Study on a Modelling Method to Predict the Corrosion Product Behavior in PWR Primary System

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1. Introduction

Corrosion products are released from the surface of structure material of the primary circuit, which contacts with the coolant. These corrosion products become mixed with primary coolant due to a continuous interaction of turbulent flow of water with the corroded surfaces of primary coolant circuits. Some of the corrosion products remain in the dissolved state, while the others become insoluble, forming a particle, whereas some materials become deposited on the surfaces of structure. As corrosion products between coolant and system surfaces have been exchanged continuously, the corrosion products are accumulated progressively on the system surfaces. Consequently, the primary coolant system is contaminated and effect on the fuel performance [1].

Many models have been developed for the prediction of corrosion product transport in PWRs. These include empirical or semi empirical models containing empirical coefficients that must be derived from experimental data. In this paper, several important computer codes have been discussed and the modelling method of new model for prediction of corrosion product has been studied.

2. Modeling

2.1 Analysis code for corrosion products

The several analysis codes to predict for corrosion product were modeled in based on the following assumption. First, both radioactive and non-radioactive crud transport depends on the mechanism of particulate removal from the structural materials and both types of crud transport and redeposits on the flow path material of coolant. Second, corrosion products come from the corrosion of stainless steel and Inconel surfaces. Third, these corrosion products deposit on all primary coolant circuit surfaces. These deposits on the in-core surfaces are tightly attached to various core surfaces by adherent form by a sintering process. Forth, some of the loosely adherent deposits on all surfaces are released by the mechanism of erosion [2].

PACTOLE CODE

PACTOLE computer code was initially developed in France, EDF & FRAMATOME. It predicts the time dependent behavior of activated corrosion products. Most of basic physical processes like corrosion product release, erosion, precipitation, dissolution of particulates and particulate transfer are included in it. A series of differential equations involving the corrosion release rate from structural materials; thermophoresis; the activities associated with soluble and particulate corrosion species; the activities of deposits and surface oxides in each zone and corresponding local dose rate are solved in the code [1,3].

CORA CODE

CORA code was developed at the Westinghouse. The crud transport mechanism is modeled semi-empirically between the indicated nodes. Nodes represent homogeneous sources and sinks of the corrosion products in a PWRs primary coolant system. The semiempirical model used in the code is based on a theoretical understanding of the physical processes involved in radiation field build-up it also uses experimental data from plants. Therefore, it is primarily useful for evaluating the effect on radiation level and crud characteristics for system or chemistry changes. Structural materials release corrosion products in soluble forms. Rate of corrosion product release is proportional to solubility. Crud is transported in various parts of primary coolant loop in both particulate and soluble forms. These crud particles are deposited on various structural materials [4,5].

CRUDSIM CODE

CRUDSIM code employs a conceptually simple model compared with the comprehensive models in CORA and PACTOLE. Only two nodes represent the PWRs primary coolant circuit. The radioactive elements considered are ⁵⁸Co and ⁶⁰Co. The crud and activity transport is predicted only on the basis of a solubility difference as a driving force. Therefore, the results of CRUDSIM calculations are used for comparisons of various coolant chemistry conditions. Good comparison with measured plant histories can be obtained using a parametrically selected transport characterization data. CRUDSIM code overlooks the effect of coolant transients on corrosion product activity build-up [1].

Aforementioned all codes consider the radioactive corrosion products released from the reactor core. Because of the main source of radiation exposure for personals carrying our repair and maintenance work on primary circuit components of PWRs. But radioactive corrosion products as well as, non-radioactive corrosion products are important to predict CRUD (chalk-river undefined deposit). The process governing the formation of radioactive corrosion products in the PWR primary system is complex and involves several interacting mechanisms. The new model considers with the several interacting mechanisms such as the uniform and generalized corrosion of base metal, the formation of double-layered oxide, the exchange of species or the release of species.

2.2 Modelling method for new model

2.2.1 Representation of the primary system

In this work, the PWR primary circuit modelling is based on the control volume approach. Therefore, there are divided into a lot of control volume or regions defined according to material property and operating characteristics. The chemical elements taken into account in the work are those composing the alloy materials found in any primary system that generate the corrosion products, as like Iron, Chromium, Nickel.



Figure 1. Control volume in PWR primary system

Figure. 1 shows the control volume, where C is concentration considered by subscripts, k is the control volume number, i is interested chemical species (soluble - Ni, Cr, Fe, particulate - these oxides), and j is the type of media.

We defined a variable mechanism that causes the corrosion product transport e.g. crystallization/ionization, dissolution/deposition, diffusion, formation etc.

