approx 3 \times 10^{20}$ n/cm² (≈ 0.45 dpa); below threshold, experimental CGRs are comparable to those of NUREG-0313
- Disposition curve for CGRs of SSs irradiated to 4 dpa: 6-8 x NUREG-0313 curve
- CGRs of SSs irradiated to 13 dpa show strong dependence of K and are a factor of 30 higher than the NUREG-0313 curve
- Fluence level above which benefit of HWC is not observed: limited data suggest, for some SSs it may be as low as $\approx 2 \times 10^{21}$ n/cm² (≈ 3.0 dpa), additional data needed to establish the threshold fluence above which irradiation effects are significant in low-DO HWC BWR or PWR environments
- Adequacy of the proposed K/size criterion for irradiated SSs needs to be examined
- Cyclic CGRs of irradiated SSs can be represented by a superposition model

Fracture Toughness of Irradiated SSs - Background

- Much of the existing fracture toughness data has been obtained in fast reactors at temperatures above 350°C
- Exposure to neutron irradiation for extended periods
 - alters the microstructure
 - increases the yield strength
 - reduces ductility
 - reduces resistance to fracture
- Fracture resistance decreases substantially for 1-10 dpa, no further decrease above saturation at ≈ 10 dpa
- Fracture toughness data needed at LWR temperatures

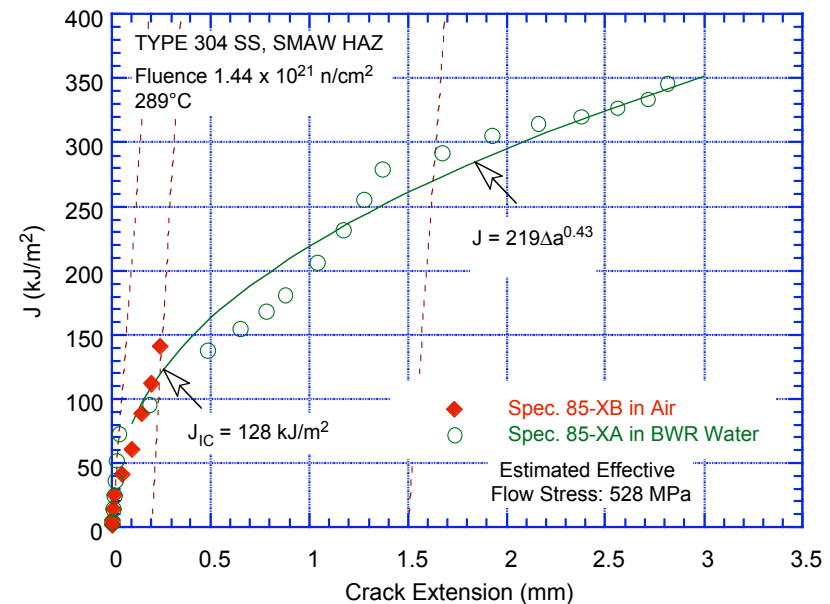
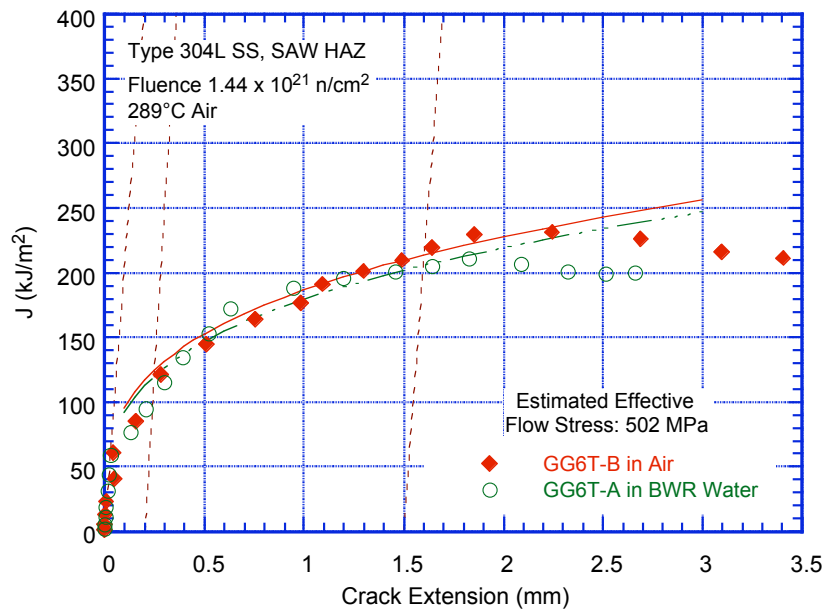


