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**TECHNICAL BASIS FOR THE
INITIATION AND CESSATION OF
ENVIRONMENTALLY-ASSISTED CRACKING OF
LOW-ALLOY STEELS IN ELEVATED TEMPERATURE
PWR ENVIRONMENTS**

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TECHNICAL BASIS FOR THE INITIATION AND CESSATION OF ENVIRONMENTALLY-ASSISTED CRACKING OF LOW-ALLOY STEELS IN ELEVATED TEMPERATURE PWR ENVIRONMENTS

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ABSTRACT

The Section XI Working Group on Flaw Evaluation of the ASME B&PV Code Committee is considering a Code Case to allow the determination of the conditions under which environmentally-assisted cracking of low-alloy steels could occur in PWR primary environments. This paper provides the technical support basis for such an EAC Initiation and Cessation Criterion by reviewing the theoretical and experimental information in support of the proposed Code Case.

INTRODUCTION

Environmentally-assisted cracking (EAC) of low-alloy steels in elevated temperature light-water reactor (LWR) aqueous environments has been a concern since the pioneering work of Kondo *et al.* (1972a, 1972b) demonstrated that corrosion-fatigue crack propagation (CFCP) rates in a Boiling Water Reactor (BWR) environment at 200°C (392°F) and 260°C (500°F) could be considerably higher than in air under the same conditions. These concerns led to a large multi-national multi-disciplinary effort to characterize and understand the EAC process (Tompkins, 1989), and considerable understanding has emerged from the efforts of this group as well as others. Much of this progress has been chronicled in a series of review articles (Tice, 1985; Scott, 1988; Bulloch, 1989; James, 1995a), each review representing an update in the state of knowledge. The result is that the basic cause-and-effect relationship is now well understood for the EAC phenomenon, and the occurrence/non-occurrence of EAC may now be quantified.

Hundreds of research papers have been written on the phenomenon of EAC of low-alloy steels in LWR environments. These papers bear witness to the fact that EAC readily occurs in autoclave experiments, provided the right conditions are met. On the other hand, the operating experience of nuclear power stations worldwide suggests that the phenomenon occurs only very infrequently in service. This apparent dichotomy has led to a delay in implementing updated

CFCP/EAC analysis procedures into standardized codes (e.g., the ASME B&PV Code) because although the EAC phenomenon can be easily initiated in autoclave experiments when the right conditions exist, there has up to now not been a reliable quantitative criterion to predict the initiation, or cessation, of EAC under more realistic service conditions. Such criteria are now available, and are under consideration by the ASME B&PV Code Section XI Working Group on Flaw Evaluation to be published as a Code Case applicable to Pressurized Water Reactors (PWR). This paper provides the technical support basis for the Code Case covering the EAC Initiation and Cessation Criteria. It should be emphasized that these criteria presently have been developed only for PWR primary system environments; there is no fundamental reason why they could not be applied to PWR secondary environments or BWR environments, but the data to support these applications are not presently available. It should be noted that the technical basis for the crack growth rate curves in the Code Case for EAC and non-EAC conditions has been documented by Eason *et al.* (1995). Therefore, the primary objective of this paper is to provide the basis for the quantitative criteria for determining the occurrence or non-occurrence of EAC. These criteria are needed in order to select the appropriate crack growth rate curve for use in fatigue crack growth analyses.

MASS TRANSPORT OF SULFIDES

It will be shown that the supply of sulfides to, and the mass transport of sulfides from, the crack-tip region, and the attendant crack-tip sulfide concentration, is the deciding factor as to whether or not EAC occurs. Since the objective of this paper is to document the technical basis for the quantitative EAC Initiation and Cessation Criteria, it will be necessary to review those aspects of the EAC phenomenon that bear upon the mass transport of sulfides. This is not, however, intended to be a general review of the EAC phenomenon; the reader is directed to the previously discussed reviews for more general information.

It has been recognized in a qualitative sense for almost two decades that high-sulfur steels were more susceptible to EAC than low-sulfur steels. However, this was quantified by VanDerSluys and Emanuelson (1993) who injected known concentrations of hydrogen sulfide (H_2S) directly into the crack-tip region of a low-sulfur steel that would otherwise not exhibit EAC in a 288°C (550°F) PWR environment. It was found that a concentration of 5 ± 2 ppm H_2S was sufficient to initiate EAC, and that the EAC could be sustained as long as the supply of H_2S was continued. Therefore, the "critical" concentration of crack-tip sulfides to cause EAC is approximately 5 ± 2 ppm.

The crack-tip sulfide concentration at any point in time is the net balance between the competing processes of supply of sulfides to the crack tip versus the loss of sulfides from the crack tip by mass transport. Metallurgical sulfides (e.g., MnS, FeS, etc.) are present as an impurity in virtually all low-alloy steels (Wranglén, 1974). In lieu of water-borne sulfur contamination which is not expected in a normal PWR environment (Wood, 1995), the only source of crack-tip sulfides is the growing crack which intersects the embedded metallurgical sulfide inclusions in the steel. Such sulfides readily dissolve in the elevated temperature water environment (Wranglén, 1974; Matsushima *et al.*, 1991) thus providing a supply of sulfide ions to the crack tip. It is clear that the higher the rate of crack extension per unit time, the higher the rate of sulfide supply to the crack tip because metallurgical sulfides are being exposed and dissolving at a higher rate.

Sulfides may be removed from the crack-tip region by any one, or more, of four mass-transport processes: 1) diffusion due to a sulfide concentration gradient from the crack tip to the crack mouth (Turnbull and Thomas, 1982; Wire and Li, 1996a), 2) ion migration due to an electrochemical corrosion potential (ECP) gradient from the crack tip to the crack mouth (Turnbull and Thomas, 1982), 3) fatigue "pumping" or advection due to cyclic motion of the crack flanks (Hartt *et al.*, 1978; Turnbull, 1983), and 4) convective flow induced within the crack enclave due to an external (to the crack) stream flow (James *et al.*, 1995b, 1997a). Each of these four mass-transport processes can, in some cases, have a profound influence upon the outcome between the competing processes of sulfide supply and sulfide loss, and hence the occurrence or non-occurrence of EAC. The four mass-transport processes are reviewed below.

