Prediction of Materials Damage History From Stress Corrosion Cracking in Boiling Water Reactors

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Over the past decade, we have developed deterministic models for predicting materials damage due to stress corrosion cracking (SCC) in boiling water reactor (BWR) primary coolant circuits. These steady-state models have been applied to fixed state points of reactor operation to yield electrochemical corrosion potential (ECP) and crack growth rate (CGR) predictions. However, damage is cumulative, so that prediction of the extent of damage at any given time must integrate crack growth rate over the history of the plant. In this paper, we describe the use of the REMAIN code to predict the accumulated damage functions for major components in the coolant circuit of a typical BWR that employs internal coolant pumps. As an example, the effect of relatively small amounts of hydrogen added to the feedwater (e.g., 0.5 ppm) on the development of damage from a 0.197-in. (0.5-cm) intergranular crack located at the exit of an internal pump was analyzed. It is predicted that hydrogen additions to the feedwater will effectively suppress further growth of the crack. We also report the first predictions of the accumulation of damage from SCC for a variable power operating cycle. We predict that the benefits of hydrogen water chemistry (HWC), as indicated by the behavior of a single crack under constant environmental conditions, are significantly muted by changes in reactor power.
[S0094-9930(00)01301-9]

Introduction

Incidences of stress corrosion cracking (SCC) of stainless steel components in the heat transport circuits (HTCs) of boiling water reactors (BWRs) are occurring with increasing frequency as nuclear power reactors age. Particular concern has arisen over the cracking of in-vessel components, such as the core shroud. Of even more concern is the possible cracking of components in the lower plenum, which contains the control rod drives, because of the safety implications and the high cost of repair.

In order to aid in component lifetime and safety evaluations, algorithms are required to predict the development of damage in a deterministic manner. A deterministic model is one that achieves prediction on the basis of known physical laws and specified future conditions. Deterministic methods emphasize mechanisms and their adherence to the natural laws, which provide constraints on the solution of the constitutive equations. Generally, deterministic models require little ''calibration'' beyond that necessary to determine values for poorly known, but physically meaningful, parameters, if they cannot be determined by separate experiment. On the other hand, empirical methods seek to extrapolate past experience into the future, frequently on the basis of models that are nonphysical and that do not adhere to the natural laws. Because empirical models are unconstrained by the natural laws, they require a large calibrating database to capture the relationships between the dependent and independent variables, which must be anticipated in advance.

In the present application, the existing database for SCC in stainless steels under BWR operating conditions does not permit the development of an empirical relationship between crack growth rate and crack length, for example. We argue later in this paper that the CGR/crack length relationship is essential to the prediction of accumulated damage. Importantly, the approach presented here captures the synergism that exists between the mechanical, chemical, and electrochemical factors in the SCC-related degradation of BWR stainless steel components. Examples of crack length tracking and prediction of the accumulated damage over the service life of a hypothetical BWR under constant load operation are presented. The impact of water chemistry on the lifetime of a component is discussed.

The Approach

Over the past decade, a number of water radiolysis codes that predict the concentrations of radiolysis products (principally H_2, O_2, and H_2O_2) around the primary coolant circuits of BWRs and in other aqueous radiolytic systems have been developed [1–8]. In our previous work [4–6,8], we combined a radiolysis code with the deterministic mixed potential model (MPM) [9] to predict ECP and with the deterministic crack growth rate algorithm (coupled environment fracture model, CEFM) [10–12] to form a general code, DAMAGE-PREDICTOR [4–6], in order to predict damage due to SCC in BWR HTCs. These steady-state models have been applied to fixed state points of operation to yield ''snap-shots'' of the developing damage in a differential sense (i.e., in terms of the CGR). However, damage is cumulative, so that prediction of the extent of damage at any given time must integrate the crack growth rate over the history of the plant, which is the innovation reported in this paper.

