

Snapshot Reviews in Emerging Fields



How irradiation promotes intergranular stress corrosion crack initiation

G.S. Was^{a,*}, C.-B. Bahn^b, J. Busby^c, B. Cui^d, D. Farkas^e, M. Gussev^c, M. Rigen He^f, J. Hesterberg^g, Z. Jiao^a, D. Johnson^h, W. Kuangⁱ, M. McMurtrey^h, I. Robertson^j, A. Sinjlawi^a, M. Song^k, K. Stephenson^l, K. Sun^a, S. Swaminathan^m, M. Wangⁿ, E. West^o

^a University of Michigan, Ann Arbor, MI, USA

^b Pusan National University, Busan, South Korea

^c Oak Ridge National Laboratory, Oak Ridge, TN, USA

^d University of Nebraska, Lincoln, NB, USA

^e Virginia Tech, Blacksburg, VA, USA

^f Johns Hopkins University, Philadelphia, PA, USA

^g Naval Nuclear Laboratory, Pittsburgh, PA, USA

^h Idaho National Laboratory, Idaho Falls, ID, USA

ⁱ Xi'an Jiaotong University, Xi'an, Shaanxi, China

^j University of Wisconsin, Madison, WI, USA

^k Shanghai Jiao Tong University, Shanghai, China

^l Material Forensics Engineering, PLLC, Seattle, WA, USA

^m Bundesanstalt fuer Materialforschung und -pruefung, Berlin, Germany

ⁿ Studsvik, Nykoping, Sweden

^o Naval Nuclear Laboratory, Niskayuna, NY, USA

ARTICLE INFO

Keywords:

Irradiation

Stress corrosion cracking

Grain boundaries

Oxidation

Austenitic alloys

Localized deformation

Dislocation channels

ABSTRACT

Irradiation assisted stress corrosion cracking (IASCC) is a form of intergranular stress corrosion cracking that occurs in irradiated austenitic alloys. It requires an irradiated microstructure along with high temperature water and stress. The process is ubiquitous in that it occurs in a wide range of austenitic alloys and water chemistries, but only when the alloy is irradiated. Despite evidence of this degradation mode that dates back to the 1960s, the mechanism by which it occurs has remained elusive. Here, using high resolution electron backscattering detection to analyze local stress-strain states, high resolution transmission electron microscopy to identify grain boundary phases at crack tips, and decoupling the roles of stress and grain boundary oxidation, we are able to unfold the complexities of the phenomenon to reveal the mechanism by which IASCC occurs. The significance of the findings impacts the mechanical integrity of core components of both current and advanced nuclear reactor designs worldwide.

* Corresponding author.

1. Introduction

1.1. Impact of irradiation assisted stress corrosion cracking

A growing concern for electric power utilities worldwide has been degradation of core components in nuclear power reactors, which make up $\sim 17\%$ of the world's electric power production. The core of a nuclear reactor presents an extremely aggressive environment consisting of high temperature water, imposed stresses, and an intense radiation field that can affect the water chemistry and the microstructure of the core components. In particular, irradiation is responsible for accelerating the intergranular cracking of austenitic alloys in reactor cores. This acceleration, termed irradiation assisted stress corrosion cracking (IASCC), is ubiquitous in that it occurs in all light water reactor (LWR) environments in numerous core components encompassing a wide range of austenitic stainless steels and nickel-base alloys. To date, over 20 different core components from over 10 stainless steel and nickel-base alloys in water reactors of all types have experienced IASCC.[1] Since it is a degradation mode that impacts *all* of the world's 400+ water reactors, and will also impact new reactor designs, understanding its origin is critical to developing mitigation strategies and avoiding its occurrence in advanced reactors being planned worldwide. While having first been observed as early as 1965[2], the process by which IASCC occurs has remained elusive. This is due in part to the synergistic nature of its occurrence, requiring an irradiated microstructure, high temperature (280–320 °C) water (corrosive), and application of stress. But also because most studies have focused heavily on correlations of environment, components or material features with cracking rather than the processes governing the cracking.

Initially, the affected components were either relatively small in size (bolts, springs, etc.) or components designed for replacement (fuel rods, control blades, or instrumentation tubes).[3] More recently, IASCC has been observed in other structural components and in a growing number of other stainless steel (and nickel alloy) core components, such as neutron source holders and control rod absorber tubes,[4] as well as instrument dry tubes and control blade handles and sheaths, which are generally in creviced locations and subject to very low stresses but higher fluences.[5,6] In the last two decades, the number of IASCC incidents has continued to grow, and there can be no question that many components in LWRs are susceptible, although large plant-to-plant and heat-to-heat differences are observed. The two most widespread cases of IASCC are boiling water reactor (BWR) core shrouds and pressurized water reactor (PWR) baffle bolts, although susceptibility clearly exists in other areas, such as control blade components, fuel components, BWR top guide, etc.[7,8] The PWR primary water environment is a low potential environment which makes the occurrence of IASCC confounding.

1.2. Characteristics of IASCC

IASCC can be categorized into radiation effects on the water chemistry and on the material properties. The effect of irradiation damage (fast neutron fluence) on IASCC has been well established. Intergranular (IG) SCC is promoted in austenitic stainless steels when a critical “threshold” fluence is exceeded. Cracking is observed in BWR oxygenated water at fluences above $2\text{--}5 \times 10^{20} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$), which corresponds to about 0.3–0.7 displacements per atom (dpa). In low potential PWR primary water, the dose at which

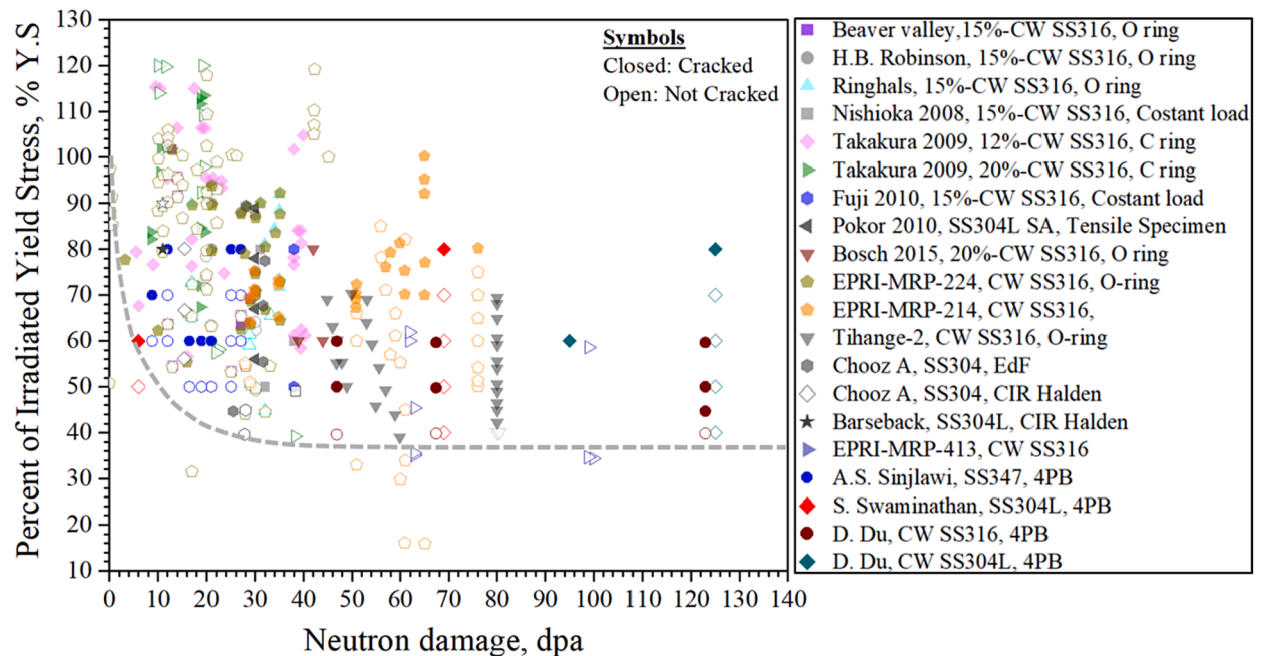


Fig. 1. Plot of % of irradiated yield stress at which cracking occurs as a function of damage level for various irradiated, austenitic stainless steels strained in simulated PWR primary water. [20–27,58] The curve represents a bounding condition on observations of cracking.

