UNITED STATES
NUCLEAR REGULATORY COMMISSION
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April 18, 1995

MEMORANDUM TO: John Austin, Chief
Performance Assessment and
Hydrology Branch

THROUGH: Norman Eisenberg, Section Leader NM for
Performance Assessment and
Health Physics Section, PAHB

FROM: Richard Codell, Sr. Hydraulic Engineer RC
Performance Assessment and
Health Physics Section, PAHB

SUBJECT: EPRI MEETING ON SOURCE TERM FOR ITERATIVE PERFORMANCE ASSESSMENTS

I attended a meeting at the Electric Power Research Institute (EPRI) in Palo Alto, CA, on February 22 and 23, 1995. The subject of the meeting was source term models for estimating release rates of radionuclides from high-level nuclear waste at the proposed Yucca Mountain repository for the purpose of performance assessment modeling. I had attended a similar meeting in December 1994 on flow and transport models. The present meeting brought together modelers who have worked on a variety of HLW performance assessments including NRC (John Walton and myself), DOE/M&O (Robert Andrews and William Halsey), AECL (Lawrence Johnson), and EPRI (Mic Apted, Robin McGuire, John Kessler, Rosa Yang, and Thomas Pigford).

We began the meeting discussing the recent changes in the high-level waste (HLW) program philosophy which will affect modeling of the performance of the site, especially the source term. John Kessler reiterated EPRI's role in repository performance assessment for Yucca Mountain (attached). Resolving the HLW repository issues is number 3 on a list of 36 priorities for nuclear power research. EPRI feels that total system performance assessment (TSPA) is the best tool for assessing the real importance of technical issues. The general goal of the workshop was to identify the components of the source term models that are essential to demonstrate the regulatory goals; e.g., (1) How complex must the models be in order to determine what is essential; (2) should conservative or best estimate approaches be used; (3) must any regulations be clarified prior to choosing the correct models; and (4) which parts of the EPRI source term model require improvements, within their limited budget and mandate?

Revised source term models must take into account the present DOE working assumptions to design the repository for a smaller areal heat load, and fewer, but more robust waste packages. One of the main incentives for the lower heat load is the basic mistrust of the complicated physics of the effects of heat on the unsaturated rock, especially the notion that heat will isolate the waste from liquid water. The sentiments expressed by some of those present was that one could not count on the long-term reliance on heat to boil away

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water, and that if you can't license to ambient conditions at the site, then you are in the wrong place.

EPRI's most recent source term model had a number of important limitations, e.g., constant infiltration rate and saturation state, no temperature dependence except as a determinant of wet/dry conditions, non-mechanistic water contact modes, and no interaction among the isotopes (NRC's Phase 2 model shares some of the same restrictions). In addition, their Phase 2 models did not take ingrowth of daughter nuclides into account. The EPRI TSPA code, IMARC, will now have transport models that include chain decay. The EPRI source term model will probably be developed by Mic Apted using the MIDAS code as a basis. I asked him for a copy of documentation and the code if possible.

Robert Andrews represented the M&O's key waste package issues as the near field environment, waste package degradation and radionuclide mobilization (attached). I presented a synopsis of NRC's recent work on source term, and the needs for further development which I took mainly from a recent paper by myself and Richard Weller (attached). John Walton (consulting at this meeting for EPRI) presented several of his recent works on processes important to the source term at Yucca Mountain (attached). Robin McGuire presented the implications of the assumptions in the source term model in EPRI's Phase 2 study of Yucca Mountain.

There were a number of interesting discussions related to source term models, some of which I will highlight by topic:

<u>Corrosion Effects</u> - John Kessler decided to limit discussion of container failures caused by corrosion, because EPRI's waste package consultant, Daniel Bullen, could not attend. However there were several related discussions on the effects of corrosion products on release of radionuclides.