Effect of Irradiation on Fracture Toughness of SSs



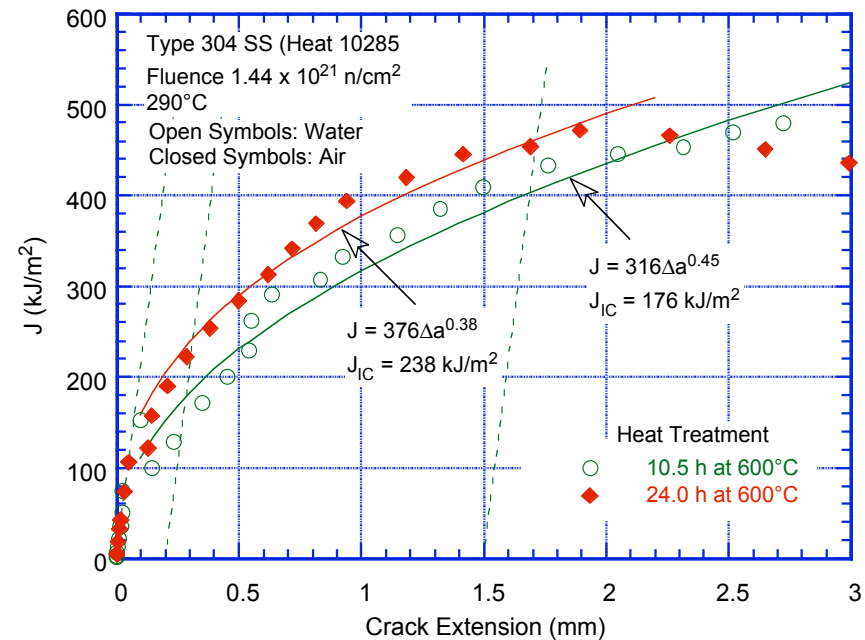
- Neutron irradiation decreases the fracture toughness of SSs
- For the same irradiation level,
 - toughness of cast CF-8M SS is lower than that of weld HAZ material, and
 - toughness of HAZ material is lower than that of sensitized material