cracking occurs is closer to 3 dpa. This “threshold” fluence depends on factors such as alloy, stress, water chemistry, corrosion potential, conductivity, temperature etc., that can affect when it occurs. But because this behavior can be replicated in post-irradiation tests, only “persistent” radiation effects (microstructural and microchemical changes) can be responsible for the “threshold-like” behavior. These radiation effects or material changes fall into three categories; (1) micro compositional effects; radiation-induced segregation (RIS) of both impurities and major alloying elements, (2) microstructural changes; the formation of dislocation loops, voids, precipitates and the resulting hardening (increase in yield strength), and (3) deformation mode; localized deformation and the formation of dislocation channels. The dislocation loop microstructure is closely tied to radiation hardening and both increase with damage until saturation occurs by ~ 5 dpa. RIS also increases with damage and tends to saturate by several dpa. All of the observable effects of irradiation on the microstructure increase with dpa in much the same manner, making isolation of, and attribution to individual contributions difficult. Although dependent on both metallurgical and environmental parameters, IASCC generally occurs over a range of dpa in which all of these microstructure features are steeply rising.

Absent irradiation, intergranular stress corrosion cracking of austenitic stainless steels requires stresses at or above the yield strength of the alloy. Even in the sensitized condition, the stress dependence curve for IG cracking of Type 304 stainless steel (304 SS) is asymptotic to the at-temperature yield strength of the sensitized alloy in 288 °C water containing 0.2 ppm oxygen. [9,10] For example, of 93 incidents of cracking on Type 304 stainless steel recirculation and core spray lines in BWRs, none were found to have occurred at stresses below the yield strength. [10] Numerous other studies have confirmed this stress dependence of cracking in austenitic stainless steels in high temperature water [11–18]. However, IASCC failures occur well below the irradiated yield strength, and the fracture surface is typically fully intergranular. Fig. 1 shows this unique aspect of IASCC – that it occurs at stresses as low as 40 % of the irradiated yield strength [19–26], which distinguishes it from IG cracking of non-irradiated stainless steels in high temperature water.

In the absence of an aqueous environment, IG cracking of irradiated stainless steel is difficult to induce at damage levels typical of components in LWR cores. While occurrences have been reported, straining experiments on irradiated stainless steels in inert environments at reactor core temperatures rarely produce IG cracking. Very high damage levels can induce transgranular (TG) cracking by channel fracture. [27] On rare occasion IG cracking has been reported, but at conditions inconsistent with IASCC, such as low temperatures, with the addition of hydrogen, or after straining into the plastic regime producing a very low percentage of IG cracking on the fracture surface [28–34].

1.3. Historical attempts to understand its occurrence

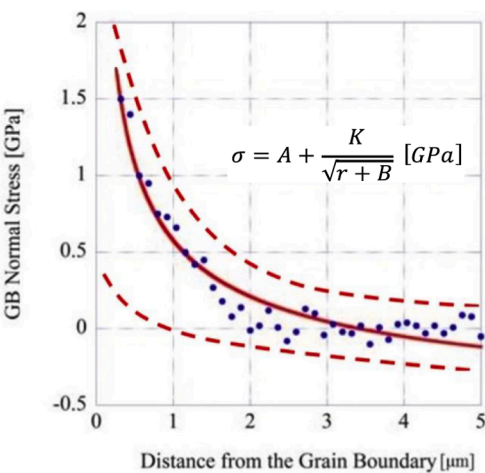
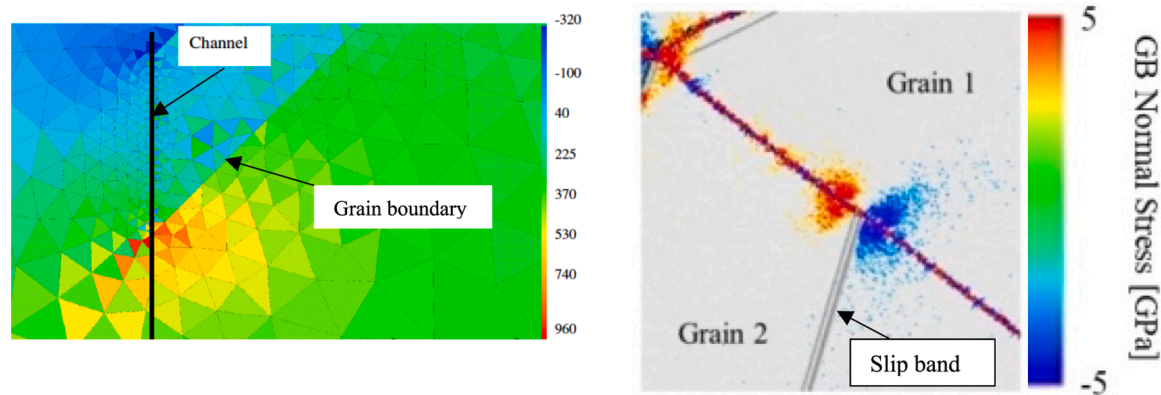
Because IASCC is a form of intergranular stress corrosion cracking, early research focused on the phenomenon of RIS of major and minor/impurity elements that could degrade the strength of the grain boundary. RIS is often implicated in IASCC of stainless steels, especially in oxidizing environments, due to the wealth of data from lab and plant operational experience with sensitized components [21,35–40]. RIS-induced grain boundary chromium depletion has been suspected as a factor because of the well-known susceptibility of sensitized stainless steels. While it correlates with IG cracking, it does not explain cracking in low potential, PWR primary water. In fact, experiments uncovered no single element that consistently correlated with increased crack initiation susceptibility [41–45] with perhaps, silicon being the one element that appeared to influence cracking. Among other features that correlate with cracking is yield strength. It is well known that yield strength is a factor in stress corrosion cracking, and in particular, the crack growth rate of austenitic stainless steels. [35,46] Irradiation causes significant increases in the yield strength by as much as 4–5 times that of the solution annealed state [47–49]. However, the physical process(es) that tie hardening to IASCC have yet to be identified.

Other mechanisms proposed include hydrogen or helium embrittlement. Transmutation of various elements causes an increase in the helium content of irradiated stainless steel and nickel alloys. At low fluence, transmutation of boron produces helium, while at higher fluence helium is primarily produced by a neutron reaction with ^{59}Ni . Because ^{59}Ni must be produced first by transmutation, the production of helium from this reaction is limited until 5 – 10 dpa. There is no evidence that helium directly affects SCC behavior, but it can produce an increase in hardness and yield strength, and may affect fracture toughness. [50] Neutron absorption by the ^{60}Ni isotope produces hydrogen that can also be absorbed from corrosion reactions with the stainless steel components. The retention of hydrogen only in those regions where measurable swelling exists correlates with the association of hydrogen with cavities. However, the large levels of hydrogen found in components such as baffle bolts cannot be generated from transmutation reactions alone but must result from corrosion reactions. [51] Nevertheless, research under the Cooperative IASCC Research (CIR) program failed to show a conclusive effect of H on IASCC. [52].

