

ANALYSIS/MODEL COVER SHEET

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**OFFICE OF CIVILIAN RADIOACTIVE WASTE
MANAGEMENT
ANALYSIS/MODEL REVISION RECORD**
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ANL-EBS-MD-000039 Rev. 00

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Initial Issue

DISCLAIMER

This contractor document was prepared for the U.S. Department of Energy (DOE), but has not undergone programmatic, policy, or publication review, and is provided for information only. The document provides preliminary information that may change based on new information or analysis, and represents a conservative treatment of parameters and assumptions to be used specifically for Total System Performance Assessment analyses. The document is a preliminary lower level contractor document and is not intended for publication or wide distribution.

Although this document has undergone technical reviews at the contractor organization, it has not undergone a DOE policy review. Therefore, the views and opinions of authors expressed may not state or reflect those of the DOE. However, in the interest of the rapid transfer of information, we are providing this document for your information per your request.

CONTENTS

	Page
1. PURPOSE	6
2. QUALITY ASSURANCE	6
3. COMPUTER SOFTWARE AND MODEL USAGE	6
3.1 COMPUTER SOFTWARE	6
3.2 MODELS	7
4. INPUTS	7
4.1 DATA AND PARAMETERS	7
4.1.1 Backfill	7
4.1.2 Quartz Saturation Indices for Various Backfill Environment Conditions	7
4.2 CRITERIA	8
4.2.1 NRC IRSR Criteria	8
4.2.1.1 Data and Model Justification Acceptance Criteria	9
4.2.1.2 Data Uncertainty and Verification Acceptance Criteria	9
4.2.1.3 Model Uncertainty Acceptance Criteria	10
4.2.1.4 Model Verification Acceptance Criteria	10
4.2.2 FEPs	11
4.3 CODES AND STANDARDS	11
4.3.1 Codes	11
4.3.2 Standards	11
5. ASSUMPTIONS	12
5.1 PROCESSES MODELED	12
5.2 GENERAL ASSUMPTIONS	12
5.2.1 Standard State of Water or Brine	12
5.2.2 Redox Conditions	12
5.2.3 Equilibrium Conditions	13
6. MODEL	13
6.1 GEOCHEMISTRY OF SILICA	14
6.1.1 Solubility of Silica Species	14
6.1.2 Dissolution and Precipitation Rates	15
6.2 CONCEPTUAL MODEL	15
6.3 MODEL DESIGN AND IMPLEMENTATION	16
6.4 MODEL RESULTS	16
7. CONCLUSIONS	19
7.1 MODELS AND RESULTS	19
7.2 UNCERTAINTY AND LIMITATIONS	19
7.3 TBV IMPACT	19

7.4	FEPs	20
8.	INPUTS AND REFERENCES.....	20
8.1	DOCUMENTS CITED	20
8.2	DATA LISTED BY DATA TRACKING NUMBER.....	22
8.3	CODES, STANDARDS, REGULATIONS, AND PROCEDURES	22

FIGURES

	Page
Figure 1. Plot of Quartz Saturation Indices for Each Case	18
Figure 2. Histogram of Quartz Saturation Indices.	18

TABLES

	Page
Table 1. Backfill Composition	7
Table 2. FEPs Potentially Applicable to the Seepage/Backfill Interactions Analysis	12
Table 3. Quartz Saturation Indices by Case.....	17
Table 4. FEPs Potentially Applicable to the Seepage/Backfill Interactions Analysis	20

ACRONYMS AND ABBREVIATIONS

ACC	accession number
AMR	Analysis/Model Report
CRWMS M&O	Civilian Radioactive Waste Management Services Management and Operations
DOE	Department of Energy
DTN	Data Tracking Number
EBS	Engineered Barrier System
f_{co2}	carbon dioxide fugacity
FEP	Features, Events, and Processes
IRSR	Issue Resolution Status Report
KTI	key technical issues
L	liter
NFE	Near Field Environment
NRC	Nuclear Regulatory Commission
PAO	Performance Assessment Operations
PT4	data0.pt4, a thermodynamic database developed in CRWMS M&O (2000)
Q^e	evaporation rate
Q^s	incoming seepage rate
R^{es}	relative evaporation rate
SI	saturation index
T	temperature
TBV	to be verified
TH	thermohydrologic
THC	thermohydrological-chemical
TIC	Technical Information Center number
TSPA-VA	Total System Performance Assessment - Viability Assessment

1. PURPOSE

As directed by written development plan (CRWMS M&O 1999a), a sub-model of seepage/backfill interactions is developed and presented in this document to support the Engineered Barrier System (EBS) Physical and Chemical Environment Model. The purpose of this analysis is to assist Performance Assessment Operations (PAO) and the Engineered Barrier Performance Department in modeling the geochemical environment within a repository drift. In this analysis, a conceptual model is developed to provide PAO a more detailed and complete in-drift geochemical model abstraction and to answer the key technical issues (KTI) raised in the NRC Issue Resolution Status Report (IRSR) for the Evolution of the Near Field Environment (NFE) Revision 2 (NRC 1999).