The first mass-transport process, diffusion, is the sole basis for the EAC Initiation and Cessation Criteria that will be discussed later. In some respects, diffusion can be the least "powerful" of the four processes, but it can always be counted upon to be operative in a surface crack exposed to water so long as there is a higher concentration of sulfides at the crack tip than at the crack mouth. Therefore, even though the other mass-transport processes may also be removing sulfides from the crack tip in a given situation, it is conservative to consider only diffusion mass transport when formulating the EAC Initiation and Cessation Criteria.

Mass transport of sulfide ions due to ion migration is not expected to be a large factor in hydrogenated PWR environments. Under such conditions, the ECP gradient from the crack tip to the crack mouth is relatively small (Gabetta and Caretta, 1987), and the migration of sulfide ions should be minimal in PWR environments. CFCP experiments employing semi-elliptical surface cracks penetrating weld-deposited cladding into the underlying steel, plus ECP measurements of the two dissimilar metals, have demonstrated that the presence of the

cladding/base metal interface does not introduce a galvanic interaction that would influence ion migration in hydrogenated low-oxygen water (James *et al.*, 1997b, 1997c). This general lack of ion migration effects in a PWR environment is, however, not the case in normal BWR environments. In normal BWR water, the relatively high dissolved oxygen content causes the crack mouth to be anodic relative to the crack tip (Gabetta and Caretta, 1987), and this produces an ECP gradient that concentrates sulfide ions at the crack tip (Andresen and Young, 1995). This increases the susceptibility to EAC in normal BWR environments, and it is for this reason that such environments are presently excluded from the EAC Initiation and Cessation Criteria discussed herein. Although the concepts employed in developing the criteria are, in theory, applicable to BWR environments (either the "normal" or the "hydrogen" water chemistries), no experimental data presently exist for initiation or cessation of EAC in BWR environments.

The third mass-transport process, fatigue "pumping" or advection, is not expected to play a significant role in most LWR cyclic stress applications. Fluids may be pumped into and out of the crack enclave by the cyclic motion of the crack flanks, but this process is most effective only at relatively high cyclic frequencies (Hartt *et al.*, 1978; Turnbull, 1983). For example, using the simple one-dimensional model of Turnbull (1983), Wire (1997a) estimated that mass transport by diffusion would dominate mass transport by advection for cyclic frequencies that occur less frequently than about one cycle each four minutes, and this is likely to be the case for most LWR cyclic stress applications.

The fourth process, convective mass transport within the crack enclave induced by external fluid flow, can be very effective in those applications where an active flow washes over a surface crack parallel to the length of the crack. Several earlier studies on compact tension-type specimens (reviewed by James *et al.*, 1995b) qualitatively demonstrated the beneficial effect of flow. However, mass-transport within the crack enclave of a compact tension specimen can be quite different from that within the enclave of the more prototypic semi-elliptical surface crack. James *et al.* (1995b, 1997a, 1997c) conducted a series of flow rate experiments, plus the associated computational fluid dynamics analyses, employing different flow velocities and semi-elliptical surface cracks of different sizes, and found that flow parallel to the crack was very effective in mitigating an active case of EAC in a high sulfur steel. This was the case both for a surface crack in a monolithic high-sulfur steel, as well as for a surface crack that penetrated weld-deposited cladding into the underlying high-sulfur steel.

The above review of sulfide mass transport processes demonstrates that it is conservative to ignore mass transport by ion migration, advection, and convection when developing the following

criteria for EAC initiation and EAC cessation in PWR environments, and base the criteria only upon diffusion mass transport.

INITIATION OF EAC IN SURFACE CRACKS

The EAC Initiation Criterion is intended to provide a quantitative relationship for predicting the conditions under which it would be necessary to employ EAC crack growth relationships (e.g., Eason *et al.*, 1995) when analyzing the in-service extension of a surface crack exposed to a PWR primary environment. Such a surface crack could either be one that had been present since the beginning of service, or it could be one that initiated sometime during service. The EAC Initiation Criterion is based on the fact that it is necessary to extend a crack at a sufficient velocity that the rate of sulfide supply exceeds the rate of sulfide loss for a sufficient period of time (or alternatively, for a sufficient crack extension, Δa) that the critical sulfide concentration for EAC is achieved at the crack tip. This criterion is based on a theoretical treatment of diffusion mass transport (Wire *et al.*, 1996a, 1997b), and was verified by experimental data (Wire and Li, 1996a; James, 1997d). The critical crack-tip velocity required to initiate EAC is designated V_{in} , and has the units mm/s (or inch/s).

The role of crack tip velocity in the production rate of sulfide ions, and the requirement to exceed a critical crack-tip velocity to cause EAC was qualitatively identified by Atkinson and Forrest (1985). Combrade *et al.* (1988) identified diffusion as the dominant mass transport process in the loss of sulfides from the crack tip under quasi-stagnant flow conditions, and theorized that under steady-state conditions, a critical time-based growth rate of about 1×10^{-5} mm/s (4×10^{-7} inch/s) was necessary to produce the sulfur concentration necessary to sustain EAC. Combrade *et al.* presented experimental data for several steels in a 288°C (550°F) PWR environment that correlated the initiation or cessation of EAC with a critical crack-tip velocity. Wire *et al.* (1996, 1997b) extended Combrade's simple steady-state analysis to account for transient diffusion conditions, and presented experimental data in support of the analysis. Van Der Sluys and Emanuelson (1993) conducted experiments in a 288°C (550°F) PWR environment whereby EAC initiation and cessation could be correlated with a critical crack-tip velocity, and these results were included in the assessment by Wire and Li.

