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Simulation of Pitting Corrosion for Ni-based Alloy Using a Cellular Automata Model

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Abstract: In order to deeply understand the pitting corrosion in Steam Generator (SG) tubing of a pressure water reactor, a Cellular Automata (CA) model was developed to simulate the pitting corrosion. The four basic elements of CA model were determined to simulate the electrochemical reactions, chemical reactions and diffusion processes. The results were qualitatively and quantitatively compared with the experimental and analog data quoted from literatures. Based on the simulation, the shapes of pitting corrosion, the corrosion current density and the effect of Cl^- on pitting corrosion can be obtained, all of which are important for predicting the structural integrity of steam generator tubing.

Key words: SG tubing; electrochemical calculation; pitting corrosion; cellular automata

Pitting corrosion is known as an evolutionary process spanning a wide range of scales from microscopic to macroscopic. Being the link between the micro-scale and macro-scale, the approaches on a mesoscopic scale are suitable for exploring the related properties and characteristics of pitting nucleation. Cellular Automata (CA) approach is exactly such a method which can be used to investigate the connections between microscopic and macroscopic world. Until now, the evolutionary process has been simulated from different point of view by CA approach [1,2]. P. Córdoba-Torres established a systematic quantitative description of corrosion process in terms of balance equations with a CA model in 2001 [3]. J. Stafiej and A. Taleb also contributed to the method for pitting corrosion analysis. They proposed another CA model to simulate the film growth and diffusion during the corrosion process, and also analyzed the mechanism how these processes contribute to the structure of the interface [4,5]. Furthermore, the edge dislocations and the grain size of material on corrosion processes were also considered in the later study [6,7]. L. Lei et al [8,9] developed a CA model to simulate the metastable and stable growth of stainless steel. The result showed that metastable growth of pitting corrosion was likely to be controlled by anodic reaction. R. M. Pidaparti [10,11] proposed a new CA approach to simulate multi-pits corrosion damage initiation and growth in aircraft aluminum materials. It was assumed that the state function was an increasing function with respect to time, because the pitting process was irreversible. In addition, some new algorithms were integrated into the CA model to meet the actual conditions. For example, D. di Caprio[12] obtained some different shapes of pitting corrosion with a stochastic CA model, which could well describe the relation between kinetics and morphologies of the corrosion front. Malki [13] proposed a CA model to study the kinetics behaviors of pit growth by the Monte Carlo (MC) method. Based on the simulation, it was found that the repassivation probability played a major role during the growth process. In general, all of the above studies laid a good foundation for the pitting corrosion simulation with mesoscopic models. However, further investigations are still necessary to get in-depth understanding of the pitting damage evolution. Since there has been little information available in literatures about the simulation of the damage evolution process for SG tubing, relevant studies were carried out in our research aiming to estimate the integrity and predict the

life of SG tubing.

This paper focused on the simulation of pitting nucleation for SG tubing using a CA model. The electrochemical reactions, solution reactions and diffusion process were simulated based on the actual conditions in a Pressure Water Reactor (PWR) to determine the four basic elements of CA model. In addition, the MC method was combined with the CA model to consider the uncertainty of particle behavior. Then the simulated results were qualitatively and quantitatively compared with the those of some closely related experimental and analog data quoted from literatures. Finally, the shapes of pitting corrosion, the corrosion current density and the effect of Cl⁻ to pitting corrosion were obtained.

1 CA Model for Pitting Corrosion of SG Tubing

CA is a dynamical system operating in discrete space-time. The discrete states of cells in a regular lattice are updated in synchrony according to a local rule. Generally, CA is made up of four elements, which form a tuple of (X, V, N, f) [14]. Here X is cellular space, and V is a nonempty finite set of automaton states. Each cell can take on only one state v (v, V) at a given time. N is the neighborhoods of the cell, and f is the state evolution rules. Considering the convenience and symmetry, a two-dimensional (2D) CA is selected. The four elements of CA model for pitting corrosion are specified in the following section.

