

# A Study of in-situ Monitoring of the Mechanical and Chemical Degradation of Primary Coolant Pipes in PWR Nuclear Power Plants

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## 1. Introduction

A new technique to detect and measure small cracks using characteristics of reactor environments is set forth in this study. Many corrosion products are produced from general corrosion and are transported throughout the RCS loop. As a result, corrosion products deposited on the nuclear fuels are irradiated by neutrons emitted from fission reactions in the fuel. The irradiation by neutrons converts some corrosion products to radioisotopes, e.g. Ni-58(n,p) Co-58 and Co-59(n,  $\gamma$ )Co-60 reactions. Radioisotopes detached by the shear force of the coolant flow and travel around RCS loops, and deposit on the surfaces of RCS loops. Due to concentration gradients between bulk coolant and internal of crevices or cracks, the radioisotopes will deposit by diffusion much more in crevices and cracks.

## 2. Environmental Qualifications

In this section some of the techniques used to model the detector channel are described. The channel model includes a SiC detector, cable, preamplifier, amplifier, and discriminator models.

### 2.1 The Corrosion Environment and Material Issues

Typical PWR nuclear power plants operate at pressure of 15.5 Mpa and the highest and lowest temperatures are approximately 326°C ( $T_{hot}$ ) and 292°C ( $T_{cold}$ ) respectively. PWRs use borated water in their primary coolant system to control the reactivity of the nuclear core by absorbing the excess neutrons. Borated water can cause significant primary water stress corrosion cracking (PWSCC) in PWR reactor coolant loops.

### 2.2 Radioisotopes in RCS loops

The dominant corrosion products which emit high energy gamma ray are Co-58, Co-60, Fe-59, Mn-54, Mn-56, Cr-51, and Zr-95. Their radiochemical reactions, half-life and the energies of characteristic gamma rays are displayed in Table 1. The soluble or insoluble activated corrosion products redistribute throughout the reactor coolant circuits. These activated corrosion products will be highly concentrated in crevices and cracks in the RCS circuits, as compared to their concentrations outside of cracks.

**Table 1 Insoluble radioisotopes and their radiation reaction and gamma energies [1]**

Nuclide	Radiochemical Reaction	Half life	$\gamma$ - Activity (MeV)
Co-58	Ni-58 (n,p) Co-58	71d	0.810, 0.865, 1.67
Co-60	Co-59 (n, $\gamma$ ) Co-60	5.26y	1.173, 1.332
Fe-59	Fe-58 (n, $\gamma$ ) Fe-59	45d	1.095, 1.292
Mn-54	Fe-54 (n,p) Mn-54	312d	0.835
Mn-56	Fe-56 (n,p) Mn-56	2.6h	0.847, 1.811, 2.110
Cr-51	Cr-50 (n, $\gamma$ ) Cr-51	27.8d	0.320
Zr-95	Zr-94 (n, $\gamma$ ) Zr-95	65d	0.724, 0.726

## 3. Proposed New Technique to Monitor the Mechanical and Chemical Degradation

Radioactive species can diffuse into the deep interior of a crack by the driving force of concentration differences between bulk coolant around the crack mouth and the crack tip. As a result of this diffusion force, the radioactive species concentrations in a crack should be high compared to other intact areas in RCS loops, and it is also the same in the case of radiation intensities emitted from the decay of radioactive species. From the analysis of the radiation spectrum, we can identify the specie atoms and their quantities using the decay equation

$$A = A_0 e^{-\lambda t} \quad (1)$$

Where A is the measured activity at the plant shut down,  $A_0$  is initial activity at operation,  $\lambda$  is decay constant, and t is time. Mass conservation must be obeyed at the crack mouth and we can establish a continuity equation at the crack mouth. The crack depth is obtained from the solution of this continuity equation. The mixed potential model (MPM), the point defect model (PDM), and the coupled environmental fracture model (CEFM) are applied to calculate the potential difference between the crack mouth and the crack tip, which is used to calculate the electrochemical crack growth rate. It is assumed in this study that the total crack length is the summation of the chemical corrosion effect of crack growth and the mechanical crack growth effect.