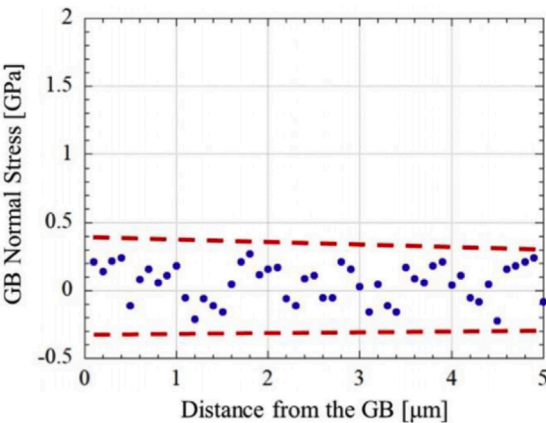
While recent reviews lay out many of the factors governing IASCC, [1,23,36,53–57] there are three processes that hold the key to the mechanism. They are: 1) Irradiated alloys deform in a completely different manner from the non-irradiated condition, 2) grain boundaries in irradiated austenitic alloys oxidize when exposed to high temperature water, and 3) silicon segregated to the grain boundary oxidizes when exposed to high temperature water. The linkage between these three factors is key to the mechanism of IASCC initiation.

2. Localized deformation

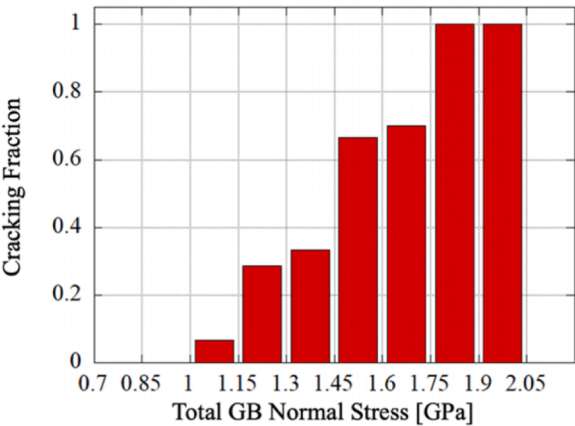
Given that fracture of irradiated stainless steels in LWR water occurs at *applied* stresses in the range 300–600 MPa, [58] (see Fig. 1), there must be some additional source of stress to fracture the grain boundary. This is where the unique deformation mode of irradiated alloys plays a key role. Irradiation produces defect clusters (defect-defect, defect-solute, dislocation loops, cavities, precipitates, etc.) that act as barriers to dislocation motion and result in hardening of the alloy with an accompanying increase in yield strength. Once dislocations overcome the obstacles, they form defect-free dislocation channels, or stacked slip planes through the solid, typically



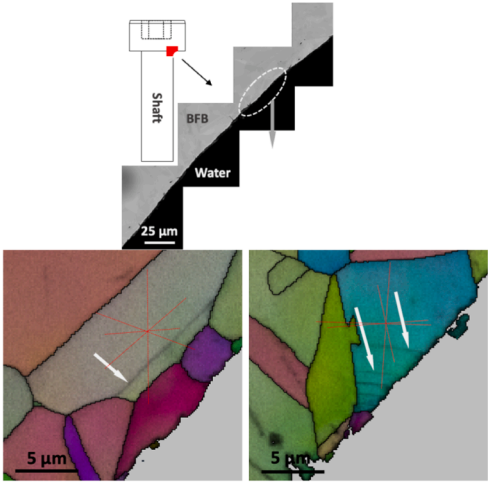
(c)



(d)



(e)



(f)

(caption on next page)

Fig. 2. Computational and experimental results on local stresses at dislocation channel-grain boundary interaction sites; a) FEA model result, [60] b) MD simulation, [61] c) stress normal to the grain boundary at 75 DDC-GB sites, [62] d) stress normal to the grain boundary at 15 CDC-GB sites,⁶⁴ (raw data is shown in blue and the fit to the Eshelby equation is shown as a solid red line, while the dashed red lines indicate upper and lower bounds for the set of data, e) probability of cracking at DDC-GB sites as a function of normal stress on the grain boundary in 304L irradiated to 5 dpa and strained in simulated BWR water, and f) EBSD IPF map overlapped with IQ map revealing a single defect-free channel (at arrow) in the near-surface BFB layer, and f) IPF + IQ maps showing multiple defect-free channels in the near-surface grain of the BFB. Red thin lines show projections of (111)-slip planes within the analyzed grain.

initiating at a grain boundary, and terminating at one, presenting two principle options. One option is the transfer of slip to a neighboring grain in which a favorably oriented slip plane is activated (continuous dislocation channel, CDC), and a second is one in which no such activation occurs and the channel terminates at the grain boundary (discontinuous dislocation channel, DDC). These two cases present very different strain/stress states at the dislocation channel-grain boundary (DC-GB) site. In fact, the intersection of discontinuous channels with grain boundaries has been observed to have a much higher probability of cracking than that for continuous DC-GB sites.[59] The cause is an increased stress at the DDC-GB site, which has been confirmed by both computation and experiment.

One of the first studies of the local stress at DDC-GB sites utilized transmission electron microscopy (TEM) and finite element (FE) analysis of a 304L stainless steel irradiated to 0.16 dpa and strained at 290 °C to initiate dislocation channels.[60] For typical values of channel aspect ratio (length/thickness), the normal stress at the grain boundary perpendicular to the loading axis was calculated to be between 4 and 8 times that of the applied stress. Given a fracture stress at high dpa of about 320 MPa,[60,21] the value of stress at the GB is then 1.3–2.6 GPa, Fig. 2a. These values agree well with large scale molecular dynamics (MD) simulations that calculated the stress on the GB at a DDC-GB site to be in the range 1.5–3.5 GPa depending on the degree of disorder of the boundary, Fig. 2b.[61,62] These computations provide a range for the stress amplification at the grain boundary at the site of DDC impact and are supported by measurements. Using high resolution electron backscattering detection (HREBSD) to determine the local strain tensor at the DC-GB sites, the local stress tensor could then be determined. A set of experiments was conducted on a proton irradiated Fe-13Cr-15Ni alloy that was deformed in high purity Ar at 288 °C to produce dislocation channels.[62] The stress normal to the grain boundary was collected for 75 DDC-GB intersection sites, and 15 CDC-GB intersection sites shown in Fig. 2c and 2d, respectively.[62] Raw data is shown in blue and the fit to the Eshelby equation[63] is shown as a solid red line. The Eshelby equation describes the stress field resulting from the pile-up of dislocations at an immovable barrier:

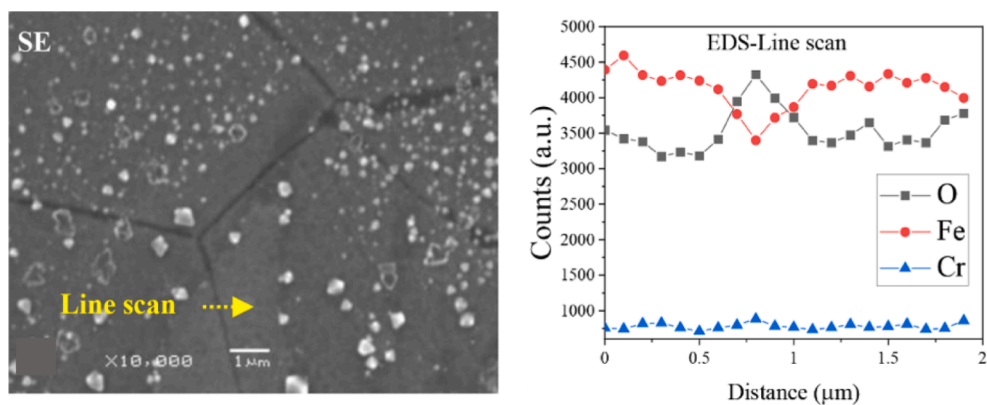
$$\sigma = A + \frac{K}{\sqrt{r+B}} \quad (1)$$

where σ denotes the grain boundary normal stress, r is the distance from the grain boundary, and K is the stress intensity factor that describes resistance to slip transfer of the grain boundary. The factor A is introduced to allow for uncertainty in the stress state of the reference pattern used during offline cross correlation analysis and B allows for uncertainty in the exact location of the grain boundary beneath the resolving limit of the EBSD step size [63]. Dashed red lines denote the upper and lower bounds of the stresses determined from HREBSD. These data confirm the MD and FE calculations of a very high local stress at DDC-GB sites, and the lack of stress enhancement at CDC-GB sites.