Effect of Environment on Toughness of Irradiated Weld HAZs



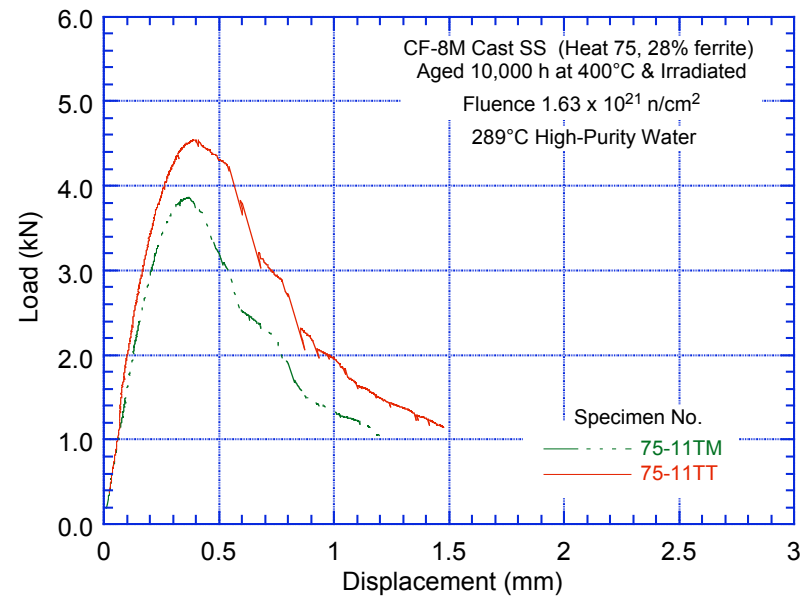
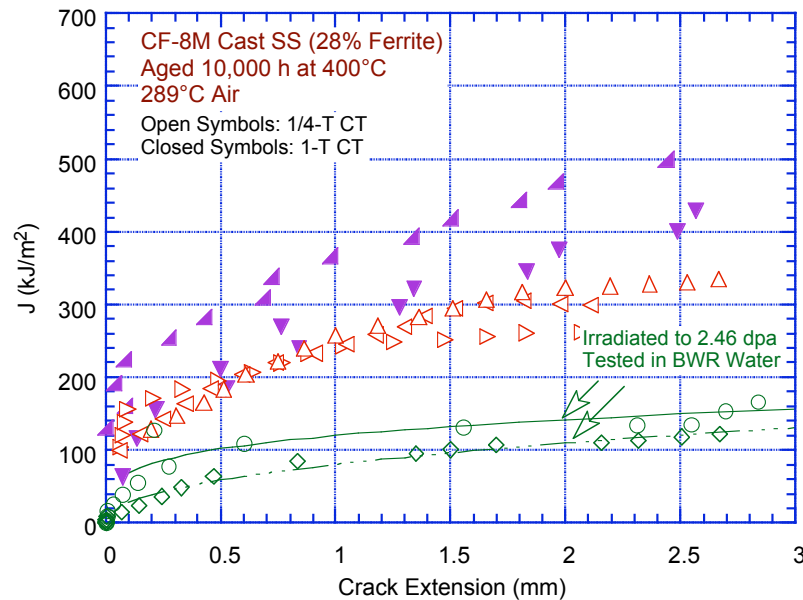
- Limited data indicate that fracture toughness of irradiated Type 304L SAW HAZ is approximately the same in air and water environments
- Complete J-R curve for Type 304 SMAW HAZ not obtained in air; large crack extension occurred at same J value both in air and water environments

Effect of Environment on Toughness of Irradiated Sensitized SSs



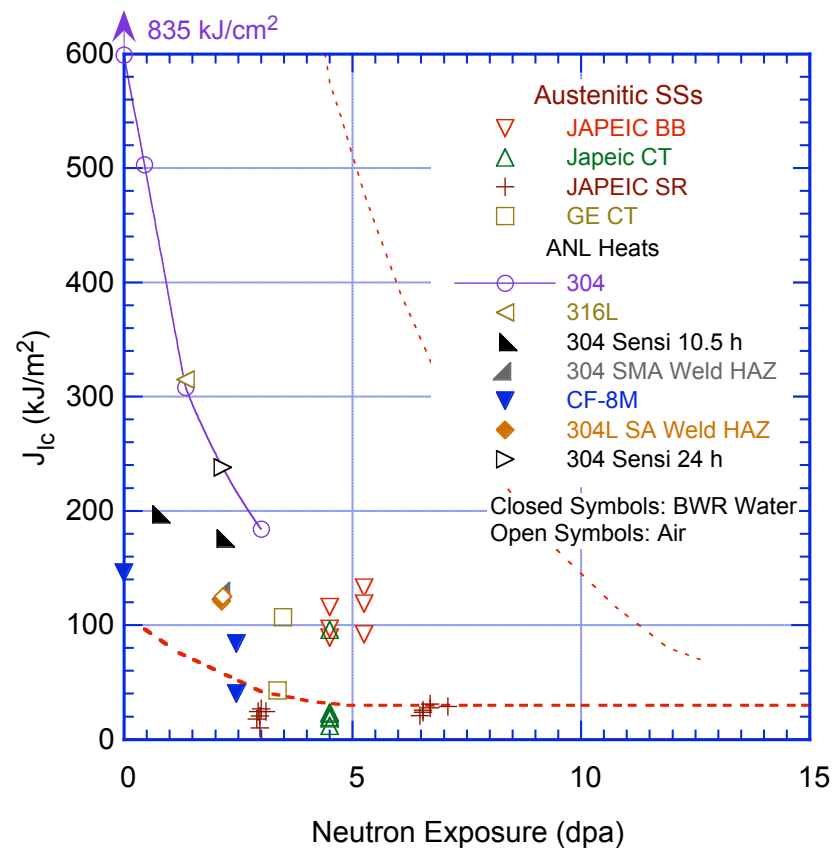
- Although material tested in air was sensitized for longer time than the material tested in water, toughness is slightly higher in air

