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T2GGM—A COUPLED GAS GENERATION MODEL FOR DEEP GEOLOGIC DISPOSAL OF RADIOACTIVE WASTE

John Avis[1], Paul Suckling[2], Nicola Calder[1], and Robert Walsh[1]

[1] Geofirma Engineering
Ottawa, Ontario, K1R 1A2, Canada
javis@geofirma.com, ncalder@geofirma.com,
rwalsh@geofirma.com

[2] Quintessa Limited
Henley-on-Thames
Oxfordshire, RG9 1AT, U.K.
e-mail: paulsuckling@quintessa.org

ABSTRACT

Deep geologic disposal of radioactive waste is being planned in a number of international programs. Within a deep geologic repository (DGR), gases are generated by corrosion of metals and degradation of organic wastes. Reactions, and thus gas generation rates, are dependent upon pressures, temperature, and the availability of water or water vapor within the repository. Furthermore, many reactions consume water. Consumption rates and repository state are not known a priori and are in fact coupled processes.

A numeric model of coupled gas generation and transport has been developed and implemented in a code (T2GGM). The code consists of a gas generation model (GGM), which calculates rates of gas generation and water consumption within the DGR due to corrosion and microbial degradation of the waste packages, integrated with the widely used two-phase flow code TOUGH2 (Pruess et al, 1999), which models the subsequent two-phase transport of the gas through the repository and into the DGR shafts and geosphere.

T2GGM has been applied to the assessment of gas transport from a proposed low- and intermediate-level radioactive waste (L&ILW) DGR and to study the impact of container corrosion in a hypothetical used fuel DGR.

INTRODUCTION

In Canada, Ontario Power Generation, assisted by the Nuclear Waste Management Organization (NWMO), is currently seeking regulatory approval for construction of a DGR for L&ILW at the site of the Bruce Nuclear Power Plant. The plan involves storing wastes in rooms constructed in extremely low-permeability sedimentary formations. The rooms will not be backfilled to allow for a large void space to minimize possible overpressures created by gas generation. Engineered seals will be emplaced in tunnels and shafts.

NWMO is also conducting evaluations of generic sites for disposal of used fuel. In one scenario under investigation, used fuel would be packaged in carbon steel containers, emplaced in disposal tunnels backfilled with bentonite, and sealed.

In both these programs, numeric modeling work has been undertaken to calculate the generation and buildup of gas in the repository, the exchange of gas and groundwater between the repository and the surrounding rock, and between the rock and the surface environment. The results are used to inform the repository design and postclosure safety assessment modeling.

Initial versions of safety assessments used TOUGH2 with the EOS3 equations of state (Pruess et al, 1999) and a standalone version of GGM. Gas generation rates were calculated based on assumptions regarding repository resaturation and repressurization rates and subsequently input to TOUGH2 as time-varying rates. Analysis of modeling results indicated that the actual repository resaturation and repressurization profiles predicted by TOUGH2 varied considerably from those assumed in developing the gas generation rates. T2GGM was developed to eliminate this discrepancy.

The GGM includes four of the key mechanisms for the generation of gas and consumption of water. Reactions are determined independently in one or more compartments. Each compartment contains a separate inventory, which could range from a panel of disposal rooms, individual rooms, or indeed individual waste containers or portions of waste containers.

Each GGM compartment comprises one or more TOUGH2 elements. At each time step, the integration code calculates compartment average values of pressure, saturation, temperature, and relative humidity, which are passed through to GGM. Gas and water generation rates for the subsequent time step are calculated by GGM and introduced into the TOUGH2 elements as sources. T2GGM data flows are shown in Figure 1.

