AREST: THE NEXT GENERATION

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ABSTRACT

Simple mass transport models using constant boundary conditions at the waste form surface and at the host rock boundary do not always result in realistic predictions of the performance of an underground repository for the disposal of high-level radioactive waste. What is needed is a model that couples the important processes that can not be modeled independently, including 1) thermal modeling, 2) geochemical modeling, 3) containment degradation, 4) waste form dissolution, and 5) radionuclide transport. Such a model is being developed by modifying the AREST code.

INTRODUCTION

The Department of Energy (DOE) has assigned the task of leading the performance assessment (PA) studies at the potential sit¿ for the underground nuclear waste repository, at Yucca Mountain, to the Civilian Radioactive Waste Management System (CRWMS) Management and Operating Contractor (M&O). In turn, the M&O has selected the AREST code as the waste package model for analyzing the engineered barrier system (EBS) of the potential repository. The AREST code was developed for the DOE at the Pacific Northwest Laboratory (PNL).2

Within the next few years, the Yucca Mountain Site Characterization Project (YMP) will be examining alternative EBS concepts that cannot, however, be analyzed with the current version of the AREST code or any other existing DOE model. The limits of the capabilities of the fundamental structure of the AREST code has been reached. It relies on relatively simple analytical models to describe release and transport of radionuclides in the waste package of the EBS. Consequently, a fundamental restructuring of the AREST code is needed to implement robust but computationally efficient models for EBS and waste package performance assessment.

This report describes the modeling capabilities for the development of the next generation of the AREST code. Defining the requirements for the capabilities of the AREST code is an iterative process, with this report describing the latest iteration.

A. Current Version

The current version of the AREST code incorporates a sophisticated graphical user interface to simplify and organize problem setup and provide visualization of the computational results. The AREST code was developed to perform probabilistic analyses of the EBS; thus the process models are simplified from the more rigorous models that are needed in a detailed waste package model. The current version of the AREST code contains several analytical models and a simple one-dimensional finite difference numerical model for calculating nuclide transport to the host rock.

The models use constant boundary conditions for both the waste form surface and the backfill/host rock interface. The detailed processes of the near-field environment are modeled external to the AREST code, in support code analyses, with the results being input into the AREST code as look-up tables and transfer functions. The processes which are modeled externally to the AREST code includes the following: 1) radiolysis, 2) water mass transfer, 3) heat transfer, 4) rock mechanics, 5) geochemistry, and 6) the waste/barrier/rock interactions. These processes were assumed to be independent of waste form dissolution, corrosion, and transport processes and thus were modeled externally.

B. Analysis

To illustrate the need for the next generation of the AREST code, which will utilize coupled chemical reactive transport models, a simple analysis is presented. In this analysis, the mass transport properties of a 0.5 meter thick backfill surrounding a glass waste form are varied. A detailed description of this analysis has been presented elsewhere. For this analysis the diffusion coefficient has been varied over two orders of magnitude from $D_r = 10^{-5}$ to $10^{-7}$ cm²/sec.
The predicted release rate of $^{243}$Am is shown in Figure 1. In this analysis, a fixed temperature-dependent solubility model (FSM) for the boundary condition at the waste form surface and a coupled process model (CPM) for glass dissolution are used. Analyzing the results, we see that the FSM over estimates the release rate for $^{243}$Am for both diffusion coefficients. This over estimation occurs because the americium will actually be controlled by the dissolution of the waste and not by its solubility. The FSM estimates are conservative in this example, but a more realistic analysis can be accomplished using the CPM.

![Figure 1](image)

Figure 1. Comparison of Release Rates for $^{243}$Am for a Fixed Solubility Release Model (FSM) and a Coupled Process Release Model (CPM).

Figure 2 illustrates the results for $^{235}$U using both boundary condition methods (FSM and CPM) and both diffusion values. In this analysis, we see that both the FSM and the CPM agree for a $10^{-5}$ diffusion coefficient. However, by decreasing the diffusion coefficient to $10^{-7}$, the release rate actually increases for the CPM. This is counter-intuitive and is caused by a redox front that develops due to the slower mass transfer, caused by the lower diffusion coefficient, and a build up of $O_2$ at the waste form surface. Thus, the coupling between the mass transport properties and changes in solution chemistry are shown to have significant consequences on the prediction of the performance of the waste package and the EBS, illustrating the need for coupled process models.

