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NM5507

OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT
ANALYSIS/MODEL COVER SHEET
Complete Only Applicable Items

1. QA: QA

2. ☒ Analysis — Check all that apply

Type of Analysis	☐ Engineering ☒ Performance Assessment ☐ Scientific
Intended Use of Analysis	☐ Input to Calculation ☐ Input to another Analysis or Model ☒ Input to Technical Document ☒ Input to other Technical Products

Describe use:

Input to TSPA-SR, TSPA-LA, and WF PMR

3. ☒ Model — Check all that apply

Type of Model	☒ Conceptual Model ☒ Abstraction Model ☐ Mathematical Model ☐ System Model ☐ Process Model
Intended Use of Model	☐ Input to Calculation ☐ Input to another Model or Analysis ☒ Input to Technical Document ☐ Input to other Technical Products

Describe use:

Only provide basis for this analysis

4. Title:

Summary of Dissolved Concentration Limits

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OFFICE OF CIVILIAN RADIOACTIVE WASTE MANAGEMENT
ANALYSIS/MODEL REVISION RECORD

Complete Only Applicable Items

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ACRONYMS AND ABBREVIATIONS

AMR	Analysis and Models Report
CPU	Central Processing Unit
CRWMS	Civilian Radioactive Waste Management System
CSNF	Commercial Spent Nuclear Fuel
DOE	U.S. Department of Energy
DTN	Data Tracking Number
LANL	Los Alamos National Laboratory
LLNL	Lawrence Livermore National Laboratory
M&O	Management and Operating Contractor
NEA	Nuclear Energy Agency
NEPO	Natural Environment Program Operations
NRC	U.S. Nuclear Regulatory Commission
PAO	Performance Assessment Operations
PNNL	Pacific Northwest National Laboratory
QA	quality assurance
RN	radionuclide
S	solubility limits
TBD	to be determined
TBV	to be verified
TSPA	Total System Performance Assessment
TSPA-LA	Total System Performance Assessment-License Application
TSPA-SR	Total System Performance Assessment-Site Recommendation
TSPA-VA	Total System Performance Assessment-Viability Assessment
WP	Waste Package
WPO	Waste Package Operations

1. PURPOSE

According to the Development Plan titled "Summary of Dissolved Concentration Limits" (hereafter known as the Development Plan) (CRWMS M&O 1999a), the purpose of this study is to perform abstraction on solubility limits of radioactive elements based on the process-level models provided by Natural Environment Program Operations (NEPO) and Waste Package Operations (WPO). This analysis is to produce solubility limits as functions, distributions, or constants for all transported radioactive elements identified by Performance Assessment Operations (PAO) radioisotope screening. The results of this analysis and conceptual models will feed to the performance assessment for Total System Performance Assessment - Site Recommendation (TSPA-SR) and Total System Performance Assessment - License Application (TSPA-LA), and to the Waste Form Degradation Process Model Report section on concentration limits.

Results from an expert elicitation for solubility limits of most radioactive elements were used in the previous Total System Performance Assessments (TSPAs). However, the elicitation conducted in 1993 does not meet the criteria set forth by the U.S. Nuclear Regulatory Commission (NRC) due to lack of documentation and traceability (Kotra et al. 1996, Section 3). Therefore, at the Waste Form Abstraction Workshop held on February 2-4, 1999, at Albuquerque, New Mexico, the Yucca Mountain Site Characterization Project (YMP) decided to develop geochemical models to study solubility for the Monitored Geologic Repository. WPO/NEPO is to develop process-level solubility models, including review and compilation of relevant thermodynamic data. PAO's responsibility is to perform abstractions based on the process models and chemical conditions and to produce solubility distributions or response surfaces applicable to the repository.

In the history of the YMP, this is the first round of solubility evaluation activity that is based on detailed geochemical modeling. Revisions to solubility limits are expected as more is learned and understood about the repository conditions and as more data become available.

2. QUALITY ASSURANCE

This analysis was prepared in accordance with the Civilian Radioactive Waste Management System (CRWMS) Management and Operating Contractor (M&O) quality assurance (QA) program. The information provided in this analysis will be used for evaluating the post-closure performance of the Monitored Geologic Repository waste forms and engineered barrier system. The PAO responsible manager has evaluated the technical document development activity (CRWMS M&O 1999b) in accordance with QAP-2-0, *Conduct of Activities*. The QAP-2-0 activity evaluation has determined that the preparation and review of this technical document are subject to *Quality Assurance Requirements and Description* (DOE 2000) requirements. In accordance with AP-2.13Q, *Technical Product Development Planning,* a development plan (CRWMS M&O 1999a) was developed, issued, and utilized in the preparation of this document.

The documentation of this analysis is in accordance with the guidance given in AP-3.10Q, *Analyses and Models*.

2.1 DEVIATION FROM THE DEVELOPMENT PLAN

As the work progressed, this analysis inevitably deviated from the Development Plan (CRWMS M&O 1999a) in a few aspects. They are:

- Sr was not planned to be analyzed by TSPA. However, its solubility is requested by the TSPA group. As time constraints prevent performance of a modeling study, it is suggested that the conservative approach be used, i.e., setting Sr solubility to 1.0 mol/L (which will allow the waste inventory to control Sr release) for the TSPA-SR until a detailed modeling study generates a realistic value.

- This analysis utilized two industrial standard software applications (Microsoft Excel and SigmaPlot). Their utilization is necessary to obtain the desired result but was not discussed in the Development Plan.

- Since this analysis has to be conducted in parallel with the in-package chemistry study, the information generated by the latter is not fully considered, as required by the Development Plan.

The above deviations have little or no consequences to the technical adequacy of this AMR.

2.2 TBV CLAIM

Unqualified data were used in this analysis. Therefore, the QA status of the analysis results should be designated "to be verified" (TBV). Analysis results used as inputs must be identified and tracked as TBV in accordance with appropriate QA procedures.

3. COMPUTER SOFTWARE AND MODEL USAGE

The major geochemical modeling tool for this analysis is EQ3/6 Version 7.2b (Wolery 1992a, 1992b; Daveler and Wolery 1992, CSCI: LLNL: UCRL-MA-110662). The software is approved by the YMP for quality affecting work and was obtained from Software Configuration Management.

All of the EQ3NR calculations described in this AMR were conducted on two computers. One is a Hewlett-Packard workstation (central processing unit [CPU] #112515) running the HP-UX B.10.20 operating system, which is located at the Las Vegas, Nevada, CRWMS M&O facility. The proper installation was verified in accordance with the Installation Guide (Wolery 1992a) and QA Procedure AP-SI.1Q, *Software Management*. An installation and test report was prepared and submitted to Software Configuration Management. All the simulations reported in this analysis were made after the proper installation. This computer was used to conduct calculations for uranium (U), neptunium (Np), and plutonium (Pu).

The other computer is located at Lawrence Livermore National Laboratory. It is a Gateway Pentium II 400 MHz desktop PC, running Windows 98, with a CPU # 6476036 (LLNL/DOE). This computer was used to conduct EQ3NR calculations for Am solubility.

The EQ3/6 package consists of several components: EQ3NR, EQ6, EQPT, and EQLIB. EQ3NR is designed for analyzing water chemistry and solubility calculation. EQ6 is for reaction path calculation. EQPT is a database pre-processor, and EQLIB is the supporting library. Since no reaction-path calculation was performed in this analysis, EQ6 was not utilized. This software is appropriate for the application and has not been used outside the range of parameters for which it has been verified.

Other software used in this analysis is Microsoft Excel 97 and SigmaPlot Version 4.0 for Windows 95, NT and 3.1, which are installed on a Dell PowerEdge 2200 PC (CPU # 112378) running Windows NT Version 4.0. Excel was used for spreadsheet analyses, only its internal functions were utilized, and no application/macro was developed. According to Section 2.1.1 of AP-SI.1Q, *Software Management*, it is thus exempted from this procedure. The Excel spreadsheets used in this analysis are included in the electronic media as listed in Attachments I, II, and III. SigmaPlot was used for both regression analysis, which is part of its spreadsheet functions, and as a graphic tool. Since only its internal functions were utilized, and no application/macro was developed, it is exempted from the procedure AP-SI.1Q, *Software Management*. The SigmaPlot spreadsheets used in this analysis are included in the electronic media as listed in Attachments I and II.

4. INPUTS

This analysis takes inputs from two process-level AMRs:

1. *Pure Phase Solubility Limits--LANL* (CRWMS M&O 2000a)
2. *Summary of In-Package Chemistry for Waste Forms* (CRWMS M&O 2000b)

The first AMR provides process level information on solubility limits of radioactive elements, and the second one provides chemical conditions within waste packages (WPs). Based on the information contained in them, this analysis is to generate solubility limits, be it response functions or distributions, which are applicable to the proposed Repository. These data were developed under QA procedures and are thus qualified data, ultimately. However, since the first AMR is still in the development stage, the QA status of the data should be "to be verified/to be determined" (TBV/TBD). The draft of the report has been transmitted from WPO to PAO according to AP-3.14Q (CRWMS M&O 2000a).

4.1 DATA AND PARAMETERS

Two database files for EQ3/6 were used in this study. One file, data0.com.R2, is distributed with the EQ3/6 package (Data Tracking Number [DTN]: MO99SPATHD62.002). The other, data0.an1.V8.R6 (DTN: LA9912WR831372.004) (renamed to data0.an4.R6 [see Attachment II] on HP workstation bhima, CPU # 112515) is a modified version of the data0.com file. The new data compiled in the AMR *Pure Phase Solubility Limits - LANL* (CRWMS M&O 2000a, Tables

2 and 3 for Np and Pu) are incorporated into this file. Table 1 gives a summary of those two database files.

Table 1. Sources of EQ3/6 Database Files

Database File Name	Source/Modifier	Element	Directory	Status	DTN
Data0.com.R2 (see Attachment I)	Released with EQ3/6 Version 7.2b	U, Am	~chen/bin/EQ36/bin	TBV-3916	MO9911SPATHD62.002
Data0.an1.V8.R6 (Data0.an4.R6) (see Attachment II)	CRWMS M&O 2000a, Tables 4, 5, and 6 (AMR F0085)/LANL	Np, Pu, Tc	~chen/bin/EQ36/bin	TBV-4168	LA9912WR831372.004

An activity having the objective of building a uniform and accepted EQ3/6 data file for the YMP (i.e., data0.ymp) is underway. Several geochemists of the YMP are involved in this activity. Unfortunately, this analysis was unable to use the uniform data file, as it is still undergoing development. Nonetheless, the uniform, accepted data file will be used to revise this analysis once it is approved by the Project (expected later this year).

Table 2 gives the source and its DTN for in-package chemical conditions.

Table 2. Source of In-Package Chemical Conditions

Parameters	Source	DTN	Status
Eh, pH, I, Ca, Si	CRWMS M&O 2000b, Table 3	SN9911T0811199.001 and SN9911T0811199.002	TBV4533, TBV-4534
Na, Cl, SiO_2, etc.	Harrar et al. 1990	LL980711104242.054	TBV-3615

Table 3 lists the sources of solubility limits used either directly or indirectly by this analysis for some elements.

Table 3. Sources of Solubility Limits for Some Elements

Parameters	Source	DTN	Status
Solubility ranges for Nb, Ni, Ra, Sn, and Zr	CRWMS M&O 2000a, sections 6.3.5-6.3.10	ACC: MOL.20000217.0217	TBV-4537
Solubility Calculations for Am	Wolery et al., 2000	LL000112051023.004	Qualified

Atomic weights given in the Periodic Table of Elements (Langmuir 1997, inside front cover page) were used to convert solubility limits from units of mol/L to mg/L. They are deemed accepted data (approved by Assistant Manager, Office of Project Execution).

4.2 CRITERIA

One of the objectives of this analysis is to address Sub-issue 3 in the NRC's Issue Resolution Status Report, Key Technical Issue: Container Life and Source Term (NRC 1999); i.e., the rate at

which radionuclides in SNF are released from the EBS through the oxidation and dissolution of spent fuel. This important document gives acceptance criteria to be applied for reviewing the U.S. Department of Energy's (DOE's) License Application. Another NRC Issue Resolution Status Report Key Technical Issue: Total System Performance Assessment and Integration (NRC 1998) has additional acceptance criteria on radionuclide (RN) solubility. Although the ultimate goal is to meet all those acceptance criteria, they are not expected to be fully satisfied in this first round of analysis. This is especially true for the qualification of the thermodynamic databases used in solubility calculations. Data acceptance activity is underway ~~to~~ partially address the data qualification issues (cf. Section 4.1). It is planned that this analysis will be revised once the datafile of data0.ymp is accepted by the Project, and fully satisfying the criteria is expected in the revision.

4.2.1 Transparency and Traceability, Quality Assurance (NRC 1999, Section 4.0)

- The collection and documentation of data, as well as development and documentation of analyses, methods, models, and codes, shall be accomplished under approved quality assurance and control procedures and standards.

- Sufficient data (field, laboratory, and natural analogue) shall be available to adequately define relevant parameters for the models used to evaluate performance aspects of the sub-issues.

- Model outputs shall be validated through comparisons with outputs of detailed process models, empirical observations, or both.

4.2.2 Solubility Controlled Release of Radionuclides (NRC 1999, Section 5.3.4.2)

The solubility limits shall be reevaluated as the near-field environment inside the WP becomes better known.

4.2.3 Solubility Controlled Release of Radionuclides (NRC 1998, Section 4.1.1.1.4)

- Sufficient data (field, experimental, and/or natural analogue data) shall be available to adequately define relevant parameters and conceptual models necessary for developing RN release rates and solubility limits abstracted in TSPA.

- Parameter values, assumed ranges, and probability distributions and/or bounding assumptions used in the RN release rates and solubility limits abstraction, such as the pH, temperature, and amount of liquid contacting the waste forms, shall be technically defensible and reasonably account for uncertainties and variabilities.

