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# Probabilistic Prediction Model for SCC Initiation Time on Alloy 600 CRDM Nozzle

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#### Abstract

The life of components made of alloy 600 due to PWSCC can be expressed by the sum of initiation and crack propagation time. Since PWSCC initiation mechanism is not fully understood, the time can be determined probabilistically with a Weibull model. Relatively the crack propagation rate is well defined as a function of the stress intensity factor.

From this relation the propagation time can be obtained. A life prediction method has been developed with the model on the crack initiation time. The Weibull model is integrated over the entire surface of a nozzle in order to determine the probability of crack initiation as function of time. To obtain larger number of data, the intra-specimen method is shown to be useful. The size requirement for an intra-specimen is developed to establish the statistical equivalence between intra-specimens. A proof of principle test has been conducted in a sodium tetrathionate solution at 40 °C loaded by the four-point bending method. Results show that the method can be developed through high temperature tests for the application to PWR Control Rod Driving Mechanism (CRDM).

### I. Introduction

H, Coriou revealed the first laboratory occurrence of "Primary (or Pure) Water Stress Corrosion Cracking (PWSCC)" on alloy 600 sheets in 1959, [1] Since 1980's PWSCC has been occurred in many alloy 600 parts, then PWSCC has received world-wide interest from the view points of both basic research and industrial remedy development, In Korea, it was required in regulatory process that the life time of the CRDM nozzles made of alloy 600 must be shown to exceed the plant design life, [2~3]

The prelude to a failure due to PWSCC can be divided into two steps, the crack initiation and the crack propagation. The correlation between the stress intensity factor and the crack propagation rate is well established in contrast to the poor understanding about the crack initiation mechanism. Because the mechanism of a crack initiation is barely understood, it is very difficult to deterministically obtain the crack initiation time. The crack initiation is influenced by various factors and the initiation time has large scatter even in a nominally identical condition. Therefore a new procedure for the probabilistic evaluation of crack initiation time is developed in this work.

### II. Probabilistic Life Prediction for CRDM

### 2.1 The Intra-Specimen

For the probabilistic approach, statistically significant amount of data is necessary to build a quantitative model. Many data points obtained from periodical inspections of operating plants allow for fairly reliable prediction of failures as function of time for PWSCC in PWR steam generator tubes, [4] For CRDM nozzles, however, the number of PWSCC data is limited. Due to its size, laboratory test is usually insufficient for a probabilistic study.

So the method of intra-specimen which can produce multiple failure time data using single specimen was proposed to generate many data. It must be designed to assure the statistical equivalence between intra-specimens.

Because the PWSCC takes place at grain boundaries the normal stress at the most favorably oriented grain boundary in an intra-specimens is dependent on the area under macroscopically uniaxial stress condition. If the uniaxial stress applied for test is  $\sigma$ , the angle between applied stress and grain boundary is  $\theta$ , and the angle between a surface and a grain in a depth direction is  $\varphi$ , the grain opening stress is equal to  $\sigma_{\rm applied} \times \cos \theta \times \cos \varphi$ . Because of the random orientation of grains, the value of  $\cos \theta$  and  $\cos \varphi$  can be taken for random number from zero to one. It is also assumed that a shape of grain is a hexagon and each side is shared by two grains. Therefore for a given grain size, the difference of effective stress is calculated as function of an intra-specimen size, i, e, number of grains,

The grain size of sensitized alloy 600 used for the proof of principle test is determined to be about 27  $\mu$ m, Figure 1 show the result of random number generation [5] on difference in the maximum resolved normal stress at a grain boundaries as function of a width of a square-shaped intra-specimen, If the intra-specimen size is 2mm×2mm, the stress difference is expected to be insignificant,

# 21.1. Area-Compensated Weibull Model

Weibull distribution has been successfully used to model crack initiation probability as function of time, A three parameter Weibull distribution is like following,

$$F(t) = 1 - \exp\left[-\left(\frac{t - t_0}{\theta - t_0}\right)^b\right]$$

where

t = time

 $\theta = \text{characteristic time or scale parameter}$ 

(1)

b = slope or shape parameter

 $t_0 = time delay or initiation time parameter(generally zero)$ 

F = cumulative probability distribution function of failure

A lot of crack initiation data can be obtained using the intra-specimen

method. Since the full specimen with the greater number of grain boundaries has the greater chance to crack initiation, the crack initiation time will be varied with the surface area of a specimen. The area effect is derived in the present work from the weakest link theory of Weibull,  $[6 \sim 7]$ 