The development plan calls for a sub-model that evaluates the effect on water chemistry of chemical reactions between water that enters the drift and backfill materials in the drift. The development plan specifically requests an evaluation of the following important chemical reaction processes: dissolution-precipitation, aqueous complexation, and oxidation-reduction. The development plan also requests the evaluation of the effects of varying seepage and drainage fluxes, varying temperature, and varying evaporation and condensation fluxes. Many of these effects are evaluated in a separate Analysis/Model Report (AMR), *Precipitates Salts Analysis AMR* (CRWMS M&O 2000), so the results of that AMR are referenced throughout this AMR.

2. QUALITY ASSURANCE

The Quality Assurance (QA) program applies to the development of this analysis. The Performance Assessment Operations responsible manager has evaluated the technical document development activity in accordance with QAP-2-0, *Conduct of Activities*. The QAP-2-0 activity evaluation, *Conduct of Performance Assessment* (CRWMS M&O 1999b), has determined that the preparation and review of this technical document is subject to *Quality Assurance Requirements and Description* (QARD) DOE/RW-0333P (DOE 2000) requirements. Preparation of this analysis did not require the classification of items in accordance with QAP-2-3, *Classification of Permanent Items*. This activity is not a field activity. Therefore, an evaluation in accordance with NLP-2-0, *Determination of Importance Evaluations* was not required.

3. COMPUTER SOFTWARE AND MODEL USAGE

3.1 COMPUTER SOFTWARE

The software used in this analysis include:

- Microsoft Excel 97, a commercially available spreadsheet software package. This software was used to tabulate and chart results. No calculations or data manipulations were performed in the spreadsheets. Validation of the Excel spreadsheet application was done by comparing the input data (Table 3 in Section 6.4) to the output (Figure 1 and Figure 2, which are graphic representations of the input data). Visual inspection confirms that the spreadsheet application provided correct results.

The EQ3/6 code is discussed in Section 4.1.2 but not used in this AMR. Only EQ3/6 results from the Precipitates/Salts AMR (CRWMS M&O 2000) are used.

3.2 MODELS

The conceptual model used for this analysis and its validation is documented in *Precipitates Salts Analysis AMR* (CRWMS M&O 2000). The conceptual model documented in *Precipitates Salts Analysis AMR* (CRWMS M&O 2000) is considered appropriate and valid for this analysis because the model is applied to a specific case of the possible dissolution of quartz backfill in that system with the understanding that quartz stability is already quantified in the model calculations.

4. INPUTS

4.1 DATA AND PARAMETERS

The Seepage/Backfill Interactions analysis uses the following input: 1) the composition of backfill and 2) a set of data developed in *Precipitates Salts Analysis AMR* (CRWMS M&O 2000). These are described below.

4.1.1 Backfill

The EBS design backfill is composed of more than 99 percent quartz sand. The non-quartz fraction has predominantly feldspar mineralogy, and the dry backfill material contains less than 0.1 percent combined halides and less than 0.1 percent organic material (CRWMS M&O 1999c). Current specifications are displayed in Table 1. These input data remain to be verified (TBV).

Table 1. Backfill Composition

Property	Value
Quartz (SiO ₂) content (by mass)	> 99 percent
Combined total content of Al ₂ O ₃ , Na ₂ O, K ₂ O, and CaO (by mass)	< 1 percent

(CRWMS M&O 1999c)

4.1.2 Quartz Saturation Indices for Various Backfill Environment Conditions

The quartz saturation index data used in the current AMR are obtained from a set of EQ3/6 input/output files (DTN: MO0003MWDTAB45.013) that was developed in *Precipitates Salts Analysis AMR* (CRWMS M&O 2000). These data remain to be verified (TBV). From these files, the saturation indices (defined in Section 6.3) for quartz were obtained from the steady state results of every combination of temperature (T), fugacity of carbon dioxide (f_{CO_2}), and relative evaporation rate (R^{es} , defined below) simulated. These cases included the following parameter values: T varied between four values (95°C, 75°C, 45°C, and 25°C), f_{CO_2} varied between 10^{-1} , 10^{-3} , and 10^{-6} , and R^{es} varied between 0, 0.1, 0.5, 0.9, 0.99, and 0.999.

Only quartz saturation indices were retrieved because the backfill is greater than 99 percent quartz. The saturation indices of other silica minerals that might precipitate were not obtained because precipitation is not a seepage/backfill interaction. Precipitation is addressed in the Precipitates/Salts AMR (CRWMS M&O 2000). According to the development plan, the scope of the *Seepage/Backfill Interactions* AMR is limited to effects on water chemistry caused by seepage/backfill interactions.

The relative evaporation rate (or flux) (R^{es}) refers to the steady state evaporation flux (Q^e) divided by (i.e., relative to) the incoming seepage rate (or flux) (Q^s). R^{es} , which may apply to a specific location and time, may be expressed as:

$$R^{es} = \frac{Q^e}{Q^s} \quad (\text{Eq. 1})$$

Representative water from well J-13 was used as the composition of the incoming seepage in the data obtained from CRWMS M&O (2000).

The software used to generate the results referenced in this AMR was EQ3/6 v7.2b [CSCI: URCL-MA-110662 V7.2b, Wolery 1992a and 1992b, Wolery and Daveler 1992] with the solid-centered flow-through addendum [CSCI: URCL-MA-110662 V7.2b, MI: 30084-M04-001 (Addendum Only) (CRWMS M&O 1998)]. This software was obtained from Configuration Management for *Precipitates Salts Analysis AMR* (CRWMS M&O 2000) and installed on an IBM-compatible computer. It was appropriate for the application in that AMR and was used only within the range of validation in accordance with AP-SI.1Q, *Software Management*.

