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Development of Reactive Transport Model for Analyzing General Corrosion of Spent Nuclear Fuel Disposal Canister under Oxidizing Condition

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1. Introduction

Canister lifetime is the most important parameter affecting safety evaluation because every hazardous radionuclides are contained in disposal canister and release of radionuclides would be started after canister failure. To evaluate canister lifetime, three kinds of corrosion experiment had been conducted and diffusion only corrosion model was developed to analyze corrosion experiment. Several limitations of diffusion only corrosion model listed below has been identified during model verification. The reactive transport model for analyzing general corrosion of copper SNF disposal canister under oxidizing condition has been developing to overcome these limitations.

- Different corrosion behavior of various kinds of Cu
- Dependency of corrosion on temperature and degree of saturation
- Surface reaction like electrochemical reaction, growth of corrosion films and so on
- Reaction in bentonite eg., precipitation, dissolution, adsorption and desorption
- Formation and extinction of corroding agent such as HS^- formation by SRB
- Microbial activities and so on…

2. Methods and Results

2.1 Model

To develop reactive transport model, copper corrosion model for uniform corrosion in sedimentary rock developed by F. King[1] had been selected as a reference and development target model. Major phenomena considered in the King's model are listed below.

- Interfacial electrochemical reactions
- Redox process
- Precipitation and dissolution process
- Sorption and desorption
- Microbial effect
- Mass transport of dissolved and gaseous species

The schematic diagram adopted in King's model is shown in the Figure 1. The species considered in the model and initial conditions are listed in the Table 1. And Table 2 shows boundary condition applied in the model.

The reactive transport model for analyzing general corrosion of copper under oxidizing condition is embodied in COMSOL based on King's general copper corrosion model. Identical assumptions, phenomena, species, boundary conditions, and initial conditions are applied in the reactive transport model. However, constant temperature and degree of saturation are applied. Some input parameters and geometry are different with original King's corrosion model due to rack of data.

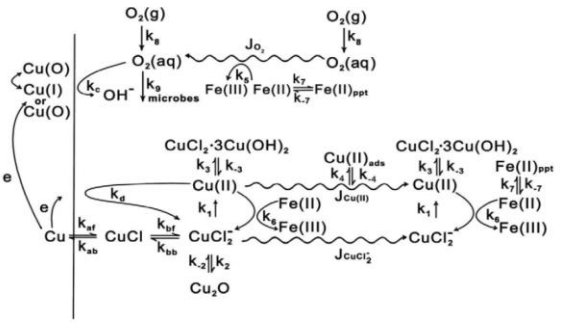

Figure 1. The schematic diagram adopted in King's general copper corrosion model [1]

Table 1. Initial conditions for each of the species included in King's general copper corrosion model [1]

Notation	Species	Initial Condition
c_A	$O_2(g)$	$c_A(x,0) = c_{AL}$
c_0	$O_2(aq)$	$c_0(x,0) = c_{0L}$
c_1	$CuCl_2^-$	$c_1(x,0) = 0$
c_2	Cu_2O	$c_2(x,0) = 0$
c_3	Cu^{2+}	$c_3(x,0) = 0$
c_4	$CuCl_2·3Cu(OH)_2$	$c_4(x,0) = 0$
c_5	Cu(II)(ads)	$c_5(x,0) = 0$
c_6	Cl^-	$c_6(x,0) = c_{6L}$
c_7	Fe(II)(aq)	$c_7(x,0) = c_{7L}$
c_8	Fe(II)(ppt)	$c_8(x,0) = 0$
T	-	$T(x,0) = T_{rock}$

Table 2. Boundary conditions for King's general copper corrosion model [1]

Species	Left-hand Boundary Condition	Right-hand Boundary Condition
$O_2(g)$	Zero flux	Zero flux
$O_2(aq)$	Electrochemical	Constant concentration
$CuCl_2^-$	Electrochemical	Zero concentration
Cu^{2+}	Electrochemical	Zero concentration
Cl^-	Electrochemical	Constant concentration
Fe(II)(aq)	Zero flux	Constant concentration
T	Time dependent	Time dependent

* Non-diffusing species are $Cu_2O(s)$, $CuCl_2.3Cu(OH)_2(s)$, Cu(II)(ads) and Fe(II)(s).