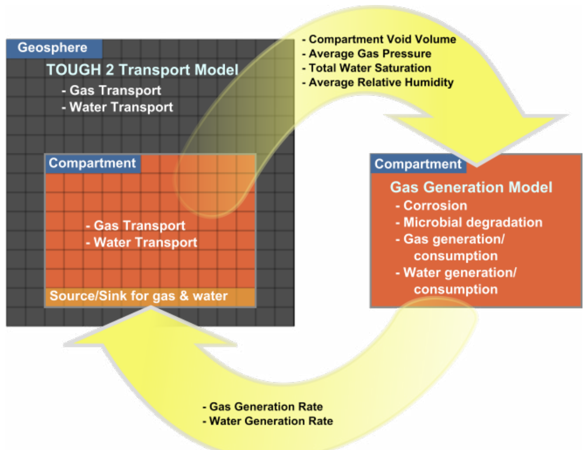

Figure 1. Data flows within the T2GGM code.

GAS GENERATION MODEL

The GGM tracks the production and consumption of the key chemical species (e.g., metals, organic wastes, gases, water) in the repository, and the fluxes of the water and gases into and out of the repository. It is designed to fully conserve Fe, C, and water, and to ensure that reactions are limited by the availability of water. Some elements are conservatively assumed not to be limiting and are not tracked to complete mass balance (e.g., N needed to support microbial reactions).

A simplified version of the complete gas generation model (GGM) is presented here. The full GGM additionally includes: enhanced corrosion of metallic wastes due to high CO_2 partial pressures; oxidation-reduction reactions for the consumption of O_2, NO_3^-, Fe(III), and SO_4^{2-} terminal electron acceptors; and a model for biomass growth, death, and recycling as an additional source of organic waste.

The organic and metallic wastes are classified into a number of waste streams. This allows the degradation/corrosion of each waste stream to be modeled independently and assigned different reaction rates. Organic wastes are classified into three groups: cellulosic wastes, ion-exchange resins, and plastics and rubbers. Metallic wastes are classified into four groups: carbon and galvanised steels, passivated carbon steels, stainless steels and nickel alloys, and zirconium alloys. The GGM includes three key mechanisms for the generation of gas and consumption of water:

- The microbial degradation of organic wastes;
- Methanogenesis via the microbial hydrogen mechanism; and
- The corrosion of metallic wastes.

These processes may occur in either the saturated (water submerged) or vapor phases. Gases are modeled as partitioning between the saturated and vapor phases according to Henry's Law. The relative humidity of the vapor phase is calculated by TOUGH2 and provided as an input to the model. Microbial activity in the vapor phase is dependent on the relative humidity and ceases below a specified threshold (default value of 0.6).

Microbial Degradation of Organic Wastes

Cellulosic wastes are modeled as cellulose that degrades in the presence of water according to the following reaction:

$$C_6H_{10}O_5 + H_2O \rightarrow 3CO_2 + 3CH_4 \quad (1)$$

The limiting reaction rate is that for cellulose hydrolysis. Ion exchange resins, plastic, and rubber are modeled in a similar fashion.

Methanogenesis via the Microbial Hydrogen Mechanism

Significant amounts of hydrogen may be produced via anaerobic corrosion. This can be consumed anywhere in the system via microbial hydrogen oxidation. During the final methanogenic stage, the relevant reaction is:

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O \quad (2)$$

provided that there is sufficient humidity to support microbial processes. The rate for this reaction is modeled as first order with respect to CO_2 concentration, but limited by the availability of hydrogen.

Corrosion of Metallic Wastes

Separate corrosion models are used for L&ILW and used fuel containers (UFC). The L&ILW model assumes isothermal conditions, whereas the UFC model allows for high temperature corrosion of the steel containers.

Corrosion of L&ILW metallic wastes and container materials occurs within the saturated (water submerged) and vapor phases. Corrosion processes in the vapor phase are dependent on the relative humidity, and cease below a threshold of 0.6. Galvanized and carbon steels are treated as a single metallic source, represented by the corrosion of Fe as carbon steel (C-steel). The overall reaction for the anaerobic corrosion of C-steel is given below:

$$3Fe + 4H_2O \rightarrow Fe_3O_4 + 4H_2 \quad (3)$$

The nonisothermal UFC corrosion model includes four phases of corrosion processes, each of which occurs under different conditions (rather than in strict sequence) based on the conceptual model for the corrosion: (1) Dry Air Oxidation, (2) Aerobic Unsaturated Corrosion, (3) Anaerobic Unsaturated Corrosion, and (4) Anaerobic Saturated Corrosion.