The thermodynamic database used to generate the referenced results is the PT4 database developed in the In-Drift Precipitate/Salts AMR (CRWMS M&O 2000). This is a Pitzer database that allows calculation of equilibrium chemistry at high ionic strength, but it is not direct input for the current AMR. It is cited here for information purposes only. For more information on its development and use, refer to CRWMS M&O (2000).

4.2 CRITERIA

Section 4.2.1 provides a summary of the applicable NRC review and acceptance criteria outlined in the Issue Resolution Status Report (IRSR) that apply to model development for the following NFE KTI sub-issue effects: (a) coupled thermal-hydrologic-chemical processes on the waste package chemical environment, (b) coupled thermal-hydrologic-chemical processes on the chemical environment for radionuclide release, and (c) coupled thermal-hydrologic-chemical processes on radionuclide transport through engineered and natural barriers (NRC 1999). A listing of the project features, events, and processes (FEPs) that potentially apply to this report is provided in Section 4.2.2.

4.2.1 NRC IRSR Criteria

The wording for the IRSR criteria listed below is generally taken from Section 4.3.1 of NRC (1999).

4.2.1.1 Data and Model Justification Acceptance Criteria

1. Consider both temporal and spatial variations in conditions affecting coupled thermohydrological-chemical (THC) effects on the chemical environment for radionuclide release. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
2. Consider site characteristics in establishing initial and boundary conditions for conceptual models and simulations of coupled processes that may affect the chemical environment for radionuclide release. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
3. Collect sufficient data on the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for conceptual models and simulations of THC coupled processes that affect the chemical environment for radionuclide release. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
4. Use sensitivity and uncertainty analyses (including consideration of alternative conceptual models) to determine whether additional new data are needed to better define ranges of input parameters. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
5. If the testing program for coupled THC processes on the chemical environment for radionuclide release from the engineered barrier system is not complete at the time of license application, or if sensitivity and uncertainty analysis indicate the additional data are needed, identify specific plans to acquire the necessary information as part of the performance confirmation program. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]

4.2.1.2 Data Uncertainty and Verification Acceptance Criteria

1. Use reasonable or conservative ranges of parameters or functional relations to determine effects of coupled THC processes on the chemical environment for radionuclide release. Parameter values, assumed ranges, probability distributions, and bounding assumptions must be technically defensible and reasonably account for uncertainties. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
2. Consider uncertainty in data due to both temporal and spatial variations in conditions affecting coupled THC effects on the chemical environment for radionuclide release. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
3. Properly consider in the evaluation of coupled THC processes the uncertainties in the characteristics of the natural system and engineered materials, such as the type, quantity, and reactivity of material, in establishing initial and boundary conditions for conceptual models and simulations of THC coupled processes that affect the chemical environment for radionuclide release. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]

4. Use available data to ensure consistency in the initial conditions, boundary conditions, and computational domain used in sensitivity analysis involving coupled THC effects on the chemical environment for radionuclide release. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
5. Assess in the performance confirmation program whether the natural system and engineered materials are functioning as intended and anticipated with regard to coupled THC effects on the chemical environment for radionuclide release from the EBS. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]

4.2.1.3 Model Uncertainty Acceptance Criteria

1. Use appropriate models, tests, and analyses that are sensitive to the THC couplings under consideration for both natural and engineering systems as described in the following examples. The effects of THC coupled processes that may occur in the natural setting or due to interactions with engineered materials or their alteration products include (i) thermohydrologic (TH) effects on gas and water chemistry; (ii) hydrothermally driven geochemical reactions, such as zeolitization of volcanic glass; (iii) dehydration of hydrous phases liberating moisture; (iv) effects of microbial processes; and (v) changes in water chemistry that may result from interactions between cementitious, or WP, materials and groundwater, which, in turn, may affect the chemical environment for radionuclide release. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
2. Investigate alternative modeling approaches consistent with available data and current scientific understanding, and appropriately consider their results and limitations. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
3. Provide a reasonable description of the mathematical models included in its analyses of coupled THC effects on the chemical environment for radionuclide release. The description should include a discussion of alternative modeling approaches not considered in its final analysis and the limitations and uncertainties of the chosen model. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]

4.2.1.4 Model Verification Acceptance Criteria

1. The mathematical models for coupled THC effects on the chemical environment for radionuclide release must be consistent with conceptual models based on inferences about the near-field environment, field data and natural alteration observed at the site, and expected engineered materials. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
2. Appropriately adopt accepted and well-documented procedures to construct and test the numerical models used to simulate coupled THC effects on the chemical environment for radionuclide release. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]
3. Abstracted models for coupled THC effects on the chemical environment for radionuclide release must be based on the same assumptions and approximations shown to be appropriate for closely analogous natural or experimental systems. Abstracted

model results are verified through comparison to outputs of detailed process models and empirical observations. Abstracted model results are compared with different mathematical models to judge robustness of results. [NRC (1999), Sections 4.1.1, 4.2.1, 4.3.1, and 4.4.1]

4.2.2 FEPs

Table 2 lists the Yucca Mountain Project FEPs (CRWMS M&O 1999d) that are addressed in this analysis. Any resolution of these FEPs is discussed in Section 7.4.