We emphasize that our approach is largely deterministic in nature, in that the outputs of the component models are constrained by known physicochemical laws [13]. This algorithm has been successfully used in modeling eleven BWRs operating in the US and Europe. Two of these reactors (Duane Arnold and Dresden-2) were included in the original study of Ruiz et al. [3], who showed that the reactors exist at the opposite extremes in the spectrum of reactor response to hydrogen water chemistry (HWC). Indeed, DAMAGE-PREDICTOR was initially calibrated on data from only Dresden-2, by adjusting liquid/steam transfer constants for

Contributed by the Pressure Vessels and Piping Division and presented at the Pressure Vessels and Piping Conference, San Diego, California, July 26–30, 1998, of THE AMERICAN SOCIETY OF MECHANICAL ENGINEERS. Manuscript received by the PVP Division, September 16, 1998; revised manuscript received October 11, 1999. Associate Technical Editor: D. M. Fryer.

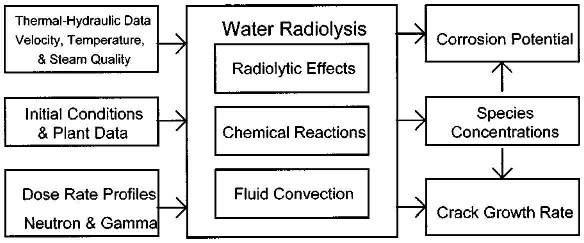

Fig. 1 Structure of the algorithm

H_2 and O_2 and the rate constant for the thermal decomposition of H_2O_2 within physically reasonable bounds or within the ranges of values that had been observed experimentally. The predicted values for $[O_2]$ and $[H_2]$ in the main steam line and in the liquid (recirculation system) phase, and the predicted ECP, as measured in autoclaves connected to the recirculation piping, were in good accord with plant data that had been reported by Ruiz et al. [3] and Indig and Nelson [14]. Additionally, the radiolysis/ECP/crack growth rate model (DAMAGE-PREDICTOR) has been used to model BWRs containing external coolant pumps in ''blind'' mini-tests (e.g., on the KKL BWR in Switzerland [15]), where the plant results (ECP, oxygen concentration) were not known to us in advance. Subsequent comparison of the predicted and measured data has shown that the code provides for accurate simulation of reactor coolant electrochemistry. Importantly, no recalibration of DAMAGE-PREDICTOR has been necessary when simulating reactors across the entire HWC response spectrum.

The code REMAIN that is used in the present work was developed to model BWRs that contain internal main coolant pumps. This code also contains deterministic models for calculating radiolytic species concentrations, ECP, and crack growth rate, but has been optimized for speed of execution, so that calculations may be performed in ''real time.'' Thus, by employing an optimized algorithm for solving simultaneous ordinary differential equations, and by writing the code in C++, that time of execution on a 200 MHz PC, compared with that of DAMAGE-PREDICTOR, has been reduced by a factor of 50 to 100. This enhanced code permits a single state point to be simulated in 1–2 min, thereby making practical iterative calculations over a locus of state points, as reported in this paper.

The main body of the algorithm (Fig. 1) is the water radiolysis model, which yields the concentrations of radiolysis products from the decomposition of water under neutron and gamma irradiation, coupled with homogeneous and heterogeneous chemical reactions, liquid/steam transfer of volatile species (H_2 and O_2), and fluid convection. After the species concentrations have been determined in the whole heat transport circuit under steady-state conditions, the ECP is calculated using an optimized mixed potential model (MPM) and the crack growth rate is estimated using an optimized coupled environment fracture model (CEFM). Auxiliary input parameters, such as flow velocity, coolant temperature, steam quality, and neutron and gamma dose rates in the coolant are obtained from other codes (e.g., thermohydraulic data from RETRAN).