- Alternative waste form dissolution and RN release modeling approaches consistent with available data and current scientific understanding shall be investigated, and results and limitations appropriately factored into the RN release rates and solubility limits abstraction.

- RN release rates and solubility limits abstraction output shall be verified through comparison to output of detailed process models and/or empirical observations (laboratory testing or natural analogues, or both).

- Important design features, physical phenomena and couplings, and consistent and appropriate assumptions shall be incorporated into the RN release rates and solubility limits abstraction.

4.3 CODES AND STANDARDS

Not applicable.

5. ASSUMPTIONS

Assumptions made in this AMR, their basis, and where they are used are as follow:

5.1 It is assumed that there is no direct interaction between radioactive elements and Cr and Ni. The basis for this assumption is that the EQ3/6 databases do not contain species such as RN(CrO4-) or RN(Ni). The only exception to this is the element lead (Pb), which is known to form PbCrO4(s) (crocoite). Since only aqueous species could increase the solubility of an element, the existence of PbCrO4(s) will not make the solubility calculations for Pb non-conservative.Thus, the effect of high Cr and Ni concentrations on solubility is indirect, through ionic strength to their activity coefficients. This assumption needs to be verified in the future and a TBV number (TBV-4535) has been assigned. This assumption is utilized in Section 6.2.

5.2 For many radioactive elements, such as Pa and Cm, the identification of controlling solids by experiments has yet to be reported. For situations like this, a conservative approach is, as suggested by Bruno et al. (1997, p. 81), to assume the amorphous solids (oxide or hydroxide) as their controlling solids. The basis for this conservative assumption is the Ostwald Step Rule (Langmuir 1997, p.324). Another reason to do so is that irradiation associated with spent nuclear fuel could damage the lattice structure of solids and make it less crystalline (Rai and Ryan 1982, p.216). This assumption is utilized in Sections 6.3 and 6.5.

5.3 It is assumed that there are no solubility-controlling solids under the repository conditions for technetium (Tc), carbon (C), iodine (I), chlorine (Cl), cesium (Cs), and strontium (Sr). The basis for this assumption is that those elements are very soluble. This is a conservative bounding condition and does not need further verification. This assumption is utilized in Sections 6.12, 6.13, 6.14, 6.19, 6.22, and 6.23.

5.4 It is assumed that schoepite is the solubility-controlling mineral for U. The basis for this assumption is that schoepite appears in both natural analog sites and laboratory experiments. This is a conservative bounding condition and does not need further verification. This assumption is used in Section 6.3.

5.5 It is assumed that $Pu(OH)_4(am)$ is the solubility-controlling mineral for Pu. The basis for this is that $Pu(OH)_4(am)$ appears in Pu solubility experiments (CRWMS M&O, 1999b). This is a conservative bounding condition and does not need further verification. This assumption is used in Section 6.5.

6. ANALYSIS/MODEL

In accordance with the Development Plan (CRWMS M&O 1999a, Tasks 2, 3, and 4), this analysis should produce solubility limits for 21 radioactive elements. Depending on their contributions to radiation dose, they are classified into two groups. Group 1 is elements with great impact on dose calculation and calls for detailed study, while Group 2 consists of less important elements (CRWMS M&O 1998a, Appendix C). The Development Plan requires that for three elements in Group 1, i.e., Np, U, and Am, their solubility limits be presented as functions of environmental conditions (pH, redox potential [Eh], temperature [T], and CO_2 fugacity [f_{CO2}]). For the rest of Group 1 and Group 2 elements, stochastic distributions will be developed, as shown in Table 4. As suggested in *Pure Phase Solubility Limits–LANL* (CRWMS M&O 2000a, Section 6.3.3), for the purpose of solubility evaluation, Am can be used as an analogue for curium (Cm) and samarium (Sm). Actinium (Ac) is also considered as analogous to Am (CRWMS M&O 1998a, Section 6.4.1.3) Thus, no separate solubility evaluation for Ac, Cm, and Sm has been conducted.

Table 4. Radioactive Elements to be Analyzed

Group		Element	Solubility Type
Group 1	1A	uranium (U), neptunium (Np), and americium (Am) (actinium [Ac], curium [Cm], samarium [Sm])	Response Surfaces
	1B	plutonium (Pu), thorium (Th), niobium (Nb), technetium (Tc), iodine (I), and carbon (C)	Distributions/bounding values
Group 2		nickel (Ni), zirconium (Zr), radium (Ra), tin (Sn), cesium (Cs), chlorine (Cl), protactinium (Pa), lead(Pb), and strontium (Sr)	Distributions/bounding values

Before geochemical models and derivation of solubility limits are discussed element by element, a brief discussion is provided on some common issues about model building and solubility limits derivation.

6.1 TECHNICAL ISSUES IN SOLUBILITY EVALUATION

A meaningful solubility evaluation involves several technical aspects. First, one has to have a good thermodynamic database to work on, along with a geochemical modeling tool. Second, the environmental conditions for which solubility is to be evaluated should be well defined. Third, one has to build a model based on environmental information and chemical properties of RNs using the geochemical modeling tool. Finally, one needs to derive solubility limits based on the model.

As the first and second aspects are just input to this analysis, this discussion will focus on the last two aspects, i.e., how to build a solubility model and how solubility limits are generated and presented. Several technical issues that are common to solubility evaluation will be addressed below. Specific issues related to certain elements will be discussed in relevant sections.

6.1.1 The Definition of Solubility

From the viewpoint of laboratory chemistry, solubility is defined as the concentration of a substance when the solution is saturated with that substance (Atkins 1994, p. 312). This definition implies: 1) solubility is defined in terms of thermodynamics, and 2) solubility is the maximum concentration the substance can reach in solution at equilibrium. In other words, solubility is the concentration of a substance when the substance is at equilibrium with the solution. For this case, the substance is an RN-bearing solid, called solubility-controlling solid.

Performance assessments are more interested in the solubility of specific elements in waters than the solubility of a substance. The solubility of an element, the maximum concentration that the element can reach in solution at equilibrium, is called elemental solubility. In general, elemental solubility strongly depends on water chemistry and varies as physical/chemical conditions change.

Solubility limits (S) are fundamental input for TSPA analyses. They are used to constrain the maximum RN concentrations. In the TSPA model, inventory concentrations (C_{in}) are calculated according to waste form dissolution rate, water volume, and RN inventory. Then, the inventory concentrations are compared against S. The real concentrations that are available for transport are the lesser of C_{in} and S.

Besides solubility, there are many other mechanisms, such as solid solutions and sorption, that control the concentrations of RNs in solutions. Those mechanisms cannot make the concentrations of RNs higher than their solubility limits. As a matter of fact, the net effects of those mechanisms are to lower RN concentrations in solutions. This study limits itself to pure-phase solubility, and exclude solid solution and sorption from current consideration, because it is conservative to do so.

6.1.2 Identification of the Controlling Solid

As discussed above, for a radioactive element, solubility is defined with respect to a solid. To evaluate solubility for a repository, one has to identify the controlling solid. Since solubility depends strongly on the solid phase, the outcome would be quite different (orders of magnitude) when different solids are chosen.

Laboratory experiments and observations of natural systems provide the basis for choosing the controlling phase. For example, experiments with Np in J-13 Well water (Harrar et al. 1990) show $Np_2O_5 \cdot xH_2O$ is the controlling solid (Efurd et al. 1998, p. 3896) for the time scale of the

experiments. Unfortunately, laboratory evidence and field observations are not always available for all the RNs. Moreover, a controlling solid for one environment may not be the controlling solid for another environment.

From the viewpoint of thermodynamics, one would tend to choose the most stable solid as the controlling phase, because the thermodynamically less stable phase would be ultimately replaced by the most stable phase. However, due to kinetic effects, a thermodynamically most stable solid may not appear in the expected repository conditions. This fact makes it unreliable to identify the controlling solid purely from thermodynamic considerations.

In fact, a geochemical rule, the Ostwald Step Rule, is a better summary for such situations. The Ostwald Step Rule says that unstable or metastable minerals form first, followed by progressively more stable minerals (Langmuir 1997, p. 324). The formation of $Np_2O_5 \cdot xH_2O$ in Np experiments is a perfect example of the Ostwald Step Rule. The more stable phase is, according to thermodynamic calculation, $NpO_2(s)$ (s stands for solid) (CRWMS M&O 1997a, pp.19-20; Efurd et al. 1998, Figure 5). It is believed that precipitation kinetics is behind the Ostwald Step Rule. In other words, during the process of waste corrosion, more stable minerals may be prevented from precipitating because less stable minerals are kinetically favored. Another good example of the Ostwald Step Rule is the formation of secondary uranyl minerals during the process of spent fuel dissolution. There, less stable schoepite precipitates first; then it is replaced by more stable uranyl silicates (Wronkiewicz et al. 1992, Section 4.2).

The Ostwald Step Rule has significant implications for choosing the controlling phase. To use a more stable phase–rather than the first formed, less stable phase–as the controlling phase for solubility calculations, it is necessary to demonstrate that the less stable mineral will be replaced by the more stable mineral(s) in a shorter period than the characteristic time scale of the problem. Specifically, since the time scale of repository performance for regulatory purpose is 10^4 years, the time scale for more stable mineral(s) to form should be less than 10^2-10^3 years. Simply arguing that the more stable phase will ultimately replace less stable minerals is not convincing, because under certain conditions it may take infinite time for a more stable phase to replace a less stable phase. For example, the mixture of hydrogen and oxygen gases at room temperature is thermodynamically unstable with respect to water, but water will never form from the mixture, unless it is ignited by flame or other means.

For many radioactive elements, such as Pa and Cm, the identification of controlling solids by experiments has yet to be reported. For situations like this, a conservative approach is, as suggested by Bruno et al. (1997, p. 81), to choose the amorphous solids (oxide or hydroxide) as their controlling solids. The Ostwald Step Rule is the main reason to choose an amorphous phase. Another reason to do so is that irradiation associated with spent nuclear fuel could damage the lattice structure of solids and make it less crystalline (Rai and Ryan 1982, p.216).

For some very soluble elements, solids are not expected to precipitate from waters under the repository conditions. The transport of those elements may not be solubility-controlled. An arbitrary large number may be assigned to their S so that their release will be controlled by waste inventory.

6.1.3 Variation and Uncertainty

In general, the solubility of an element for a repository is not a constant. Instead, it changes over a certain range. The spread is caused by two factors. One is the variation in chemical conditions, the other is called uncertainty. Although both variation and uncertainty have similar effects on solubility limits, distinguishing them from each other is beneficial.

As environmental conditions within a repository vary widely, so does solubility of RNs. A meaningful solubility evaluation should account for the variation in solubility caused by the changes in environmental conditions. As long as the ranges of environmental conditions are known (as inputs to this analysis), the ranges of solubility variation can be calculated readily. It is useful to understand the effects of changes in environmental conditions on solubility limits. For example, with that understanding, one can tell how a proposed repository design feature would affect solubility limits and ultimately the repository performance by analyzing its effects on environmental conditions.

Unlike variation, uncertainty is due to lack of understanding of a phenomenon. For performance assessment, it should be reduced as much as possible, although it cannot be totally eliminated.

Uncertainty arises from all of the steps in solubility evaluation. For example, it can be from the identification of a solubility-controlling solid. It can also come from the thermodynamic data used for the calculation. Furthermore, it can be introduced from uncertainty in environmental conditions. Distinguishing uncertainty from variation and understanding the major sources of uncertainty are the prerequisites to reducing uncertainty.

6.1.4 Response Surface vs. Probability Distribution

In performance assessment, it is a common practice to represent solubility limits as statistical distributions. However, statistical distribution is not the only way to represent solubility. As always, there is another way to do so, i.e., to use deterministic models. With a deterministic approach, solubility is represented as a deterministic function of environmental conditions or a response surface. Both statistical distribution and response surface approaches have pros and cons.

The advantages with a response surface approach are: 1) it directly tells how solubility depends on environmental conditions, and 2) variations due to changes in environmental conditions are built into the model; therefore, variation and uncertainty are somewhat separated. However, how to present uncertainties with response surfaces is a question that remains.

Statistical distribution approach presents a picture from statistical point of view. However, it lumps variation and uncertainty together and masks the dependent relationship of solubility on environmental conditions.

For those elements with well-known chemical properties, it is preferred to use a response surface approach to represent their solubility limits. On the other hand, if knowledge of their chemical properties is relatively poor, or if uncertainty may be large, a probability approach may be chosen. In this study, one or the other approach will be used for each element.

6.1.5 Distribution Type and Parameters

If a probability approach is to be used for an element, the next question is how to derive the distribution type for solubility (e.g., uniform or log uniform, or log normal?). In the history of the YMP, the answer to this question has depended on professional judgments. In the solubility evaluation, professional judgment may not be totally eliminated, but it is desirable to have as much objective basis as possible.

To determine the distribution type of a population, one needs to sample the population first, then plot the sampled data in histograms. Visual examination could suggest some distribution types over others. A statistically more rigorous way is to conduct statistical testing of certain hypotheses regarding distribution types. With a certain confidence level, some of the hypotheses will pass the testing while others will be rejected. Due to time constraints, this analysis utilizes a histogram/visual-examination approach to determine distribution type and leave the more rigorous approach for the next revision.

6.1.6 The Role of Empirical Data

Experiments can be classified into two types: one is to obtain fundamental data (designated as Type 1 experiments); the other is to investigate phenomena and processes (Type 2). In general, the data obtained from Type 1 experiments can be applicable to situations that deviates from the conditions from which they are obtained, with certain restrictions. An example of a Type 1 experiment is measurement of Gibbs free energy of certain materials under standard conditions. The purpose of a Type 2 experiment is not to obtain fundamental, universally applicable data. Rather, it is to investigate specific phenomena or processes. The data obtained from Type 2 experiments is empirical data, which may not be universally applicable.