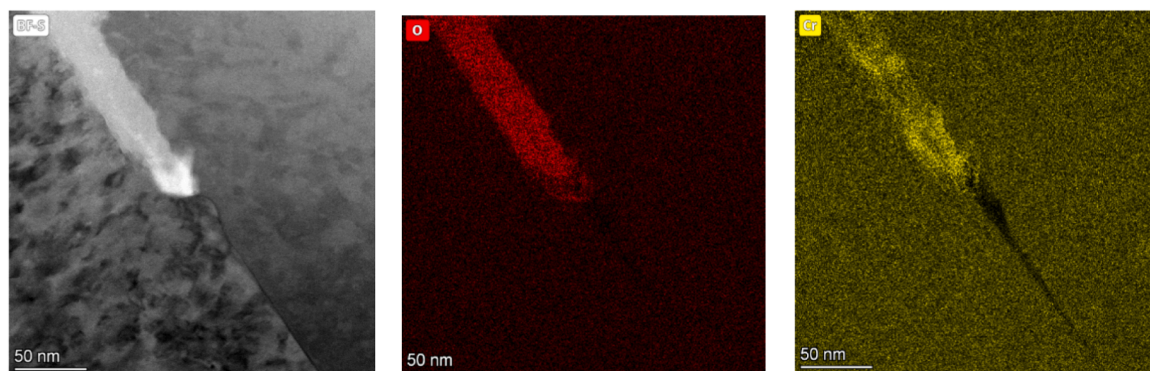
That the high local stress translates into IG cracking was demonstrated by subsequent slow strain rate experiments conducted on the sample in 288 °C water. Each of the 90 grain boundaries was inspected for cracks and only the boundaries intersected by DDCs cracked. The cracking probability is plotted as a function of the normal tensile stress on the grain boundary in Fig. 2e. Note that cracking occurs when the normal stress exceeds ~ 1.0 GPa (at 0.5 μ m from the boundary) and the probability increases monotonically with stress reaching a value of unity at ~ 1.8 GPa.

As evidence of the role of dislocation channels inducing grain boundary cracking, Penders et al.[64,65] observed numerous cases of discontinuous channels intersecting cracked grain boundaries in O-ring tests on flux thimble tube samples exposed in-reactor. While there is increasing evidence for dislocation channels acting as sources of cracks in post-irradiation tests, recent results show that they are active in-reactor as well. A baffle former bolt (BFB) was investigated after reaching ~ 41 dpa in service.[66] BFBs are used to maintain the integrity of baffle plates in the reactor core and are often reported to suffer from IASCC.[67] Although the investigated bolt did not show detectable macroscale cracks, it retained an oxide layer that was formed while in service, suggesting that the near-surface microstructure was not disturbed. Fig. 2f shows the region of the bolt examined using EBSD. Patterns from the circled region are shown in the EBSD inverse pole figure (IPF) map overlapped with an image quality (IQ) map. The channels are localized in the near-surface layer of the sample subjected to elevated local stresses while in service.[67] The fine channels shown are spaced ~ 1 μ m apart, which is typical for strain-induced features in irradiated austenitic steels. The defect-free channel appearance is distinctly different from the plastic deformation expected for nonirradiated steel (smooth misorientation gradients).[68] Channels were easy to distinguish from occasional scratches because the channels follow traces of (111)-planes (dislocation slip planes in face-centered cubic [fcc]-crystals), the projections of which are shown as thin red lines. The channel number, appearance, and distribution resemble results from post-irradiation, slow-strain rate tensile tests when the specimens were loaded at stresses below yield stress.[69] Such early channels suggest that the component experienced a stress level of at least 0.4 yield strength (YS) at least once during service, consistent with the data shown in Fig. 1.

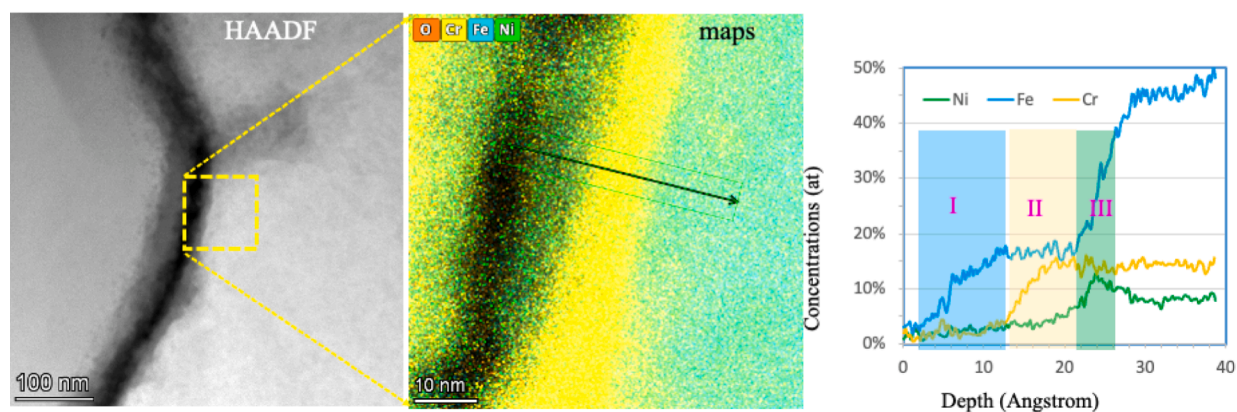
While DDCs are clearly the source of increased stress on the grain boundary, by themselves, they rarely result in IG fracture as discussed in section 1.2. Rather, there must be some additional factor causing the grain boundary to be susceptible to cracking in



(a)



(b)



(c)

Fig. 3. Examples of oxidized grain boundaries; a) planer view with composition profile across a boundary in irradiated 304L SS [85] b) cross sectional view with element maps of a boundary in 304L SS, and c) HAADF image, composition map, and composition profile showing the triple layer structure of the oxidized grain boundary with an Fe-rich oxide over a Cr-rich oxide on a thin Ni-rich layer adjacent to the metal in irradiated 347 SS.