According to the CEFM, crack advance occurs via the slip-dissolution-repassivation mechanism at the crack tip. The dissolution of metal (crack growth) takes place when the oxide layer ruptures, due to elastic and plastic deformation at the crack tip, and a bare metal surface is exposed to the crack environment. Provided that the crack aspect ratio (x/a, where x is the crack length and a is the crack mouth opening displacement) is greater than about 5, which is certainly the case for a long, tortuous intergranular crack in a sensitized stainless steel, particularly when oriented transverse to the fluid flow direction, the crack internal environment is hydrodynamically quiescent and the CEFM can be used to accurately calculate the crack growth rate [16]. The physico-electrochemical condition that must be satisfied by a growing crack is the conservation of charge, which requires that the net positive current exiting the crack mouth be consumed quantitatively on the external surfaces by various cathodic reactions (e.g., reduction of oxygen and hydrogen peroxide). Thus, the CEFM emphasizes that the crack internal and external environments are strongly coupled, a postulate that has been demonstrated unequivocally by experiment [17]. Indeed, the CEFM predicts that modifying the electrocatalytic properties of the external surface may mitigate crack growth. Thus, we have shown theoretically that, if the exchange current densities of the redox reactions (reduction of oxygen and hydrogen peroxide and the oxidation of hydrogen) are increased in the presence of a stoichiometric excess of hydrogen, or are decreased with no restriction on the ratio of hydrogen to oxidizing specie concentrations, the ECP is displaced in the negative direction and the crack growth rate is decreased. The first case is the basis of noble metal-enhanced hydrogen water chemistry (NMEHWC), as achieved by using noble metal coatings on the stainless steel surfaces external to the crack [18–20]. Recent work [21] indicates that this strategy increases the exchange current densities for the hydrogen electrode reaction (HER) and the oxygen electrode reaction (OER) at 288°C by factors of about 20 and 3, respectively, as determined by fitting DAMAGE-PREDICTOR to plant data. NMEHWC is now undergoing trials in operating BWRs. The disadvantages of NMEHWC are the cost of the noble metal, the need to store hydrogen on site, coolant circuit activation due to Co^{60} transport and the formation of N^{16}, and the uncertainty (and unknown negative impact) associated with the introduction of a new material into the HTC on future reactor operation. This latter factor may require that the reactor continue to operate on HWC for the remainder of its life, or at least until a strategy is devised for removing unreactive noble metals from the surfaces. On the other hand, inhibition of the redox reactions that occur on the stainless steel surfaces external to the crack is predicted and found [22] to strongly decrease the crack growth rate, even in the absence of HWC. Inhibition may be achieved by a variety of means, including the deposition of a dielectric coating on the steel surface, as has been recently demonstrated in the laboratory [22].

Crack Growth History and Component Lifetime

The crack length, x_N, over the anticipated service time of a component, T, is obtained by an accumulation of the crack advances over N periods of time $\Delta t_1, \ldots, \Delta t_i, \ldots, \Delta t_N$

$$X_i = X_{i-1} + \mathrm{CGR}_i \cdot \Delta t_i, \quad i = 1, \ldots, N \tag{1}$$

$$T = \sum_{i=1}^{N} \Delta t_i \tag{2}$$

The crack growth rate, CGR_i, is presumed to be time-independent for each interval, Δt_i, but of course it depends on the crack length (through K_I and because of changes in the current and potential distributions in the crack internal and external environments), and hence changes from increment to increment. The initial crack length, X_0, corresponds to the depth of a pre-existing crack (detected during an inspection or assumed for a safety analysis scenario). The algorithm outlined in the foregoing for calculating the accumulated damage is essentially identical to that employed recently [23] to predict the lifetime of disks in low-pressure steam turbines under conditions where thin electrolyte films precipitate on the steel surface. Finally, we note, parenthetically, that the present calculations have been made possible by enhancing the mathematical and computational procedures to achieve reductions in the execution time of the code by a factor of about 100. Thus, a typical simulation involves a value of N ranging from 40 to 100. The nonenhanced DAMAGE-PREDICTOR typically requires 1–2.5 h to execute a single state point on a 200-MHz computer to yield a total simulation time of up to 250 h (i.e., more than 11 d). However, the enhanced code (in this case, REMAIN) performs