Phase 1—Dry Air Oxidation occurs under the initial high temperature, low relative humidity aerobic environment. This is represented by

$$4Fe + 3O_2 \rightarrow 2Fe_2O_3 \quad (4)$$

As the relative humidity increases, Phase 2, Aerobic Unsaturated Corrosion, will start to dominate, which is represented by

$$4Fe + 3O_2 + 2H_2O \rightarrow 4FeOOH \quad (5)$$

Eventually, conditions will become anaerobic, and saturation will remain low in the backfill close to the canister. Phase 3, Anaerobic Unsaturated Corrosion, is represented by Equation (3).

Finally, water may reach the canister. In the presence of water containing carbonates from the surrounding clay, the dominant corrosion mechanism becomes

$$Fe + CO_3^{2-} + 2H_2O \rightarrow FeCO_3 + H_2 + 2OH^- \quad (6)$$

This is referred to as Phase 4, Anaerobic Saturated Corrosion.

Each of these corrosion processes has its own temperature-dependent corrosion-rate expression.

Implementation

GGM consists of first-order coupled differential equations given by the mass-balance equations and nondifferential equations, such as Henry's law. These equations are evolved forward in time using a modified Euler time stepping scheme, and the nondifferential equations are solved for the remaining variables at each time step.

TOUGH2 INTEGRATION

GGM capabilities are invoked by using the GGMIN keyword in the T2GGM input file, typically generated using the mView pre-processor (Avis et al. 2012). Control flags are read and GGM elements and compartment numbers are specified. Connections from GGM compartments to the geosphere are defined, and GGM basic rate constants are specified. Compartment inventories are defined in separate individual input files. A pre-run check confirms consistency of entered data and echoes input to the standard output file.

After each time step is executed, average compartment pressures, saturations, and relative humidity (RH) are calculated. These are passed to GGM, which then calculates gas generation and water consumption for each compartment. Internally, GGM tracks generation and consumption of six gases: H_2, H_2S, CH_4, CO_2, N_2, and O_2. However, TOUGH2/EOS3 uses a single gas, air. Modifications have been made to TOUGH2/EOS3 to use other single gases (currently, H_2, CH_4, CO_2, He, Ne). GGM sums the individual molar generation rates of the six GGM gases to result in a total molar generation rate for each compartment, which is converted to a mass rate for the equivalent number of moles of the single specified TOUGH2 gas.

Simulations were conducted under two GGM scenarios: (1) non-water-limited (NWL), which assumed that there was always sufficient water available to support reactions and water consumed by reactions was not removed from the repository; and (2) water-limited (WL), which enforced a mass balance of water and limited reaction rates to available water inflow rates.

Total compartment gas generation and water consumption rates (for WL simulations) are then allocated to each element in a compartment using one of several available allocation schemes. The rates are incorporated in the TOUGH2 matrix assembly in subroutine MULTI in a manner similar to TOUGH2 subroutine QU.

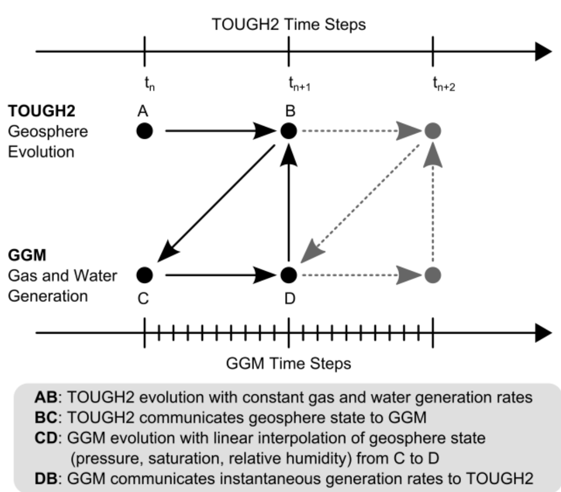

Figure 2. Time-stepping approach

Time stepping for GGM and TOUGH2 are largely decoupled. As shown in Figure 2, GGM will typically take multiple time steps during each TOUGH2 time step. GGM suggests a maximum time step size for TOUGH2 based on rates of change of calculated generation and consumption rates.