Geochemical model calculations for solubility evaluation need many fundamental data obtained from Type 1 experiments. However, empirical data from natural analogue studies and spent fuel dissolution experiments can provide useful information for solubility evaluation, too. For example, concentrations of RNs in natural systems can be used as a counter-example to check the upper bound of solubilities. The paragenesis sequence of uranium (or other elements) minerals can help in choosing the right controlling mineral(s) for model calculations. Concentrations of RNs observed in natural analogues and laboratory experiments can also be used as corroborating data and to increase confidence in modeling results.

Another way to utilize empirical data is to directly use the RN concentrations measured in laboratory experiments. Compared to solubility limits obtained from conventional geochemical modeling work, they are generally lower and probably more realistic. For example, in the previous TSPAs, the solubility limit for Cm is based on the steady-state concentrations of Cm in spent fuel leaching experiments carried out at Pacific Northwest National Laboratory (PNNL) (CRWMS M&O 1998a, Section 6.4.1.3; Wilson 1990a, Section 3.2.4, 1990b, Section 3.6). Another attempt at using empirical data in solubility evaluation was made by Sassani and Siegmann (CRWMS M&O 1997a). In their interim evaluation of Np solubility, Np concentrations measured in various spent fuel corrosion experiments are utilized. However, their argument is not well received by the regulator and a couple of reviewers (Bell 1998, Item 9; Budnitz et al. 1999, p. 91; NWTRB 1998, p.6).

The fact that this analysis is not well received does not mean empirical data cannot be used; rather, it means that it is a more challenging task to utilize empirical data. What should be remembered is that solubility analyses are bounding analyses. If empirical data are to be used directly with solubility analyses, the bounding property of empirical data should be established with thorough work.

Several steps should be taken to establish the bounding property for empirical data. At first, the principal mechanism that controls the concentrations should be clearly identified. If it is controlled by a pure phase, then standard procedures of pure-phase solubility study will yield the answer. If it is limited by other mechanism(s), then it must be demonstrated that the mechanism(s) controlling the concentration in the experiments still operates in repository conditions over a long period. This step is the most difficult to achieve. However, considering the potential benefits it may bring to the calculated performance of the repository, the efforts to meet the challenge may be well worthwhile.

6.1.7 Summary

Although geochemical calculation plays an important role in solubility evaluation, the final product is a blend of quantitative analyses and professional judgment. There are different recipes to derive solubilities, and in each recipe, the proportion of quantitative analysis and professional judgment varies. For the YMP to progress to license application the proportion of quantitative analysis should be increased and the proportion of professional judgment should be reduced as much as possible. The above discussion seeks to address several technical issues and various approaches that have been proposed to achieve that objective.

As discussed in Section 6.1.2, the conceptual models of solubility evaluation (i.e., the identification of solubility-controlling solid) are conservative and, thus, are adequate to use for performance assessment. They will not be validated at this time.

6.2 IN-PACKAGE CHEMICAL CONDITIONS

A detailed modeling study of in-package chemical conditions has been conducted and is documented in an AMR titled "*Summary of In-Package Chemistry for Waste Forms*" (CRWMS M&O 2000b). It models chemical reactions among the solution and WP materials using the function of solid-centered flow-through of EQ6 (CRWMS M&O 1998b). While a certain amount of water (dripping water) is added to the system, the same amount of solution (reacted water) is withdrawn from the system. The study assumes that "the solutions that drip into the package will have the composition of J-13 well water for ~50,000 years" (CRWMS M&O 2000b, Section 3.1). Due to the reactions among the water and WP materials, the water composition changes.

Unfortunately, this analysis had to be conducted in parallel with the in-package chemistry analysis, due to time constraint. Although the preliminary results of in-package chemistry were informally supplied to this analysis as they became available, most of the results were not available until modeling calculations for this AMR have been done. As a result, this analysis was unable to fully utilize all of the information generated by the in-package chemistry study. This section summarizes the results generated by the in-package chemistry study and also gives

the chemical conditions used for solubility evaluation. Comparing them and applying geochemical reasoning will provide a general idea about the impacts of deviation in chemical conditions. For a few important elements, formal impact analyses will be conducted. However, the only way to fully address this issue is to revise this analysis, if necessary, after the in-package chemistry study is completed.

The in-package chemistry study covers two types of WPs: Civilian Spent Nuclear Fuel (CSNF) WP and Co-disposal WP. Because CSNF is the dominant waste and the resources for this analysis are constrained, this analysis will just consider the results about CSNF WPs.. Table 5 lists the ranges of several important variables given by the study. Also listed in Table 5 are J-13 well water composition and chemical conditions.

Table 5. Ranges of In-Package Fluid Composition of CSNF WPs

Variable	CSNF WPs	J-13 Water (Harrar et al. 1990, Table 5.2)
pH	3.6 - 8.1	7.41
Eh (V)	0.7 - 1.0	0.34
Ionic Strength I (mol/kg)	0.003 - 1.7	0.003
Ca (mol/kg)	3.9E-6 - 3.2E-4	3.25E-4
Si (mol/kg)	3.7E-10 - 1.9E-4	1.01E-3

A comparison between the modeling results (CSNF WPs column in Table 5) and J-13 well water shows the in-package fluid exhibits the following major changes from J-13 well water:

- pH may decrease as low as 3.6, though it may increase to 8.1.
- Eh ranges from 0.7 to 1.0 V. That difference from J-13 well water is due to the conservative assumption of being open to the atmosphere.
- Major cations of J-13 well water, such as Ca and Si, decrease.
- Ionic strength (I) can be as high as 1.7 mol/kg. The major contributors for high ionic strength are Cr and Ni, the corrosion products of stainless steel (CRWMS M&O, 2000b, p.23).

Based on the above observations regarding the in-package chemistry, the following conclusions can be drawn:

- The fluid composition within CSNF WPs has a pH range from 3.6 to 8.1.
- Its Eh is higher than 0.34 V, the measured Eh for J-13 well water.
- The concentrations of major cations of J-13 well water either do not change much or decrease. Therefore, the measured concentrations of those cations for J-13 well water can be used for solubility calculation.

- Since the EQ3/6 databases do not contain species such as $RN(CrO_4^-)$ or RN(Ni), it is assumed that there is no direct interaction between radioactive elements and Cr and Ni. The only exception to this is the element lead (Pb), which is known to form PbCrO4(s) (crocoite). Since only aqueous species could increase the solubility of an element, the existence of PbCrO4(s) will not make the solubility calculations for Pb non-conservative. Thus, the effect of high Cr and Ni concentrations on solubility is indirect, through ionic strength to their activity coefficients.

The last bullet regarding ionic strength demands further elaboration of the effects of ionic strength on solubility and, from a practical point of view, how it should be treated.

The influence of ionic strength on solubility is called salting-in effect (for neutral species, it is called salting-out effect). In other words, higher ionic strength would increase the solubility of electrolyte substances (salting-in) and decrease the solubility of non-electrolyte substances (salting-out).

The salting-in effect can be quantitatively represented through activity correction. There are several models to calculate the activity coefficient of aqueous species. The most widely used formula is the so-called "B-dot" equation (Wolery 1992b, Section 3.3). However, it becomes less accurate when ionic strength is higher than 0.7 mol/kg. Pitzer's model (Wolery 1992b, Section 3.5) is developed for high ionic strength situations, but its application is limited due to lack of data.

To evaluate the effects of ionic strength on solubility calculations, several questions regarding activity correction need to be answered. The first question is, "What are the options?" It may be agreed that Pitzer's model is not practical for most of the problems, because there are not enough Pitzer's coefficients to utilize the model. What remains is the Debye-Hückel equation and its variations, including B-dot equation. They all have the same problem of less accuracy at high ionic strength. The consensus may be that the best choice is the B-dot equation.

Without other choices, a few questions about the B-dot equation must be answered. One is how inaccurate the B-dot equation is for ionic strength as high as 1.7 mol/kg. Another is what is the effect of the inaccuracy on solubility calculation. The absolute answers, based on current knowledge, are not available. However, an estimation of the reliability of the solubility calculations would be helpful.

The valid range of the B-dot equation implemented in EQ3/6 has been studied by Paul Cloke in 1997 (CRWMS M&O 1997b, Appendix D). By comparing activity coefficients calculated by EQ3/6 and those measured by experiments, it was concluded that the EQ3/6 results up to an ionic strength of about 4.0 can be used qualitatively. Since the maximum ionic strength to be considered for solubility evaluation is 1.7, that conclusion is applicable for this analysis. On the other hand, a sensitivity analysis shows that for J-13 well water, increasing its ionic strength to 0.97 mol/kg only increases Np solubility by a factor of 1.7 (see Section 6.4).

Therefore, the conclusion is that the inaccuracy in solubility calculations introduced by a less accurate activity correction equation at a high ionic strength situation is not significant, compared to other uncertainties in solubility model calculations. Moreover, the inaccuracy in solubility

calculation introduced by omitting high ionic strength is small. Thus, although the composition of in-package fluid has some unique characteristics, provided that the pH and Eh ranges are covered, using J-13 well water as the reference water for solubility calculation will not introduce significant errors.

A modeling study regarding the CO_2 composition of the ambient repository atmosphere indicates that, for most of the time, f_{CO2} is about 10^{-3} bar (DOE 1998, Section 3.3.3.1). This is also the value used in the in-package chemistry study (CRWMS M&O, 2000b, Section 5). The range of f_{CO2} in the following analyses is set from $10^{-2.5}$ to $10^{-4.0}$ bar to cover its potential variations.

Due to decay heat from the waste, it is expected that the temperature within WPs will increase from the ambient temperature. Immediately after the emplacement of WPs, their temperature can go as high as 200°C (DOE 1998, Figure 3-22). However, only the temperature below the boiling point of water is interesting to this study. Therefore, the temperature in the following study is set from 30°C to 90°C, which should cover the most likely range in the Repository below the boiling point of water.

Based on the above discussion and the average composition of J-13 well water (Harrar et al. 1990, Table 5.2), the synthesized composition of the reference water to be used for solubility calculations is listed in Table 6.

Table 6. Chemical Composition of Reference Water

Variable	Composition (mg/L)[a]	Source
Na	45.8	DTN: LL980711104242.054
Cl	7.14 (adjustable)[b]	DTN: LL980711104242.054
SiO_2(aq)	60.97	DTN: LL980711104242.054
Ca	13.0	DTN: LL980711104242.054
K	5.0	DTN: LL980711104242.054
Mg	2.01	DTN: LL980711104242.054
Li	0.048	DTN: LL980711104242.054
F	2.18	DTN: LL980711104242.054
NO_3^-	8.78	DTN: LL980711104242.054
SO_4^{--}	18.4	DTN: LL980711104242.054
pH[c]	3.6 - 8.5 standard units	CRWMS M&O, 2000b
Eh	> 0.34 V	CRWMS M&O, 2000b
Log f_{CO2}	-2.5 - -4.0 (bar)	DOE, 1998
T	30°C - 90°C	DOE, 1998

[a]except as noted.

[b]EQ3 has an option of balancing charges automatically by adjusting the concentration of an aqueous species specified by the user. In the following EQ3NR calculations, this feature is utilized so that electrical neutrality is maintained for the aqueous solutions. The adjustable species in most cases is Cl^-. In cases where electrical neutrality could not be reached by adjusting Cl^-, Na^+ is specified.

[c]As mentioned before, the information generated by the study of In-package Chemistry on pH was not fully utilized in this study. In most of the calculations in this study, pH varies from 4.5 to 8.5.

6.3 URANIUM (U) SOLUBILITY

6.3.1 Thermodynamic Data

After several years of extensive review, the Nuclear Energy Agency (NEA) published "Chemical Thermodynamics of Uranium" in 1992 (Grenthe et al.). Lawrence Livermore National Laboratory (LLNL) has submitted that set of data to the Technical Data Management System (TDMS).

EQ3/6 Version 7.2b has several companion thermodynamic databases (DTN: MO9911SPATHD62.002). One of them is the *.NEA data file. This data file contains the above mentioned data set. However, the equilibrium constant of schoepite in that data file does not change with temperature. Another data file that contains the NEA dataset is the *.COM file, in which the equilibrium constant of schoepite is temperature dependent. Since it is well known that U solubility is temperature dependent (Murphy 1997), the data file of data0.com.R2 is chosen for this calculation to include temperature effects.

6.3.2 Controlling Mineral

Once the WPs are breached and water enters, spent nuclear fuel will react with the incoming water or water vapor. As a result, uranyl (UO_2^{2+}) minerals will precipitate under oxidizing conditions. Laboratory experiments and field observations show that the most common secondary uranyl minerals expected to form in the Repository are schoepite, soddyite, uranophane, and/or Na-boltwoodite. Schoepite is chosen as the controlling mineral for this analysis for several reasons:

1. Schoepite is the first mineral to be formed during the process of spent fuel corrosion (Wronkiewicz et al. 1992, Section 4.2),
2. Field observations and modeling study show that schoepite can last more than 10,000 years, albeit thermodynamically unstable for the repository conditions (Pearcy et al. 1994, p. 718-719; Finch et al. 1996, Table 1),
3. Schoepite is less stable than the other above mentioned minerals; using it as the controlling phase will make this analysis conservative for U solubility, and
4. The temperature dependency of the equilibrium constant of schoepite is known.

The conceptual model that schoepite is the solubility-controlling mineral for U is conservative and, thus, adequate for use in TSPA and does not require validation this time.