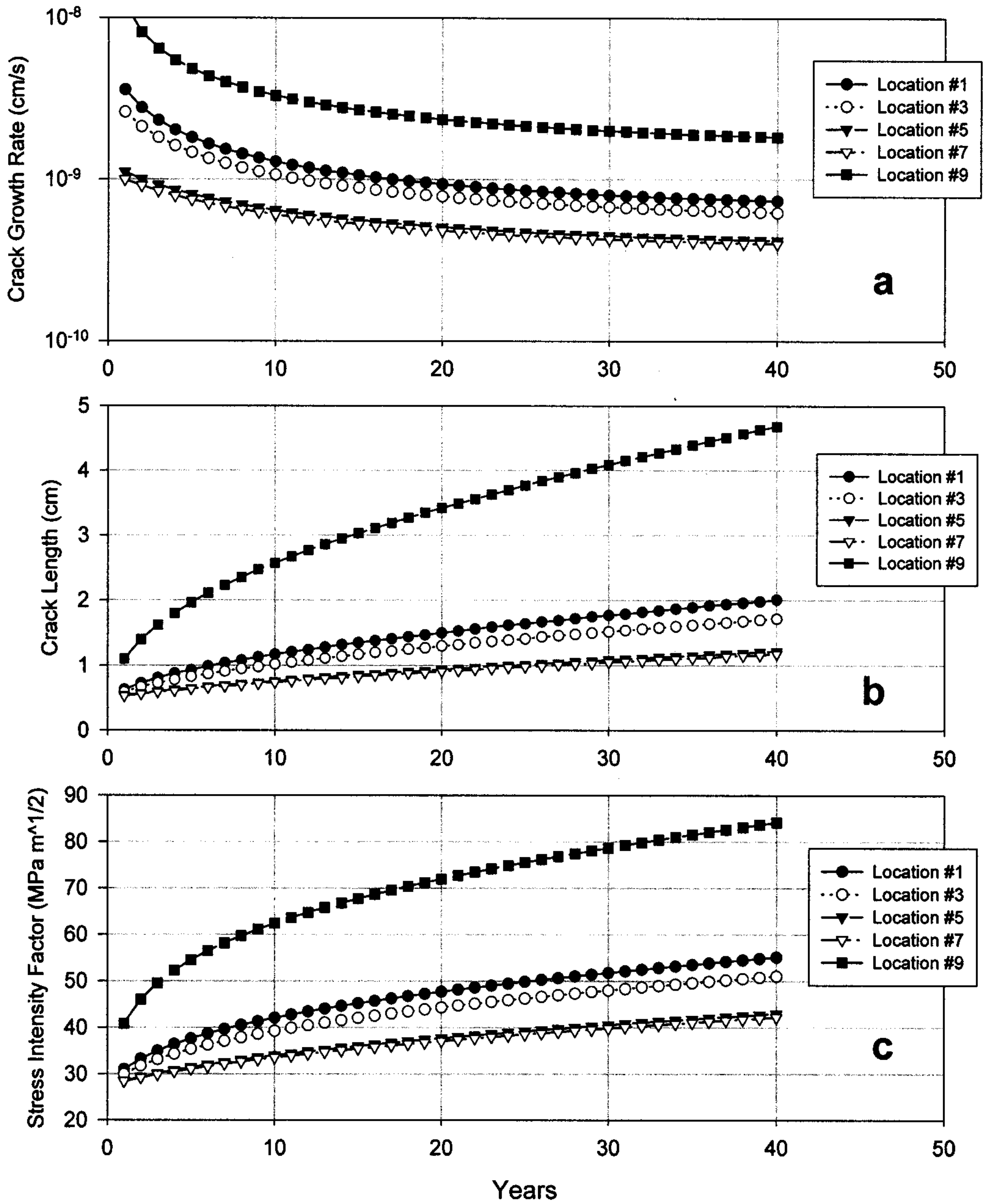

Fig. 2 Predicted histories of growing cracks in the lower plenum of an operating boiling water reactor. Location no. 1 is at the bottom of the internal pump, and location no. 9 is at the bottom (entrance) to the core. Intermediate locations (nos. 3, 5, and 7) represent regions near the bottom of the reactor vessel and at various elevations along control rod drive structures.

the same calculation in about 1.5 min per state point to yield a much reduced, and more practical, simulation time of 1.0 to 2.5 h.