### 3.1 Measurement of the Radiation Energies and Intensities

Exact concentrations of the radioisotope species in the bulk coolant at the interest area are needed to establish the mass-conservation (continuity) equation at

the crack mouth of interest. The rate of change of a single activated corrosion product in the primary coolant is defined by Mirza et al. as:[1]

$$\frac{dn_w}{dt} = \sigma \phi_c N_w - \left\{ \sum_j \frac{\epsilon_j Q_j g(t)}{V_w} + \sum_k \frac{I_k g(t)}{V_w} + \lambda \right\} n_w + \frac{K_p g(t)}{V_w} n_p + \frac{K_c g(t)}{V_w} n_c \quad (2)$$

where the first term quantifies the generation of radioactive nuclei, the second is for the removal of radioactive nuclei from the purification system, and the third and fourth terms mean that the removal of activated isotopes from deposit on the pipes and cores, respectively.

The concentration of chemical species  $i$ ,  $C_i$ , obeys the following continuity equation,

$$\frac{\partial C_i}{\partial t} = D_i \nabla^2 C_i + z_i U_i \nabla (C_i \nabla \phi) + R_i \quad (3)$$

where  $D_i$  is the effective diffusion coefficient of species  $i$ ,  $z_i$  the charge number,  $\phi$  is the electrostatic potential in the solution,  $R_i$  represents the rate of production or depletion of species  $i$  by chemical reaction and  $U_i$  is the mobility. The potential distribution in the second term in Equation 3 can be obtained from the MPM [2] and the PDM [3]. Once we obtain the potential value in the crack, we can solve Equation 3. The solution of this equation expresses the concentration of species  $i$  in terms of crack depth  $x$  [4].

Now we obtain the concentration profile of species  $i$  along the crack depth. The next step is to integrate the concentration profile with respect to the crack depth,  $x$ , then the crack depth can be expressed as a function of concentration of species  $i$ . Finally the crack depth can be obtained by substituting the concentration of species  $i$  obtained from the radioactive scanning around the welding joint in the RCS pipes. The expected calculation result and concentration profile is shown in Figure 1.

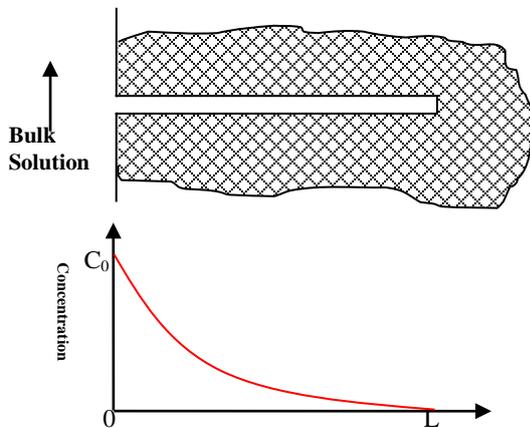


Figure 1 Expected Isotopes profile along crack depth

### 3.2 Modeling of the chemical degradation

The MPM, the PDM, and the CEFM are based on the physical condition that charge conservation must be obeyed at a metal surface when a corrosion process is in progress. The charge conservation constraint is

$$\sum_{j=1}^N i_{R/O,j}(E) + i_{corr}(E) = 0 \quad (4)$$

where  $i_{R/O,j}$  is the partial current density due to the  $j^{\text{th}}$  redox couple in the system and  $i_{corr}$  is the corrosion current density of the material.

For Type 304 SS, the steel oxidation current density,  $i_{corr}$ , was modeled by the point defect model (PDM) which was developed by Macdonald [3]:

$$I_{corr} = \Gamma F \left[ k_2^0 e^{a_2 V} e^{b_2 L} e^{c_2 p H} + k_4^0 e^{a_4 V} e^{c_4 p H} + k_7^0 e^{a_7 V} e^{c_7 p H} \left( \frac{C_{H^+}}{C_{H^+}^0} \right)^n \right] \quad (5)$$

The anodic current density,  $I_{corr}$ , is the integration of the anodic current component extracted from the PDM on the passive film.