The crack initiation probability of a small area element  $\delta A$  at time t is  $\delta \xi$  (t), that can be expressed as follows:

$$\partial \xi(t) = \partial A \int_0^t g(t)dt \tag{2}$$

$$\xi(t) = -\ln[1 - F(t)] = A \int_{0}^{t} g(t)dt$$

$$g(t) = \frac{\xi'(t)}{A}$$
(3)

where g(t) is the probability of cracking in a unit surface area at time between t and t+dt and F(t) is the cumulative probability distribution function (CDF) at time t. The probability of crack initiation, is assumed to increase linearly with area A. Hence,  $\xi(t)$ , g(t) is reduced to be independent of area. It is assumed that the area element  $\delta$  A is very small value, and the CDF probability, F(t), for an area A can be derived using Eq.s (1~2) as follows:

$$F(t) = 1 - \exp\left[-\int_A dA \int_0^t g(t)dt\right] \tag{4}$$

If  $\xi$ (t) can be calculated from a phenomenological model obtained from experimental data fitting function, then g(t) is directly attained from  $\xi$ (t). If the form of function  $\xi$ (t) is taken to be  $\frac{1}{A} \left( \frac{t-t_0}{\theta} \right)^{\delta}$ , the CDF becomes the area compensated Weibull distribution, [8]

The area compensated Weibull distribution can be used to fit crack initiation data of intra-specimens each with area of A<sub>i</sub>, The CDF about a larger area can be expressed in terms of g(t) as it is, From intra-specimens, it is possible to obtain the CDF of any real size component with total area of A, as follows:

$$F(t) = 1 - \exp\left[-\frac{\int_A dA'}{A_i} \left(\frac{t - t_{0_k}}{\theta_i}\right)^{b_k}\right]$$

where

F = cumulative failure probability function(total surface area = A) (5)  $A_i = \text{surface area of an intra} - \text{specimen}$  $t_0, \theta_i, b_i \Rightarrow \text{ the values obtained from intra} - \text{specimens}$ 

$$g(t) = \frac{\xi(t)}{A_i} = \frac{1}{A_i} \left(\frac{t - t_{0i}}{\theta_i}\right)^{b_i} \tag{6}$$

## 2.2 The Probabilistic Initiation Model

It is widely accepted that the crack initiation time is governed by temperature and stress at the surface, as follows:  $[9\sim10]$ 

$$t_I \propto \sigma^{-n} \cdot \exp\left(\frac{Q}{RT}\right)$$
where
 $t_I = crack \ i \ nitiation \ time$ 
 $\sigma = surface \ tensile \ stress$ 
 $n = stress \ exponent(4~5), \ supposed \ to \ be \ 4$ 
 $Q = PWSCC \ acivation \ energy \ for \ initiation \ stage \ (50 \ kcal/mole)$ 
(7)

As the temperature and stress are varied in a CRDM nozzle, it is necessary to modify the crack initiation distribution to take into account the effect of temperature and stress. The shape parameter, b, representing the degree of a scatter of a crack initiation time may be almost independent of environmental variables. The characteristic time,  $\theta$ , corresponding to the time for 63,2% crack initiation probability is assumed to follow Eq. (7).

Therefore the characteristic time may be corrected for local temperature and stress, as a following.

$$\theta(T_{CRDM}, \sigma_{CRDM}) = \left(\frac{\sigma_{EXP}}{\sigma_{CRDM}}\right)^4 \frac{\exp(Q/RT_{CRDM})}{\exp(Q/RT_{EXP})} \theta_{EXP} \tag{8}$$

Since the Weibull distribution is obtained from intra-specimens, the area correction is also necessary.

$$F_{CRDM}(t) = 1 - \exp\left[-\left(\frac{\int_{A_{CRDM}} dA'}{A_i}\right) \left(\frac{t}{\theta_{CRDM}}\right)^b\right] \tag{9}$$

Using Eq. (9), the probability of crack initiation of a CRDM nozzle is obtained from area integration of a CRDM.