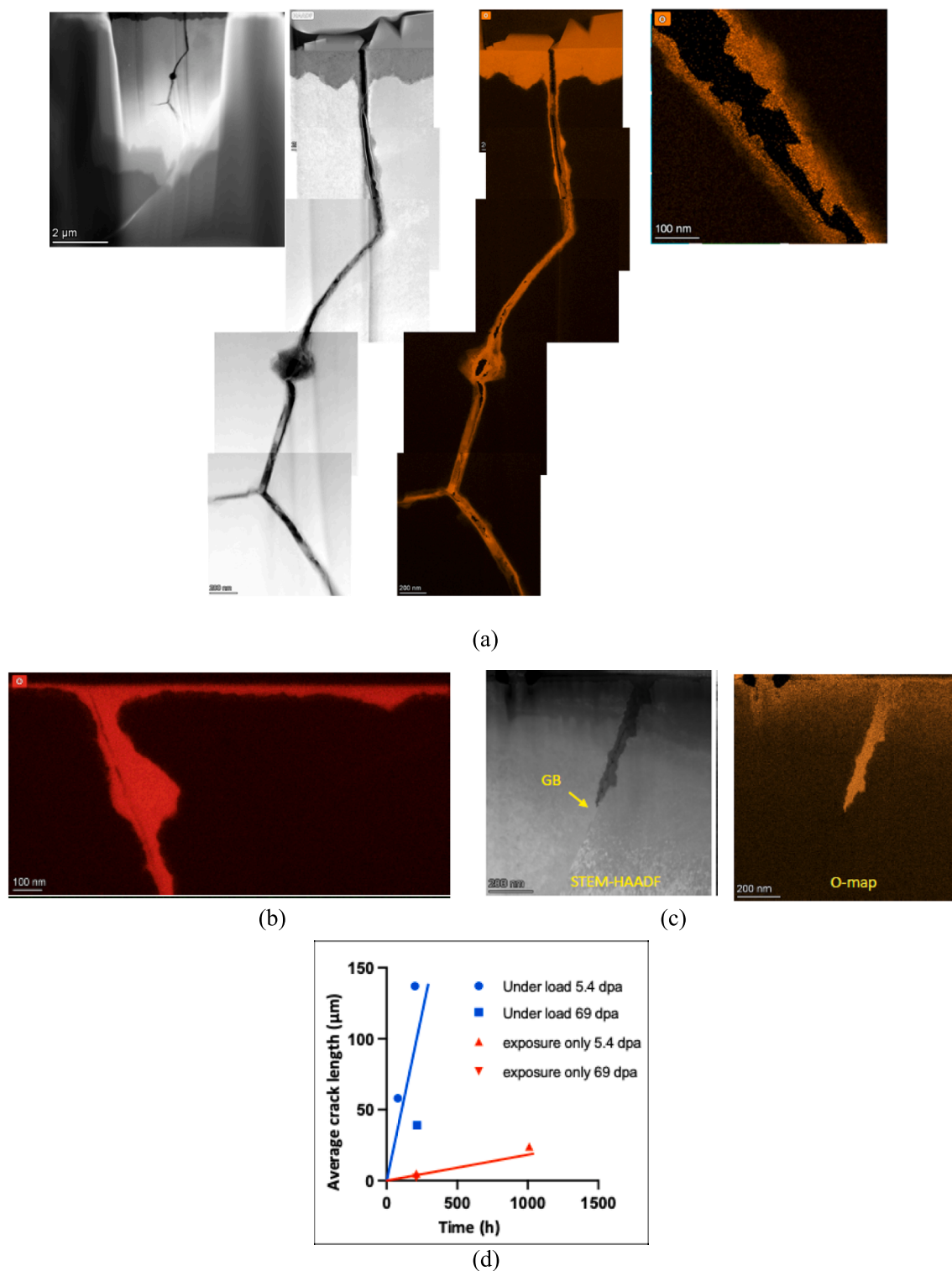
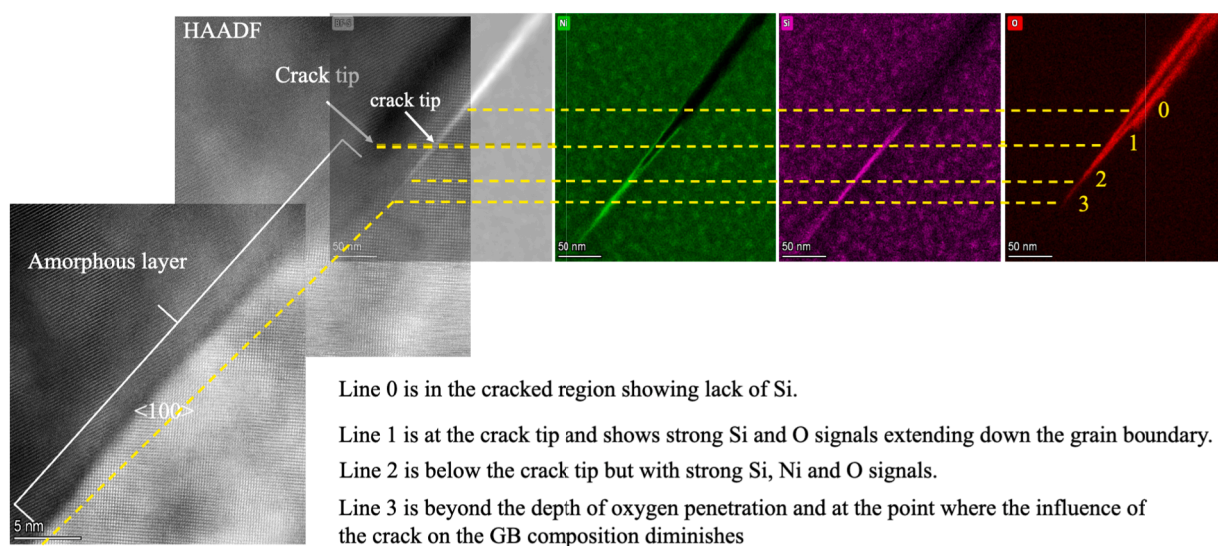
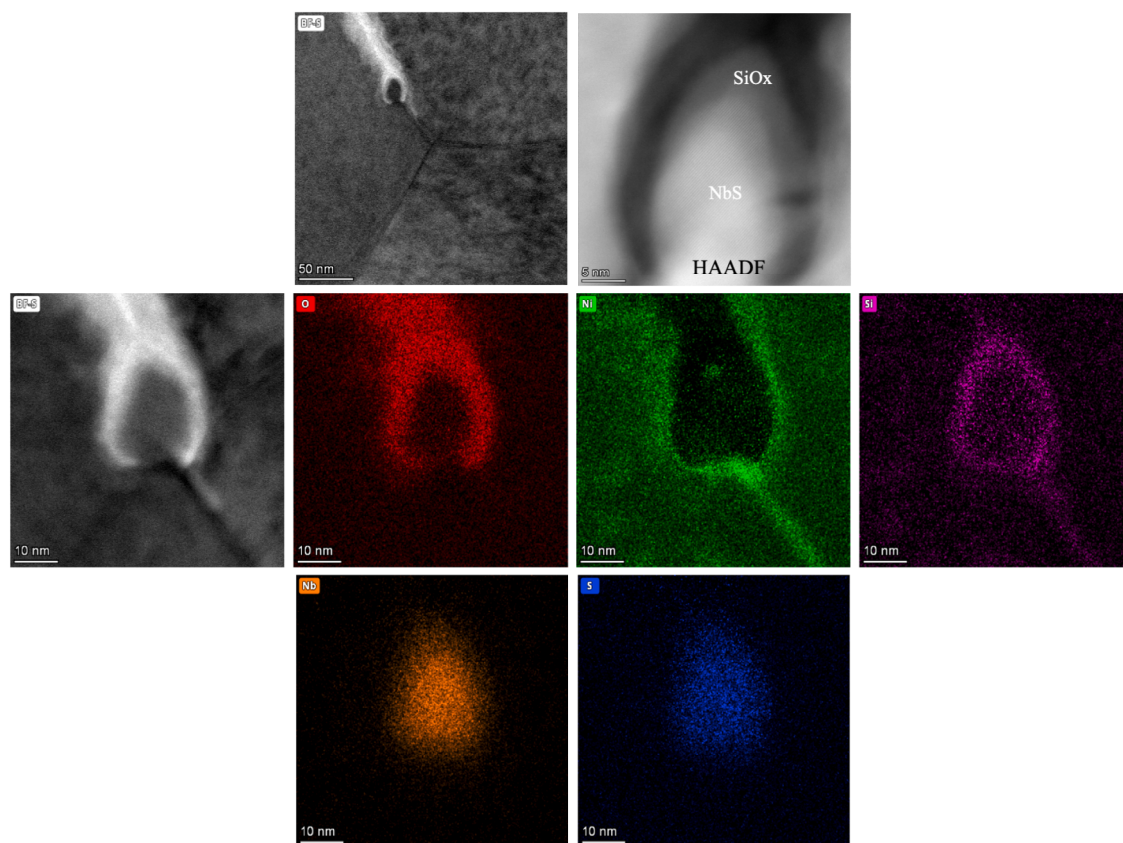