4.3 CODES AND STANDARDS

4.3.1 Codes

This AMR was prepared to comply with the DOE interim guidance (Dyer 1999) which directs the use of specified Subparts/Sections of the proposed NRC high-level waste rule, 10 CFR Part 63. Relevant requirements for performance assessment from Section 114 of that document are: "Any performance assessment used to demonstrate compliance with Sec. 113(b) shall: (a) Include data related to the geology, hydrology, and geochemistry ... used to define parameters and conceptual models used in the assessment. (b) Account for uncertainties and variabilities in parameter values and provide the technical basis for parameter ranges, probability distributions, or bounding values used in the performance assessment. ... (g) Provide the technical basis for models used in the performance assessment such as comparisons made with outputs of detailed process-level models"

4.3.2 Standards

ASTM C 1174-97, *Standard Practice for Prediction of the Long-Term Behavior of Materials, Including Waste Forms, Used in Engineered Barrier Systems (EBS) for Geological Disposal of High-Level Radioactive Waste*, was used as guidance in the preparation of this model.

Table 2. FEPs Potentially Applicable to the Seepage/Backfill Interactions Analysis

YMP FEP Number	NEA Category	FEP Name
2.1.04.01.00	2.1.04u	Preferential pathways in the backfill
2.1.04.02.00	2.1.04au	Physical and chemical properties of backfill
2.1.04.02.09	2.1.09ay	Water chemistry, tunnel backfill
2.1.04.03.00	2.1.04r	Erosion or dissolution of backfill
2.1.04.04.00	2.1.04az	Mechanical effects of backfill
2.1.04.04.00	2.1.07w	Mechanical impact/failure, buffer/backfill
2.1.04.05.00	2.1.04b	Backfill evolution
2.1.04.06.00	2.1.11g	Small pieces of backfill undergo phase changes when heated and welded together
2.1.04.08.00	2.1.04t	Diffusion in backfill
2.1.04.09.00	3.2.07r	Radionuclide transport through backfill
2.1.09.05.04	3.2.03s	Sorption on filling material
2.2.10.06.07	2.1.09j	Precipitates from dissolved constituents of tuff and repository materials form by evaporation during thermal period

ACC: MOL.19991214.0518; MOL.19991214.0519

5. ASSUMPTIONS

5.1 PROCESSES MODELED

As explained in the introduction to Section 6, the only seepage/backfill interaction evaluated in this AMR is the potential dissolution of quartz backfill.

5.2 GENERAL ASSUMPTIONS

5.2.1 Standard State of Water or Brine

Water in this analysis is assumed to be standard state. That is, the properties of the water are assumed to be unaffected by interface curvature or other effects resulting from contact with solid surfaces. For a more detailed explanation, refer to *Precipitates Salts Analysis AMR* (CRWMS M&O 2000, Section 5.2.1). This assumption, used throughout, does not affect the uncertainty in the model and is therefore not designated TBV.

5.2.2 Redox Conditions

The redox conditions within the drift are expected to be oxidizing at all times except possibly for the first several hundred years after the waste packages would be covered by the backfill. During these early times, it is possible that reducing conditions could occur. However, reducing conditions actually reduce corrosion rates for the drip shield and waste package container. In addition, they do not affect interactions with quartz backfill because silica is not sensitive to redox conditions. For these reasons, redox conditions are assumed to be oxidizing at all times.

This assumption, used throughout, is reasonable and conservative for the scope of this AMR and is therefore not TBV.

5.2.3 Equilibrium Conditions

The model assumes equilibrium chemical conditions except for a few minerals that are not allowed to precipitate. Chemical reactions are assumed to occur rapidly compared to anticipated seepage and evaporation rates. The assumption of equilibrium conditions will not affect the uncertainty in the model and is therefore not designated TBV. This assumption is used throughout.

Several slow-forming minerals in the database are not allowed to precipitate. The suppressed minerals include albite high, albite low, albite, corundum, cristobalite(alpha), cristobalite(beta), quartz, tridymite, diaspore, dolomite, hercynite, ice, illite, k-feldspar, maximum microcline, and spinel. For an explanation, see Section 5.4 of *Precipitates Salts Analysis AMR* (CRWMS M&O 2000). These mineral suppressions are TBV in the referenced AMR. The only suppressed minerals that became supersaturated in the calculations were quartz, maximum microcline, K-feldspar, albite, albite low, tridymite, diaspore, and dolomite. In a separate set of EQ3/6 calculations documented in CRWMS M&O (2000), albite low was allowed to precipitate. However, this change affected the predicted concentrations by less than one percent for any of the aqueous components.

6. MODEL

As stated in Section 1, the development plan calls for an evaluation of the effects of seepage/backfill interactions on water chemistry resulting from the following important processes and parameters:

- Dissolution-precipitation,
- Aqueous complexation,
- Oxidation-reduction,
- Seepage and drainage fluxes,
- Temperature variation, and
- Evaporation and condensation fluxes.

Most of these processes and parameters are incorporated in the development of the Precipitates/Salts Model documented in *Precipitates Salts Analysis AMR* (CRWMS M&O 2000). The model is used in that AMR to assess the effects of these processes and parameters (except for oxidation-reduction and reaction with backfill) on water chemistry within the drift. Thus, output from that AMR is used to assess the additional potential interaction with the backfill. In this way, dissolution, precipitation, aqueous complexation, and varying temperatures, seepage fluxes, and evaporation fluxes are evaluated in a coupled manner. Oxidation-reduction is addressed separately in Section 5.2.2.