APPLICATION TO L&ILW DGR

Detailed gas modeling (Geofirma and Quintessa, 2011) of the proposed DGR was conducted to support regulatory submissions (OPG 2011a, 2011b). The modeling was performed for a number of parameter and conceptual-model sensitivity cases, over a 1 Ma time frame starting at repository closure. All cases were derived from a reference case characterization of the system that assumes a constant present day climate, with no change in boundary conditions during the 1 Ma period.

Data

Most of the data used in the modeling are specific to the DGR system and have been taken from the waste characterization, site characterization, and repository engineering programs. The overall DGR program has been structured such that the safety assessment has been produced in multiple iterations, in synchronization with the inventory, design, and geosciences programs.

The repository is planned to be excavated at a depth of about 680 m in low permeability Ordovician age limestone. Several hundred meters of low permeability shales and limestones separate the repository from the nearest permeable formations above and below. Site characterization results indicate that the Ordovician formations are underpressured and show evidence of the presence of a partial gas phase. Porewater within the host rock is extremely saline, as are groundwaters in the underlying and overlying medium permeability formations.

Capillary pressure and relative permeability data are represented using the modified van Genuchten equations, with parameter values derived from site characterization data for geosphere materials and external sources for shaft-sealing materials. Parameters are adjusted within the excavation damaged zones (EDZ) to reflect increased permeability in these zones.

The inventory of approximately 200,000 m^3 of waste packages includes organic (2.2×10^7 kg) and metallic (6.6×10^7 kg) materials.

Discretization and Property Assignment

The model discretization uses a simplified representation of the repository panels, access tunnels and shaft system. The planned repository will have two shafts (main and ventilation) that are located in close proximity. For modeling purposes, these were combined to form a single shaft with the effective cross-sectional area equivalent to that of the two designed shafts. Access tunnel layout was also simplified and individual emplacement rooms in each panel combined. The resulting model grid consists of 126,000 nodes and 390,000 connections. Figure 3 is an illustration of the model discretization.

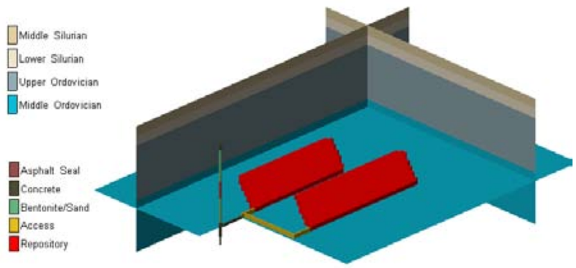

Figure 3. Model discretization showing repository panels, access tunnels, and combined shaft.

The emplacement rooms and repository tunnels are not backfilled, so as to allow void space for expansion of gas. The two waste panels form a single GGM compartment.

<u>Reference Case (NE-RC) Results</u>

The NE-RC case assumes 10% gas saturations within an underpressured Ordovician geosphere, consistent with site characterization results.

The operational phase of the facility was simulated as a 60-year period when shaft and repository pressures were set to atmospheric and gas saturations were set to 100%. At closure, the monolith (emplaced concrete seal within access tunnels and the bottom part of the shafts) and shaft seals are assumed to be placed instantaneously at specified gas saturations.

From this point forward, gas forming from degradation and corrosion reactions starts to pressurize the repository. The first few years involve sustained gas and water consumption due to aerobic degradation. There is a sharp peak in the rate of gas consumption (primarily hydrogen) and water production during the ferric iron and sulphate reduction stages, due to hydrogen oxidation.