# 2.3 Crack propagation model

A crack is assumed to have been initiated where the stress and temperature is the highest according to the Eq. (7). The propagation time from initiation to an allowable limit size can be deterministically calculated by employing a deterministic model, such as P. Scott's model,  $[11 \sim 12]$ 

$$\frac{da}{dt} = 2.8 \times 10^{-12} (K_I - 9)^{1.16} \text{ m/sec} \quad (at 330^{\circ} C)$$
 (10)

Temperature dependence of the crack propagation rate is usually described by an Arrhenius-type Eq. [13];

$$\frac{da}{dt} = 2.56 \times \exp\left(\frac{Q}{RT}\right) \times (K_I - 9)^{1.16}$$
where  $K_I = \text{crack tip stress intensity factor } [MPa\sqrt{m}] \text{ can be}$ 

$$determined \text{ from Newman-Raju's calculatoin}$$

$$Q = 33[\text{kcal/mol-K}]$$
(11)

Then, the crack propagation time, tp, can be determine, as follows:

$$t_{P} = \int_{a_{h}}^{a_{f}} \frac{1}{\left(\frac{da}{dt}\right)} da \tag{12}$$

When the initiation crack size, a<sub>i</sub>, is determined by grain size of a material and the allowable crack size, a<sub>f</sub>, can be specified from regulatory limits such as ASME Section XI.

### III. Proof-of-Principle Experiment

### 3.1 Experiment

A commercial plate with a 1,6 mm thickness is obtained form Metals Technology (USA) in a mill annealed condition. The second material is obtained from the EAC-J round robin test program, [14]

A SCC test with sensitized alloy 600 is accomplished to prove the validity of the intra-specimen method. The specimen was cut into 33 mm  $\times$  170 mm and then sensitized by a treatment at 704 °C for 30 min in a air, [15] The SCC test specimen is applied about 333 MPa with a four-point bending apparatus, (Fig. 1) [16] The stress distribution of specimen is analyzed with both the theoretical model given in ASTM G 39 and a FEM analysis by ANSYS version 5.2, [17]

The size of an intra-specimen is determined by Monte Carlo simulation to contain the variation in the maximum grain boundary stress within 0.1%, as shown in Fig. 2. The number of intra-specimens (2 mm  $\times$  2 mm) in a four-point bending specimen is 30 in a 5 $\times$ 6 rectangular array, as shown in Fig. 1. A 0.1 M solution of sodium tetrathionate (Na<sub>2</sub>S<sub>4</sub>O<sub>6</sub> · 2H<sub>2</sub>O) is used to cause a SCC. [17] The SCC test is conducted in a thermostat at a temperature of 40±0.2 °C. Using a video microscope with a magnification of 350 $\times$  the surface is inspected.

# 3.2. Result of Proof of Principle Test

Seven sets of intra-specimens including two type of specimen (a commercial plate: Figure 4, the EAC-J round robin material: Figure 5) have been tested. For up to 400 min, the data show a tread that follows Weibull statistics well. After 400 minutes the fitted Weibull distribution overestimates the measured failure probability. As an initiated crack grows, the increasing crack opening displacement will reduce local the tensile stress on the surface. This is believed to be the main reason why the failure rate is decreased compared with an early stage. The time shift of a specimen to specimen is rather large. It is attributed to the variation in the material susceptibility among specimens due to relatively short sensitization time.

In order to examine the area effect on the crack initiation time within a specimen, the adjacent  $2 \text{ mm} \times 2 \text{ mm}$  intra-specimens are coalesced into large intra-specimens with 2 times or 3 times the individual area. Crack initiation

data up to 400 minutes are taken to avoid the stress relaxation effect, Supposing a two parameter Weibull distribution, the Eq. (9) can be altered into the linear form of Weibull distribution,

$$\ln \ln \left(\frac{1}{1-F}\right) = b_i \ln t - b_i \ln \theta + \ln \frac{A}{Ai}$$
 (13)