6.3.3 Sensitivity Analyses

The Development Plan (CRWMS M&O 1999a) specifies that U solubility should be a function of Eh, fugacity of CO_2, temperature, and pH, , and. However, under certain circumstances, U solubility may not be sensitive to all the four variables. Sensitivity analyses would show whether all of the parameters should be included in the uranium solubility model or whether some of them can be omitted.

Four groups of calculations have been carried out. The chemical composition of the water for those calculations is J-13-like water. The controlling mineral is schoepite, unless otherwise noted. One of the four parameters (Eh, f_{CO2}, and T, and pH) varies in each group of calculations. Table 7 lists the calculated results.

Table 7. Results of Sensitivity Calculations for Uranium Solubility

Input File	Sensitivity Analyses On	Value	U Solubility (mg/L)	Note
Usens01.3i	Eh (V)	0.34	4.31	
Usense1.3i		0.76	4.31	
Usense2.3i		0.10	4.31	
Usense3.3i		-0.10	1417.9	
Usense4.3i		-0.10	14.02	Controlling phase: UO_2(am)
Usens01.3i	Log f_{CO2} (bar)	-3.0	4.31	
Usensf1.3i		-2.0	102.03	
Usensf2.3i		-4.0	1.00	
Usens01.3i	T (°C)	30	4.31	
Usenst1.3i		60	0.44	
Usenst2.3i		90	0.15	
Usens01.3i	pH	7.41	4.31	
Usensp3.3i		6.51	1.23	
Usensp5.3i		8.31	172.28	

The table shows that the solubility of schoepite in J-13-like water is not sensitive to Eh change between 0.76 and 0.10 V. It is expected that the Eh for the repository is most likely falling into the 0.1 to 0.76 V range. Hence, Eh could be excluded from this U solubility model. The results also show that schoepite solubility depends on pH, temperature, and the fugacity of CO_2.

6.3.4 Independent Variables and Their Ranges

The water composition used for U solubility calculation is the reference water listed in Table 6, with pH, temperature, and fugacity of CO_2 changing over the given ranges independently. Table 8 lists the data points for each variable that are used to calculate U solubility. The data matrix consists of 9x4x4 = 144 data points. However, a few data points are out of the range the code

can handle, i.e., the calculation cannot generate a converging solution. Thus, the number of data points is less than 144 (128).

The convergence problems occur at either high pH or low pH, due to computational limits of the geochemical modeling tool. It imposes restrictions on the ranges of conditions the model calculations can be performed. It in turn limits the valid ranges of the response surface.

Table 8. Data Matrix

Parameter	Number of Grid Points	Grid Points
pH	9	5.0, 5.5, 6.0, 6.5, 7.0, 7.5, 8.0, 8.5, 9.0
T (°C)	4	30, 50, 70, 90
Log f_{CO2} (bar)	4	-2.5, -3.0, -3.5, -4.0

6.3.5 Results and Response Surfaces

Table 9 lists the calculated U solubility for 128 data points, along with the conditions. The calculated U solubility ranges from 0.11 mg/L (4.69E-7 mol/L) to 101.95 mg/L (4.28E-4 mol/L), with an average of 2E-5 mol/L. Based on the data, more than 14 non-linear regression analyses have been performed using SigmaPlot Version 4.0. Based on the r^2 values, the following 4 regression equations are those that best fit among those regression analyses (see computer file of Regression.JNB of Attachment I).

Table 9. Calculated U Solubility (mg/L)

	Log f_{CO2} = -2.5 (bar)			
pH	T=30° C	T=50° C	T=70° C	T=90° C
7.0	5.37	1.04	0.35	0.19
6.5	2.22	0.60	0.27	0.18
6.0	1.68	0.65	0.36	0.26
5.5	3.60	1.59	0.92	0.68
5.0	14.94	7.50	4.41	3.29
7.5	20.61	3.79	0.94	0.33
8.0	(a)	42.83	7.42	1.44
8.5	(a)	(a)	(a)	66.42
9.0	(a)	(a)	(a)	(a)
	Log f_{CO2} = -3.0			
pH	T=30° C	T=50° C	T=70° C	T=90° C
7.0	2.04	0.50	0.21	0.14
6.5	1.23	0.42	0.21	0.15
6.0	1.31	0.57	0.32	0.23
5.5	3.42	1.53	0.88	0.88
5.0	14.82	7.45	4.38	3.26
7.5	5.28	0.98	0.31	0.16
8.0	23.04	4.06	0.95	0.31
8.5	(a)	101.95	12.34	2.02
9.0	(a)	(a)	(a)	(a)

	Log f_{CO2} = -3.5			
pH	T=30° C	T=50° C	T=70° C	T=90° C
7.0	1.11	0.36	0.17	0.12
6.5	0.92	0.37	0.20	0.14
6.0	1.20	0.54	0.31	0.22
5.5	3.36	1.51	0.87	0.64
5.0	14.78	7.43	4.37	3.25
7.5	1.98	0.46	0.19	0.12
8.0	5.36	0.99	0.30	0.16
8.5	31.32	5.06	1.11	0.35
9.0	(a)	(a)	32.14	4.38
	Log f_{CO2} = -4.0			
pH	T=30° C	T=50° C	T=70° C	T=90° C
7.0	0.83	0.32	0.16	0.12
6.5	0.83	0.36	0.19	0.14
6.0	1.16	0.53	0.30	0.22
5.5	3.34	1.51	0.87	0.64
5.0	14.76	7.43	4.37	3.25
7.5	1.07	0.34	0.16	0.11
8.0	2.01	0.47	0.19	0.12
8.5	5.76	1.08	0.34	0.18
9.0	(a)	8.32	1.62	0.49

a: No data because the EQ3NR calculations do not converge.

Equation 1 is:

$$\log[U] = -7.3133 + 5.5614pH + 0.4298pH^2 + 1.9285\log f_{CO2} - 7.3377\log(1/T) + 0.4164pH \times \log f_{CO2} + 3.9531pH \times \log(1/T) + 1.6967\log f_{CO2} \times \log(1/T) \quad \text{(Eq. 1)}$$

Equation 2 is:

$$\log[U] = -19.5221 + 5.2794pH + 0.4290pH^2 - 2.3196\log f_{CO2} - 12.2087\log(1/T) + 0.4112pH \times \log f_{CO2} + 3.8444pH \times \log(1/T) \quad \text{(Eq. 2)}$$

Equation 3 is:

$$\log[U] = 7.9946 - 2.6963pH + 0.4292pH^2 - 1.6286\log f_{CO2} + 0.0095T + 0.4161pH \times \log f_{CO2} - 0.0051pH \times T - 0.0022\log f_{CO2} \times T \quad \text{(Eq. 3)}$$

Equation 4 is:

$$\log[U] = 14.455 - 6.1474pH + 0.4303pH^2 - 3.0984\log f_{CO2} - 1077.7099/T + 0.4166pH \times \log f_{CO2} + 575.3238pH/T + 247.7642(\log f_{CO2})/T \quad \text{(Eq. 4)}$$

where $[U]$ is the total concentration of uranium in mg/L, f_{CO2} is the fugacity of CO_2 in bar. T is the temperature in Kelvin. Temperature is in units of Kelvin, as in most thermodynamics relations (same for Equations 2 to 4).

Some of the statistical parameters for these four equations are summarized in Table 10. Table 10 reveals that statistically, all of these 4 equations fit the data quite well ($r^2 > 0.90$, $P < 0.0001$). The differences among them are minor, except that Equation 3 passes the normality test while the others do not. Since normal distribution is an inherent condition of SigmaPlot, the normality test should be regarded as an important criterion. Therefore, Equation 3 may be a better fit than the others.

Table 10. Statistical Summary of Regression Analyses

Parameter[a]	Equation 1	Equation 2	Equation 3	Equation 4
r^2	0.9264	0.9248	0.9237	0.9285
Standard Error	0.1903	0.1915	0.1937	0.1876
P	<0.0001	<0.0001	<0.0001	<0.0001
Normality Test	Failed	Failed	Passed	Failed
Constant Variance Test	Passed	Passed	Passed	Passed

[a]The exact meanings of each statistical parameter can be found in the Reference Manual of Transform & Regressions, SigmaPlot 4.0 for Windows, SPSS Inc., 1997.

6.3.6 Impact Analysis for Low pH

The response surface obtained in the above is based on calculations for pH from 5.0 to 9.0. Later, it is predicted by the in-package chemistry study that pH can go as low as 3.6 for CSNF WPs. Although it is believed that the probability for the extreme low pH to occur is very low, an impact analysis was conducted to see if it is adequate to use Equation 3 for the low pH situations.

Table 11 lists the predicted U solubility for 3 different pHs using Equation 3, along with the calculated U solubility by EQ3NR. The temperature is set to 30°C, and $\log f_{CO2}$ is set to -3.0. The table shows that U solubility predicted by the response surface is higher than that calculated by EQ3NR. The difference decreases with the increase in pH. It is concluded that Equation 3 gives a conservative prediction for U solubility for low pH situations. Thus, it is adequate for use in TSPA-SR. Although the difference is just about 50 percent at the extreme situation, it may be desirable to revise the response surface to include low pH conditions.

Table 11. Comparison of Predicted U Solubility (mg/L) by Equation 3 with that of EQ3NR

	Predicted by Equation 3	Predicted by EQ3NR
pH = 3.6	3607.06	2278.4
pH = 4.0	463.08	292.82
pH = 4.5	55.52	51.50

6.3.7 Summary and Discussion

Using the EQ3/6 software package and its companion database data0.com.R2, and assuming schoepite is the controlling mineral, uranium solubility is obtained as a function of pH, temperature, and f_{CO2}, as previously identified in Equation 3:

$$\log[U] = 7.9946 - 2.6963pH + 0.4292pH^2 - 1.6286\log f_{CO2} + 0.0095T + 0.4161pH \times \log f_{CO2} - 0.0051pH \times T - 0.0022\log f_{CO2} \times T$$

where $[U]$ is the total concentration of U in mg/L (or g/m^3), T is the temperature in Kelvin, and f_{CO2} is the fugacity of CO_2 in bar.

The model is conservative because the more stable uranyl phases are not used for the calculation. Therefore, the model is adequate for use in TSPA-SR. To be more realistic (e.g., using uranophane as the controlling mineral for the calculation), more accurately measured thermodynamic data are required for this mineral, such as its formation energy and heat capacity and a detailed model to account for the effects of the precipitation of uranophane itself on the concentrations of Si and Ca, etc.

6.4 NEPTUNIUM (Np) SOLUBILITY

6.4.1 Thermodynamic Data

Thermodynamic data measurements, evaluations, and compilation were documented in the AMR titled *Pure Phase Solubility Limits--LANL* (CRWMS M&O 2000a, AMR F0085). The data have been submitted to TDMS and have a DTN: LAWR831372AN99.002. An EQ3/6 data file (data0.an1.V8.R6) was created by the authors of the report by modifying data file data0.com.V8.R6 and was used in this analysis (DTN: LA9912WR831372.004). The data file was renamed as data0.an4.R6 (which is submitted on a CD-ROM and listed in Attachment II) on an HP workstation (CPU # 112515).

6.4.2 Controlling Mineral

There has been lengthy discussion about Np-bearing phase(s) that could form under the Repository conditions (cf. CRWMS M&O, 1998b). Thermodynamically, NpO_2 is the stable phase (CRWMS M&O 1997a, pp. 19-20. However, it has not been observed in solubility experiments, except for some unusual conditions. It is believed that a kinetic barrier prevents NpO_2 from precipitating.

$NaNpO_2(CO_3)\cdot xH_2O$ was observed in experiments (Nitsche et al. 1993a, p. 37). However, a detailed analysis by Runde in *Pure Phase Solubility Limits–LANL* (CRWMS M&O 2000a) found that $NaNpO_2CO_3\cdot 3.5H_2O$ is stable only when $[Na^+]$ is greater than 0.05 molar at neutral pH. At pH 10, $[Na^+]$ greater than 0.5 molar is required for $NaNpO_2CO_3\cdot 3.5H_2O$ to be stable. For the Repository, where Na^+ content is expected to be in the millimolar range, $NaNpO_2CO_3\cdot 3\cdot 5H_2O$ is not expected to precipitate.

By further analyzing the stability field for Np(V) solid phases (Np_2O_5, $NpO_2(OH)$, and $NaNpO_2CO_3 \cdot 3 \cdot 5H_2O$), the report concludes that Np_2O_5 is the solubility controlling phase in J-13 well water. However, if Eh is below 0.12 V, $Np(OH)_4$(am) will be the solubility controlling mineral for Np. In the following discussion, Np_2O_5 (or $Np(OH)_4$(am) for reducing conditions) will be assumed as the solubility controlling mineral.

The conceptual model that Np_2O_5 is the solubility controlling mineral for Np is based on laboratory observations and thermodynamic considerations. It is conservative and, thus, adequate for use in TSPA. No validation is needed.

6.4.3 Sensitivity Analyses

The Development Plan (CRWMS M&O 1999a, Section 1, Task 2) specifies that Np solubility should be a function of temperature, pH, fugacity of CO_2, and Eh. However, under certain circumstances, Np solubility may not be sensitive to all of the four parameters. Sensitivity analyses would show whether all of the parameters should be included in the response surface or some of them can be omitted.

In fact, as observed from experiments, Np solubility is not sensitive to temperature changes (CRWMS M&O 2000a, p. 23). It was reported that "With increasing temperature a slight decrease in solubility is observed at pHs 7 and 8.5, while at pH 6 the neptunium solubility remains approximately constant." Therefore, the temperature effects on the thermodynamic data were not accounted for in the AMR. Consequently, this analysis will not consider solubility dependency on temperature.

Three groups of calculations have been carried out. The chemical composition of the water for those calculations is J-13-like, as given in Table 6. The controlling mineral is Np_2O_5. One of the three parameters (pH, Eh, and f_{CO2}) varies in each group of calculations. Table 12 lists the calculated results.