![Figure 2](image)

Figure 2. Comparison of Release Rates for $^{235}$U for a Fixed Solubility Release Model (FSM) and a Coupled Process Release Model (CPM).

CODE USES

In being selected by DOE (YMP) as the waste package model, the AREST code will be used to respond to questions and concerns by the DOE about the waste package and EBS. The possible uses of the code could include addressing regulatory criteria, calculating source-term release for total systems performance assessment, potential repository design considerations, and sensitivity and uncertainty modeling. To address these concerns the AREST code must be capable of supplying the following data:

- flux as a function of space and time
- concentration as a function of space and time
- cumulative fluxes
- fractional release rates
- time-dependent container inventories
- waste container failure times.
SUPPORT CODES

Engineered barrier system performance cannot be modeled with the AREST code without information that describes the near-field environmental conditions of the waste package. Figure 3, illustrates the coupled processes in the near-field environment of the waste package. The solid arrows represent a strong coupling between processes. The near-field computer codes that are used to simulate the physical and chemical processes are complex and require extensive computing time. Therefore, it is not practical for all of these codes to be incorporated directly into the AREST code. Thus, process-specific codes are used to simulate the near-field environment and provide this information to the AREST code. The use of near-field support codes in this fashion preserves the computational efficiency of the AREST code without the loss of modeling accuracy that would result from the adoption of simplified models.

The AREST code will contain radiological, hydro-thermal, thermal, mechanical, and geochemical input modules that are designed to link the near-field parameters with the main components of the AREST code. Figure 4 illustrates the support code data that are supplied by each model.

A. Radiological Modeling

Radiological source-term analyses are needed to evaluate the radionuclide inventory and decay heat characteristics of the waste package. The heat generation rate and radionuclide inventory characteristics are important for modeling thermal and radionuclide release rate performance. Inventories for each nuclide, at the time of the potential repository closure, will be an input parameter for the AREST code.

B. Near-Field Hydrothermal Modeling

The near-field environment surrounding the waste package will significantly affect container corrosion and radionuclide dissolution processes. A primary assumption associated with the AREST code, however, will be that container corrosion and radionuclide processes do not

![Figure 3. Coupled Near-Field Environmental Processes. Solid Arrows Imply a Strong Coupling.](image)

![Figure 4. AREST and AREST Support Code Modeling.](image)
influence the near-field thermal and hydrologic environment. This assumption allows the complex modeling of the thermal and hydrologic environment to be performed independently of the waste package model. One flaw of this approach is that the corrosion processes will eventually alter the thermal and geometric characteristics of the waste package, which in turn will influence heat and mass transport from the container. The following two sections describe the hydrothermal support code modeling of fluid flow, heat transport, and mass transport for the AREST code.

1. Subsurface Fluid Flow and Heat Transport. The prediction of the hydrothermal environmental conditions in the immediate vicinity of the waste package is critical to predicting container corrosion and radionuclide dissolution processes. The undisturbed environment at the horizon of the potential repository at Yucca Mountain is believed to be dominated by highly fractured tuffaceous rock with relatively high saturations. Fault structures are assumed to transect the potential repository horizon. Previous numerical predictions have demonstrated that emplacement of waste package containers with radioactive decay heat sources significantly alter the surrounding thermal and hydrologic environment. The thermal and hydrologic fields that develop in response to the thermal loading from the potential repository are characterized by complex three-dimensional heat pipe type flow structures and resaturation zones surrounding the waste packages. These phenomena are strongly dependent on the thermal and hydrologic properties of the tuffaceous rock and the potential repository thermal loading.

Numerical simulators capable of modeling the complex fluid and heat transfer phenomena characteristic of those anticipated for the potential repository environment will be used to predict subsurface environmental parameters such as temperature, moisture content, relative humidity, liquid and vapor flow rates, and heat transfer rate. Ultimately, numerical simulators of this nature should include capabilities for modeling multiphase subsurface flow and heat transport considering fracture and matrix disequilibrium. These codes support the AREST code by providing specific thermal and hydrologic environmental data for specified thermal loading scenarios. Conca and Wright have shown that some transport properties (e.g., diffusion coefficient, hydraulic conductivity, and retardation factor) can be modeled as functions of the hydrologic parameter data predicted by the subsurface flow and transport support codes.