Effect of Environment on Toughness of Irradiated Cast SSs



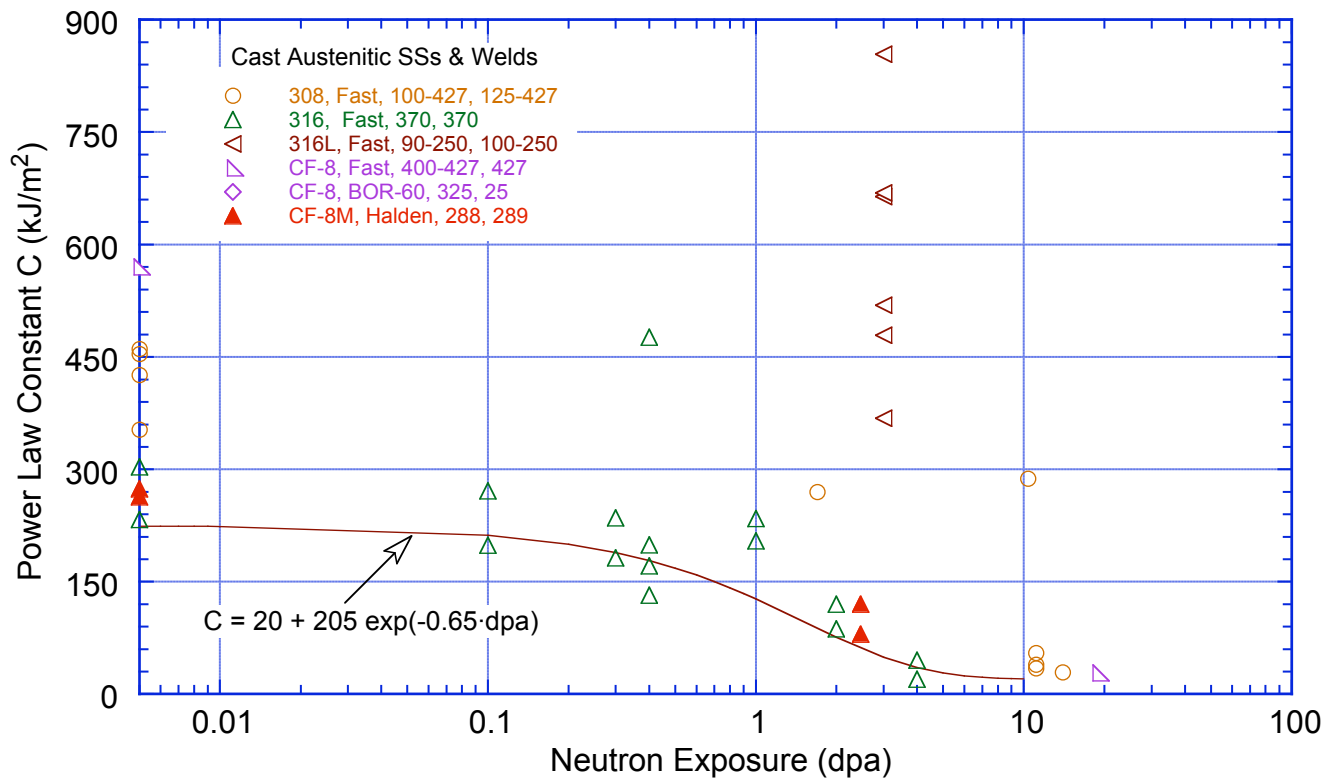
- Tests on thermally aged and irradiated cast SS in air were not conducted
- Both tests in water show large load drops and 0.5-1.0 mm crack extension; such behavior is typically not observed during tests in air