Table 12. Sensitivity Analysis for Np Solubility

Input File	Sensitivity Analysis on	Value	Np Solubility (mg/L)	Note
Npsens01	Eh (V)	0.34	3.92	Controlling mineral: Np_2O_5
Npsens04		0.76	3.92	
Npsens06		0.90	3.92	
Npsens01	Log f_{CO2} (bar)	-3.5	3.92	pH = 7.41
Npsensf1		-2.5	4.28	pH = 7.41
Npsensf2		-4.5	3.88	pH = 7.41
Npsensf3		-3.0	57.05	pH = 9.5
Npsensf4		-3.5	6.80	pH = 9.5
Npsens01	pH	7.41	3.92	
Npsensp1		8.41	0.77	
Npsensp2		6.41	38.80	

Npsens01	Ionic Strength (mol/L)	0.002	3.92	Base value = 1.0
Npsensl1		0.02	4.34	Increased by 1.11 (to base value)
Npsensl2		0.20	5.45	Increased by 1.39
Npsensl4		0.94	6.74	Increased by 1.72

The table shows that the solubility of Np_2O_5 in J-13-like water is not sensitive to Eh change between 0.34 and 0.90 V. Thus, Eh is excluded from the Np solubility model.

The results also show that Np solubility strongly depends on pH, but its dependency on CO_2 fugacity is not significant. As one can expect, this dependency is stronger at higher pH than at neutral pH. From this sensitivity study alone, it is not clear if f_{CO2} should be excluded from the model or not. Hence, f_{CO2} is included for this study.

The fourth group of calculations listed in Table 12 investigates the effects of ionic strength on Np solubility. Np solubility under varying ionic strengths (by increasing the concentrations of Na^+ and Cl^-) were calculated while the other conditions were unchanged. The results clearly show that Np solubility increases with ionic strength. While ionic strength increases from 0.002 to 0.94 mol/L, solubility increases at most 1.72 times, which is quite small compared with its changes caused by other variations. Therefore, ionic strength is not included in the model.

6.4.4 Independent Variables and Their Ranges

The water composition listed in Table 6 is used for Np solubility calculations. Temperature is fixed at 25°C and Eh is fixed at 760 mV. However, pH and fugacity of CO_2 will be changed independently. The fugacity of CO_2 will change from $10^{-2.5}$ to $10^{-4.0}$ (cf. Table 6), and pH will change from 4.5 to 8.5. Table 13 lists the data points for each variable. The data matrix consists of 9x4 = 36 data points.

Table 13. Ranges of Parameters for Np Solubility

Parameter	Number of Grid Points	Grid Points
pH	9	4.5, 5.0, 5.5, 6.0, 6.5, 7.0, 7.5, 8.0, 8.5
Log f_{CO2}	4	-2.5, -3.0, -3.5, -4.0

6.4.5 Results and Response Surfaces

Table 14 lists the calculated Np solubility for 36 data points, along with the conditions.

Table 14. Calculated Np Solubility (mol/L)

	Log f_{CO2}			
pH	-2.5	-3.0	-3.5	-4.0
4.5	0.015	0.015	0.015	0.015
5.0	4.40E-3	4.40E-3	4.40E-3	4.40E-3

5.5	1.36E-3	1.36E-3	1.36E-3	1.36E-3
6.0	4.28E-4	4.28E-4	4.28E-4	4.28E-4
6.5	1.35E-4	1.35E-4	1.35E-4	1.35E-4
7.0	4.32E-5	4.28E-5	4.27E-5	4.26E-5
7.5	1.54E-5	1.41E-5	1.37E-5	1.35E-5
8.0	1.08E-5	6.23E-6	4.88E-6	4.46E-6
8.5	7.09E-5	8.41E-6	3.33E-6	1.97E-6

The calculated Np solubility ranges from 2E-6 to 1.5E-2 mol/L, with an average of 2.3E-3 mol/L.

The table shows that at pH < 7.5, the change in f_{CO2} has a very small effect on Np solubility. Only for pH = 8.5 does Np solubility changewith f_{CO2}. This is because Np(V) carbonate complexes are not important until pH 8 (CRWMS M&O 2000a, Figure 6; Silva and Nitsche 1995, Figure 8). As the model calculation for in-package chemistry shows that the maximum pH is just 8.1, f_{CO2} could be excluded from the Np solubility model.

Therefore, for pH less than 8 and oxidizing conditions, Np solubility depends only on pH, provided that $Np_2O_5(s)$ is the controlling mineral. In other words, [Np] = f(pH). This function can be derived from equilibrium conditions. The reaction controlling Np solubility can be written as:

$$Np_2O_5(s) + 2H^+ = 2NpO_2^+ + H_2O \qquad \text{(Eq. 5)}$$

The equilibrium constant for this reaction is $10^{5.2}$ (CRWMS M&O 2000a, Table 4, DTN: LAWR831372AN99.002).

Another important Np species for the conditions of interest is $NpO_2OH(aq)$ (aq stands for aqueous). One of its speciation reactions is:

$$NpO_2OH^-(aq) + H^+ = NpO_2^+ + H_2O \qquad \text{(Eq. 6)}$$

which has an equilibrium constant of $10^{11.3}$ (this equilibrium constant is obtained from the equilibrium constant of another reaction of species $NpO_2OH(aq)$ given in CRWMS M&O 2000a, Table 4, DTN: LAWR831372AN99.002).

At equilibrium, $Np_2O_5(s) + 2H^+ = 2NpO_2^+ + H_2O$ yields

$$\frac{[NpO_2{}^+]^2}{[H^+]^2} = 10^{5.2} \qquad \text{(Eq. 7)}$$

where [] denotes molal concentration,

or:

$$[NpO_2^+] = 10^{2.6}[H^+] \qquad \text{(Eq. 8)}$$

From Equation 6:

$$\frac{[NpO_2^+]}{[NpO_2OH][H^+]} = 10^{11.3} \qquad \text{(Eq. 9)}$$

or:

$$[NpO_2OH] = \frac{[NpO_2^+]\cdot 10^{-11.3}}{[H^+]} \qquad \text{(Eq. 10)}$$

Thus, the total concentration of Np is:

$$\begin{aligned}[Np] &= [NpO_2^+] + [NpO_2OH] = 10^{2.6}[H^+] + \frac{[NpO_2^+]\cdot 10^{-11.3}}{[H^+]} \\ &= 10^{2.6}[H^+] + \frac{10^{2.6}[H^+]\cdot 10^{-11.3}}{[H^+]} \\ &= 10^{-8.7} + 10^{2.6}[H^+] = 2.0E-9 + 398[H^+]\end{aligned} \qquad \text{(Eq. 11)}$$

The above equation was obtained under the assumption of ideal solution. Since ionic strength correction is too complicated to calculate by hand, the geochemical modeling tool, EQ3NR, will be used for that purpose.

In fact, simply fitting the data in Table 14 using SigmaPlot with the constraint that the constant term is bigger than zero yields that:

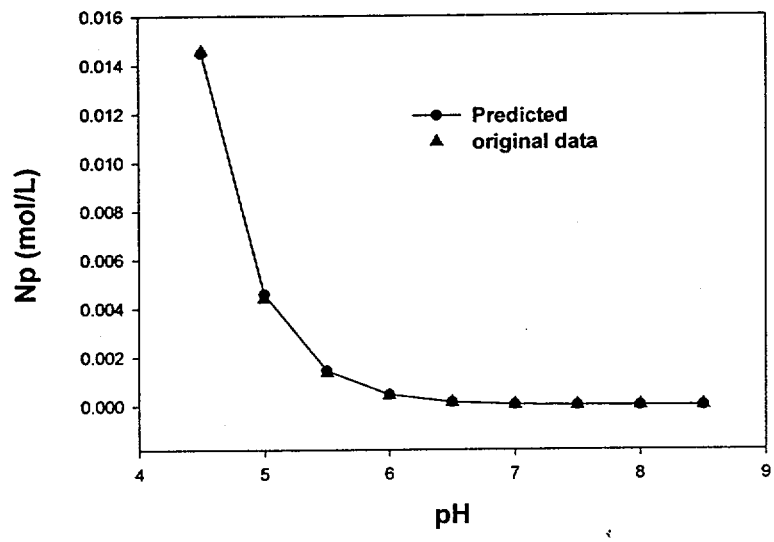
$$[Np] = 3.18E-13 + 458.19 \times a_{H^+} \qquad \text{(Eq. 12)}$$

where a_{H+} is the activity of H^+, [Np] is Np concentration in mol/L.

The r^2 value for this regression equation is 0.9997 (cf. the computer file of "regression-2.JNB" listed in Attachment II). In other words, 99.97 percent of data variance is represented by the regression equation. The good match of the response surface to the original data can be also seen from Figure 1.

If the concentration of Np is in units of mg/L, then Equation 12 can be converted, provided that the density of the solution is 1.0:

$$[Np](mg/L) = 7.538E-8 + 1.086 \times 10^{8-pH} \qquad \text{(Eq. 13)}$$



NOTE: The response surface matches the original data very well.

Figure 1. Comparison of Np Solubility Predicted by the Response Surface and the Original Data

6.4.6 Impact Analysis for Low pH

The response surface obtained above is based on calculations for pH from 4.5 to 8.5. Later, it is predicted by the in-package chemistry study that pH can go as low as 3.6 for CSNF WPs. Although it is believed that the probability for the extreme low pH to occur is very low, an impact analysis was conducted to see if it is adequate to use Equation 13 for the low pH situations.

Table 15 lists the predicted Np solubility at 2 different pHs using Equation 13, along with the calculated Np solubility by EQ3NR. The $\log f_{CO2}$ is set to -3.0. The table shows that Np solubility predicted by the response surface is about 10 percent lower than that calculated by EQ3NR. Thus using the equation to calculate Np solubility when 3.6 < pH < 4.0, an error of 10 percent may be introduced. This equation will be revised when this AMR is revised.

Table 15. Comparison of Predicted U Solubility (mg/L) with that of EQ3NR

pH	Predicted by Equation 13	Predicted by EQ3NR
3.6	2.73E+04	3.0E+04
4.0	1.09E+04	1.2E+04

6.4.7 Summary and Discussion

Equation 13 is recommended for TSPA analyses. This model is valid for pH from 4.5 to 8.5, and for f_{CO2} from 10^{-4} to $10^{-2.5}$ bar. When $3.6 < pH < 4.0$, the equation may understate Np solubility by 10 percent.

6.5 PLUTONIUM (Pu) SOLUBILITY

6.5.1 Thermodynamic Data

Thermodynamic data measurements, evaluation, and compilation were documented in the AMR titled *Pure Phase Solubility Limits--LANL* (CRWMS M&O 2000a). An EQ3/6 data file (data0.an1.V8.R6) was created by the authors of the report by modifying data file data0.com.V8.R6 and was used in this analysis (DTN: LA9912WR831372.004). The data file was renamed as data0.an4.R6 (see Attachment III) on the HP workstation.

6.5.2 Controlling Mineral

Despite numerous studies regarding Pu solubility, understanding of the stable fields of Pu solids still bears a large uncertainty. The most studied Pu solids are PuO_2 and $Pu(OH)_4$(am) (or $PuO_2 \cdot xH_2O$, where am stands for amorphous). The experiment conducted at Los Alamos National Laboratory (LANL) with J-13-like water does not find the formation of plutonium carbonates (Efurd et al. 1998, p. 3897). Although a Pu-bearing phase has been detected by transmission electron microscopic analyses of samples from drip tests on ATM103 fuel (CRWMS M&O 2000c, Section 6.4), its formula and thermodynamic properties are unknown. Thus, it is not possible at the present to use it as the controlling phase for solubility analysis. Nonetheless, it provides the basis for an alternative model for Pu solubility, as discussed in the end of this section.

Solids precipitated from LANL's over-saturation experiments (CRWMS M&O 2000a, Section 6.1) have a color of dark green, which is characteristic of Pu(IV) solid phases. X-ray diffraction data match the data reported for PuO_2. However, the diffuse and broad X-ray diffraction peaks suggest poor crystalline structures. Nonetheless, precipitates at higher temperature (90°C) have a sharper X-ray pattern than solids of lower temperature.

In another Pu solubility experiment for Yucca Mountain waters, similar results were obtained (Nitsche et al. 1993a, p.63). In that study, at least two solid phases have been observed for experiments at 90°C. One is a yellow-green powdery phase, probably non-crystalline. The other is darker green clumps. Nitsche et al. (1993a, p. 63) believe that "such a combination of crystalline and amorphous materials in this solid can explain the observed powder pattern, which is composed of both very sharp and diffuse lines."

Therefore, it appears that the solubility-controlling minerals in those laboratory experiments are "plutonium hydroxides and/or plutonium colloids, aging towards $PuO_2 \cdot xH_2O$" (CRWMS M&O 2000a, Section 6.1). The value of X should vary from 2 to zero. For X = 2, it is $Pu(OH)_4$, the amorphous end member. For X = 0, it is PuO_2, the crystal end member. Since the crystalline

phase has been formed within laboratory time scale, it is reasonable to assume that over geological time, plutonium hydroxides will convert to PuO_2(c) (c stands for crystalline), and it should be used as the solubility-controlling mineral for Pu for the repository.

However, another process makes the selection of the controlling mineral for Pu solubility calculation less straightforward. This process is the damage of the crystal structure of Pu solids caused by α-decay of Pu isotopes. Rai and Ryan (1982) reported that in 1300 days, $^{238}PuO_2$(c) was found to convert to a less crystalline form of PuO_2, denoted as PuO_2(lc). On the other hand, over the same period, they observed Pu hydroxide gradually converted to anhydrous crystalline material. They concluded that a steady state material that has the properties between those of the true crystalline PuO_2 and those of amorphous hydroxide will form over time and control Pu solubility. Unfortunately, no quantitative properties, such as Gibbs free energy, of this less crystalline material have been reported. Thus, it cannot be used as the solubility-controlling mineral for Pu for this solubility calculation. In order to be conservative, $Pu(OH)_4$(am) will be used as the controlling solid for Pu solubility calculation.