The longest sustained period of gas generation occurs during the methanogenic stage up to 4000 years, primarily due to the production of hydrogen through the corrosion of the carbon and galvanized steels. Methane generation via the microbial metabolism of carbon dioxide and hydrogen, and the exhaustion of the metallic wastes, causes a decline in the amount of hydrogen present after this time. The NE-RC case parameters maximize microbial activity and gas generation by assuming that microbes are active if there are organics and water present, regardless of salinity or other factors, and that essentially complete degradation occurs.

Gas generation and water consumption rates both continue to decline as the waste packages become fully degraded. Figure 4 shows predicted rates, while Figure 5 presents the evolution of gas composition within the repository.

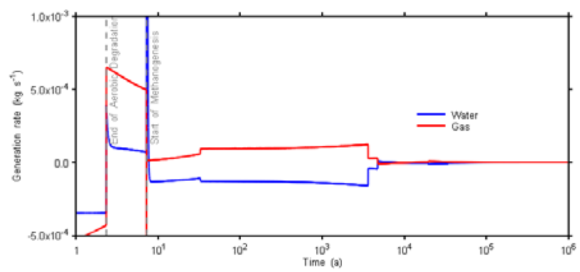

Figure 4. Non-water-limited reference case gas generation and water consumption rates in repository

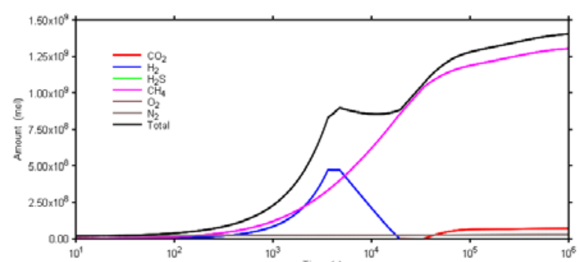

Figure 5. Amounts of gas in the vapor phase within the repository (Non-water-limited reference case)

Pressures rise in the repository as gas is generated. The low permeability of the seal system prevents pressurized gas from traveling up the shaft. The reference case assumes that gas is present in the geosphere, albeit in small amounts due to the low gas saturations (10%) and low porosities of the rock mass. In addition to gas generated within the repository by degradation/corrosion, formation gas flows into the repository and contributes to the increase in pressure. For the NWL scenario, small amounts of formation liquid enter the repository during the period up to approximately 80 ka when pressure in the repository is lower than the liquid pressure in the surrounding geosphere. In the WL scenario, all water initially present in the repository is consumed within 500 years, after which reactions are limited by inflow rates. In both scenarios, any residual water is expelled as pressures in the repository continue to increase. The evolution of repository pressure and saturation is shown in Figure 6. Repository saturation (the fraction of the repository volume that is filled with liquid) peaks at less than 0.01 or 1%. For the WL scenario, a minimum repository liquid saturation (10^{-4}) is enforced for numeric reasons during the period of liquid inflow. At the end of the 1 Ma simulation period, the repository is essentially dry.

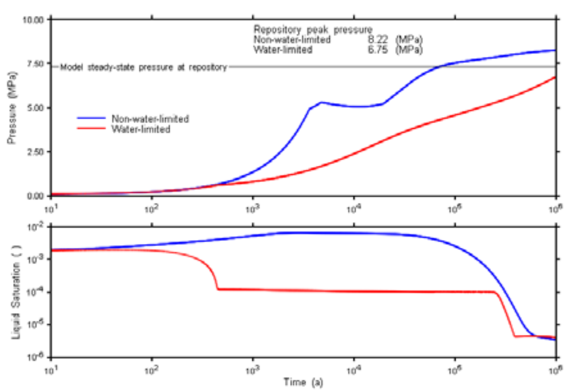

Figure 6. Evolution of reference-case repository pressure and liquid saturation for water-limited and non-water-limited scenarios

Other Cases Results

Figures 7 and 8 present the repository pressure and saturation evolution for all calculation cases for the NWL scenario.