The theoretical analyses and experiments in support of EAC initiation were all based on the assumption that the crack tip was initially relatively free of sulfides. This is a very reasonable

assumption both for pre-existing surface cracks, or for surface cracks that initiate in service. In the case of pre-existing surface cracks, it is almost certain that they would be relatively free of crack-tip sulfides even before the plant went into service. Diffusion of sulfides from the crack occurs anytime a surface crack is exposed to water, and not just when the plant is operating. All primary system internal surfaces would be wetted for many months prior to placing the plant in service (e.g., pre-service testing), and diffusion times for such cracks are considerably shorter than the pre-service testing times (Wire *et al.*, 1996a, 1997b). In addition, any water flow that might occur during pre-service testing would further decrease the time necessary to reduce crack-tip sulfide concentrations to relatively low levels. Hence, it is reasonable to assume that any pre-existing cracks would enter service with crack tips relatively free of sulfides. This will be demonstrated in an experiment to be discussed later.

In the case of cracks that initiate in service at the wetted surface, the very shallow depth of such an initiating crack will lead to very short diffusion times (Wire and Li, 1996a). In addition, convective mass transport due to any active flow at the surface will also be effective on a shallow crack. Hence, crack-tip sulfide concentrations in such shallow cracks will be relatively low, and non-EAC growth can be assumed at least to the depth where linear-elastic fracture mechanics (LEFM) analysis methods start to be viable; e.g., crack depths beyond about 1.3 mm (0.05 inch) to 2.5 mm (0.10 inch). Beyond that depth, the EAC Initiation Criterion can be employed to assess whether the use of EAC crack growth relationships is necessary or not. Because it is based on crack-tip velocities and mass transport, the use of LEFM analysis methods in the EAC Initiation Criterion is, in theory, not necessary; elastic-plastic analysis methods could be employed if desired. However, with the present state of analysis methods, the use of LEFM is certainly the most convenient.

The EAC Initiation Criterion proposed by Wire and Li (1996a), and recommended for use in the Code Case, was formulated using high-sulfur EAC-susceptible ASTM A302-B plate steels (UNS K12022). The criterion is conservative for such steels, and the degree of conservatism increases with decreasing steel sulfur content; i.e., the crack-tip velocity required to initiate EAC, V_{in} , will increase with decreasing steel sulfur content. Because the rate of sulfide supply to the crack tip is proportional to the product of the crack velocity times the metallurgical sulfide volume fraction, it follows that a higher crack-tip velocity will be required to initiate EAC in a steel with a lower volume fraction of metallurgical sulfides. The high-sulfur A302-B plate steels employed in the criterion development were among the most EAC-susceptible steels ever tested. Hence, the EAC initiation velocities reported herein should be conservative for almost any steels in service, even without the imposition of additional safety factors.

The average crack velocity in service, $\bar{V}$, is defined as the crack extension over a given period of time, divided by the total time in that period. The total period of time includes the cyclic periods for fatigue loadings, dwell periods between or during fatigue cycles, and even time when the plant is not operating but the crack surfaces would be wetted. In this respect, $\bar{V}$ differs from other time-based growth rates like the time-based air rate, $\dot{a}_b$, as proposed by Shoji *et al.* (1981, 1983) and incorporated into crack growth relationships by Eason *et al.* (1995). The definition of $\bar{V}$ is $\Delta a / \Delta T$ where Δa is the crack extension over a given time increment and ΔT is that total time increment. In contrast, the definition of $\dot{a}_b$ is $(da/dN) / T_R$ where T_R is the load-rise time for that cycle. $\dot{a}_b$ is related to the crack-tip strain rate in a fatigue event, is the correct mechanical crack-driving parameter for CFCP, and indeed provides an excellent basis for correlating CFCP results over a wide range of parameters (e.g., ΔK , R , T_R , etc.) as shown by Eason *et al.* (1995). Eason's correlations were based entirely on results from continuous-cycling fatigue experiments. However, the use of the load-rise time in the formulation of $\dot{a}_b$ does not reflect the total time for diffusion of sulfides from the crack tip during operation. Fatigue load-rise times may amount to a small proportion of total operational time for typical LWR operation, yet diffusion mass transport occurs on a full-time basis. Eason *et al.* (1995) have correlated EAC threshold values on an $\dot{a}_b$ basis, but have acknowledged that many of the data employed in the correlation were not true EAC thresholds. Because of the differing bases for $\bar{V}$ and $\dot{a}_b$, and because much of the data employed by Eason did not represent true EAC thresholds, the proposed EAC Initiation Criterion is not based on, nor closely related to, the EAC thresholds proposed by Eason although a small portion of the data employed in developing both formulations are common. However, the steady-state non-EAC and steady-state EAC crack growth relationships developed by Eason *et al.* (1995) will be employed to calculate crack extension in the EAC Initiation Criterion, as they represent the best relationships available.

In the EAC Initiation Criteria, EAC initiates when the mean crack-tip velocity, $\bar{V}$, exceeds the critical velocity to initiate EAC, V_{in} , and does so for a sufficient minimum amount of crack extension, Δa_{cr} :

$$\bar{V} \geq V_{in} \quad \text{and} \quad \Delta a \geq \Delta a_{cr}$$

Note that $\bar{V} \geq V_{in}$ is necessary but not sufficient to initiate EAC; both parts of the criterion must be satisfied. Physically, this means that the mean crack-tip velocity must not only equal or exceed V_{in} , but it must do so for a sufficient period of time (or alternatively, expose sufficient embedded sulfides with a minimum Δa) that the critical crack-tip sulfide concentration to cause EAC is achieved. Therefore, EAC will not initiate during an isolated single cyclic event that causes the instantaneous crack-tip velocity to exceed V_{in} . This is because the momentarily higher rate of sulfide supply will not be sustained, and the small, momentarily higher, increase