Recognizing that the crack opening displacement, a, and stress intensity factor, K_I, will grow with time as the crack advances, assuming a constant load, one can specify that failure of a component will occur during the ith time interval if the accumulated damage, X_i, exceeds a limiting value, $X_{\lim}$ which is termed the critical dimension, or if the stress intensity, $K_{I,i}$, exceeds the critical value for fast, unstable fracture (K_{IC}, which for stainless steel is 60–65 MPa$\sqrt{\text{m}}$). We refer to these two cases as being ''damage-controlled'' and ''stress-controlled'' failures, respectively. In the demonstration case, the stress intensity is assumed to increase with $X^{1/2}$, short crack effects are ignored, and the crack opening displacement, a, is assumed to be proportional to the length of the growing crack, X (i.e., we assume that the aspect ratio is independent of the crack length). Because the present calculations assume an active, preexisting crack of 0.197 in. (0.5 cm) in length, no account of initiation is incorporated into the model. We note, however, that the theories of passivity breakdown and pit growth are now well developed [16,24], and the transition of pits into cracks was incorporated into recent lifetime estimations for low-pressure steam turbine disks [23].

As noted in the foregoing, the present calculations have been carried out on a hypothetical BWR employing internal coolant pumps operating under both normal water chemistry (NWC) and hydrogen water chemistry (HWC) conditions. The HWC operating conditions assume that 0.5 ppm of hydrogen is continuously injected into the feedwater. The present simulations further assume that the reactor operates continuously for 40 yr at full power. This assumption is, of course, unrealistic, because it ig-

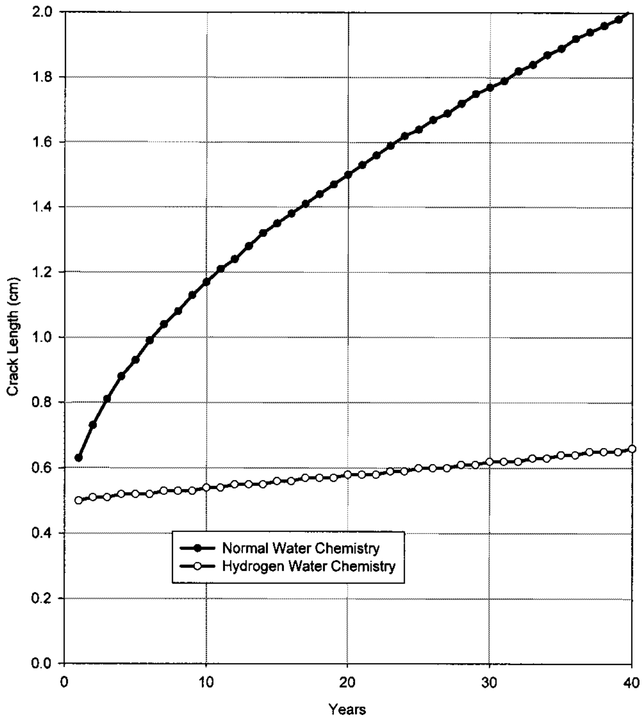

Fig. 3 Predicted effect of HWC (feedwater $[H_2]=0.5$ ppm) on the accumulated damage. The initial value of the crack length (0.5 cm, 0.197 in.) may be considered to be the depth of a detected crack, and the time in years as the remaining (extended) service time after introducing HWC.

nores scheduled fueling outages and unscheduled outages for unanticipated maintenance. Accordingly, the time is better identified (but only approximately so) with the number of effective full power days (40 yr = 14,600 EFPDs). However, scheduled and unscheduled outages are easily incorporated into the calculation, as are changes in power level, stress state, and water chemistry (including changes in conductivity and the implementation of HWC and/or EHWC). The system parameters, which will be published at a later date, were chosen to be generic for a class of reactors, and do not describe any particular plant, simply because we merely wish to illustrate the computational process. Nevertheless, plant-specific parameters are readily incorporated into the simulation to predict the response of SCC damage in any given reactor to future operating scenarios.