In a case of an extension to larger specimen, the linear form of a cumulative failure probability has the same slope and shifts in direction of y axis according to the area ratio. Weibull distribution parameters are obtained by the least-square fitting of three intra-specimen data sets, i. e.; 2×2 mm, 4×2 mm and 6×2 mm respectively. Their slopes on Weibull plot are found to be almost equal, however, the amount of shift in the characteristic time due to an area alteration is smaller than the prediction by Eq. (13). The amount of graphs is plotted and fitted according to the area ratio so as to find the exact amount of shift of graph. Using this result, we can modify Eq. (9) to correct of the area compensated Weibull distribution according to the area dependence, as follows (Figure 6):

$$F(t) \approx 1 - \exp\left[-\left(\frac{A}{A_i}\right)^{0.63} \times \left(\frac{t}{\theta_i}\right)^{b_i}\right]$$
where
$$F(t) = \text{Cumulative failure probability density function for } A$$

$$A_i = \text{Area of the intra-specimen}$$

$$b_i = \text{Shape parameter from intra-specimens}$$

$$\theta_i = \text{Scale parameter from intra-specimens}$$
(14)

As the empirical exponent can be obtained from experiment data, we may improve the crack initiation model, Eq. (9) can be modified as Eq. (15).

$$F_{CRDM}(t) = 1 - \exp\left[-\left(\frac{\int_{A_{CRDM}} dA'}{A_i}\right)^s \left(\frac{t}{\theta_{CRDM}}\right)^b\right]$$
where s is empirical exponent for the area—compensation (15)

The proof-of-principle experiment showed that the intra-specimen method combined with Eq. (15) can lead to the development of a probabilistic model

for CRDM life prediction, by repeating the experiment in the PWR primary water,

### V. Conclusion

To identify an appropriate probabilistic model, crack initiation time has been measured for a sensitized alloy 600 plate loaded by a four point bending technique in a sodium tetrathionate solution at 40°C. Statistically significant number of data has been acquired using the intra-specimen method that allows for multiple data production using single specimen. The experiment is designed to assure the statistical equivalence between intra-specimens. To make an equivalent condition among intra-specimens, within a specimen, an intra-specimen has a minimum area to include at least a grain boundary aligned normal to the applied stress. It is shown that a two parameter Weibull model can describe the scatter of SCC initiation time that is defined as the time to initiate an intergranular crack, With the validation of intra-specimen method, it can be applied to high temperature test in order to acquire a large number of data, Finally CRDM life is predicted by the sum of crack initiation time and crack propagation time.

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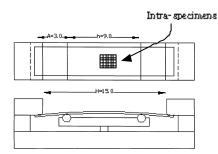


Fig. 1. Four-point bending apparatus for proof of principle test



Fig. 3, SCC test in the thermostat at  $40 \,^{\circ}\mathrm{C}$ 

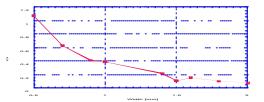


Fig. 2. The Maximum resolved stress(MRS) difference as function of the size of intra-specimen for SNU material (Grain size: ASTM # G = 7.5)

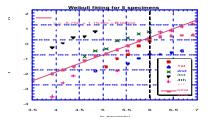


Fig. 4 Test result for sensitized alloy 600 plate tested in 0.1M sodium tetrathionate solution at 40°C

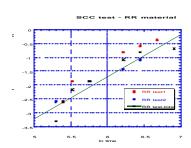


Fig. 5. Test result of EAC-J RR material in 0.001M Sodium Tetrathionate solution at 40°C

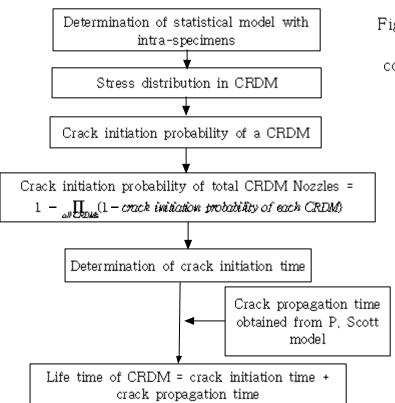
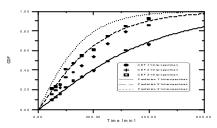
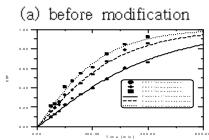


Fig. 7. The Prediction Scheme of Life Time of CRDM Nozzles





(b) after modification Fig. 6. Weibull distribution for variable area considering modified area effect