2. Subsurface Mass Transport. Multiphase subsurface transport codes will interact with the AREST code in two ways. The first interaction involves the prediction of the thermal and hydrologic environment surrounding the waste package. This provides the AREST code with environmental data that is critical for predicting container corrosion and radionuclide dissolution processes. Given these data, the AREST code will make numerical predictions concerning the release rates of various radionuclides.

The second interaction that multiphase subsurface transport codes have with the AREST code involves the prediction of mass transport rates of the released radio- nuclides to the accessible environment. Transport of radio- nuclides to the accessible environment (e.g., ground surface and water table) will be predicted using the thermal and hydrologic flow fields from the subsurface fluid flow and heat transport analyses. Transport of radionuclides to the accessible environment is strongly dependent on the subsurface flow fields and geochemical environment. Radio- nuclides can be transported to the accessible environment by the mechanisms of advection and diffusion through either the gas or aqueous phases. The geochemical environment determines the equilibrium partitioning state of the radionuclide between the three phases: solid, aqueous, and gas. To accurately predict the transport of radio- nuclides to the accessible environment, the support code must have the capabilities of modeling multiphase fluid flow, heat transfer, and mass transport within the fractured rock environment of the potential site for the repository at Yucca Mountain. The mass transport capabilities should include solid-liquid-gas partitioning, liquid and gas transport, and radioactive decay. Secondary capabilities will include nonlinear transport properties and radioactive decay chain modeling. An example of multiphase mass transport analyses applied to the potential site for the repository at Yucca Mountain has been reported for the migration of 14C to the accessible environment.

C. Mechanical Modeling

Detailed modeling of the host rock is important in defining boundary conditions. The condition of the rock, fractured or unfractured, and the stress that the rock places on the waste package can be important when analyzing groundwater flow, radionuclide release, and corrosion processes. These results will be entered into the AREST code as boundary conditions as described below.

1. Borehole Stability. Discontinuum stress analyses are needed to evaluate a rock mass that is so sparsely fractured (i.e., jointed) that individual fractures cannot be neglected. In the disturbed rock zone, fractures need to be modeled to determine the stability of the underground openings and load conditions on the waste package. This information will be used as boundary conditions at the host rock interface by the AREST code.

2. Structural Integrity. Continuum stress analyses are needed to evaluate the structural integrity of waste package component materials. Continuum stress codes can also be used to model a rock mass that is continuous (i.e., unfractured) or a rock mass that is so pervasively fractured that individual fractures can be neglected. The load conditions on the waste package container are important for predicting stress-induced corrosion and will be input into the AREST code.

D. Geochemical Modeling

Speciation and mass transfer analyses are needed to evaluate the near-field geochemistry and waste/barrier/rock hydrothermal interactions. The near-field geochemistry will significantly affect the corrosion of metallic containers,
stability of a clay backfill, dissolution of the waste form, and solubility of the radionuclides.

1. Initial Groundwater. The primary geochemical model needed to support the AREST code will be a near-field groundwater speciation model. The groundwater speciation model will be used to predict the potential repository groundwater species concentrations relative to changes in near-field hydrothermal conditions. The AREST code will input an initial groundwater composition, as a function of temperature, as calculated by the geochemical model. The initial groundwater composition will then be used to initiate groundwater modeling internal to the AREST code. The groundwater speciation model represents the very near-field geochemical conditions at the waste package container surface.

2. Radiolysis. Radiolysis analyses are needed to evaluate the dissociation of water and any chemical constituents present in the water as a result of exposure to radiation. Radiolysis could potentially alter the groundwater chemistry near a waste package which could affect container corrosion rates, waste form dissolution rates, and radionuclide solubilities. If radiolysis is shown to have a significant effect on the EBS performance, then models will be developed to incorporate these effects.

PROCESS MODEL DESCRIPTION

The current version of the AREST code was developed using a process-level structure, with the processes calculated sequentially. This type of structure will not work for the new version of the AREST code due to the inner process coupling that will be modeled. The important coupled processes shown in Figure 4 are described in the following sections.

A. Water Flow

The water that enters into the waste package system will be modeled in the hydrologic support code analysis. Saturation and groundwater flows will be calculated as a function of temperature for different heat-loading scenarios. These data will be read into the AREST code as boundary conditions and used to estimate the hydrothermal conditions of the waste package. Rewetting will occur when temperature of the waste package drops below the boiling point (about 95°C at the potential site at Yucca Mountain).

