Multiphysics modeling of CRUD and boron hideout during PWR operations

Seungjin Seo, Nakkyu Chae, Samuel Park, Richard I. Foster, Sungyeol Choi*
Department of Nuclear Engineering, School of Engineering, Seoul National University
**Choisys7@snu.ac.kr*

1. Introduction

Axial Offset Anomaly (AOA) has caused serious risk to economics and safety during the operation of Nuclear Power Plants (NPPs). The AOA has been regarded as the coupled mechanism between Corrosion Related Unidentified Deposit (CRUD) and boron hideout. Observations have suggested that the CRUD was deposited on the upper span of the fuel rod dominantly, where power decrease was significant. Also due to the porous structure with numerous steam chimneys, soluble species such as boron and lithium can be concentrated within the structure, via a wick-boiling structure. Still, it is a challenge to measure the boron hideout within that kind of porous structure in high-temperature and high-pressure conditions, therefore modeling research is required to understand and predict the boron hideout phenomena. In this research, a new model was developed to predict CRUD generation and boron hideout within the CRUD layer, for given operation conditions. Analysis for the saturation behavior for CRUD deposition and erosion was conducted. Also, the relationship between pH driven by chemistry and radiolysis and solubility of lithium metaborates was analyzed. Sensitivity studies for the operation conditions, wick-boiling structure and concentration of corrosion product were conducted.

2. Method

2.1. Generation of CRUD

2.1.1. Deposition and erosion of CRUD

Particulate oxide species of nickel and iron such as Fe3O4, NiFe2O4, and NiO were considered the source of CRUD, with a fixed concentration in bulk coolant. The size of the oxide particles was assumed as a minimum particulates value of 0.25 μm.

The deposition and erosion model was adapted from S. K. Beal's model, which used general particulate deposition and erosion model with revision to match the erosion rate through turbulent flow, as shown in equation (1) [1].

$$\dot{\delta}_{CRUD} = \phi_p \frac{M_p}{\rho_w \varepsilon} - \alpha \delta_{CRUD} \quad (1)$$

where, $\dot{\delta}_{CRUD}$ is growth rate of CRUD (mm/s), δ_{CRUD} is thickness of CRUD (mm), ϕ_p is particulate flux toward surface (mol/mm^{2}s), M_p is molecular weight of particles (g/mol), ρ_w is water density (g/mm^3), ε is porosity, α is erosion coefficient (1/s).

2.1.2. Heat transfer and capillary flow within the CRUD

Heat and mass transfer within the porous CRUD was considered as equation (2) and equation (3), via wick-boiling structure. For heat transfer within the CRUD, thermal conduction with effective CRUD conductivity, convection via capillary flow, and boiling-induced heat sink were considered, with Newton's convection law and Chen's pool boiling boundary conditions [2]. For the capillary flow, Darcy's law for porous liquid flow was applied with a boiling-induced mass sink, with an assumption of perfect removal of vapor via the steam chimney, equation (2) – (3).

$$\nabla\left(\frac{k_{th}^{eff}}{C_v^{eff}}\nabla T\right) + \tau\left(\frac{\kappa}{\mu\varepsilon}\nabla P\right)\nabla \mathrm{T} - \varepsilon\frac{q_{SNB}}{C_v^{eff}} = 0 \quad (2)$$

$$\nabla^2 P - \varepsilon q_{SNB}\frac{\mu}{H_{fg}\rho_w\kappa} = 0 \quad (3)$$

where, k_{th}^{eff} is effective thermal conductivity of CRUD considering coolant and solid composition (W/mmK), C_v^{eff} is effective heat capacity (J/gK), T is temperature (K), τ is tortuosity, κ is permeability (mm^2), μ is viscosity (bar s), P is pressure (bar), q_{SNB} is heat flux caused by Sub-cooled Nucleate Boiling (W/mm^3), and H_{fg} is vaporization enthalpy (J/g).

2.2. Mass transfer and chemistry within CRUD

2.2.1. Mass transfer and chemistry

Time-dependent change of species concentration via diffusion, convection, and chemical reactions was considered as equation (4). The list of chemical reactions of boron and lithium within the CRUD was summarized in Table 1[3]. A radiolysis reactions of water in high temperature and pressure suggested by H. Christensen was used in this model [4].

$$\frac{\partial C}{\partial t} = D\nabla^2 C - v\nabla C + \left(\frac{\partial C}{\partial t}\right)_R \quad (4)$$

where, C is concentration of species (mol/mm^3), D is diffusivity of species (mm^2/s), and v is velocity of capillary flow (mm/s).

Table 1. Chemical reactions of boron and lithium were used in this model.

1	$B(OH)_3 + OH^- \leftrightarrow B(OH)_4^-$
2	$2B(OH)_3 + OH^- \leftrightarrow B_2O(OH)_5^- + H_2O$
3	$3B(OH)_3 + OH^- \leftrightarrow B_3O_3(OH)_4^- + 3H_2O$
4	$3B(OH)_3 \leftrightarrow B_3O_3(OH)_3 + 3H_2O$
5	$Li^+ + H_2O \leftrightarrow LiOH + H^+$

2.2.2. Solubility and precipitation

The solubility of lithium-metaborates was calculated using pH, concentration, and temperature distribution within the CRUD. The precipitation was implemented such that, when the concentrations of boron and lithium exceeds the solubility limit of metaborates, lithium metaborates was precipitated within the CRUD layer. In addition, it was assumed that the precipitation reaction was governed by the mass transfer rate of boron and lithium because it has been reported that the dissolution rate of metaborate is fast enough so it was regarded that all boron precipitation was dissolved out during the shutdown period of PWR.