It can be seen that pressures never exceed 10 MPa and are thus well below the estimated lithostatic pressure at the repository horizon (17 MPa). Repository saturations do not exceed 15%, except for cases with no gas generation (NE-NG1 and NE-NG2) and the disruptive event shaft failure cases (SF-BC and SF-ED).

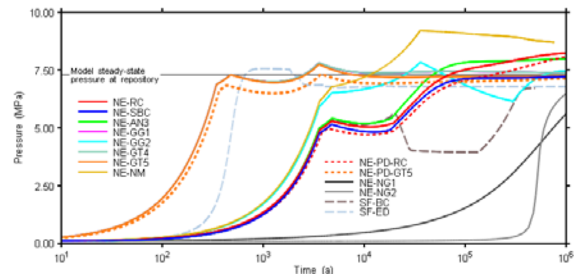

Figure 7. All Cases: Evolution of repository pressure (non-water-limited scenarios)

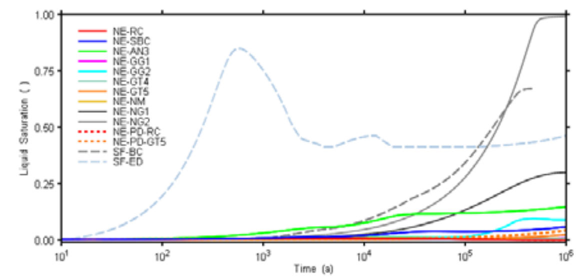

Figure 8. All Cases: Evolution of repository saturation (non-water-limited scenarios)

There are five normal evolution cases where pressures in the repository are sufficient to initiate flow up the shaft. These are high gas generation rate cases (NE-GG1 case or variants NE-GT4, NE-GT5, NE-PD-GT5), or the high gas pressure case (NE-NM). Shaft gas flow for normal evolution cases exits the shaft at the overlying, more permeable formation, with no free gas reaching potable groundwater. Only the shaft seal failure cases have the potential for free gas to directly reach the shallow groundwater.

NWL results were conservative, leading to generally higher repository pressures and saturations. For most WL simulations, the repository remained virtually dry for the majority of the 1 Ma simulation period.

Conclusions

Normal evolution and sensitivity cases indicate that the repository will perform as designed, isolating the waste and generated gases for the 1 Ma performance period.

APPLICATION TO NEAR-FIELD UFC CORROSION

The goal of this ongoing work is to assess room resaturation, gas pressures, and container corrosion for an all-steel UFC in a low-permeability sedimentary host rock. While copper shell UFCs are the reference design, steel UFCs are being evaluated as a design option consistent with some sedimentary rock DGR concepts in Europe. Unlike a UFC with a copper shell, the steel canister will corrode over time, producing hydrogen gas.

Simulations to date have focused on the scale of the UFC/placement room, assessing the near-field response of the geosphere and engineered sealing materials to steel used-fuel-container corrosion and consequent gas generation. Only water-limited scenarios have been assessed. With focus on a single container, the model accounts for nonisothermal behavior, two-phase flow, variable saturation, and variable corrosion. As a conservative simplification for this work, the placement room is assumed to be fully sealed—continuity of the excavation damaged zone (EDZ) beyond the placement room is not considered.

Discretization and Property Assignment

The near-field geometry of this model, called the quarter-container model, is shown in Figure 9.

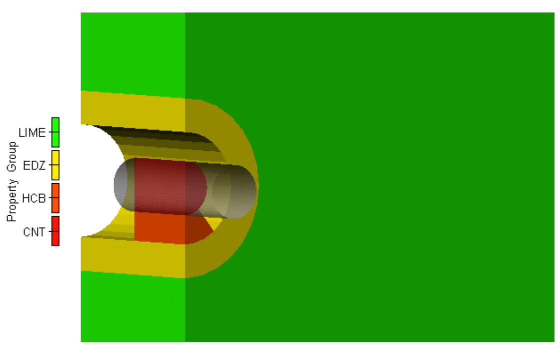

Figure 9. Close-up views of container, EDZ, highly compacted bentonite pedestal, and limestone host rock.