in crack-tip sulfide concentration will diffuse away before another significant cyclic event occurs. Only when steady almost-continuous relatively high-amplitude cyclic loadings occur that drive the crack tip to high enough average velocities for a significant period of time (as occurs in many autoclave experiments), is EAC likely to occur. This is, in fact, why EAC is relatively easy to initiate in high-sulfur steels in autoclave experiments, yet is relatively infrequent in the same class of steels in service. Combrade *et al.* (1988) observed that most CFCP specimens are precracked in an air environment prior to insertion into an autoclave. This air precracking exposes a relatively large inventory of undissolved metallurgical sulfide inclusions on the crack flanks, and these inclusions start to dissolve when water is introduced into the autoclave. Under such conditions, crack-tip sulfide concentrations can easily exceed the critical level. Scott (1990) has published recommended test procedures for CFCP experiments in autoclaves, and these procedures suggest starting the fatigue cycling "within several hours" of having stabilized the autoclave temperature and water chemistry. Hence, many autoclave experiments in the past were predisposed toward exhibiting early EAC, and this EAC was generally sustained by continuous cycling at cyclic frequencies high enough to result in relatively high crack velocities. In contrast, the EAC initiation experiments performed by Wire and Li (1996) and James (1997d) were "cleaned" of crack-tip sulfides before fatigue cycling commenced.

A typical EAC initiation test from Wire and Li (1996a) is illustrated in Figure 1. This experiment was conducted on a 1T-CT specimen of high-sulfur A302-B steel. The specimen had been precracked in air to a precrack depth of about 2.54 mm (0.10 inch), and then soaked in an autoclave of quasi-stagnant low-oxygen water at 243°C (470°F) for 2-3 weeks prior to starting fatigue cycling. The specimen was held under a static load ($\approx 75\%$ of the maximum fatigue load) to ensure that it was at least partially open during the soak period. It is clear that the 2-3 week soak period was sufficient to allow sulfides to diffuse from the crack enclave at least to a concentration below the critical concentration, because EAC was not observed in the first or second test phases. Crack extension predictions are also shown in Figure 1 for three possible scenarios: mean air behavior (Eason *et al.*, 1995), mean non-EAC behavior (James, 1994a), and mean EAC behavior (James, 1994a). It is clear that during the first two test phases of the experiment shown in Figure 1, the observed crack growth rates were not even as high as mean non-EAC rates ($\approx 1.7 \times$ air rate). Three test phases were employed, and the same load-rise times ($T_R = 25.5$ s) and load-fall times ($T_F = 4.5$ s) were employed in each phase. However, differing dwell times (T_D) at the minimum load between each active fatigue cycle were employed in each test phase. Thus, the total cyclic period in any test phase was $T_R + T_F + T_D$. As seen in Figure 1, the total cyclic periods were 566 s, 230 s, and 30 s, for Test Periods 1, 2, and 3, respectively. The observed mean crack velocity ($\bar{V}$) was 1.32×10^{-7} mm/s (5.20×10^{-9} in./s) in Test

Phase 1, and 2.40×10^{-7} mm/s (9.46×10^{-9} in./s) in Test Phase 2. Only when the average crack velocity was increased to 4.22×10^{-6} mm/s (1.74×10^{-7} in./s) in Test Phase 3 did EAC initiate as shown in Figure 1.

The Figure 1 example employed dwell times between the active fatigue cycles to achieve the desired crack velocities. Other EAC initiation tests presented by Wire and Li (1996a) employed a continuous cycling waveform at different cyclic frequencies to achieve the equivalent effect of modifying crack velocities, and the results are the same. Figure 2 shows the dataset employed by Wire and Li to arrive at an EAC initiation velocity (V_{in}) for high-sulfur A302-B steels (two different heats with sulfur contents in the range 0.024% to 0.035%). Note that the lowest average crack velocity at which EAC initiated was about 5×10^{-7} mm/s (2×10^{-8} in./s). Most of the specimens represented in Figure 2 were "1T-CT" compact tension specimens with precracks of approximately 2.54 mm (0.10 inch) from the notch. Because diffusion mass transport is an inverse linear function of the crack depth, it is necessary to incorporate the crack depth into the relationship for V_{in} :

$$V_{in} = (5 \times 10^{-7})(2.54/a) = 1.27 \times 10^{-6}/a$$

(V_{in} = mm/s, a = mm)

or

$$V_{in} = (2 \times 10^{-8})(0.1/a) = 2.0 \times 10^{-9}/a$$

(V_{in} = in./s, a = inch)

The other part of the EAC Initiation Criterion that must be satisfied is the requirement for the minimum crack extension at a given $\bar{V}$ to exceed Δa_{cr} . Wire and Li (1996a) found that the minimum observed Δa_{cr} to be approximately 0.3 mm (0.012 inch). Hence, the complete EAC Initiation Criterion becomes:

$$\bar{V} \geq V_{in} = 1.27 \times 10^{-6}/a \quad (\text{mm/s})$$

$$\Delta a \geq \Delta a_{cr} = 0.3 \text{ mm}$$

or

$$\bar{V} \geq V_{in} = 2.0 \times 10^{-9}/a \quad (\text{in./s})$$

$$\Delta a \geq \Delta a_{cr} = 0.012 \text{ inch}$$

The above criterion was developed mainly using CT-type specimens, although Wire and Li (1996) did report results from one surface-cracked specimen. To provide additional verification with the more prototypic surface crack geometry, James (1997d) conducted two experiments employing semi-elliptical surface cracks in high-sulfur A302-B steels. In one case, the surface crack penetrated only the high-sulfur steel, while in the other case the crack penetrated weld-deposited cladding into the underlying high-sulfur steel. In the first case of the monolithic steel specimen, EAC initiated locally at the deepest penetration of the crack ($a = 13.82$ mm, $2c = 30.48$ mm) when $\bar{V}/V_{in} = 1.73$ and $\Delta a/\Delta a_{cr} = 3.0$. This shows that the EAC Initiation Criterion was

conservative in this case because V_{in} is the minimum average crack velocity at which EAC initiation has been observed. In the case of the second experiment where the semi-elliptical crack ($a = 15.78$ mm, $2c = 30.28$ mm) penetrated the cladding into the steel, EAC did not initiate when $\bar{V}/V_{in} = 1.08$, and $\Delta a/\Delta a_{cr} = 1.6$; again, a conservative result. Note also that the crack depths in the two surface-crack experiments were significantly different than the 2.54 mm (0.10 in.) used in the criterion development.