The conceptual model that $Pu(OH)_4$(am) is the solubility-controlling mineral for Pu is conservative and, thus, adequate for use in TSPA-SR. It does not require validation.

6.5.3 Independent Variables and Their Ranges

The water composition used for Pu solubility calculation is the reference water listed in Table 6. The effects of several environmental variables, including temperature, pH, Eh, and fugacity of CO_2, should be considered in Pu solubility calculation. However, temperature dependency of the equilibrium constant for the solubility controlling mineral (i.e., $Pu(OH)_4$(am)) is not available. Without that information, the inclusion of temperature effects into Pu solubility models is not very meaningful, so it is omitted for this analysis. If the data become available later, then this analysis will be revised.

Table 16. Ranges of Parameters for Pu Solubility

Parameter	Number of Grid Points	Grid Points
pH	5	4.0, 5.0, 6.0, 7.0, 8.0
Eh (V)	3	0.34, 0.55, 0.76
Log f_{CO2}	2	-3.0, -3.5

Variations in pH, Eh, and the fugacity of CO_2 are considered. Table 16 lists the data points for each variable. The data matrix consists of 5x3x2 = 30 data points. These data points are uniformly distributed along the ranges of the three variables.

6.5.4 Results and Distribution

Table 17 gives the calculated Pu solubility (in units of mg/L) using $Pu(OH)_4$(am) as the controlling solid. One data point is missing, because the calculation does not converge. The maximum solubility is 49 mg/L = 2.04E-4 mol/L, while its minimum is 1.04E-10 mol/L.

Table 17. Calculated Pu Solubility (mg/L) with $Pu(OH)_4$(am)as the Controlling Mineral

	Log f_{CO2} = -3.0			Log f_{CO2} = -3.5		
pH	Eh = 0.34	Eh = 0.55	Eh = 0.76	Eh = 0.34	Eh = 0.55	Eh = 0.76
4.0	3.53	2.60E-03	4.89	3.53E+00	2.60E-03	4.89E+00
5.0	4.08E-04	1.43E-03	4.89	4.08E-04	1.43E-03	4.89E+00
6.0	4.91E-05	1.42E-03	4.90	4.91E-05	1.42E-03	4.90E+00
7.0	2.75E-05	1.41E-03	5.86	2.75E-05	1.40E-03	5.24E+00
8.0	2.52E-05	2.73E-03	(a)	2.49E-05	1.83E-03	4.90E+01

a: No data because the calculation does not converge.

Attempts have been made to find a distribution pattern by plotting the above data in histograms with different bins, both in linear concentration scale and in log(concentration) scale. However, no patterns can be easily recognized, probably due to the small number of calculations. In linear concentration scale, the above 29 data points are concentrated in a narrow range, while they distribute more evenly on log(concentration) scale. Therefore, a log-uniform distribution is proposed for Pu solubility, with a maximum of 49 mg/L =2.0E-4 mol/L and a minimum of 2.49E-5 mg/L =1.0E-10 mol/L.

6.5.5 Summary and Discussion

A logarithm uniform distribution is presented to represent Pu solubility limits, with a range from 1.0E-10 to 2.0E-4 mol/L. A response surface to represent Pu solubility is more appropriate and will be developed in REV 01.

6.6 AMERICIUM (Am) SOLUBILITY

The solubility analysis for Am was originally conducted in LLNL (Wolery et al., 2000). This section is a shortened version of the original document.

6.6.1 Thermodynamic Data

After several years of extensive review, the Nuclear Energy Agency (NEA) published "Chemical Thermodynamics of Americium" in 1995 (Silva et al., 1995). LLNL has submitted that set of data to TDMS. This set of data has been incorporated into EQ3/6 databases. The data0.com.R2 data file is chosen for this calculation.

6.6.2 Controlling Mineral

$AmOHCO_3$ was chosen as the controlling solid phase in all calculations. The choice of this mineral is based on the studies of Nitsche et al. (1993a, 1993b p.1494), which identify $AmOHCO_3$ as the solid phase precipitated from J-13 water at a pH range from 5.9 to 8.4, and temperatures from 25 to 90°C. This is the most likely controlling phase under the range of environmental variables of interest to this analysis. However, other possible controlling phases may exist. As discussed in Section 6.6.6, using $AmOHCO_3$ is conservative, in the sense that it

may overpredict Am solubility in the certain ranges of environmental conditionsof interest to this analysis.

The conceptual model that $AmOHCO_3$ is the solubility-controlling mineral for Am is conservative and, thus, adequate for use in TSPA-SR. It does not require validation.

6.6.3 Water Composition

All calculations reported here are based on J-13 groundwater, as reported by Harrar et al. (1990), except as noted here. J-13 water contains Al, Li, Fe, Mn, and Sr at concentrations between 30 and 50 μg/L. These component concentrations are sufficiently small that they do not affect the computed groundwater chemistries and were ignored.

6.6.4 Independent Variables

6.6.4.1 Temperature

The temperature dependence of americium solubility is poorly understood. Nitsche (1986) and Nitsche et al. (1993a, 1993b) show differences in Am solubility between 30 and 90°C, but with no identifiable trend. These results were mainly obtained with $AmOHCO_3$ as the solubility-controlling phase. The thermodynamic database data0.com.R2 contains no data pertinent to the temperature dependence of the thermodynamic stability of any Am aqueous or solid species. This problem traces back to Silva et al. (1995), the source of these Am species data, which nonetheless remains the definitive work on the subject. Hence all calculations were made only for 30°C, using thermodynamic data for 25°C. The use of these calculations to represent Am solubility at higher temperature (90-100°C) is likely not to be overly problematic as long as the pH input to the response formula is the quench (25-30°C) pH, not the "at temperature" pH. Nevertheless Am solubility at higher temperatures remains not well understood, and the use of the response function at such temperatures introduces additional uncertainty.

6.6.4.2 Eh

The only important oxidation state of Am in natural systems is Am(III) (Langmuir, 1997, Table 13.8, Silva and Nitsche, 1995, Fig. 4). Thus it is expected that the impact of Eh on Am solubility would be minimal and will not be analyzed in this study.

6.6.4.3 pH

A peculiarity of using a carbonate mineral such as $AmOHCO_3$ as a solubility-controlling solid with a specified CO_2 fugacity is that such solutions only exist over a rather limited range of pH. That range is denoted here as (a, b), where "a" is the minimum pH value and "b" the corresponding maximum. In more practical terms, "a" is the lowest pH for which the code calculations will converge, while "b" is the highest such pH.

6.6.4.4 CO_2 Fugacity

The calculations were made for CO_2 fugacities of 10^{-2} bar (higher than ambient), 10^{-3} bar (near ambient), and sub-ambient values of 10^{-4}, 10^{-5}, 10^{-6}, 10^{-7}, and 10^{-8} bar (10^{-4}-10^{-8} are sub-ambient).

6.6.5 The Calculated Data

The actual Am solubility and ionic strength data obtained from the EQ3NR output files is given in Table 18.

Table 18. Results from the EQ3NR Calculations of Am Solubility, Based on $AmOHCO_3$ as the Solubility-Controlling Mineral

log f_{CO2}	pH	Am (molal)	log f_{CO2}	pH	Am (molal)
-2	5.45	2.602E-02	-5	10	2.738E-08
-2	6	8.911E-05	-5	11.09	3.137E-03
-2	7	5.458E-07	-6	6.77	4.974E-02
-2	8	1.570E-07	-6	7	2.225E-03
-2	9	1.035E-04	-6	8	1.427E-05
-2	9.54	7.313E-02	-6	9	8.806E-07
-3	5.78	2.861E-02	-6	10	8.563E-08
-3	6	1.260E-03	-6	11	2.913E-07
-3	7	1.801E-06	-6	11.59	9.651E-04
-3	8	6.610E-08	-7	7.09	8.945E-02
-3	9	2.568E-07	-7	8	1.434E-04
-3	10	7.017E-03	-7	9	8.768E-06
-3	10.08	3.542E-02	-7	10	8.441E-07
-4	6.11	3.225E-02	-7	11	1.045E-07
-4	7	1.447E-05	-7	12	5.566E-05
-4	8	1.862E-07	-7	12.09	3.105E-04
-4	9	2.266E-08	-8	7.41	1.865E-01
-4	10	3.438E-06	-8	8	1.544E-03
-4	10.59	1.127E-02	-8	9	8.774E-05
-5	6.44	3.824E-02	-8	10	8.438E-06
-5	7	1.469E-04	-8	11	1.010E-06
-5	8	1.465E-06	-8	12	3.207E-07
-5	9	9.282E-08	-8	12.59	1.078E-04

The calculated Am solubility ranges from 2.27E-8 to 0.187 mol/kg, with an average of 0.0128 mol/kg.

The following fit was obtained using Sigmaplot 4.0:

$$\log[Am] = a + b*pH + c*pH^2 + d*\log(f_{CO2}) + e*(\log f_{CO2})^2 + \quad \text{(Eq. 13)}$$

$$+ g*(pH^2)*\log(f_{CO2}) + h*pH*(\log f_{CO2})^2$$

where:

[Am] is the Am solubility in units of mg/L, f_{CO2} is the fugacity of CO_2 in atmospheres, and a= 58.0335, b= -18.9422, c= 1.4744, d= -6.0032, e= -0.7005, g= 0.1162, and h= 0.1146.

An important result is that for any value of log f_{CO2}, this formulation collapses to one that contains terms only in pH and pH^2. This collapsed formulation is parabolic and upwardly concave. That assures that outside the (a, b) field, the calculated solubility values will always rapidly trend higher from values in the field. Thus, should any out-of-bounds values be computed in performance assessment, they should be very high and lead to a mass-limited calculated value of dissolved Am.

Some of the calculated data points, as well as the collapsed Am solubility curves under chosen f_{CO2} were plotted in Figure 2. The figure shows that in general, the fit is good. However, the parabolic curves are overly symmetrical compared with the actual data.

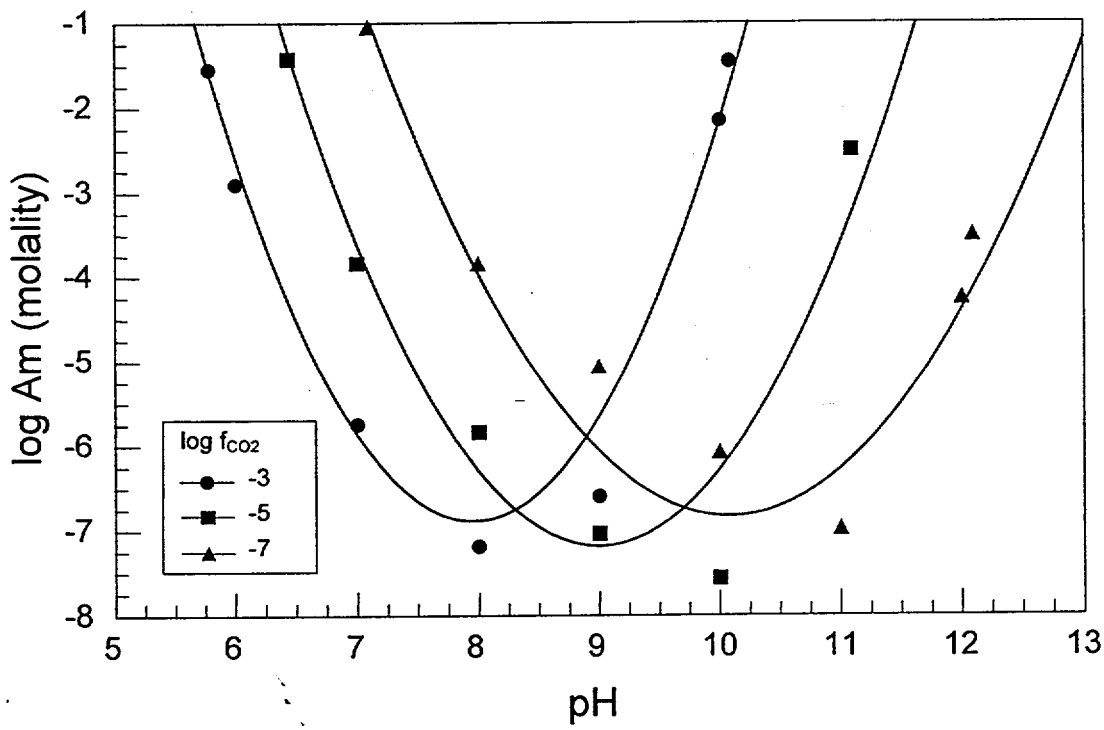


Figure 2. Comparison of the Fitted Response Surface (Solid Curves) with the Original EQ3NR Calculated Solubilities of Am (molal) for f_{CO2} Values of 10^{-3}, 10^{-5}, and 10^{-7} bar

6.6.6 Discussion and Summary

The geochemical modeling code package EQ3/6 (Wolery, 1992a) was used to calculate the solubility of americium in J-13-like water. Although thermodynamic data were only available for 25°C, the calculations were performed for 30°C assuming that the thermodynamic data would be essentially unaffected. The main calculations were made using the EQ3NR speciation-solubility code (Wolery, 1992b). All solubility calculations assumed that $AmOHCO_3$ was the solubility-controlling solid.

The calculations suggested that AmO_2 might be the actual solubility-controlling phase in a repository system, as most of the solutions in the calculations were supersaturated with this phase. Its lack of appearance in experimental solubility studies may be due to kinetic limitations, as are known or suspected in the case of other actinide(IV) oxides. If there is a problem in performance assessment with Am solubilities based on $AmOHCO_3$ control being too high, the possible control by AmO_2 should be considered, as this phase should yield lower solubilities.