The model extends 500 m above and 1000 m below the repository horizon. Horizontally, the model extends 10 m from the center of the canister, assuming that emplacement drifts are spaced on 20 m centers. The large vertical extents were required to appropriately model heat dissipation over one million years. The reference case assumes that the void space surrounding the canister will be filled with bentonite pellets. A single layer of elements in contact with the container are connected to the GGM, as shown in Figure 10.

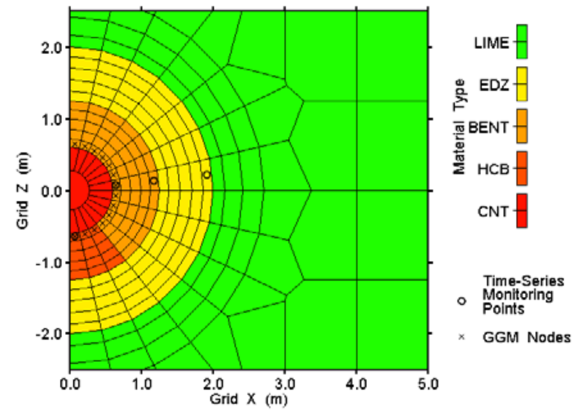

Figure 10. Discretization in container plane.

Preliminary Results

Initial results of the modeling include corrosion, thermal, and saturation response (Figure 11). Although preliminary and conservative, these results provide an example of the value of this coupled approach to modeling long-term degradation of waste canisters in low-permeability sedimentary environments.

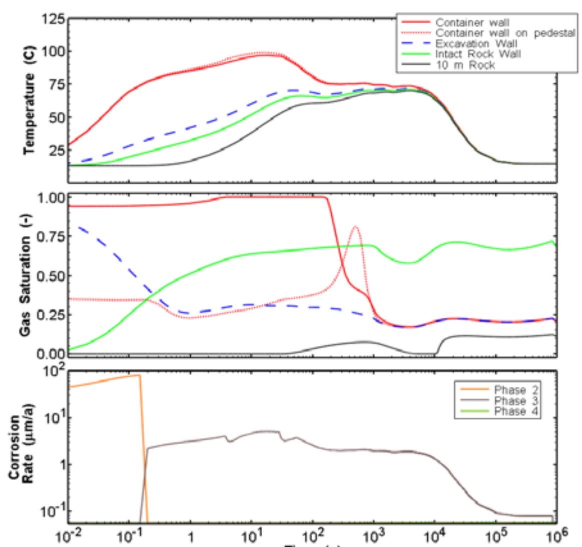

Figure 11. Time series results for temperature, gas saturation, and corrosion rate.

The bentonite pellets adjacent to the container are fully gas saturated at approximately 3 years and begin to resaturate with water at approximately 200 years, as the temperature drops. By 2000 years, gas saturations are approximately 20%, and remain so for the remainder of the simulation. Gas saturations in the EDZ are higher due to reduced capillary pressures, when compared to the host rock and bentonite.

Corrosion results show that Phase 1 never occurs: repository relative humidity is at 100% from the time of emplacement with 6% liquid in the gap fill and 65% in the pedestal. Phase 2 corrosion occurs for a period of approximately 0.2 years, after which all oxygen is exhausted. Phase 3 corrosion is initiated, and continues under water-limited conditions until the canister is consumed completely at 850 ka. Beyond 2000 years, as the bentonite resaturates and the incoming water flow rate is reduced, the rate of Phase 3 corrosion gradually moderates, reaching a new equilibrium that persists until 850 ka.

CONCLUSIONS

The T2GGM code allows for an assessment of system behavior that couples gas generation processes with repository pressure and saturation states. It has been applied to L&ILW and used fuel DGR assessments in low-permeability media. The water-limited scenario results show the impact of limiting reaction processes to available resources. T2GGM provides a new capability to include important coupled repository processes in TOUGH2 simulations.

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