All of the above examples of EAC initiation were for high-sulfur A302-B steel tested in hydrogenated low-oxygen water at 243°C (470°F). Other examples of EAC initiation may be found in the literature, and these will be discussed below. For example, Figure 3 provides an illustration of two experiments (one on a 1T-CT, and one on a 2T-CT specimen) conducted on a high-sulfur A302-B steel in hydrogenated low-oxygen water at 149°C (300°F) by James (1994a). In both experiments, EAC was turned on from a non-EAC state by increasing the crack velocity (by frequency changes and/or ΔK changes) above a critical value. Also, in both experiments, EAC was turned off when the crack velocity was reduced (again by frequency and/or ΔK changes) below the same critical value: about 1.0×10^{-6} mm/s (3.94×10^{-8} in./s). The subject of EAC cessation will be discussed in greater detail later. Note that EAC behavior at 149°C (300°F) does not exhibit the same "plateau" or "upper slope" behavior as EAC at higher temperatures. The lack of a "plateau" at 149°-150°C can be observed in the results of several studies (e.g., Cullen *et al.*, 1983; Tice *et al.*, 1993; James, 1994a; Auten *et al.*, 1993, 1995).

Additional examples of EAC initiation (and cessation) at a critical velocity were presented by Combrade *et al.* (1988) as the basis for their proposal for a critical time-based crack growth rate for EAC initiation. They employed a high-sulfur ($\approx 0.013\%$ S) ASTM A508-3 forging (UNS K12042) in a PWR environment at 288°C (550°F). The experiments were conducted at a constant $\Delta K = 40$ MPa $\sqrt{m}$ (36.4 ksi $\sqrt{in.}$), and different crack velocities were achieved by cyclic frequency changes. The results are plotted in Figure 4. Combrade *et al.* employed different waveforms, both symmetric and asymmetric, as well as different frequencies, and these were not completely specified in their paper. Therefore, it was not possible to convert their results into crack velocities as defined herein, and the plot of Figure 4 employs $\dot{a}_p$ as the abscissa, and $\dot{a}_c$ as the ordinate. Nevertheless, the values shown in Figure 4 are probably within a factor of two of actual crack velocities, and they clearly show that EAC initiation and cessation occur at a constant value of $\dot{a}_c \approx 6.8 \times 10^{-6}$ mm/s (2.7×10^{-7} in./s).

VanDerSluys and Emanuelson (1993) conducted an experiment wherein the cyclic frequency was switched from 0.0005 Hz to 0.0001 Hz, and subsequently back to 0.0005 Hz. The experiment

was conducted on a high-sulfur ($S = 0.013\%$) ASTM A533-B steel (UNS K12539) in a PWR environment at 288°C (550°F). The results, given in Figure 5, show that EAC was observed in the first and third test phases at 0.0005 Hz , but not in the second test phase at 0.0001 Hz . Note, however, that there was a 208 hour delay into the second test phase before EAC turned off, and a 282 hour delay into the third test phase before EAC re-initiated. These delay periods are quite consistent with the transient diffusion mass-transport theories of Wire *et al.* (1996a, 1997a, 1997b). The average crack velocity during the first test phase when EAC was on was $\bar{V} = 5.5 \times 10^{-6}\text{ mm/s}$ ($2.17 \times 10^{-7}\text{ in./s}$). When the frequency was changed to 0.0001 Hz at the start of the second test phase, the mean velocity became $\bar{V} = 1.1 \times 10^{-6}\text{ mm/s}$ ($4.33 \times 10^{-8}\text{ in./s}$) with EAC still temporarily on. However, this new crack velocity was now lower than the EAC initiation velocity, V_{in} , and EAC ceased after about 208 hours; i.e., $V_{in} \geq 1.1 \times 10^{-6}\text{ mm/s}$. Since the rate of sulfide loss through mass transport was now greater than the rate of sulfide supply through crack growth, EAC was now unsustainable and the 208 hour delay was the time necessary for diffusional mass transport to remove sufficient sulfides from the crack tip to result in EAC cessation. In a similar fashion, the crack tip was relatively free of sulfides during the non-EAC portion of the second test phase at 0.0001 Hz . The mean crack velocity during the non-EAC portion of the second test phase was $\bar{V} = 2.82 \times 10^{-7}\text{ mm/s}$ ($1.11 \times 10^{-8}\text{ in./s}$), and this became $\bar{V} = 1.41 \times 10^{-6}\text{ mm/s}$ ($5.55 \times 10^{-8}\text{ in./s}$) when the cyclic frequency was increased to 0.0005 Hz at the start of the third test phase. This new higher crack velocity was now greater than V_{in} and, after a delay of 282 hours, EAC initiated; i.e., $V_{in} \leq 1.41 \times 10^{-6}\text{ mm/s}$. The delay period was that time necessary for the crack-tip sulfide concentration to build up to the critical level to initiate EAC. It is therefore clear that, for the VanDerSluys and Emanuelson experiment, $1.10 \times 10^{-6} \leq V_{in} \leq 1.41 \times 10^{-6}\text{ mm/s}$ ($4.33 \times 10^{-8} \leq V_{in} \leq 5.55 \times 10^{-8}\text{ in./s}$). Note also in Figure 1 that the Δa from the start of the third test phase to the initiation of EAC exceeded the $\Delta a_{cr} = 0.3\text{ mm}$ (0.012 in.) requirement.