3. Simulation conditions

3.1. Operation conditions

History of cladding heat flux and distribution of turbulent kinetic energy was from the data of Seabrook plant, Unit #1, Cycle 5 [5]. The coolant temperature within the core was calculated through the heat flux data. Particulate concentration in the plant conditions was suggested by D. J. Walter.

3.2. Numerical approach

MATLAB code via the Finite Differential Method was developed to satisfy the dramatic increase of mesh number for the mass transfer calculation within the thick CRUD layer. The adaptive time step was applied for the time-dependent simulation, and the time step for the CRUD deposition and heat transfer was unit of day order and that of mass transfer and chemistry was unit of seconds because the mass transfer and chemistry required high-order tolerance to be converged.

4. Results and discussion

4.1. CRUD behavior

CRUD behavior was analyzed during three cycles for understanding the refueling stage, and during five cycles for checking saturation behavior for further operation. As shown in Figure 1, the results of CRUD deposition and erosion showed saturation behavior along with an increase in operation times, up to 2 cycles. After the point, the point where CRUD thickness reached to saturation thickness became extended. The saturation thickness was well matched to the suggested thickness of the previous report, lower than 100 μm. Sensitivity studies for the cladding heat flux and wick-boiling parameters such as porosity and chimney density were conducted to validate of these results.

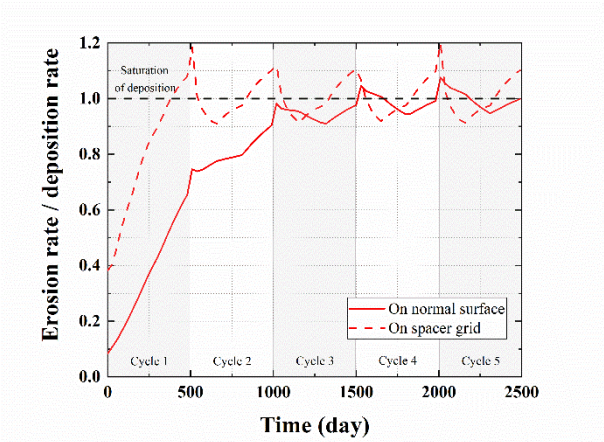

Figure 1. Simulated CRUD behavior in this model during five cycles simulation.

4.2. pH behavior

The pH within the CRUD layer was calculated by considering the radiolysis, water ionization, and boron-lithium chemistry, for the relatively higher temperature region compared to that of bulk coolant. As shown in Figure 2, pH near the cladding increased up to 8.2, where the solubility of lithium metaborate decreased. It was analyzed that the increase in temperature near bulk coolant temperature affected the radiolysis and water ionization to reduce pH, but for the higher temperature region, the increase in temperature leads to an increase in pH due to the behavior of water ionization.

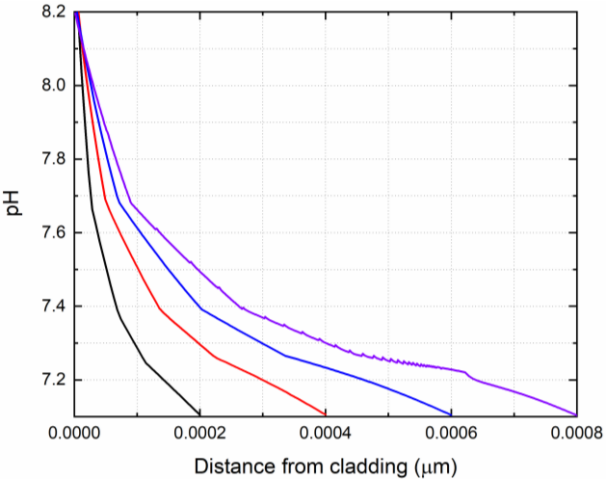

Figure 2. Simulated pH within the CRUD, during one cycle simulation along with CRUD thickness.

4.3. Boron behavior and neutron absorption

An increase in chemical species concentration via wick-boiling structure was observed due to continuous boiling and capillary flow. The concentration factors of the species follow the inverse proportion of their diffusivity. For this result, the ideal wick-boiling structure having 80 % porosity and 5000 chimneys/mm^2 with uniformly distributied boron and lithium concentration factor showed that the concentration of these species can exceed the solubility limit of lithium metaborate.

When the precipitation was governed by the mass transfer rate, the dominant precipitatied species were lithium tetraborate, which has relatively lower solubility than lithium monoborate [13]. As a future work, the accurate precipitation rates for the two species; lithium tetraborate and lithium monoborate will be covered.

5. Summary and future work

A Multiphysics model for CRUD and boron hideout was developed and a 1-cycle simulation was conducted through the model. The behavior of CRUD generation, pH change, boron hideout, and neutron absorption were analyzed. Future work for analyzing several assumptions was necessary for the improvement of the accuracy and detail of the model.

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