Fig. 4. Oxygen maps showing cracks in oxidized grain boundaries in a) irradiated 347 SS, b) irradiated 316 SS, and c) irradiated 304L stainless steel, all strained in simulated PWR primary water at 320 °C, and d) the effect of stress on the depth of the grain boundary oxide [85] for the cases of straining in 320 °C water (blue symbols), and exposing in 320 °C water and subsequently straining in 320 °C argon (red symbols).

service. That factor is the degradation of grain boundary strength by oxidation.



(a)



(b)

Fig. 5. High resolution transmission electron microscopy image and accompanying composition maps of a crack tip in a) 347 SS irradiated to 26.4 dpa and strained to 60 % of the irradiated yield stress in KOH at 320 °C showing amorphous silicon oxide (a-SiO_x) extending over 50 nm beyond the crack tip and down the grain boundary, b) in the same sample, a-SiO_x formation at the top of a NbS particle where the crack arrested, and c) 304 SS (low Si) irradiated to 5.4 dpa and strained in LiOH at 320 °C to 60 % showing absence of SiO_x and Si above the crack tip.

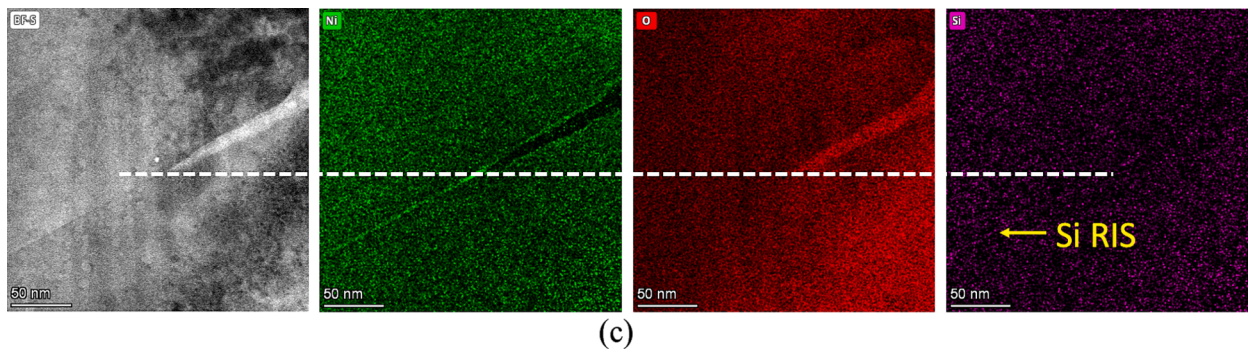


Fig. 5. (continued).

3. Grain boundary oxidation

The exposure of irradiated austenitic alloys to high temperature water results in oxidation of the grain boundaries. Grain boundary oxidation is commonly observed in many nickel-base alloys in the non-irradiated state [70–77], but rarely in stainless steels. Fig. 3a and 3b show planar and cross-section images of oxidized grain boundaries from irradiated stainless steels. The grain boundary oxide formed in irradiated stainless steels in high temperature water is an iron-chromium spinel consisting of a multi-layered structure; iron-rich over chromium-rich over nickel-rich at the oxide-metal interface as shown in the HAADF image and composition map in Fig. 3c [78–82].

The oxide degrades grain boundary strength to varying degrees depending on its structure and porosity. Abad et al. measured the fracture stress of iron-chromium spinel to be 1.4–1.6 GPa,[83] and Dugdale et al. conducted a finite analysis of an experiment on oxidized grain boundaries in nickel-base alloy 600 that yielded a value of 1.35 GPa.[74] These values fall within the range of the normal stress caused by discontinuous dislocation channel impingement on the grain boundary shown in Fig. 2e. Fujii has shown a progressive reduction in GB fracture strength in micro-tensile tests of alloy 600 with exposure time in high temperature water.[70] While only semi-quantitative, measurements consistently show that the stress to fracture a grain boundary progressively drops with increasing exposure to high temperature water, bringing its value into the range measured by HREBSD.[84] This leads to fracture of the grain boundary oxide resulting in oxidation followed by fracture further down the boundary. If grain boundary oxidation indeed lowers the fracture stress, then once oxidized, grain boundaries should be susceptible to cracking without aid of the environment. Given that grain boundaries in irradiated stainless steels are not susceptible to IG fracture in an inert environment, as discussed earlier, fracture of an oxidized grain boundary in an inert environment would establish oxidized grain boundaries as susceptible to IASCC initiation.

To verify the role of oxidation on fracture, a set of experiments was performed to decouple stress and corrosion by exposing samples of irradiated 304 SS in simulated PWR primary water at 320 °C without application of stress.[85] Samples were subsequently strained to stresses below yield in an inert (ultra-high purity argon) environment, and companion samples of the same alloy were strained in the same water environment and at the same temperature. Cracking was observed in both sets of samples (exposed then strained in Ar, or strained in water) and at roughly the same fraction of the irradiated yield stress. The only difference was in the degree of cracking which was greater in the sample that was strained in water.

Fig. 4a-c show several cracks along grain boundaries in irradiated alloys 347, 316 and 304, respectively, strained incrementally in simulated PWR primary water containing 1000 ppm B as H_3BO_3 , 2 ppm Li as LiOH and 3 ppm H at 320 °C. Note the tortuous fracture path through the oxide, as opposed to the oxide-metal interface, indicating a highly brittle oxide with weak tensile strength. While grain boundaries can oxidize without the aid of stress, the application of stress greatly increases the extent of grain boundary oxidation [85–87]. Exposure under stress also results in longer cracks than exposure alone for an equivalent time (followed by loading) as shown by the crack length vs. exposure time graph in Fig. 4d for irradiated 304L stainless steel as a function of time in the environment. Since irradiated stainless steels are generally not susceptible to IG cracking in inert gas below the yield stress, the oxidized state of the grain boundary must be a key factor in IASCC.

4. The role of silicon

There is yet an additional factor in the mechanism of IASCC that can enhance the cracking susceptibility of some irradiated stainless steels. As noted earlier, data exists to suggest that silicon may play a role in IASCC. Silicon readily segregates to grain boundaries in austenitic alloys under irradiation. The grain boundary concentration can be significant, exceeding 10 at% in some cases,[88,89] as profiles are very narrow indicating a type of monolayer segregation.