B. Container Lifetime

Mathematical models are required for the physical processes of pitting corrosion, general (uniform) corrosion, stress corrosion cracking (transgranular and intergranular), crevice corrosion, intergranular attack, and galvanic corrosion. However, some of these models may be eliminated in the future, depending on the material(s) chosen for the waste container. For example, a single-material container would not be expected to be susceptible to intergranular attack, but a coated material may be susceptible to crevice effects. For this version of the AREST code it is suggested that the model requirements be based on the assumption that the chosen material will be susceptible to the following:

- Fitting - depending on future (unknown) changes in repository chemistry
- General (Uniform) - to account for oxidizing conditions that may occur naturally (geological upsets) or by human intrusion
- Stress Corrosion - because pits, etc., may evolve into cracks which could be exacerbated by mechanical loads
- Crevice - because geological motion or backfill collapse may create local cells between the container and rock.

C. Nuclide Mobilization

The current version of the AREST code contains models for nuclide mobilization/dissolution, including 1) constant concentration for solubility-limited nuclides, 2) temperature-dependent solubilities, 3) UO₂ solubility function, 4) constant reaction rate of the waste form, and 5) a glass dissolution model.

For spent fuel, models based on experimental data will be developed to analyze the following situations: 1) the instant release of the inventories of soluble nuclides in the fuel/cladding gap and the grain boundaries of the fuel, 2) temperature-dependent concentrations for solubility-limited nuclides, 3) new mechanistic models for the dissolution kinetics of spent fuel which will control the rate of mobilization of soluble nuclides contained in the UO₂ matrix of the spent fuel, and 4) release of gaseous nuclides, ^14C.

The current version of the AREST code contains a mechanistic model for the dissolution of a glass waste form. This model couples the dissolution of the glass with the groundwater composition and mass transport into a backfill material. Since mass transport of the waste away from the waste form surface is a key process of this model, the model will have to be adapted to fit the numerical transport models that will be incorporated into the AREST code.

D. Waste/Barrier/Rock Interactions

The waste form dissolution models of the AREST code will utilize kinetic reaction-path calculations. The glass model, for instance, will use affinity and elemental concentration data calculated as a function of temperature, reaction progress, and iron content in the groundwater. Because this interaction must be coupled with mass transport and corrosion of the container, it will be modeled internally as a function of the groundwater composition, the dissolution of the waste, the corrosion of the overpack, and the transport of the waste away from the system.
E. Nuclide Transport

Numerical models will be developed to predict mass transport of the dissolved waste through the backfill and into the host rock. Spacial discretization will include a two-dimensional orthogonal grid with provisions to extend the model to a three-dimensional non-orthogonal grid. The models will check for solubility limits, radionuclide decay, and decay chain ingrowth at each node and time step throughout the EBS.

The new transport models will utilize parameters that are dependent on the groundwater composition, thermal conditions, and saturation value, including 1) diffusion and retardation coefficients as a function of saturation, 2) sorption values as a function of groundwater composition, and 3) solubilities as a function of temperature and groundwater composition for solubility limits/precipitation.

At the potential site at Yucca Mountain, diffusion should dominate the transport through the backfill and into the host rock. Thus the majority of the effort in the model development stage will be concentrated in the area of diffusive transport. However, diffusion/advection models will be developed to model transport through the backfill and into a fractured host rock. These models will be used to analyze disruptive events and periodic infiltration events.

F. Uncertainties

Uncertainties will be incorporated in each process model: from container corrosion to waste form dissolution to transport into the host rock. Propagation of uncertainties in models obtained from experiments, i.e., if a model was obtained by a regression analysis of experimental data, will include the uncertainty of the model as estimated by the experimental data. The goal is to estimate release at any specified time with a single performance parameter (e.g., average release rate) and an estimated uncertainty measure (i.e., a standard deviation or variance).

CONCLUSION

It has been illustrated in a simple analysis that the current models in the AREST code, which utilize fixed boundary conditions, are not sufficient, and in some cases not conservative, in the prediction of the performance of the waste package and EBS. Detailed coupled chemical processes with numerical transport models are needed by DOE to analyze the potential site for the repository, at Yucca Mountain, Nevada.

This report briefly describes the capabilities and requirements for the next generation of the AREST code, which has been selected by DOE (YMP) as the waste package model for the analysis at the potential site at Yucca Mountain. The report describes the process models that will be incorporated into the AREST code and the support code analyses that are needed to support the modeling.

REFERENCES