Although many hundreds of CFCP/EAC experiments have been conducted over the years and input into the EPRI Database for Environmentally Assisted Cracking (EDEAC, Mindlin, *et al.*, 1986), very few experiments have been conducted with the express objective of studying the kinetics of EAC initiation; i.e., starting with a crack that is relatively "clean" of sulfides, and systematically changing the crack velocity to initiate EAC. Therefore, the experiments discussed above represent those conducted with EAC initiation as the test objective. These experiments are summarized in Table I. It will be noted in Table I that, in general, there is relatively good agreement between the critical EAC initiation velocities observed in the different studies. The two largest parameter variables between the experiments in Table I are steel sulfur content and test temperature. Note that the lowest value of V_{in} listed in Table I is for the steel with the highest

sulfur content, A302-B plate, while the highest initiation velocity was for an A508-3 forging with roughly half the sulfur content; James (1995a) points out that forgings are often less susceptible to EAC than plate steels with the same sulfur content. It is expected that V_{in} should be a function of sulfur content, and should increase with decreasing sulfur levels. That is because the sulfide supply rate to the crack tip is proportional to the crack extension rate per unit time (velocity) times the volume fraction of sulfides in the steel. Therefore, for steels of lower sulfur content (i.e., lower volume fraction or area fraction), it is necessary to drive the crack at a higher velocity in order to achieve the critical sulfide concentration at the crack tip for EAC initiation (Wire and James, 1996b). In addition, forgings often have lower sulfide area fractions and mean sulfide sizes than plates of the same sulfur content, and this also decreases EAC susceptibility (Wire et al., 1996b, 1997b). Therefore, the value of $V_{in} = 5.0 \times 10^{-7}$ mm/s for a 2.54 mm deep crack listed in Table I, is the minimum observed in this very high sulfur steel, and use of this value will be increasingly conservative as steel sulfur contents decrease from the 0.024%-0.035% level.

CESSATION OF EAC FOLLOWING THE BREAKTHROUGH OF EMBEDDED CRACKS

Three examples of the cessation of EAC have already been discussed in the foregoing discussion of EAC initiation, and it is clear that EAC will be unsustainable and cease if the mean crack velocity ($\bar{V}$), including EAC effects, falls below the EAC Initiation Velocity, V_{in} (see Figures 3-5). However, this phenomenon requires further discussion because of the important scenario of a relatively large embedded crack in a high-sulfur steel breaking through to the wetted surface sometime during service. In such a scenario, the embedded crack could have a relatively large number of undissolved metallurgical sulfide inclusions exposed on the crack flanks, and the introduction of water could produce the same result as the previously-discussed pre-cracking of specimens in air: the initially high crack-tip sulfide concentration could predispose the crack toward exhibiting EAC. Wire (1997a) has addressed this possibility and has performed transient diffusion calculations to show that as long as the crack velocity (including EAC effects) remains below V_{in} , any EAC following breakthrough will be transitory, and longer-term crack growth will be at the non-EAC level. In a related experimental study, Li (1997) has conducted several experiments on high-sulfur steels with cracks of differing depths, and demonstrated EAC cessation when $\bar{V} < V_{in}$. This section will review the various analyses and experiments that support the concept of EAC cessation.

Figures 3, 4, and 5 all demonstrate that an active case of EAC will cease if the mean crack velocity drops below the EAC initiation velocity. As shown in Figure 5, relatively short delay

times* may accompany the transition from a state of EAC to a non-EAC state. However, the steady-state condition will remain at a non-EAC state so long as mean crack velocities are not increased above V_{in} .

There have been a few cases where EAC has been rather persistent, yet EAC cessation did occur. An example is an experiment conducted by James (1994) on a high-sulfur A302-B steel, and shown in Figure 6. This experiment, conducted at constant $\Delta K = 16.49 \text{ MPa}\sqrt{\text{m}}$ (15 ksi $\sqrt{\text{in}}$), started at a cyclic frequency of 1 Hz, and progressed successively to continuous cycling frequencies of 0.1, 0.01, 0.001, 0.0005, and 0.0001 Hz; i.e., the experiment started in the upper right-hand corner of Figure 6, and progressed toward the lower left-hand corner. Consequently, about 1000 hours were spent at very high crack velocities and/or EAC conditions. This is a loading history that is unlikely to occur in service, but it undoubtedly produced a crack-tip sulfide concentration at the solubility limit. Wire (1997a) has shown that, under such conditions, EAC cessation can be delayed (but not stopped) by the dissolution kinetics of exposed metallurgical sulfides within the crack enclave; i.e., additional sulfide inclusions cannot dissolve until diffusion mass transport has lowered the sulfide concentration below the solubility limit. Figure 7 illustrates that EAC cessation was indeed in progress during the last phase of testing at 0.0001 Hz. Testing was terminated before total cessation had occurred, but it is clear from Figures 6 and 7 that crack velocities were indeed diminishing. Note in Figure 7 that a delay time of about 460 hours was observed before crack growth rates started to decrease. This experiment was conducted on a "2T-CT" specimen ($a_{\text{notch}} \approx 30.48 \text{ mm}$), and the crack had a mass-transport depth (as measured from the notch) of about 33.45 mm at the start of the 0.001 Hz test phase. Therefore, for a mass-transport crack depth of 33.54 mm, the minimum EAC initiation velocity would be expected to be

$$V_{in} = (1.27 \times 10^{-6}) / 33.45 = 3.80 \times 10^{-8} \text{ mm/s}$$

The mean crack velocity during the last half of the 0.0001 Hz test phase was $\bar{V} = 4.06 \times 10^{-8} \text{ mm/s}$, a value slightly above $V_{in} = 3.80 \times 10^{-8} \text{ mm/s}$, yet EAC cessation was already in progress. This again illustrates that V_{in} is the minimum value for EAC initiation, and hence is conservative.

Li (1997) conducted a series of experiments designed to simulate the breakthrough of an embedded crack and the subsequent EAC cessation. "1T-CT" specimens of a high-sulfur A302-B steel were precracked in air to a series of different depths beyond the notch: 3.48, 4.65, 7.15,

* A few hundred hours of delay time is probably not significant when compared to the total life of a plant.