The analysis of crack advance and accumulated damage was performed for several locations in the lower plenum of the primary coolant circuit. The first location shown in this analysis (Location no. 1) corresponds to the bottom (exit) of an internal pump, while the last location (no. 9) is at the bottom (entrance) of the core. The intermediate locations (nos. 3, 5, and 7) represent examples of regions near the bottom of the reactor vessel and at various elevations along the control rod drive structures. Because more highly oxidizing conditions, due to radiolytically generated oxygen and hydrogen peroxide, exist closer to the core, the crack growth rate is predicted to vary significantly among the locations chosen for this simulation; Fig. 2(*a*). As a result the damage (crack length) that accumulates over the assumed service time (T is arbitrarily taken as 40 yr) is predicted to vary strongly with position along the lower plenum; Fig. 2(*b*). Once the length of a crack at a given location exceeds the critical value, failure of the component occurs. The lifetime for a component at location m can then be estimated from the point of interception of the $X_m(t)$ function and a horizontal line at $X=X_{\rm lim}$, where $X_{\rm lim}$ is component-dependent critical crack depth. Since crack growth will also alter the stress intensity at the crack tip, as noted in the foregoing, the lifetime of certain components may be controlled, not by the penetration of the crack through a component dimension (e.g., a pipe wall), but by the value of the stress intensity after a given period of service exceeding K_{IC}. In this latter case, the service life is determined by the intercept of $K_{Im}(t)$ and $K_I = K_{IC}$ in Fig. 2(*c*). It is important to note that, although the stress intensity is predicted to increase monotonically with time, the crack growth rate is predicted to decrease over the same period. This is a consequence of the impact of increasing crack length on the current and potential distributions within the internal and external crack environments, as predicted by the CEFM [11]. The impact that increasing crack length has on the crack growth rate is predicted to be a function of conductivity, ECP, flow rate (even for a high aspect ratio crack), and stress intensity [12]. Accordingly, the accumulated damage that is predicted becomes a sensitive function of the operating history of the reactor, in a manner that is unlikely to be captured by empirical models.

The impact that hydrogen water chemistry (HWC) is predicted to have on the accumulated damage is illustrated for one case (Location no. 1) in Fig. 3. Thus, the addition of 0.5 ppm of H_2 to the feedwater, under HWC operating conditions, is predicted to decrease the increment in accumulated damage at the bottom (exit) of an internal pump after 40 yr of continuous operation (or approximately $0.75\times40\times365=10{,}950$ EFPDs, assuming an availability of 75 percent), by a factor of almost ten from $2.0-0.5=1.5$ cm (0.591 in.) to $0.655-0.50=0.155$ cm (0.061 in.). This is a substantial reduction in the extent of damage that could not have been estimated from the crack growth rates at a single state point alone (because of the different, nonlinear dependencies of crack growth rate on time for the two cases shown in Fig. 3) or by using a model that fails to recognize a dependence of crack growth rate on crack length. In this particular case, neither stress-controlled failure (i.e., $K_I>K_{IC}$) nor damage-controlled failure ($X>X_{\rm lim}$) is predicted to occur, provided that $X_{\rm lim}>2$ cm (0.787 in.).

Summary and Conclusions

The concept of accumulated damage (i.e., the damage function) has been introduced for the purpose of analyzing SCC-induced degradation of stainless steel components in the primary heat transport circuits of operating boiling water reactors. In the present work, accumulated damage is estimated by calculating the crack growth rate at closely spaced state points and then integrating over time to yield the crack length. This approach illustrates the synergism that exists between the mechanical, chemical, and electrochemical factors in SCC-related degradation processes that occur in stainless steel components in BWR coolant circuits.

Prediction of the accumulated crack length was performed for several locations within the lower plenum of a generic BWR that has internal coolant pumps, by specifying an initial crack length (corresponding to a detected crack) and a future operating schedule. Examples of component lifetime estimations are presented for both damage-controlled and stress-controlled failures. Furthermore, the impact of hydrogen water chemistry (HWC) on the accumulated damage, due to the growth of a crack at the exit of an internal pump, has been explored. The addition of 0.5 ppm of hydrogen to the reactor feedwater is predicted to reduce the accumulated damage over 40 yr from the point of application of HWC by a factor of almost ten, compared with the damage that would have been incurred under NWC operation.

Nomenclature

Δt_i = ith increment in time
T = service time
CGR_i = crack growth rate for ith time increment
X_i = crack length at end of ith increment in time
K_I = stress intensity
K_{IC} = critical stress intensity

$X_m(t)$ = crack length at location m for time t
$X_{\lim}$ = component-dependent critical crack length
$K_{\lim}(t)$ = component-dependent stress intensity at time t
X_o = initial crack length

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