Seepage/backfill interaction under varying conditions is investigated by examining the quartz saturation indices (defined in Section 6.3) predicted by the EQ3/6 Pitzer model developed in

Precipitates Salts Analysis AMR (CRWMS M&O 2000). The backfill is composed of more than 99 percent quartz sand (Table 1). A potentially important seepage/backfill interaction that might significantly affect water chemistry in the backfill is the dissolution of this quartz. Quartz saturation indices (defined in Section 6.3) from the referenced data set indicate whether quartz dissolution is thermodynamically favored over a full range of possible temperature, seepage, and evaporation conditions and possible carbon dioxide fugacities within the backfill.

Seepage interaction with the non-quartz fraction of the backfill is not evaluated in this AMR because of insufficient information on the quantity and mineralogy of this fraction. The non-quartz fraction is less than one percent of the backfill mineralogy (Table 1) and thus may not be abundant enough to significantly affect the water chemistry in the backfill such that performance of the EBS is affected. Nevertheless, an analysis of the seepage interaction with this small fraction should be performed when more precise information on the composition and quantities of this fraction becomes available.

According to the development plan, the scope of the *Seepage/Backfill Interactions* AMR is limited to effects on water chemistry caused by seepage/backfill interactions. Potential seepage/backfill interactions other than dissolution of quartz backfill are not evaluated. These interactions include adsorption, hydrolysis of minerals such as feldspar in the non-quartz fraction of the backfill, and others. Before these additional interactions can be appropriately evaluated, additional characterization of the properties and composition of the backfill is needed. Effects on backfill permeability are outside of the defined scope.

6.1 GEOCHEMISTRY OF SILICA

6.1.1 Solubility of Silica Species

The solubility of quartz and other silica minerals is affected by pH, temperature, pressure, and ionic strength (Dove and Rimstidt 1994, pp. 262-265). At 25°C, 1 bar atmospheric pressure, and a pH between 1 and 9, the dominant aqueous silica species is $\text{SiO}_2(\text{aq})$ (also written $\text{SiO}_2\cdot\text{H}_2\text{O}$ or $\text{H}_4\text{SiO}_4^\circ$) (Dove and Rimstidt 1994, p. 264; Langmuir 1997, p. 246; Drever 1988, p. 100). Above pH 9, the solubility of silica increases by several orders of magnitude as the soluble species H_3SiO_4^- and $\text{H}_2\text{SiO}_4^{2-}$ become increasingly stable. In the pH range of approximately 9.82 to 13.1, H_3SiO_4^- is the dominant silica species, followed by $\text{H}_2\text{SiO}_4^{2-}$ at pH values greater than 13.1 (Langmuir 1997, p. 246). For comparison, another source indicates that the pH values of these transitions are closer to 9.9 and 11.7, respectively (Drever 1988, p. 100). The differences are due to differences in the adopted dissociation constants at 25°C.

In addition to the uncertainty of the values of the dissociation constants, these constants are affected by changes in temperature and pressure. As illustrated in Krauskopf and Bird (1995, p. 88) at pH values less than 9, a temperature increase from 25°C to 100°C causes the solubility of $\text{SiO}_2(\text{aq})$ to increase from about 0.002 molal to about 0.006 molal for a solution equilibrated with amorphous silica. This figure also shows that for a solution equilibrated with quartz, the same temperature increase causes the solubility of $\text{SiO}_2(\text{aq})$ to increase from about 0.0001 molal to about 0.0008 molal. At temperatures over 200°C, solutions are in equilibrium with quartz (Dove and Rimstidt 1994, p. 267). Below this temperature, factors such as flow rate and precipitation rate are important and may cause sustained supersaturation with respect to quartz

due to the slow formation of quartz relative to other silica minerals (Dove and Rimstidt 1994, pp. 267-268; Rimstidt 1997, p. 490-491). Silica solubility is increased by Na^+ and/or Cl^- (Dove and Rimstidt 1994, p. 265). The effects of temperature and solution composition on solubility are accounted for by equilibrium geochemical models such as EQ3/6. These effects factor into the calculated saturation indices for each mineral phase (see section 6.4).

6.1.2 Dissolution and Precipitation Rates

Quartz dissolution and precipitation rates are controlled by temperature, solution composition (catalysis by alkali cations, ionic strength, trivalent metals) and pH (Dove and Rimstidt 1994, pp. 278-287). An increase in temperature causes an increase in the dissolution rate. For example, a rate increase of three orders of magnitude occurs as the temperature increases from 25°C to 100°C. An increase in dissolved salts also increases the dissolution rate. For relatively dilute solutions, dissolution rates increase by a factor of five to eight with the addition of sodium and potassium chloride for neutral pH and temperatures below 70°C. The changes in dissolution rates are cation-dependent. In 0.09 to 1.0 molal NaCl, silica maintains its dissolution rate for a pH of 5 to 8 over a temperature range of 60°C to 120°C. However, dissolution rates increase by an order of magnitude as the solution reaches 4 molal NaCl (Dove and Rimstidt 1994, pp. 281-282).