10.85, and 11.42 mm^{**}. Fatigue cycling commenced as soon as the autoclave temperature and water chemistry had equilibrated; generally within about a day. Hence, the initial crack-tip sulfide concentrations were likely to be elevated. In this respect, these experiments were little different from the vast majority of CFCP/EAC experiments previously conducted on low-alloy steels. However, Li employed a one-hour dwell period ($T_D = 3600$ s) between each active fatigue cycle. Such loadings are more realistic than continuous cycling, and have the effect that the mean crack velocity is reduced significantly. An example of such an experiment is given in Figure 8. In this case, the initial mass-transport crack depth was 4.65 mm (as measured from the notch). Hence, EAC would be expected to initiate at

$$V_{in} = (1.27 \times 10^{-6}) / 4.65 = 2.73 \times 10^{-7} \text{ mm/s}$$

As shown in Figure 8, accelerated crack growth rates were observed in the early portion of the experiment, and the mean crack velocity observed was $\bar{V} = 1.86 \times 10^{-7}$ mm/s. However, because $\bar{V} \leq V_{in}$, the accelerated cracking was not sustainable, and after about 283 hours the EAC ceased. Subsequent CFCP growth rates were comparable to the non-EAC behavior. Note that if the continuation of EAC had been predicted on the basis of the time-based crack growth rate, $\dot{a}_b$, elevated crack growth rates would have been predicted to continue indefinitely. Using $T_R = 42.5$ s, $\dot{a}_b$ would be predicted to be 9.90×10^{-7} mm/s, a value well within the EAC regime on a time-domain plot, but over two orders of magnitude higher than the $\bar{V} = 6.96 \times 10^{-9}$ mm/s observed in the latter stages of this experiment.

The other EAC cessation experiments presented by Li (1997) had similar outcomes to the above example in Figure 8. One of these experiments featured a crack penetrating about 6 mm of weld-deposited cladding into the underlying high-sulfur steel to a total mass-transport crack depth of 11.42 mm. EAC cessation was observed, and the presence of cladding had no apparent effect upon the mass transport of sulfides or the cessation of EAC.

Another example of EAC cessation that can occur when fatigue cycling is interrupted for a period of time is shown in Figure 9. Both EAC initiation and cessation are demonstrated in this experiment that was conducted on a high-sulfur A302-B steel. The initial air precrack (3.20 mm measured from the notch) was "cleaned" of sulfides by soaking under static load for about two weeks prior to commencing active fatigue cycling. The observed mean crack velocity very early in the experiment was $\bar{V} = 6.38 \times 10^{-7}$ mm/s, a value somewhat higher than the predicted initiation velocity $V_{in} = (1.27 \times 10^{-6}) / 3.20 = 3.97 \times 10^{-7}$ mm/s, but only mildly enhanced cracking was

^{**} The total precrack depth of 11.42 mm includes about 6mm of weld-deposited Alloy EN82H (UNS N06082).

observed initially: $\bar{V}/V_{\text{air}} \approx 4$. At an intermediate stage of this first test phase, $\bar{V}/V_{\text{air}}$ had increased to about 8.7, and by the end of the first test phase, EAC had initiated with values of $\bar{V}/V_{\text{air}} \approx 17$. At this point, fatigue cycling was stopped and the crack tip was again "cleaned" of sulfides by soaking for about five weeks under a static load. When fatigue cycling resumed, using a cyclic period twice that of the first test phase, EAC cessation had occurred. Observed mean crack velocities in the second test phase ranged between 1.45×10^{-7} and 2.19×10^{-7} mm/s; values below the predicted mean EAC initiation velocity for this test phase: $V_{\text{in}} = 2.95 \times 10^{-7}$ mm/s. This experiment simulated the scenario where EAC had been active, but fatigue cycling had been interrupted for a period of time (e.g., a plant outage), following which EAC did not resume.

Finally, all of the EAC cessation experiments discussed above were conducted on either "1T-CT" or "2T-CT" test specimens. In order to verify that the results on compact tension specimens represented the more prototypic surface crack, an EAC cessation experiment was conducted on a high-sulfur A302-B steel specimen containing a relatively large semi-elliptical surface crack (see James and Wilson, 1994b, for the specimen design). The initial semi-elliptical crack (pre-cracked in air) was relatively large: crack depth $a = 10.16$ mm, crack length $= 2c = 23.37$ mm. Active fatigue cycling commenced as soon as the autoclave temperature and water chemistry had equilibrated; hence, a relatively high crack-tip sulfide concentration would be expected. The first test phase was conducted at $R = 0.7$ with $T_R = 42.5$ s, $T_F = 7.5$ s, and $T_D = 3600$ s. Because active instrumentation could not be used to infer crack dimensions as the test progressed, a second test phase was employed at a different stress ratio in order to "beach-mark" the fracture face and delineate the first phase crack extension in a post-test examination. The second test phase was conducted at $R = 0.1$, with $T_R = 304$ s, $T_F = 54$ s, and $T_D = 0$ s. The results are shown in Figure 10. The value of V_{in} at the point of deepest crack penetration at the start of the test was predicted to be $V_{\text{in}} = (1.27 \times 10^{-6})/10.16 = 1.25 \times 10^{-7}$ mm/s. The total cyclic period of 3650 s was designed to produce a mean crack velocity below V_{in} , and hence, EAC would be expected to cease after some relatively short period of time (e.g., a few hundred hours based on earlier tests on CT specimens). On the other hand, if the time-based crack growth rate $\dot{a}_b = 2.58 \times 10^{-6}$ mm/s were used as the basis of prediction, EAC would be expected to continue. Predictions of crack extension for mean EAC and mean non-EAC behavior are shown in Figure 10 for both test phases, as well as the pre-test and post-test measured crack depths. It is clear that any accelerated cracking that may have occurred early in the experiment ceased relatively rapidly, and that no significant EAC occurred over the entire testing period of about 2143 hours.