Constant extension rate tests were conducted on a high purity heat of Fe-18Cr-12 N-1Mn-0.02C and one of the same composition with the addition of 1.05 wt% Si. Both were irradiated with 3.2 MeV protons to 5.5 dpa at 360 °C.[42] Samples were strained at a rate of $3.5 \times 10^{-7} \text{ s}^{-1}$ in simulated BWR water at 288 °C or PWR primary water (PW) at 320 °C. In BWR water, the Si-containing sample had almost double the %IG on the fracture surface, and 4 times the number of cracks on the gage section over the high purity sample. In

PWR PW, the Si-containing sample had 54 IG cracks vs. none on the high purity sample. In another experiment, de-segregation of grain boundary Si following post-irradiation annealing correlated well with a reduction in IASCC susceptibility.[90] Yonezawa et al. created a series of heats of stainless steels and nickel-base alloys with varying Si content and conducted SSRT experiments in simulated PWR primary water.[91,92] Results revealed that in a 15Cr-50Ni-xSi alloy, the %IG on the fracture surface increased dramatically at a Si concentration of about 1 wt%. Silicon had the same effect in crack growth tests on 304L SS containing various levels of Si conducted in high temperature water. Andresen et al. studied the effect of Si as a function of corrosion potential.[93] At low potential, an increase in Si from 2 % to 6 % resulted in an increase in crack growth rate by more than a factor of 10. In another study, CGR tests of a Fe-28Ni-12Cr-xSi alloy revealed that an increase in Si from 0 to 2.5 wt% produced an increase in the CGR by a factor of 22.[94] These cases illustrate the significant effect that Si can have in promoting IG cracking in a low potential, PWR primary water environment, in which IASCC is known to be less severe than in a high potential environment. However, the process by which Si affects IASCC has not been explained to date.

High resolution TEM analysis of crack tips in high Si, Type 347 SS strained in PWR primary water provides insight into the process. Fig. 5a shows a high resolution image of the crack tip of a 347 sample strained in 320 °C simulated PWR primary water containing KOH. Below the crack tip is an oxidized region approximately 2–3 nm wide and extending ~ 50 nm down the grain boundary. Penders et al.[64,65] also observed an oxidized region beyond the crack tip in fractured O-ring samples of CW 316L SS. Here we have identified the oxide as an amorphous silicon oxide, $a\text{-SiO}_x$, adjacent to a $\langle 100 \rangle$ matrix lattice. The EDS maps of Ni, Si and O show that beyond the depth of penetration of oxygen (line 3), the boundary is enriched in both Ni and Si due to radiation induced segregation. Above the tip (line 0), Ni is enriched at the oxide-matrix interface and Si is absent. The amorphous SiO_x region lies between lines 1 and 2 (about 50 nm depth), as evidenced by the very strong Si and O signals and the partitioning of Ni to the oxide-metal interface. The absence of silicon above the crack tip is due to its high solubility in high temperature water [95,96] Its dissolution potentially provides for enhanced crack initiation and also creates a pathway for oxygen diffusion down the grain boundary followed by subsequent oxidation and cracking.

A second example from the same sample, Fig. 5b, shows an intergranular crack arrested at a NbS particle with $a\text{-SiO}_x$ above the particle and just below the crack tip. Note that Si is absent in the crack here as well. Fig. 5c shows that in a low Si 304L SS alloy that was also susceptible to IASCC, there is minimal Si segregation to the grain boundary and no evidence of amorphous silica at the crack tip.

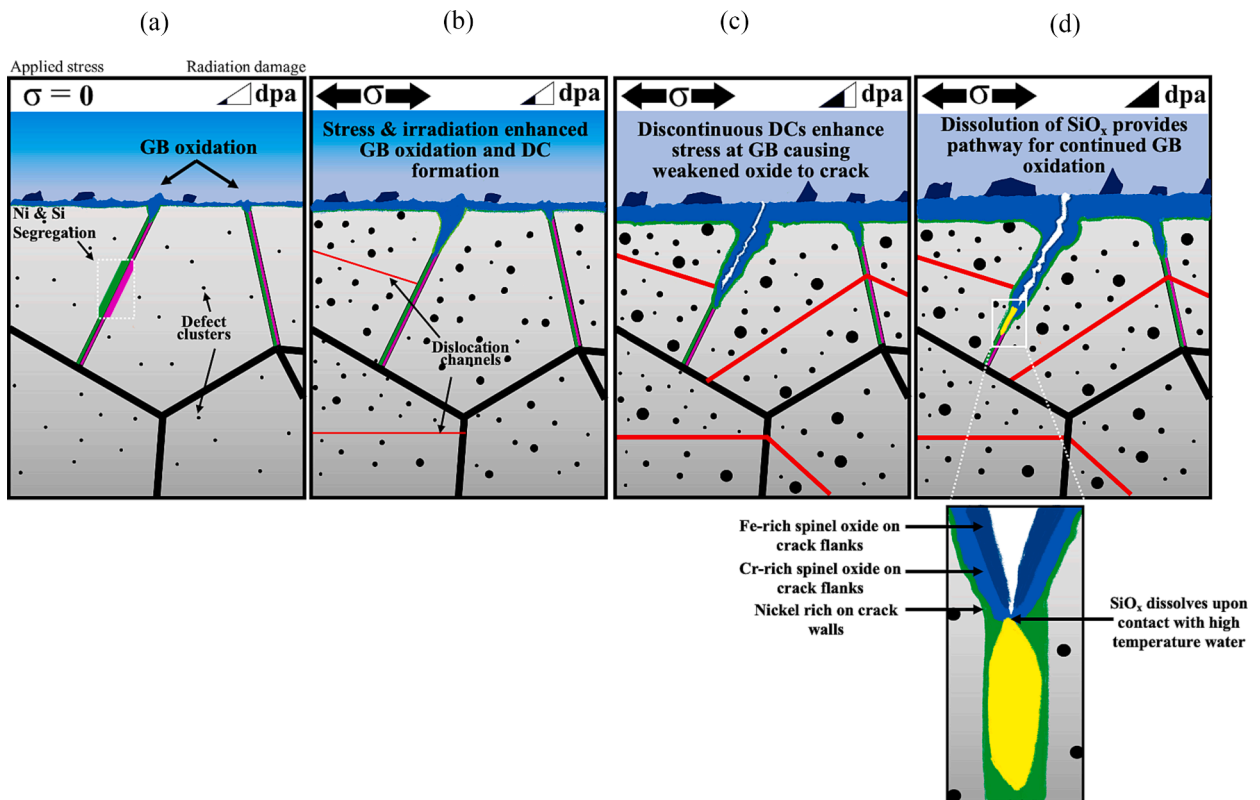


Fig. 6. Schematic of the processes driving IASCC: a) irradiation-induced defect cluster formation, segregation of minor and major alloying elements to grain boundaries, and grain boundary oxidation at low damage level, b) application of stress combined with increased irradiation damage enhances grain boundary oxidation and induces formation of dislocation channels, c) dislocation channels impinging on GBs near the surface cause fracture of the weak oxidized grain boundaries, and d) exposure of Si-enriched GBs to water oxidizes the Si to amorphous SiO_x that dissolves, providing a pathway for continued oxidation of the boundary, setting up the conditions for crack growth.

While not required for IASCC, silicon enrichment at the grain boundary augments the susceptibility of the alloy to IASCC initiation and perhaps, propagation.[97,98] Thus, silicon enhances the susceptibility to IASCC but is not necessary for it to occur.

5. The mechanism of irradiation assisted stress corrosion cracking

The processes just described provide for a coherent picture of the IASCC mechanism, depicted in Fig. 6. The key processes that drive IASCC in irradiated stainless steels interact in the following way: High temperature water combined with irradiation results in oxidation of grain boundaries, Fig. 6a. Applied tensile stress enhances the rate of grain boundary oxidation, which weakens the boundary and reduces the fracture strength to the ~ 1 GPa regime and potentially lower, Fig. 6b. Although the applied stress can be well below the yield strength of the alloy, plastic flow occurs on a very localized scale in properly oriented grains. The localized deformation in the form of dislocation channels that terminate at the grain boundary create local stresses at the DC-GB site of ~ 1 GPa and higher, sufficient to nucleate a crack in the near surface region of an oxidized (weakened) grain boundary, Fig. 6c. The fracture of the oxide allows for oxidation of the boundary beneath the crack tip, providing for continued growth of the crack. In cases where Si is enriched at the grain boundary by radiation induced segregation, the silicon oxidizes to form an amorphous silica layer at the boundary several nm wide and tens of nm below the crack tip. By virtue of its solubility in high temperature water, a-silica dissolves, aiding the extension at the crack tip and providing for oxidation of Si further down the boundary, Fig. 6d.