The precipitation or dissolution of lower order silica oxides, such as chalcedony, is not greatly inhibited by kinetic factors over a temperature range of 25°C to 100°C; however, the dissolution of quartz is very slow (Klein and Hurlbut 1999, p.571). At pH 5 and 25°C, the predicted mean lifetime of a 1-millimeter cube of quartz is 24 million years (Langmuir 1997, p. 233). This prediction is based upon a dissolution rate of $10^{-13.39}$ mol/m²/s (Langmuir 1997, p. 233). As pH increases to 12, the rate remains low but may rise to approximately $10^{-10.5}$ mol/m²/s (Langmuir 1997, p. 78). Thus, the lifetime of the same cube of quartz would decrease to about 35,000 years.

Quartz is a relatively stable mineral at low temperatures and pressures (Klein and Hurlbut 1999, pp. 526-532). It melts at over 1728°C but may undergo polymorphic transitions above 573°C at atmospheric pressure. Based upon estimates of repository conditions, quartz will not undergo solid-solid transitions to its polymorphs.

6.2 CONCEPTUAL MODEL

The conceptual model for the potential dissolution of quartz is essentially identical to the EQ3/6 Pitzer model developed in *Precipitates Salts Analysis AMR* (CRWMS M&O 2000) except that the stability of quartz sand is monitored. Seepage/backfill interactions cannot be adequately investigated without incorporating the effects of evaporation on the composition of the water in contact with the backfill. The only difference in the current application is that the potential dissolution of the quartz backfill is investigated.

Dissolution of the quartz backfill is investigated by examining the quartz saturation indices (defined in Section 6.3) for each of the cases simulated by the EQ3/6 Pitzer model. These cases include the set of all simulations for conditions of 85-percent relative humidity and higher (DTN: MO0003MWDTAB45.013). Lower relative humidity cases were ignored because the increased

evaporation at lower relative humidity favors precipitation of silica and continued saturation or supersaturation of quartz (CRWMS M&O 2000). Although silica salts are predicted to form at relative humidity values below 85 percent, precipitation of these salts is not considered a seepage/backfill interaction (see Section 4.1.2).

6.3 MODEL DESIGN AND IMPLEMENTATION

For the design, implementation, and evaluation of the EQ3/6 Pitzer model used to investigate the potential for backfill dissolution, the reader is referred to *Precipitates Salts Analysis AMR* (CRWMS M&O 2000). Saturation indices for quartz were tabulated and evaluated with respect to backfill dissolution.

The saturation index (SI) is defined as the logarithm of the ratio of the reaction quotient (or ion activity product) to the equilibrium constant (Langmuir 1997, p. 8). A zero value for SI indicates equilibrium with respect to the mineral. Strictly interpreted, a positive value indicates the water is supersaturated with respect to the mineral and a negative value indicates undersaturation. Supersaturation favors precipitation of the mineral and undersaturation favors dissolution. However, because there is some uncertainty in the equilibrium constants, the EQ3/6 code interprets an SI value between -0.2 and +0.2 as an indication of approximate saturation with respect to the mineral.

6.4 MODEL RESULTS

Based upon results of the Precipitates/Salts AMR (CRWMS M&O 2000), over 97% of the predictions for various conditions with J-13 incoming water have a quartz saturation index greater than zero. Part of this result stems from suppression of quartz and tridymite, which allows chalcedony to be the precipitating phase that controls silica solubility. The predictions are shown in Table 3, Figure 1, and Figure 2. In 84 simulations of J-13 water, only two results predicted a quartz saturation index less than zero. In these cases the quartz saturation indices were -0.092 and -0.052. In each case, the temperature was 95°C and the fugacity of carbon dioxide was 10^{-6} which are consistent with predictions of conditions within the first several hundred years. In these two cases, the seepage rate was very high relative to the evaporation rate (R^{es} of 0 and 0.1, respectively). However, these low evaporation rates relative to seepage rates are not likely to occur at early times. The likelihood of such conditions is predicted in other AMRs. Regardless, these quartz saturation indices are within the range that EQ3/6 associates with saturated conditions (see Section 6.3).

These results indicate that none of the likely combination of conditions in the backfill will create a considerable tendency for quartz to dissolve. This observation is consistent with the fact that evaporation tends to favor silica precipitation, not dissolution. For the unlikely case of zero evaporation, incoming seepage water is expected to already be approximately saturated with respect to silica (as is the case for average J-13 well water (Table 1)), which will also inhibit quartz dissolution.

Table 3. Quartz Saturation Indices by Case.

Temperature (°C)	R^{es}	Quartz Saturation Index ($f_{CO_2} = 10^{-6}$)	Quartz Saturation Index ($f_{CO_2} = 10^{-3}$)	Quartz Saturation Index ($f_{CO_2} = 10^{-1}$)
25	0 ^a	0.271	0.271	0.271
25	0.1	0.271	0.271	0.271
25	0.5	0.271	0.271	0.271
25	0.9	0.271	0.271	0.271
25	0.99	0.271	0.271	0.271
25	0.999	0.271	0.271	0.271
25	≥ 1 ^b	0.271	0.271	0.271
45	0	0.191	0.254	0.254
45	0.1	0.243	0.254	0.254
45	0.5	0.254	0.254	0.254
45	0.9	0.254	0.254	0.254
45	0.99	0.254	0.254	0.254
45	0.999	0.254	0.254	0.254
45	≥ 1	0.254	0.254	0.254
75	0	0.018	0.232	0.232
75	0.1	0.055	0.232	0.232
75	0.5	0.232	0.232	0.232
75	0.9	0.232	0.232	0.232
75	0.99	0.232	0.232	0.232
75	0.999	0.232	0.232	0.232
75	≥ 1	0.232	0.232	0.232
95	0	-0.092	0.066	0.136
95	0.1	-0.052	0.110	0.220
95	0.5	0.160	0.220	0.220
95	0.9	0.220	0.220	0.220
95	0.99	0.220	0.220	0.220
95	0.999	0.220	0.220	0.220
95	≥ 1	0.220	0.220	0.220