2.2 Results

From COMSOL base reactive transport model developed in this study, various outputs such as corrosion potential and corrosion current density, concentration of each species and flux at time and space, time dependent consumption of the initial inventory of oxygen, and corrosion depth can be obtained. Figure 2 to Figure 5 shows simulation results on corrosion potential, concentration, consumption of oxygen, and corrosion depth respectively. Some of major outputs of King's model can be obtained using this model. However, there were differences between behaviors of each result from King's model and COMSOL model. These differences might be caused by excluded dependency on temperature and degree of saturation and some different input parameters.

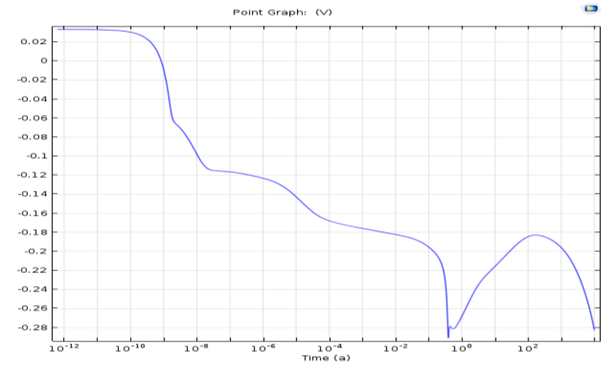

Figure 2. Corrosion potential results from COMSOL reactive transport model

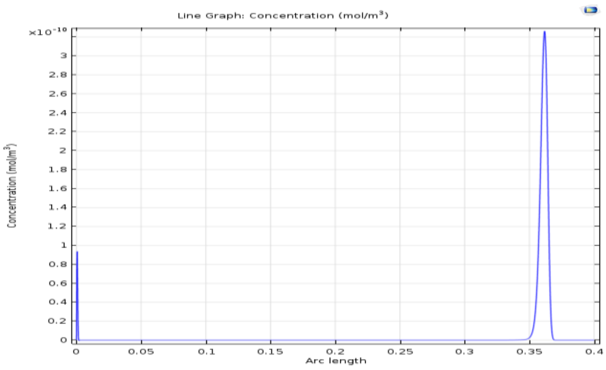

Figure 3. Concentration distribution results from COMSOL reactive transport model

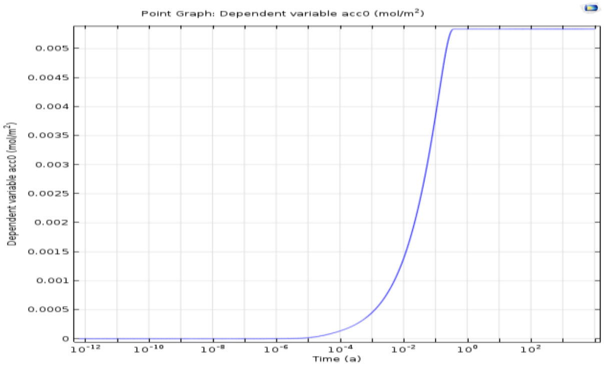

Figure 4. Evaluated consumption of oxygen from COMSOL reactive transport model

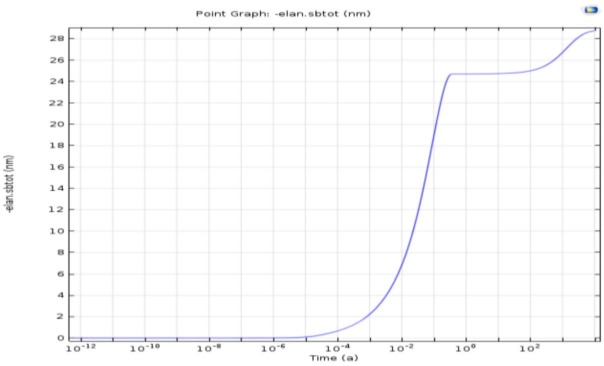

Figure 5. Evaluated corrosion depth using COMSOL reactive transport model

3. Conclusions

Reactive transport model for analyzing general corrosion of copper is developed using COMSOL in this study and major outputs of King's general copper corrosion model can be obtained using developed model. Some limitations of diffusion only corrosion model could be solved using this reactive transport model. Important revisions for some parameters and application of dependencies on temperature and degree of saturation are necessary to analyze corrosion experiment conducted by KAERI. Modification on major phenomena, boundary condition and initial condition and input data must be conducted considering different underground circumstance between Korea and Canada as well.

REFERENCES

[1] Fraser King, Theory Manual for the Copper Corrosion Model for Unifor Corrosion in Sedimentary Rock CCM-UC 1.1, NWMO, 2008