The current density ( $i_{R/O}$ ) for a redox couple (e.g.  $O_2/H_2O$ ,  $H^+/H_2$ ,  $H_2O_2/H_2O$ ) [5]



(where  $R$  is the reduced species and  $O$  is the oxidized species) can be expressed in terms of a generalized Butler-Volmer equation as

$$i_{R/O} = \frac{e^{(E-E_{R/O}^e)/b_a} - e^{-(E-E_{R/O}^e)/b_c}}{\frac{1}{i_{0,R/O}} + \frac{1}{i_{l,f}} e^{(E-E_{R/O}^e)/b_a} - \frac{1}{i_{l,r}} e^{-(E-E_{R/O}^e)/b_c}} \quad (7)$$

where  $i_{0,R/O}$  is the exchange current density,  $i_{l,f}$  and  $i_{l,r}$  are the mass-transfer limited currents for the forward and reverse directions of the redox reaction, respectively, and  $b_a$  and  $b_c$  are the anodic and cathodic tafel constants.  $E_{R/O}^e$  is the equilibrium potential as computed from the Nernst equation:

$$E_{R/O}^e = E_{R/O}^0 - \frac{2.303RT}{nF} \log \left( \frac{a_R}{a_O} \right) \quad (8)$$

where  $a_R$  and  $a_O$  are the thermodynamic activities of  $R$  and  $O$ , respectively, and  $E_{R/O}^0$  is the standard potential, which is readily calculated from the change in standard Gibbs energy for the cell reaction ( $\Delta G_{R/O}^0$ );  $E_{R/O}^0 = -\Delta G_{R/O}^0 / nF$ .

The crack growth rate (CGR) is calculated using Faraday's law:

$$\frac{dL}{dt} = \frac{MI_{corr}}{\rho_m zFA_{ct}} \quad (9)$$

where  $M$  is the atomic weight of the metal,  $\rho_m$  is the metal density,  $z$  is the oxidation number, and  $F$  is Faraday's constant. The cracks are assumed to grow over the entire service time of a component, therefore the crack depth is obtained by an accumulation of the crack advances over certain time periods,  $\Delta t$

$$x_i = x_{i-1} + CGR_i \cdot \Delta t \quad (10)$$

The calculated crack depth  $x_i$  is a crack growth caused by the purely electrochemical effect during the service time of interest and  $x_{i-1}$  is the crack depth before the accumulation of the crack growth in this operation cycle. Here this electrochemical crack growth is named by electrochemical crack length (ECL) [6].

### 3.3 Modeling of the mechanical degradation

It is recognized that the crack growth rate depends upon crack length, independent of the stress intensity factor [6]. Thus, it is necessary to differentiate between the mechanical crack length (MCL), which, together with the stress, establishes the stress intensity, and the electrochemical crack length (ECL), which partly controls the potential distribution between the crack tip and the external surface [6].

It has been observed experimentally that crack growth rate becomes independent of potential at sufficiently negative ECP value (-230 mV<sub>SHE</sub> recommended by US NRC) [7], and that the fracture morphology changes from intergranular brittle fracture to ductile failure. Wilkinson and Vitek suggested a crack propagation model by the nucleation and growth of cavities in advance of the crack tip. The average CGR is given by [8]:

$$\frac{da}{dt} = cf \quad (11)$$

where  $c$  is the dimension of an event and  $f$  is the microevent frequency. It is expected that this MCL is a part of the total crack depth measured and calculated in this study. As a matter of fact, the total crack depth should be equal to the summation of both MCL and ECL.

## 4. Conclusions and Discussions

The proposed technique is comparatively simple compared to the existing NDE methods, and it is very accurate. Traditional NDE methods depend on human skill to decipher the measured signal. This is why many electric utilities have introduced a performance demonstration (PD) policy to evaluate and enhance the NDE engineer's skills. The radiation scan technique (RST) is a considerably fast test to find possible places

where flaws exist. When this RST is used together with UT and ECT to find flaws, the inspection will be very accurate and it will reduce the inspection time.

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