DTN: MO0003MWDTAB45.013

^a no evaporation but seepage occurs^b evaporated to an ionic strength of 10 molal

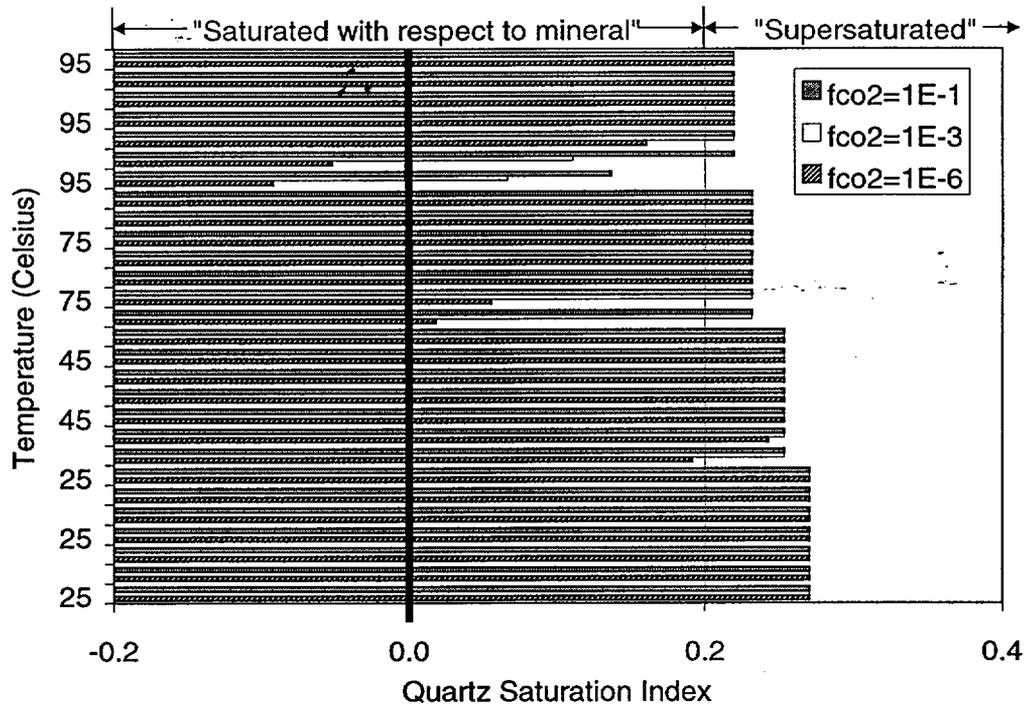


Figure 1. Plot of Quartz Saturation Indices for Each Case

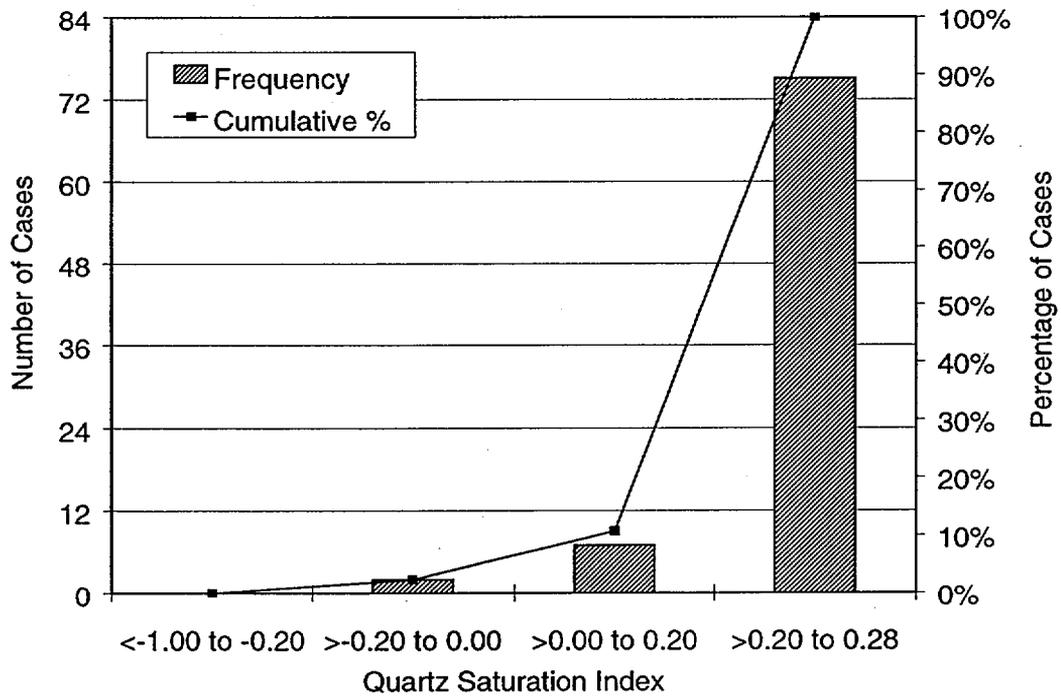


Figure 2. Histogram of Quartz Saturation Indices.