The resulting data was fit to an equation of the form:

$$\log[Am] = a + b*pH + c*pH^2 + d*\log(f_{CO2}) + e*(\log f_{CO2})^2 +$$

$$+ g*(pH^2)*\log(f_{CO2}) + h*pH*(\log f_{CO2})^2$$

The resulting fit is expected to be applicable at pH values from about 5 to 12, and for f_{CO2} from 10^{-8} to 10^{-2} bar.

6.7 ACTINIUM (Ac) SOLUBILITY

No process level information for actinium solubility was available to this analysis. In the previous TSPAs, Ac is considered analogous to Am, as suggested by the project experts, though no basis was provided to support that suggestion (CRWMS M&O 1998a, Section 6.4.1.3). Like Am, only trivalent states of Ac are stable in water (Baes and Mesmer, 1976, p.129) This suggestion is adopted and a response surface similar to Equation 13 is recommended (with a difference of a constant) as its solubility limit, as follows:

$$\log[Ac] = a + b*pH + c*pH^2 + d*\log(f_{CO2}) + e*(\log f_{CO2})^2 + \qquad \text{(Eq. 14)}$$

$$+ g*(pH^2)*\log(f_{CO2}) + h*pH*(\log f_{CO2})^2$$

where:

[Ac] is the Ac solubility in units of mg/L, f_{CO2} is the fugacity of CO_2 in bar, and a= 58.00378, b= -18.9422, c= 1.4744, d= -6.0032, e= -0.7005, g= 0.1162, and h= 0.1146.

6.8 CURIUM (Cm) SOLUBILITY

As indicated by *Pure Phase Solubility Limits--LANL*, AMR F0085 (CRWMS M&O 2000a, 6.3.3), Cm is expected to behave like Am, and using the Am database to calculate Cm solubility is technically reasonable. Therefore, a response surface similar to Equation 13 as Cm solubility is suggested, as follows:

$$\log[Cm] = a + b*pH + c*pH^2 + d*\log(f_{CO2}) + e*(\log f_{CO2})^2 + \qquad \text{(Eq. 15)}$$

$$+ g*(pH^2)*\log(f_{CO2}) + h*pH*(\log f_{CO2})^2$$

where:

[Cm] is the Cm solubility in units of mg/L, f_{CO2} is the fugacity of CO_2 in bar, and a= 58.0404, b= -18.9422, c= 1.4744, d= -6.0032, e= -0.7005, g= 0.1162, and h= 0.1146.

6.9 SAMARIUM (Sm) SOLUBILITY

As indicated by *Pure Phase Solubility Limits* - LANL AMR (CRWMS M&O 2000a, Section 6.3.3), Sm is expected to behave like Am, and using the Am database to calculate Sm solubility is technically reasonable. Therefore, a response surface similar to Equation 13 as Sm solubility is suggested, as follows:

$$\log[Sm] = a + b*pH + c*pH^2 + d*\log(f_{CO2}) + e*(\log f_{CO2})^2 + \qquad \text{(Eq. 16)}$$

$$+ g*(pH^2)*\log(f_{CO2}) + h*pH*(\log f_{CO2})^2$$

where:

[Sm] is the Sm solubility in units of mg/L, f_{CO2} is the fugacity of CO_2 in bar, and a= 57.82495, b= -18.9422, c= 1.4744, d= -6.0032, e= -0.7005, g= 0.1162, and h= 0.1146.

6.10 THORIUM (Th) SOLUBILITY

The AMR titled *Pure Phase Solubility Limits--LANL* (CRWMS M&O 2000a, Section 6.3.8) does not provide a Th solubility range. The major reason for this omission is that the EQ3/6 current databases do not include thorium-carbonate species. As experiments have demonstrated that Th solubility may increase dramatically if the concentration of carbonate is high enough (Rai et al. 1995), model calculations without thorium-carbonate species are not meaningful.

As an alternate solution, a constant solubility of 1.0E-5 mol/L is proposed for Th. A small amount of corroborating data can support this proposed value: 1) the 1993 Expert Elicitation of the Project gives the Th solubility range of 10^{-10} to 10^{-7} mol/L; 2) NRC's total-system performance assessment (TPA) code version 3.2 (Greeves 1999, p. A-38) uses 2.3E-4 kg/m^3 = 10^{-6} mol/L as the solubility limit for Th, with consideration of thorium-carbonate species. In fact, this proposed value is one to two orders of magnitude higher than these corroborating data, and, thus, is conservative and adequate for use in TSPA-SR.

The deficiency in Th thermodynamic data will be fixed in the acceptance activity of the "data0.ymp" datafile. Once the data file is accepted by the Project, modeling calculations for Th solubility will be conducted and new values of Th solubility limits will be generated.

6.11 NIOBIUM (Nb) SOLUBILITY

A study of Nb solubility has been conducted and documented in *Pure Phase Solubility Limits LANL* (CRWMS M&O 2000a, Section 6.3.5) using EQ3NR. It suggests that Nb solubility range from 1.0E-15 to 1.0E-7 mol/L. It is recommended that TSPA-SR use a constant solubility of 1.0E-7 mol/L for Nb. This abstraction is conservative and, thus, adequate for use in TSPA-SR.

6.12 TECHNETIUM (Tc) SOLUBILITY

Under the repository conditions, it is expected that no solubility-controlling solid exists for Tc. Therefore, Tc solubility is arbitrarily set to 1.0 mol/L, which will allow the waste inventory to control Tc release.

6.13 CARBON (C) SOLUBILITY

Although under neutral or high pH, calcite may control the solubility of C, under pH as low as 3.6, calcite is not stable (Langmuir, 1997, p.202, Fig. 6.6). Therefore, C solubility is arbitrarily set to 1.0 mol/L, which will allow the waste inventory to control C release.

6.14 IODINE (I) SOLUBILITY

Under the repository conditions, it is expected that no solubility-controlling solid exists for I. Therefore, I solubility is arbitrarily set to 1.0 mol/L, which will allow the waste inventory to control I release.

6.15 NICKEL (Ni) SOLUBILITY

A brief EQ3NR calculation carried out in the AMR titled *Pure Phase Solubility Limits--LANL* (CRWMS M&O 2000a, Section 6.3.6) gives the range of Ni solubility for Yucca Mountain waters as 1.4E-6 to 3.1 mol/L, provided that bunsenite (NiO) is the solubility controlling mineral. No effort has been made to generate the distribution type for Ni solubility. A log-uniform distribution is proposed for Ni solubility for TSPA-SR, with a minimum of 1.4E-6 and a maximum of 3.1 mol/L.

6.16 ZIRCONIUM (Zr) SOLUBILITY

Another brief EQ3NR calculation documented in the AMR titled *Pure Phase Solubility Limits - LANL* (CRWMS M&O 2000a, Section 6.3.10) generates the range of Zr solubility for Yucca Mountain waters as 2.1E-14 to 6.8E-10 mol/L. It is suggested that TSPA-SR use a constant of 6.8E-10 mol/L for Zr solubility.

6.17 RADIUM (Ra) SOLUBILITY

Radium solubility has also been studied briefly in *Pure Phase Solubility Limits--LANL*, AMR F0085 (CRWMS M&O 2000a, Section 6.3.7). For Yucca Mountain waters, Ra solubility ranges from 1.1E-6 to 2.3E-6 mol/L. A constant solubility of 2.3E-6 mol/L is recommended for Ra.

6.18 TIN (Sn) SOLUBILITY

The calculated solubility range of tin reported in *Pure Phase Solubility Limits--LANL* (CRWMS M&O 2000a, Section 6.3.9) for Yucca Mountain waters is very narrow, from 4.8E-8 to 4.9E-8 mol/L. Thus, a constant solubility of 5.0E-8 mol/L for Sn is suggested for TSPA-SR.

6.19 CHLORINE (Cl) SOLUBILITY

Under the repository conditions, it is expected that no solubility-controlling solid exists for Cl. Therefore, Cl solubility is arbitrarily set to 1.0 mol/L, which will allow the waste inventory to control Cl release.

6.20 PROTACTINIUM (Pa) SOLUBILITY

Due to the lack of thermodynamic data for Pa, no data compilation has been carried out in the *Pure Phase Solubility Limits--LANL* (CRWMS M&O, 1999b, section 6.3.1). Thus, it is not possible to conduct EQ3NR calculations for Pa solubility. The process AMR suggests a solubility range of 1.0E-5 to 1.0E-10 mol/L. This range is the same as Pa solubility values used in TSPA-93, which was obtainedby the Project experts elicitation process (Wilson et al. 1994, Table 9-2b, DTN: SN9911T0410194.001). The project experts further suggested a log-uniform distribution over that range. Therefore, for TSPA-SR, a log-uniform distribution for Pa solubility is recommended, with a minimum of 1.0E-10, a maximum of 1.0E-5, and a mean of 3.2E-8 mol/L.

6.21 LEAD (Pb) SOLUBILITY

In carbonate containing waters, $Pb_3(CO_3)_2(OH)_2$ is believed to be an major important solid phase for constraining Pb solubility (CRMWS M&O, 2000a, Section 6.3.11). Unfortunately, EQ3/6 databases do not contain thermodynamic data for this solid. The process AMR (CRWMS M&O, 2000a, Section 6.3.11) claims that, without thermodynamic data for that solid, calculated Pb(II) solubility is suspect. Based on reported Pb(II) solubility range in carbonate containing waters, the process AMR suggests a solubility range of 1.0E-10 to 1.0E-5 mol/L in Yucca Mountain waters.

The above range is very close to the solubility range of Pb (1.0E-8 to 1.0E-5 mol/L, log-beta distribution) used in the TSPA-93 (Wilson et al. 1994, Table 9-2b, DTN: SN9911T0410194.001), which is suggested by the Project experts.

It is recommended that TSPA-SR use a log-uniform distribution, with a minimum of 1.0E-10, and a maximum of 1.0E-5 mol/L, to constrain Pb solubility.

6.22 CESIUM (Cs) SOLUBILITY

Under the repository conditions, it is expected that no solubility-controlling solid exists for Cs. Therefore, Cs solubility is arbitrarily set to 1.0 mol/L, which will allow the waste inventory to control Cs release.

6.23 STRONTIUM (Sr) SOLUBILITY

Sr is quite soluble. The most likely solids to precipitate under the repository conditions are carbonate or sulfate. As Sr is an element added to the TSPA-SR RN list at the last minute, time did not permit an analysis. Therefore, it is assumed that under the repository conditions, no solubility-controlling solid exists for Sr. Thus, Sr solubility is arbitrarily set to 1.0 mol/L, which will allow the waste inventory to control Sr release. Its solubility will be modeled when this AMR is revised.

7. CONCLUSIONS

As the first systematic and comprehensive solubility study in the history of YMP, solubility limits have been derived for 21 elements. Of these, Pa and Sr were not in the original plan. Table 18 gives response surfaces for U, Np, Am Ac, Sm, and Cm (in units of mg/L). Table 19 gives solubility distributions for the rest (in units of both mol/L and mg/L.)

The degree of analysis detail varies from element to element. As resources were limited, priority was given to certain important elements, such as Np, Pu, U, and Am. For some very soluble elements, there is not an adequate basis to specify a solubility-controlling solid, so they are assumed to be infinitely soluble. Elements in this category are Tc, C, I, Cs, and Cl. Due to time constraint, for some elements, no detailed analysis beyond the work in *Pure Phase Solubility Limits--LANL* (CRWMS M&O 2000a) has been done.

Unqualified data were used in this analysis. Therefore, the QA status of the analysis results should be designated "to be verified" (TBV). Analysis results used as inputs must be identified and tracked as TBV in accordance with appropriate QA procedures.

Table 18. Summary of Solubility Limits - Response Surfaces

Element	Atomic-Weight (Langmuir 1997, Inside Front Cover)	Response Surface (mg/L)
U	238.0	$\log[U] = 7.9946 - 2.6963pH + 0.4292pH^2 - 1.6286\log f_{CO2} + 0.0095T + 0.4161pH \times \log f_{CO2} - 0.0051pH \times T - 0.0022\log f_{CO2} \times T$
Np	237.0	$[Np] = 7.538E-8 + 1.086 \times 10^{8-pH}$
Am, Ac, Cm, and Sm	243.13, 227.03, 150.4, 247, respectively	$\log[Am] = a + b * pH + c * pH^2 + d * \log f_{CO2} + e * (\log f_{CO2})^2 + g * pH^2 * \log f_{CO2} + h * pH * (\log f_{CO2})^2$

Coefficients	Am	Ac	Cm	Sm
a	58.0335	58.0038	58.0404	57.8250
b	-18.9422	-18.9422	-18.9422	-18.9422
c	1.4744	1.4744	1.4744	1.4744
d	-6.0032	-6.0032	-6.0032	-6.0032
e	-0.7005	-0.7005	-0.7005	-0.7005
g	0.1162	0.1162	0.1162	0.1162
h	0.1146	0.1146	0.1146	0.1146

Note: T is temperature in Kelvin; f_{co2} is the fugacity of CO_2 in bar; log refers to the base 10 logarithm; E is power of ten.

Moreover, through this first round of analysis, substantial experience in solubility abstraction has been gained. A set of methods to build solubility models has been developed. The approaches to obtaining response surfaces and stochastic distributions for solubility limits also set an example for future analyses.

While progress is being made, areas for improvement remain. The following six areas will be considered for future revision:

- A uniform, accepted thermodynamic database for EQ3/6.
- Necessary laboratory experiments to reduce conceptual uncertainties, especially the controlling solids for Pu.
- Better defined in-package chemistry. Not only the ranges of variables, but also their distributions should be given.
- Detailed analyses for most elements, just like the analyses done for Np, Pu, and U in this study.
- More EQ3NR calculations for better abstractions as more samples will increase the confidence level for the conclusions drawn from them.
- Evaluation of solubility limits for another type of WP, i.e., high-level waste glass.