SUMMARY AND CONCLUSIONS

A criterion has been proposed as a Code Case to the ASME B&PV Code to give guidance as to the initiation and cessation of environmentally-assisted cracking of low-alloy steels in PWR environments. This paper provides the technical basis for that proposal, and the results may be summarized below:

- The critical factors that lead to EAC were reviewed, and the consensus position is that a critical concentration of sulfides (probably $\approx 5 \pm 2$ ppm) is required at the crack tip to initiate EAC.
- The crack-tip sulfide concentration is dependent upon the balance between supply of sulfides, and loss of sulfides through any one, or more, of four mass-transport processes: diffusion, ion migration, advection, or convection.
- EAC will initiate from a crack that is initially "clean" of sulfides when the mean crack velocity, $\bar{V}$, exceeds the EAC initiation velocity, V_{in} , for some minimum amount of crack extension Δa_{cr} . Based on experimental data for very high-sulfur A302-B steels, $V_{in} = (1.27 \times 10^{-6})/a$ mm/s, and $\Delta a_{cr} = 0.3$ mm, where "a" is the mass-transport crack depth.
- The EAC Threshold relationship developed by Eason *et al.* (1995) is not employed in the EAC Initiation Criterion. However, the relationships developed for steady-state non-EAC and steady-state EAC by Eason *et al.* are the best available, and are utilized in the EAC Initiation Criterion for cases of steady-state non-EAC and steady-state EAC.
- Pre-existing surface cracks, or those that initiate on the surface in service, will be free of sulfides early in life. Therefore, EAC will initiate from surface cracks only when $\bar{V} \geq V_{in}$ and $\Delta a \geq \Delta a_{cr}$. Satisfying only one of these conditions is not sufficient for EAC initiation.
- If an embedded crack in a high-sulfur steel breaks through to the wetted surface during service, EAC may initiate following the breakthrough. However, the EAC will be only transitory and will cease if the resulting mean crack velocity ($\bar{V}$), including EAC effects, is less than V_{in} .
- The above relationships are based on very high-sulfur steels, and are conservative for that class of steels. Applications to steels with lower sulfur contents will become progressively more conservative as the sulfur content decreases.
- The above relationships are based on diffusion mass-transport processes only. Mass transport by ion migration is thought to have little effect in PWR environments. It is conservative to ignore the beneficial effect of advection and convection mass transport in the formulation of these relationships.

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TABLE I.
SUMMARY OF CRITICAL EAC INITIATION VELOCITIES

<u>INVESTIGATOR</u>	<u>ALLOY</u>	<u>SULFUR CONTENT</u> (% by WEIGHT)	<u>TEMPERATURE</u> (°C)	<u>CRITICAL INITIATION</u> <u>VELOCITY (mm/s)</u>
Wire & Li (1996a)	A302-B	0.024 - 0.035%	243° (470°F)	5.0×10^{-7} (a)
James (1994a)	A302-B	0.024 - 0.027%	149° (300°F)	1.0×10^{-6} (b)
James (1997d)	A302-B	0.024 - 0.027%	243° (470°F)	1.57×10^{-7} (c)
VanDerSluys (1993)	A533-B	0.013%	288° (550°F)	$\sim 1.26 \times 10^{-6}$ (d)
Combrade (1988)	A508-3	0.013-0.014%	288° (550°F)	$\sim 3.4 \times 10^{-6}$ (e)

- a) Velocity shown is for a crack depth of 2.54 mm. Based on eleven experiments shown in Figure 2.
b) Based on two experiments shown in Figure 3.
c) EAC initiated locally at the deepest point of a semi-elliptical surface crack 13.97 mm deep.
d) Mean of 1.10×10^{-6} and 1.41×10^{-6} mm/s as discussed in the text.
e) Assumed that $V = \dot{a}_0/2$ for symmetrical waveforms (e.g., sine, isocles triangle, etc.) as shown in Figure 4.

Figure 1. A typical EAC initiation test result from Wire and Li (1996a). The crack tip is initially free of sulfides, and crack velocities are systematically increased until EAC initiates.

Figure 2. Results from a number of EAC initiation tests from Wire and Li (1996a).

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Figure 3. Demonstration of EAC turning on at crack velocities above, and turning off at velocities below, a critical crack velocity $V_{in} \approx 1.0 \times 10^{-6}$ mm/s. Experiments were conducted on a high-sulfur A302-B steel in hydrogenated low-oxygen water at 149°C (300°F). From James (1994a).

Figure 4. Demonstration of EAC turning on at crack velocities above, and turning off at crack velocities below, a critical time-based environmental rate: $\dot{a}_e \approx 6.8 \times 10^{-6} \text{ mm/s}$. Experiments were conducted on a medium-sulfur A508-3 steel in a PWR environment at 288°C (550°F). From Combrade et al. (1988).

Figure 5. The results of VanDerSluys and Emanuelson (1993) showing the initiation and cessation of EAC in a high-sulfur (0.013% S) A533-B steel tested in a 288°C PWR environment. The test was conducted at a constant $\Delta K = 66 \text{ MPa}\sqrt{\text{m}}$ with $R = 0.1$. Note the delay times involved in the transitions from EAC to non-EAC and back to EAC.

Figure 6. Time-domain plot ($\dot{a}_e = f[\dot{a}_b]$) of crack growth response of a high-sulfur A302-B steel over a wide range of crack velocities at 243°C. From James (1994a).

PVP97X.DRW

Figure 7. Crack length versus cycles plot for the two "Heat A" data points in Figure 6 at the lowest frequencies (two lowest levels of $\dot{a}_b$).

PVP97Z.DRW

Figure 8. Crack length versus cycles plot for an EAC cessation test conducted on a high-sulfur A302-B steel at 243°C. From Li (1997).

YA1S3.DRW

Figure 9. Crack length versus cycles plot for EAC initiation and cessation on a high-sulfur A302-B steel tested at 243°C. EAC cessation occurred after a five week period of fatigue inactivity.

FIG-10.DRW

Figure 10. Crack depth versus cycles plot for an EAC cessation experiment conducted on a surface-cracked high-sulfur A302-B steel at 243°C.