7. CONCLUSIONS

This document and its conclusions may be affected by technical product input information that requires confirmation. Any changes to the document or its conclusions that may occur as a result of completing the confirmation activities will be reflected in subsequent revisions. The status of the input information quality may be confirmed by review of the Document Input Reference System database.

7.1 MODELS AND RESULTS

The results indicate that dissolution of quartz backfill by seepage derived from evaporated J-13 well water is not likely to occur, and that if any dissolution does occur, it will likely be negligible. Dissolution of quartz, which makes up more than 99 percent of the backfill, is generally not favored except only weakly in the most unlikely scenarios.

Two of the 84 cases investigated are shown to have quartz saturation indices below zero (Table 3, Figure 1). The values of these indices are -0.092 and -0.052, which the code EQ3/6 generally considers to be saturated conditions, given the uncertainty in thermodynamic data. Moreover, these two cases are for conditions of high temperature (95°C) and high seepage rate relative to the evaporation rate (a factor of 10 or more higher). Such a combination of conditions is unlikely. For these reasons, dissolution of the quartz backfill is not likely and will not likely affect water chemistry within the backfill. These results and conclusions are consistent with the fact that evaporation tends to favor mineral precipitation, not dissolution.

7.2 UNCERTAINTY AND LIMITATIONS

For the analysis of quartz backfill dissolution, the greatest uncertainties are likely the predicted inputs (e.g., the seepage rate and seepage composition) that feed the Precipitates/Salts analysis that generated the input used in this analysis. The analysis was limited to a conceptual model in which J-13 well water was the seepage. However, the conclusion that quartz dissolution is not favored, except perhaps in the most unlikely scenarios, is reasonable because evaporating water tends to favor mineral precipitation, not dissolution. For the unlikely case of zero evaporation, incoming seepage water is expected to already be approximately saturated with respect to silica (as is the case for average J-13 well water), which will also inhibit quartz dissolution.

Potential seepage/backfill interactions other than dissolution of quartz are not evaluated in this AMR. These interactions include adsorption, hydrolysis of minerals such as feldspar in the non-quartz fraction of the backfill, and others. Before these interactions can be appropriately evaluated, additional characterization of the properties and composition of the backfill is needed.

7.3 TBV IMPACT

The EQ3/6 data were acquired from the results of EQ3/6 Pitzer model developed in *Precipitates Salts Analysis AMR* (CRWMS M&O 2000). These acquired data remain to be verified (TBV) until that AMR becomes qualified. Additional information on the TBV status and implications can be found in *Precipitates Salts Analysis AMR*.

The other TBV is the composition of the backfill.

7.4 FEPs

Table 4 lists the FEPs in Table 2 and the conclusions based on the Seepage/Backfill Interactions analysis.

Table 4. FEPs Potentially Applicable to the Seepage/Backfill Interactions Analysis

YMP FEP Number	NEA Category	FEP Name	AMR Conclusions
2.1.04.01.00	2.1.04u	Preferential pathways in the backfill	This FEP not addressed in this AMR.
2.1.04.02.00	2.1.04au	Physical and chemical properties of backfill	Based on the results of this AMR and the Precipitates/Salts AMR (CRWMS M&O 2000), the water chemistry within the backfill will likely be primarily controlled by evaporative processes and not by interactions with backfill. Affects of backfill on water chemistry are limited. This AMR considered the potential for dissolution of quartz backfill and the potential effects on water chemistry.
2.1.04.02.09	2.1.09ay	Water chemistry, tunnel backfill	The seepage composition considered in this AMR and the Precipitates/Salts AMR (CRWMS M&O 2000) is J-13 water. Water chemistry indicates saturation with respect to silica. These AMRs predict that backfill dissolution will likely be small or negligible.
2.1.04.03.00	2.1.04r	Erosion or dissolution of backfill	Dissolution of quartz backfill will likely be small or negligible. Erosion was not addressed by this AMR.
2.1.04.04.00	2.1.04az	Mechanical effects of backfill	This FEP is not addressed by this AMR.
2.1.04.04.00	2.1.07w	Mechanical impact/failure, buffer/backfill	This FEP is not addressed by this AMR.
2.1.04.05.00	2.1.04b	Backfill evolution	This FEP is not addressed by this AMR.
2.1.04.06.00	2.1.11g	Small pieces of backfill undergo phase changes when heated and welded together	This FEP is not addressed by this AMR.
2.1.04.08.00	2.1.04t	Diffusion in backfill	This FEP is not addressed by this AMR
2.1.04.09.00	3.2.07r	Radionuclide transport through backfill	This FEP is not addressed by this AMR.
2.1.09.05.04	3.2.03s	Sorption on filling material	This FEP is not addressed by this AMR.
2.2.10.06.07	2.1.09j	Precipitates from dissolved constituents of tuff and repository materials form by evaporation during thermal period	This FEP is not addressed by this AMR.

ACC: MOL.19991214.0518; MOL.19991214.0519

8. INPUTS AND REFERENCES

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