John Walton presented recent work on corrosion of the waste package by water films, and the lowering of vapor pressure by electrolytes. He suggests that most water in the vicinity of the waste package will evaporate and leave behind a scale deposit and concentrated solution. The concentrated brine solution will lower vapor pressure and maintain a liquid water film at elevated temperatures, leading in some cases to conditions favorable to aggressive corrosion. Some of this work was sponsored under the Engineered Barrier Element at the CNWRA. Lawrence Johnson commented that there are many experiments with the effects of carbonates and bicarbonates on the rate of corrosion of the fuel, and that the mechanistic models should include these electrolytes. Walton further discussed an analysis of the effect of products from canister metal corrosion serving as sorbing media for released radionuclides. Metal (especially steel) corrosion products may influence the release in several ways. Spent fuel and metal alteration are electrochemical processes, and the corrosion processes for each would be affected by the other. The corrosion products are highly sorptive and are initially amorphous with high surface area. However with time they may crystalize, possibly leading to the re-release of some of the attached radionuclides not incorporated into the crystals, so the effectiveness of this phenomenon is not clear. The corrosion products would be present in relatively small quantities

compared to the natural materials surrounding the canisters, so they may not have a significant effect in the long run. Under some postulated conditions the fuel and other metals could consume oxygen and render the interior of the waste package anaerobic, which would further affect oxidation processes. This would be a temporary condition that depends on a number of complicated factors such as the extent of container failure. Consumption of oxygen may be an important factor while the fuel is hot and the UO_2 oxidation rates are relatively high.

William Halsey suggested that iron ions might have a detrimental effect on glass waste, and that the effects of corrosion products from container material on glass are being studied.

Inventory of Radionuclides in Spent Fuel - Many of the numbers are calculated rather than measured, and there could be large errors in the inventories of certain radionuclides. Thomas Pigford recommended EPRI include a better basis for gap and other compartment inventories.

Lawrence Johnson commented that the "instant release" fraction of radionuclides dominated all releases from the Canadian fuel. I explored the differences and concluded that the relatively slow rate of fuel oxidation in the reducing environment of the Canadian concept would not allow large releases from the interior of the fuel, so the inventory in the gap and grain boundaries near the surface of the fuel takes on relatively greater importance. This would not be the expected case in the U.S. concept, with an oxidizing environment after waste package failure.

Spatial and Temporal Variability - John Kessler noted that the heterogeneity of the geologic media, waste, and external forces dictate that the source term is not a homogeneous concept; e.g., a "fast path" source term may be different from the "average" source term. If a waste package were located in an area with significant fracture flow, the water could contribute to early failure and large releases from the waste package, coupled with a fast fracture flow path to the accessible environment. Such a "fast-path" source term appears to be required to evaluate DOE's Groundwater Travel Time rule, 10 CFR 960, which considers significant transport along fast paths.

In my presentation on NRC's Phase 2 analysis, I commented that we had attempted to address the issue of spatial and temporal variability by choosing seven "representative" waste packages to represent the average behavior of tens of thousands of containers, but had met with only partial success. William Halsey was familiar with the problem, and said that attempts to choose representative source parameters was not successful at Livermore either. He commented that the only solution seemed to be to run enough calculations per Monte Carlo realization to cover the range of parameters representing the spatial and temporal variability, and that perhaps faster computers were the key.

Effect of Legislation on Source Term Needs - There was discussion about the various HLW bills before Congress. The Industry Bill will emphasize dose modeling. Thomas Pigford expects the bills will also emphasize a period of compliance of no longer than 10,000 years, although the National Academy of

Science panel of which he is a member will urge a 100,000 year period. The requirements of this proposed legislation, especially the compliance period, would have an influence on what is needed for TSPA modeling.

Effects of Concrete on Source - There is likely to be a significant amount of concrete in the emplacement drifts, both from the floor material and the shotcrete on the walls. The effects of the concrete on the waste will be a factor in the source term model, and I believe I heard that DOE plans to study the sorptive properties of concrete which has been degraded by the long-term effects of heat.

Heat Load Strategy - There are potential detrimental effects of the lower heat load strategy, namely the waste packages may be exposed to more aggressive corrosion because of wetter conditions. DOE is looking at the effectiveness of corrosion-resistant alloys and sacrificial overpacks to deal with corrosion. I did not detect that DOE has considered the build-up of aggressive electrolytes as portrayed in John Walton's analyses (discussed above).