6. Confirmation and impact on the industry

With an understanding of how IASCC occurs, a number of questions regarding the characteristics or observations of IASCC over decades can be addressed, such as: Why an irradiated alloy is so susceptible to IASCC? How can cracking occur below the yield point? Why does cracking correlate with yield strength? Why does cracking susceptibility seem to saturate at high dpa? How can IASCC occur in a constant load test? What about the cold worked condition in which DCs don't readily form? Why don't all boundaries crack?

What makes the irradiated condition so susceptible to IASCC is the mode of deformation that results in dislocation channels that create high local stress on an oxidized grain boundary. Cracking can occur well below the yield point because plasticity occurs very locally in the form of dislocation channels in well oriented grains. At low stresses, the density of channels is very low, but if enough are created that enhance the local stress state at oxidized boundaries, then crack formation will occur. IASCC correlates with yield strength because yield strength increases with damage level due to defect build-up, increasing the degree of localized deformation. As noted earlier, the development of dislocation channels in core components occurs both in-reactor and in post-irradiation testing at reactor temperatures. Once the irradiated microstructure saturates, so does the yield strength and therefore, the localization of deformation. In a rising load test such as the constant extension rate test (CERT) or slow strain rate test (SSRT), channel formation increases with stress producing increasing numbers of DDC-GB sites, thus increasing the probability of crack nucleation. In a constant load test, while the load is fixed, the state of the grain boundaries changes due to oxidation, progressively degrading their strength and increasing their susceptibility to IG crack initiation. Cold worked stainless steels also fail by IASCC, but dislocation channels are not often observed. The intersection of these deformation bands with grain boundaries have been found to be sites of increased oxidation and crack initiation [64,65,73,85,99] and potentially, increased stress. Thus, deformation bands formed during cold working likely serve in much the same role as DCs.

The big question is, why don't all boundaries crack. Since cracking relies on a critical stress at a susceptible (oxidized) grain boundary located near the surface, a number of conditions must be fulfilled. First, a channel must be formed, which requires a grain with a slip plane properly oriented with respect to the stress. That channel must terminate at a grain boundary rather than transmit slip to the neighboring grain. Slip termination vs. transmission is likely also a function of the grain boundary structure. The boundary must be oriented so as to maximize the local stress at the DC-GB intersection site. The boundary must be oxidized. Finally, and importantly, the channel must intersect an oxidized grain boundary near the surface to initiate an IG crack.

An additional consideration is that while dislocation channels form and impact grain boundaries in service, as shown earlier, irradiation during deformation tends to homogenize the dislocation structure compared to post-irradiation deformation,[100] reducing the number of initiation sites still further. Thus, the number of conditions that must be fulfilled means that IASCC is a rather rare occurrence such that only a small fraction of grain boundaries will be susceptible to failure for any given set of environmental conditions, hence, the long initiation time. It is not surprising that because of the interplay of so many factors, probabilistic approaches are often employed to explain IASCC.[57].

While the mechanism described here provides both qualitative and quantitative descriptions of the processes that govern IASCC in irradiated stainless steel, there remain open questions and areas that need to be explored. For example, while most austenitic stainless steels and nickel base alloys show susceptibility, the degree of susceptibility varies. Clearly there are secondary effects of composition and/or microstructure. The roles of second phases and martensite in the initiation of cracks are still not understood. Surface finish is known to have an effect on cracking susceptibility as does cold work. Finally, while this mechanism explains crack initiation, it does not treat crack growth. It may be that the factors driving the growth of an incipient crack differ from those that control crack initiation. Crack initiation in thin sections is the critical factor controlling lifetime. This was evident in the case of free span IGSCC of steam generator tubes made from nickel-base alloy 600 as the incubation time for cracking was a full decade, but growth to failure forced replacement of the steam generator within three years.[101] Similarly, most of the data shown in Fig. 1 are from O-ring tests of irradiated stainless steel flux thimble tubes in which initiation is assumed to control the time to failure. This assumption is supported by the last four cases in the legend that were conducted as incremental straining tests (similar to CERT or SSRT) and were stopped at the stress where the first crack(s) were observed. That these data agree with O-ring data indicate that the crack growth stage

was fast relative to the crack initiation stage. Nevertheless, experiments to uncover the specific processes driving crack growth have not been conducted to date.

A deeper understanding of the IASCC mechanism provides the opportunity to develop models to predict its occurrence, and strategies to mitigate its effect. The former is of considerable interest to reactor operators (utilities), and the latter to the industry supplying components for light water reactor plants and advanced reactors. With an understanding of the physical processes driving IASCC initiation, physically-based predictive models can be developed that will provide greater capabilities than probabilistic approaches used now. As for mitigation strategies, these may include eliminating or reducing dislocation channel formation, grain boundary engineering to increase the fraction of grain boundaries that transmit DCs, pre-oxidation of components to minimize GB oxidation in reactor, minimizing the Si content, etc. Understanding the fundamental mechanism behind IASCC provides for the development of mitigation measures and physically-based predictive models that may serve as important tools for the avoidance of IASCC in future generations of nuclear reactors.

CRediT authorship contribution statement

G.S. Was: Conceptualization, Data curation, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Writing – original draft, Writing – review & editing. **C.-B. Bahn:** Data curation, Formal analysis. **J. Busby:** Conceptualization, Formal analysis, Investigation, Methodology. **B. Cui:** Data curation, Formal analysis. **D. Farkas:** Conceptualization, Data curation, Formal analysis, Software, Funding acquisition. **M. Gussev:** Data curation, Formal analysis. **M. Rigen He:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software. **J. Hesterberg:** Formal analysis, Investigation, Methodology. **Z. Jiao:** Conceptualization, Formal analysis, Investigation, Methodology. **D. Johnson:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology. **W. Kuang:** Data curation, Formal analysis, Investigation, Methodology. **M. McMurtrey:** Data curation, Investigation, Methodology. **I. Robertson:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology. **A. Sinjlawi:** Formal analysis, Investigation. **M. Song:** Formal analysis, Investigation. **K. Stephenson:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology. **K. Sun:** Formal analysis, Investigation, Methodology. **S. Swaminathan:** Investigation, Methodology. **M. Wang:** Data curation, Investigation, Methodology. **E. West:** Investigation, Methodology.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

This work was supported by the Office of Basic Energy Sciences, U.S. Department of Energy awards DE-FG02-08ER46525 and DE-FG07-05ID14703; Nuclear Energy University Program, Office of Nuclear Energy, U.S. Department of Energy award DE-AC07-05ID14517; Light Water Reactor Sustainability Program, Office of Nuclear Energy, U.S. Department of Energy awards 400079632 and 4000185584; U.S. Department of Energy awards F031891, 4000136101 and 4000129492; Electric Power Research Institute awards EP-P20783/C10134, EP-P39425/C17515, 10002154, 10002164, 10003608, 10003872, 10002970; Michigan Ion Beam Laboratory; Michigan Center for Materials Characterization; and the many helpful discussions with Peter Scott, Peter Andresen and Stephen Bruemmer.

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