1.1 Cellular space X

The main cellular space includes metal, oxidation film, the solution in secondary loop, and ions in the solution, as shown in Fig.1.

Alloy 690, which is one of the most commonly used materials for SG tubing, is considered in this model. Ni, Cr and Fe are the main elements in the composition of alloy 690, which make up 98% of the weight of the material. Their atomic diameters are about 0.248, 0.254 and 0.250 nm, respectively. So each site of the CA model is considered as a square with side length of 10 nm, which stands for 40 atoms. In this case, the cellular space contains 2000×2000 cells in all. In the cellular space, each cell is a 2D position vector with the form of $x=(x_1, x_2)$.
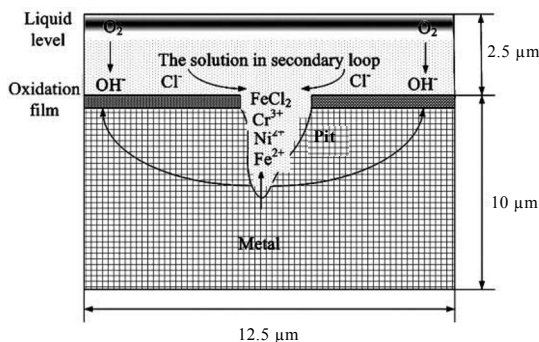

Fig. 1   Cellular space for CA simulation

1.2 Automaton states V

In order to simulate the chemical reactions at mesoscopic level, the sites standing for different ions or atoms were set as below. The sites representing the metal are labeled Me and called metal or Me sites. The passivated regions, labeled P sites, represent the sites covered with a passive layer, which is compact enough to prevent anodic dissolution. The products which are composed of hydroxides, oxides, and water are label M(aq) sites. The other lattice sites, labeled A, B, C and E, denote the acidic regions, basic regions, chloridion and neutral environment, respectively.

1.3 Neighborhoods N

N is the neighborhoods of a cell. The state of each cell depends on the states and configurations of other cells in the neighborhoods n of that cell. In the 2D model, one cell will have 4 neighborhoods at the location of the up, down, left and right. If the cell is expressed as $(0, 0)$, its neighborhoods can be expressed as $(0, 1)$, $(-1, 0)$, $(1, 0)$ and $(0, -1)$, respectively.

1.4 Evolution rules f

It's known that the CA model is built by dividing the corroding system into cells, in which each one has various possible states at a given time. The conversion among different ions or atoms stands for the different chemical and electrochemical reactions, which can simulate the raw material to be corroded gradually. There are three parts in this system, which are solution, oxidation film, and metal. The evolution rules in the different parts and their interfaces are shown in the coming section.

1.4.1 Evolution rules for metal

The metal evolution rules represent an anodic reaction of metal dissolving at the interior of a pit. In an acidic medium, the anodic dissolution can be described as follows:

$$M + nH^+ = M^{n+} + \frac{n}{2}H_2 \uparrow \tag{1}$$

In a neutral medium, the anodic dissolution can be described by the follows reaction:

$$4M + nO_2 + 2nH_2O = 4M^{n+} + 4nOH^{-1} \tag{2}$$
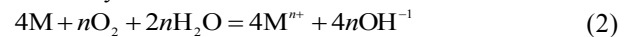
In a basic environment the surface is expected to be passivated. The anodic dissolution can be described by the reaction as follows:

$$M + OH^- = MOH_{ads} + e^- \tag{3}$$

The Me site will not change if the metal site is not in contact with the corrosive electrolyte. In this case, the metal maintains its original characteristics and does not change.

The Me site will change if the metal site is in contact with the corrosive electrolyte, which stands for anode reaction of corrosion in the interface. There are two cases for the transition: one is M(aq) site, the other is P site. The change of Me site into M(aq) site will happen with probability of 1.0 when the number of neighbors A sites are higher than the number of neighbors B sites, or with probability of p_{corr} when the number of neighbors A sites are