This document may be affected by technical product input information that requires confirmation. Any changes to the document 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.

Table 19. Summary of Solubility Limits - Distributions

Element	Variable	Distribution Type	Min (mol/L) / Min (mg/L)	Max (mol/L) / Max (mg/L)	Atomic Weight (Langmuir 1997, Inside Front cover)
Pu	log (concentration)[a]	Uniform	-10.0	-3.69	239.13
			-4.62	1.68	
Th	Concentration	Constant	1.0E-5[b]		232.0
			2.32		
Nb	Concentration	Constant	1.0E-7		92.91
			9.29E-3		
Tc	Concentration	Constant	1.0		98.91
			9.89E+4		
C	Concentration	Constant	1.0		12.01
			1.2E+4		
I	Concentration	Constant	1.0		126.9
			1.27E+5		
Ni	log (concentration)	uniform	-5.85	0.49	58.69
			-1.08	5.25	
Zr	Concentration	Constant	6.8E-10		91.22
			6.20E-5		
Ra	Concentration	Constant	2.3E-6		226.02
			0.52		
Sn	Concentration	Constant	5.0E-8		118.69
			5.93E-3		
Cl	Concentration	Constant	1.0		35.45
			3.54E+4		
Pa	log (concentration)	uniform	-10.0	-5.0	231.04
			-4.64	0.36	
Pb	log (concentration)	uniform	-10.0	-5.0	207.19
			-4.68	0.32	
Cs	Concentration	Constant	1.0		132.9
			1.33E+5		
Sr	Concentration	Constant	1.0		87.62
			8.76E+4		

[a] log refers to the base 10 logarithm

[b] E: power of ten

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SN9911T0811199.002. Results of EQ6 Calculations of Waste Package Degradation for Civilian Spent Nuclear Fuel. Submittal date: 11/15/1999.

9. ATTACHMENTS

Attachment	Title
I	**List of Computer Files for U Solubility Calculation**
II	**List of Computer Files for Np Solubility Calculation**
III	**List of Computer Files for Pu Solubility Calculation**

(The computer files will be submitted to the Record Processing Center on electronic media in the records package for this AMR. In addition, these files may be obtained through the Technical Data Management System (TDMS) under DTN: MO0004SPASOL10.002.)

ATTACHMENT I

LIST OF COMPUTER FILES FOR U SOLUBILITY CALCULATION

List of Computer Files for U Solubility Calculation

(The computer files will be submitted to the Record Processing Center on electronic media)

Volume in drive E has no label.
Volume Serial Number is ACBC-7919

Directory of E:\yueting\PA\AP-3.10Q-2\Attachments\ATTACHI

File Name	Date	Time	Size
data0.com.R2	08/30/99	10:30a	2,109,751
pHimpact.xls	11/04/99	11:22a	16,896
usensp5.3i	08/12/99	12:32p	11,200
usense1.3i	08/12/99	01:12p	11,200
usense2.3i	08/12/99	12:25p	11,200
usense3.3i	08/12/99	12:26p	11,200
usense4.3i	08/12/99	12:27p	11,200
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schsf449.3o	02/24/00	09:30a	66,519

Note: data0.com.R2 is the database for EQ3NR calculations. Files with suffix "3i" are EQ3NR input files, while those with suffix "3o" are EQ3NR output files. Two files with suffix "xls" are MS Excel spreadsheet files, which contain the summary information of EQ3NR calculations. The file of "Regression.JNB" is SigmaPlot file, which records the regression analyses on U response surfaces.

ATTACHMENT II

LIST OF COMPUTER FILES FOR Np SOLUBILITY CALCULATION

List of Computer Files for Np Solubility Calculation

(The computer files will be submitted to the Record Processing Center on electronic media)

```
Volume in drive E has no label.
Volume Serial Number is ACBC-7919

Directory of E:\yueting\PA\AP-3.10Q-2\Attachments\ATTACHII
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Npsensf2.3i	09/14/99	01:40p	11,271
Npsensf3.3i	09/17/99	02:32p	11,271
Npsensf4.3i	09/16/99	09:13a	11,270
NpsensI1.3i	10/01/99	07:25a	11,285
NpsensI2.3i	10/01/99	07:25a	11,283
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Nprss124.3i	09/16/99	12:19p	11,270
Nprss125.3i	09/16/99	12:19p	11,270
Nprss126.3i	09/16/99	12:20p	11,270
Nprss127.3i	09/16/99	12:20p	11,270
Nprss128.3i	09/16/99	12:43p	11,270
Nprss129.3i	09/16/99	12:44p	11,270
Nprss131.3i	09/16/99	12:22p	11,270
Nprss132.3i	09/16/99	12:22p	11,270
Nprss133.3i	09/16/99	12:23p	11,270
Nprss134.3i	09/16/99	12:23p	11,270
Nprss135.3i	09/16/99	12:24p	11,270
Nprss136.3i	09/16/99	12:24p	11,270
Nprss137.3i	09/16/99	12:25p	11,270
Nprss138.3i	09/16/99	12:25p	11,270
Nprss139.3i	09/16/99	12:25p	11,270
Nprss141.3i	09/16/99	12:26p	11,270
Nprss142.3i	09/16/99	12:27p	11,270
Nprss143.3i	09/16/99	12:27p	11,270
Nprss144.3i	09/16/99	12:28p	11,270
Nprss145.3i	09/16/99	12:28p	11,270
Nprss146.3i	09/16/99	12:29p	11,270
Nprss147.3i	09/16/99	12:29p	11,270
Nprss148.3i	09/16/99	12:30p	11,270
Nprss149.3i	09/16/99	12:30p	11,270

Nprssph4.3i	10/12/99	12:00p	11,270
phimpt01.3i	11/04/99	12:49p	11,288
Nprss111.3i	09/16/99	12:09p	11,270
regression-2.JNB	11/12/99	04:45p	85,504
sensana.xls	10/03/99	08:26a	16,896
data0.an4.R6	09/29/99	02:46p	3,132,754
Npsens01.3o	02/24/00	09:41a	84,142
Npsens04.3o	02/24/00	09:41a	83,241
Npsens06.3o	02/24/00	09:41a	84,146
NpsensI1.3o	02/24/00	09:42a	85,590
NpsensI2.3o	02/24/00	09:42a	85,799
NpsensI4.3o	02/24/00	09:42a	86,303
Npsensf1.3o	02/24/00	09:42a	84,318
Npsensf2.3o	02/24/00	09:42a	84,021
Npsensf3.3o	02/24/00	09:42a	87,090
Npsensf4.3o	02/24/00	09:42a	86,744
Npsensp1.3o	02/24/00	09:42a	85,186
Npsensp2.3o	02/24/00	09:42a	82,777
Nprss111.3o	02/24/00	09:44a	82,084
Nprss112.3o	02/24/00	09:44a	82,509
Nprss113.3o	02/24/00	09:44a	82,485
Nprss114.3o	02/24/00	09:44a	83,183
Nprss115.3o	02/24/00	09:44a	84,015
Nprss116.3o	02/24/00	09:44a	84,390
Nprss117.3o	02/24/00	09:44a	83,988
Nprss118.3o	02/24/00	09:44a	86,226
Nprss119.3o	02/24/00	09:44a	84,689
Nprss121.3o	02/24/00	09:44a	82,190
Nprss122.3o	02/24/00	09:44a	82,562
Nprss123.3o	02/24/00	09:44a	82,538
Nprss124.3o	02/24/00	09:44a	82,966
Nprss125.3o	02/24/00	09:44a	84,068
Nprss126.3o	02/24/00	09:44a	84,228
Nprss127.3o	02/24/00	09:44a	86,100
Nprss128.3o	02/24/00	09:44a	84,308
Nprss129.3o	02/24/00	09:44a	85,648
Nprss131.3o	02/24/00	09:44a	82,084
Nprss132.3o	02/24/00	09:44a	82,403
Nprss133.3o	02/24/00	09:44a	82,485
Nprss134.3o	02/24/00	09:44a	83,182
Nprss135.3o	02/24/00	09:44a	83,702
Nprss136.3o	02/24/00	09:44a	84,388
Nprss137.3o	02/24/00	09:44a	84,364
Nprss138.3o	02/24/00	09:44a	86,079
Nprss139.3o	02/24/00	09:44a	85,954
Nprss141.3o	02/24/00	09:44a	82,084
Nprss142.3o	02/24/00	09:44a	82,350
Nprss143.3o	02/24/00	09:44a	82,485
Nprss144.3o	02/24/00	09:44a	83,182
Nprss145.3o	02/24/00	09:44a	83,596
Nprss146.3o	02/24/00	09:44a	84,282
Nprss147.3o	02/24/00	09:44a	84,302
Nprss148.3o	02/24/00	09:44a	84,340
Nprss149.3o	02/24/00	09:44a	86,324
phimpt01.3o	02/24/00	09:44a	84,074
phimpt02.3o	02/24/00	09:44a	82,858

Note: data0.an4.R6 is the database for EQ3NR calculations. Files with suffix "3i" are EQ3NR input files, while those with suffix "3o" are EQ3NR output files. Two files with suffix "xls" are

MS Excel spreadsheet files, which contain the summary information of EQ3NR calculations. The file of "regression-2.JNB" is SigmaPlot file, which records the regression analyses on Np response surfaces.

ATTACHMENT III

LIST OF COMPUTER FILES FOR Pu SOLUBILITY CALCULATION

List of Computer Files for Pu Solubility Calculation

(The computer files will be submitted to the Record Processing Center on electronic media)

```
Volume in drive E has no label.
Volume Serial Number is ACBC-7919

Directory of E:\yueting\PA\AP-3.10Q-2\Attachments\AttachIII
```

File Name	Date	Time	Size
data0.an4.R6	09/29/99	02:46p	3,132,754
Pubase02.3i	02/24/00	11:07a	11,273
Pubase03.3i	02/24/00	11:07a	11,272
Pubase04.3i	02/24/00	11:07a	11,272
Pubase05.3i	02/24/00	11:07a	11,272
Pubase06.3i	02/24/00	11:07a	11,269
Pubase07.3i	02/24/00	11:07a	11,270
Pubase08.3i	02/24/00	11:07a	11,269
Pubase09.3i	02/24/00	11:07a	11,272
Pubased0.3i	02/24/00	11:07a	11,272
Pubased1.3i	02/24/00	11:07a	11,269
Pubased2.3i	02/24/00	11:07a	11,270
Pubased3.3i	02/24/00	11:07a	11,269
Pubased4.3i	02/24/00	11:07a	11,272
Pubased5.3i	02/24/00	11:07a	11,272
Pubased6.3i	02/24/00	11:07a	11,272
Pubased7.3i	02/24/00	11:07a	11,272
Pubased8.3i	02/24/00	11:07a	11,272
Pubased9.3i	02/24/00	11:07a	11,272
Pubasef0.3i	02/24/00	11:07a	11,272
Pubasef1.3i	02/24/00	11:07a	11,272
Pubasef2.3i	02/24/00	11:07a	11,272
Pubasef3.3i	02/24/00	11:07a	11,272
Pubasef4.3i	02/24/00	11:07a	11,272
Pubasef5.3i	02/24/00	11:07a	11,272
Pubasef6.3i	02/24/00	11:07a	11,269
Pubasef7.3i	02/24/00	11:07a	11,270
Pubasef8.3i	02/24/00	11:07a	11,269
Pubasef9.3i	02/24/00	11:07a	11,272
Pubase00.3i	02/24/00	11:07a	11,272
Pubase00.3o	02/24/00	11:07a	78,762
Pubase02.3o	02/24/00	11:07a	82,292
Pubase03.3o	02/24/00	11:07a	80,425
Pubase04.3o	02/24/00	11:07a	79,187
Pubase05.3o	02/24/00	11:07a	78,169
Pubase06.3o	02/24/00	11:07a	85,249
Pubase07.3o	02/24/00	11:07a	82,923
Pubase08.3o	02/24/00	11:07a	80,878
Pubase09.3o	02/24/00	11:07a	79,548
Pubased0.3o	02/24/00	11:07a	78,567
Pubased1.3o	02/24/00	11:07a	85,092
Pubased2.3o	02/24/00	11:07a	82,733
Pubased3.3o	02/24/00	11:07a	80,868
Pubased4.3o	02/24/00	11:07a	79,553
Pubased5.3o	02/24/00	11:07a	78,567
Pubased6.3o	02/24/00	11:07a	84,946
Pubased7.3o	02/24/00	11:07a	82,666
Pubased8.3o	02/24/00	11:07a	80,869
Pubased9.3o	02/24/00	11:07a	79,315

Pubasef0.3o	02/24/00	11:07a	78,163
Pubasef1.3o	02/24/00	11:07a	85,048
Pubasef2.3o	02/24/00	11:07a	82,927
Pubasef3.3o	02/24/00	11:07a	80,891
Pubasef4.3o	02/24/00	11:07a	79,392
Pubasef5.3o	02/24/00	11:07a	78,764
Pubasef6.3o	02/24/00	11:07a	84,155
Pubasef7.3o	02/24/00	11:07a	82,281
Pubasef8.3o	02/24/00	11:07a	80,411
Pubasef9.3o	02/24/00	11:07a	79,024
sensana.xls	02/24/00	11:20a	62,976

Note: data0.an4.R6 is the database for EQ3NR calculations. Files with suffix "3i" are EQ3NR input files, while those with suffix "3o" are EQ3NR output files. The files of "sensana.xls" is an MS Excel spreadsheet file, which contains the summary information of EQ3NR calculations.