2.2.2 Mass balance and transfer mechanisms



Figure 2. Mass transfer mechanisms

Corrosion of the metal leads to the formation of a duplex oxide layer. The formation of this outer layer is partially due to the ions coming from the metal that precipitate after mixing with the ions in the boundary layer. The dissolution of the deposit or the outer oxide layer can occur when the concentration of ions in the coolant is below the equilibrium concentration. The dissolution phenomenon is controlled by the transport kinetics of ions on the wall into the bulk of the coolant (mass transfer coefficient), by the oxide transformation kinetics, and by the concentration gradient. Ions precipitate when their concentration in the coolant is greater than the equilibrium concentration. This occurs near a precipitation site, i.e.: near already existing oxide, and thus near particles but especially near the wall. Ions therefore need to be transported near the wall or the particles before precipitating. The erosion term involves the erosion of particles composing the deposit resulting from the coolant friction forces. The diffusion mechanism occurs between the inner oxide layer and the outer oxide layer, as well as between the outer oxide layer and the deposit. This mechanism follows Fick's law. The convection term describes the transport of particles or ions in the primary coolant through the different primary system control volumes. More specifically, this term is a function of the coolant flow rate [3, 6].

The behavior of the corrosion products in each control volume is modelled by the mass balance equation for each species in each media as follow:

$$\frac{\partial C_{i,j}^k}{\partial t} + \nabla \cdot \overrightarrow{J_i^k} = R_i + S_i$$

where $C_{i,j}^k$ is the concentration of chemical species, i at kth control volume in bulk coolant, $\vec{J_i^k}$ is the material flux between 2 media, R_i^k , S_i^k is the release/sink of chemical species by chemical or physical reaction.

The main driving force for corrosion product transport in the PWR primary coolant comes from coolant temperature change throughout the system and a subsequent change in corrosion product solubility. As a coolant temperature changes around the PWR primary circuit, saturation status of the corrosion products in the coolant also changes such that under-saturation in S/G and super-saturation in the core [1].

2.2.3 Material transport equation

Material transport equation was defined to transport modeling of soluble species as follow [7]:

$$V_k C_k^{bulk} \frac{dC_k^i}{dt} = \frac{\dot{m}}{\rho_{w,k-1}} C_{k-1}^{bulk} x_{k-1}^i - \frac{\dot{m}}{\rho_{w,k}} C_k^{bulk} x_k^i - J_k A_k$$

where x_k^{bulk} is the mole fraction of metal ion in kth control volume, C_k^{bulk} is the total concentration in the bulk, m is the mass flow rate, ρ_w is the water density, A_k is the control volume surface area, J_k is the flux of soluble species to/from the surface.

In this model, simply assume that the concentration of Fe and Ni in the bulk fluid same in all control volume. This is almost correct because of the fast flow rates. Therefore steady state equation about soluble species is

$$\sum J_i^{sol}A = 0$$

where J is

$$J_i^{sol} = C_{bulk} \left[k_i \left(C_{bulk}^{sol} - C_{surf}^{sol} \right) \right]$$

 C_{bulk}^{sol} is the soluble species bulk concentration, C_{surf}^{sol} is the soluble species surface concentration, C_{bulk} is the total concentration in the bulk, k_i is the mass transport rate, i is the soluble species.

The same steady state equation from soluble transport is used again.

$$J_i^{par} = C_{bulk} k_{dep} C_k^{part} - k_{rel} \frac{M_k^{part}}{A_k}$$

where x_k is the particulate mole fraction in k^{th} control volume, C_{bulk} is the total concentration in the bulk, k_{dep} and k_{rel} are deposition and release coefficients, A_k is the control volume surface area, J_k is the molar flux of particulate to surface area A_k of the circuit, M is molar mass of particulate on surface area A_k .

The rate constant is expressed by Arrhenius equation. The Arrhenius equation gives the quantitative basis of the relationship between the activation energy and the reaction rate.

These flux equation can used to calculate the flux of corrosion products between two media when the chemical species have soluble/particulate form in kth control volume. Calculated results can use to predict the behavior of corrosion products. This new model is develop in progress.

3. Conclusions

In this work, various model related with the prediction of corrosion product transport have been reviewed and the modelling method of new model have been suggested. Modelling the behavior prediction of the corrosion products in a PWR primary system will be available in terms of now and future nuclear industry to deal with the important issue such as the process of decontaminations and decommissioning in the primary system. As the result, this method can be applied a methodology to estimate the corrosion product.

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