Change in J_{Ic} of Austenitic SSs with Neutron Dose



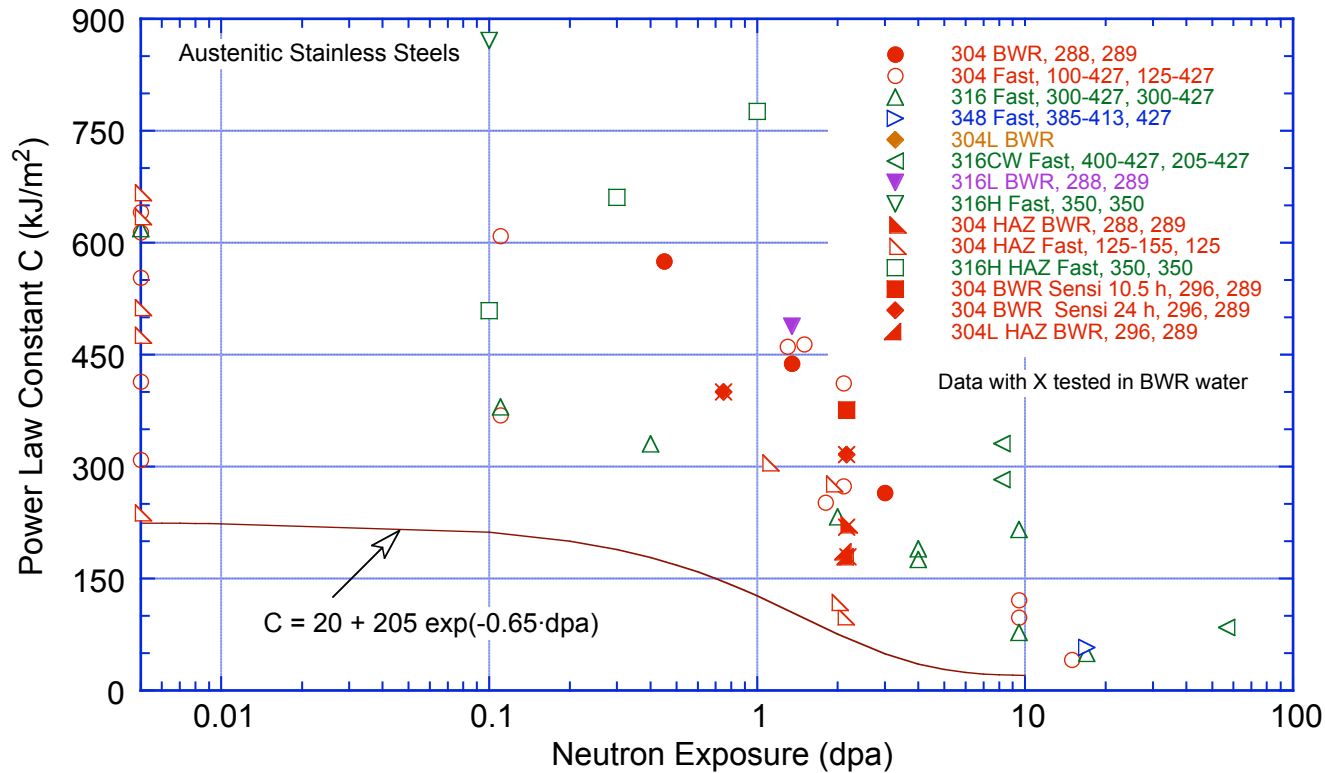
- Data for BWR-irradiated materials within the scatter band for fast reactor
- J_{Ic} can decrease to ≈ 15 kJ/m² ($K_{Ic} = 54$ MPa m^{1/2}) at 3-5 dpa
- Data obtained in BWR water are generally lower than those obtained in air

Change in Coefficient C of Power-Law J-R Curve for Cast SSs & Weld Metals with Neutron Dose