Cladding Protection - DOE is considering taking credit for cladding protection, and there are some estimates that this would be a substantial barrier for up to thousands of years. There was considerable discussion on this issue, and there was recognition that at high temperatures, oxidizing fuel could lead to unzipping of the cladding on the fuel rods. Rosa Yang noted from her experience on reactor fuel that the cladding of irradiated rods generally bonds tightly to the fuel, virtually eliminating the gap as a channel for oxygen transport. She would expect that this leads to a measure of protection of the fuel even if there was a failure at some other place along the fuel rod. However, someone commented that cladding protection may be too difficult a problem to justify including it as a barrier. There are already crevices in the cladding, and hydride reorientation (the relief of stress as hydrogen diffuses out of the crystal lattice) might lead to failures. Furthermore, the time frame of interest in repositories is much greater than that for reactors or spent-fuel storage.

Effect of Thermally Induced Stresses - William Halsey commented that there were questions about the stability of the emplacement drifts, and that the current thinking is that they would be stable during the heating phase and less stable as they cooled. There have been no full-scale thermal tests on drifts.

Thomas Pigford commented that thermal stresses would play a part in waste package failure mechanisms. Welds will be the most likely point of corrosion or stress failure. Lawrence Johnson commented that titanium is a good material because its welds remain the same material as the rest of the canister.

Ingress of Water, Contact with Waste, and Chemistry - An important factor in the source term modeling is the fraction of infiltrating water contacting the waste. Robert Andrews commented that DOE contractors are using the FEHM multiphase flow code together with FRACMAN fracture generator to try to estimate directly fracture flow from refluxing water near the waste package.

They hope through these analyses to estimate the number of waste packages that are wetted by dripping water. They noted the recent modeling work by Tom Buscheck at Livermore on the "born loser" waste package (a worst-case placement of a hot package in the path of a flow-capable fracture), with reflux of water flowing back along fracture pathways directly to the waste packages. There is an analysis by J.R. Phillip and Ben Ross (perhaps two separate papers) on dripping in tunnels. Work from Stripa in Sweden suggests that 90% of the dripping comes from 7% of the visible fractures. I commented that I was not convinced that these numbers were relevant to Yucca Mountain because most of the data were from saturated sites.

The group concluded that there is a large judgment factor for the parameters of the model such as determining the fraction of waste packages or fuel coming into contact with water. Andrews suggests assigning someone to the problem to see what has been done on this subject already, and try to avoid the use of expert judgment to find values.

Thomas Pigford commented that the estimate of the fraction of the waste contacted by liquid water at low temperatures might be approached as a "falling film" problem, which has been explored in the chemical engineering literature with regard to wetting of solid substrates in mass exchange processes. Furthermore, he had a recent Ph.D. student who estimated the conditions under which dripping from fractures might occur. I intend to follow-up on these references.

William Halsey noted that the source term models should take into account episodic flow, distributed in time and space, which he believes is much more likely than continuous flow. This would help to address the question of how much of the time the waste package and the fuel would be wetted. The model should also consider the possibility of a rise in the water table, either from above (infiltration) or below (regional rise), leading to inundation.

Thomas Pigford brought up the point that small holes in the waste package might still limit the rate of mass transfer of liquid and air and add a measure of protection of the waste. Others disagreed with the point of view that once corrosion begins, there is a relatively rapid increase in size and number of the holes. Lawrence Johnson noted, however, that under dry conditions, holes (such as natural defects) in carbon steel tend to close because the oxide formed has lower density than the metal. He thinks it is unreasonable to assume that initial defects large enough to cause trouble in the repository would not have been detected before emplacement.

We discussed the dissolution of the spent fuel by liquid water. I mentioned the Pena Blanca natural analog is evidence that the oxidized uranium might be fairly immobile because it formed stable silicate secondary minerals, and that furthermore, it was likely that much of the fission and activation products would be captured in the secondary mineralization. Tests with spent fuel seem to indicate that silica suppresses dissolution of the uranium dioxide and release of the radionuclides contained. Lawrence Johnson disagreed that this would be the case at Yucca Mountain however, because he expects that the amount of uranium present would consume the relatively small amount of silica in the water, and the oxidized form of uranium would be more of a uranium

oxide rather than a silicate. Thomas Pigford reiterated his belief that in some circumstances the oxygen getting in might be consumed by oxidizing the container metal and fuel. Pre-oxidation of fuel by air may occur at high temperatures, and there are some experimental data indicating that the pre-oxidized fuel may release radionuclides faster followed contact with liquid water. Such a scenario might be more pertinent where the waste package also includes a bentonite barrier. Lawrence Johnson commented that the Canadians are considering oxygen-consuming materials inside of the waste packages, but there would be little available free oxygen, mainly from radiolysis, in their case.