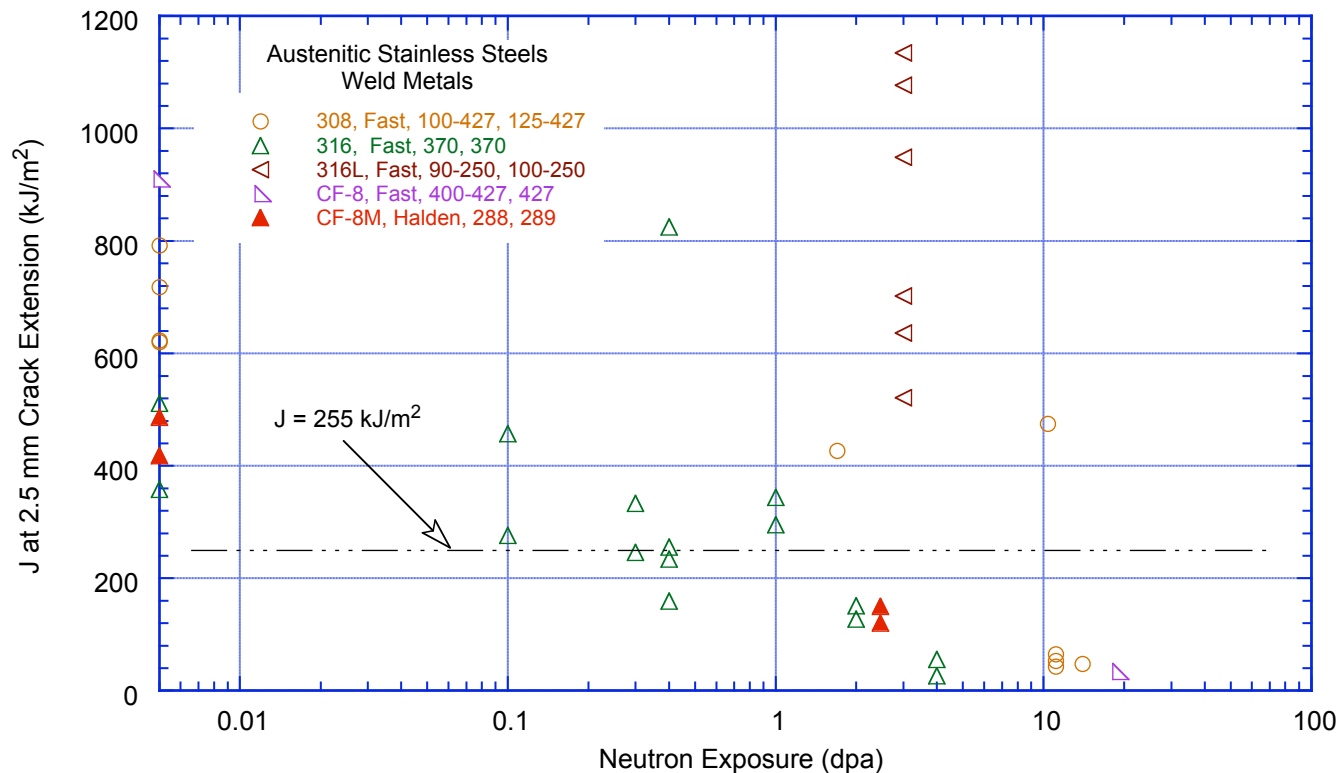
- For fluence less than 5 dpa, existing data can be bounded by a power-law J-R curve with coeff. C expressed by the curve and exponent $n = 0.37$

Change in Coefficient C of Power-Law J-R Curve for Austenitic SSs with Neutron Dose



- The power-law J-R curve expression yields a bounding C value of 225 kJ/m² (1285 in-lb/in²) for materials irradiated less than 0.5 dpa & 28 kJ/m² (160 in-lb/in²) for materials irradiated to about 5 dpa

Experimental Values of J-integral at 2.5 mm Crack Extension for SSs as a Function of Neutron Exposure



- EPRI TR-106092 proposed threshold value of $J_{2.5} = 255 \text{ kJ/m}^2$ for potentially significant reduction in toughness of thermally aged cast SSs
- For SSs irradiated up to 0.3 dpa ($2 \times 10^{20} \text{ n/cm}^2$), $J_{2.5}$ is above 255 kJ/m²

Summary

- Neutron irradiation decreases fracture toughness of austenitic SSs
- For irradiated SSs, toughness of cast SS is lower than that of weld HAZ material, and toughness of HAZ is lower than that of SA or sensitized SS
- J-R curve data for irradiated 304L SAW HAZ in water are comparable to those in air
- However, results for irradiated sensitized SS and cast SS suggest a possible effect of water environment; additional tests are needed to verify these results
- Existing data indicate little or no change in toughness below 0.5 dpa, & rapid decrease between 1 and 5 dpa to reach a saturation value
- A fracture toughness trend curve that bounds the existing data has been defined in terms of J_{Ic} vs. neutron dose or coeff. C of power-law J-R curve vs. neutron dose
- For fluence less than 5 dpa, existing data can be bounded by a power-law J-R curve with coeff. C expressed by $C = 20 + 205 \exp(-0.65 \text{ dpa})$ and exponent $n = 0.37$