John Walton suggested that CO_2 would be consumed by the carbonation of calcium hydroxide in concrete. This could lead to locally high pH near waste packages. An advanced source term model could include the dynamics of carbon transport in the water and gas in the near field. Other near-field chemistry must take into account the effect of corrosion on pH, formation of nitric acid by radiolysis of moist air, and the difference between the behavior of thin water films and bulk water caused by phenomena such as surface tension in the former.

Potential Benefits of Backfill - John Walton discussed his recently published work on the effects of salt water films on the surface of hot waste packages. While the tendency of the water at negative pressure would be to flow around the drifts or stay attached to the walls, dripping onto the waste packages could be enhanced by the deposition of minerals carried by refluxing water, leading to possibly stalactites near the waste packages. John advocated the use of a porous backfill because this would lead to more confidence in the prediction of the flow of water near the waste package. His most important point was that the models and experiments leading to accurate prediction of the release rates from spent fuel waste forms at Yucca Mountain may be many years away, and there is no evidence that the analysts are converging on an answer. Granular backfill would make the problem more tractable, because it would be relatively easier to demonstrate through models and experiments that the heat would reduce the chance that liquid water from above would ever come into contact with the waste packages. He believes that the centerpiece of waste package performance assessment should not depend on technology that cannot be demonstrated. He commented that (1) mass transfer approaches for corrosion of the waste package and the fuel should be abandoned, and (2) backfill might mitigate the effects of high, but transient infiltration by providing temporary storage. It would dry out from the repository heat between interludes of high infiltration. Furthermore, a porous backfill might inhibit the transport of colloids that might form near the waste package. However others in the room reminded him that backfill would cause problems such as higher waste package temperature and an eventual path for liquid water when the waste packages cooled enough. I also commented that uneven deposition of minerals from the dripping or capillary water might establish pathways. At Livermore, William Glassley is modeling the transport and deposition of minerals near the heat source, and the possibility of fracture plugging.

A current waste package emplacement scheme has the canisters partially buried in backfill, halfway up the sides. DOE is interested in backfill, but the

temperature of the waste packages would increase. William Halsey believes that the 350°C temperature limits currently in place in the DOE regulations is no longer pertinent and should be relaxed to accommodate possible backfilled configurations.

Mic Apted commented that crushed zeolites have been discussed as a backfill, but would be problematic in the Yucca Mountain case because they are unstable at high temperatures and contain chemically bound water. No one has proven that cooked zeolites would work as an effective backfill. The large amount of concrete in the emplacement drifts might have some beneficial sorptive properties.

Colloids - The formation and transport of colloids in the near field was a topic of much discussion. William Halsey commented that there were no DOE-funded studies of colloid underway at the labs. Jack Gauthier however may be working on a new colloid model for Sandia. While many of the other national concepts for HLW repositories employ bentonite barriers which might be effective filters for colloids, crushed tuff backfill might not be effective in the Yucca Mountain case. There were arguments for and against colloids being problematic. While many of the panel thought that colloids might well form, the research on colloids focuses on their transport. Ivars Neretniks' work on colloid transport and the effect of clay barriers on filtration will be reviewed.

Miscellaneous Topics of Discussion

Effects of Matrix Diffusion - As a sidelight on the main focus of the meeting, source term, Robert Andrews described that the M&O is looking at matrix diffusion in the transport models in an effort to decide whether or not there is a need for further Calico Hills exploration. John Kessler told me that EPRI's contractor will include matrix diffusion in their Phase 3 PA's as well. This point out a need for NRC to include this important phenomenon in the Phase 3 IPA.

National Academy of Science (NAS) Recommendations - Thomas Pigford, a member of the National Academy of Science panel on high-level waste regulations, commented to us that he expected the panel would not give any support to EPA's cumulative release limits. He expects the NAS report to be circulated soon. He commented to me that the NRC Phase 2 analysis of the dose scenario I shared with him last December was very useful, and may have had an influence on the NAS report.

Criticality - There was some discussion about criticality issues and the recent calculations by Edward Bowman of Los Alamos on the potential for nuclear explosions in the repository. Those familiar with the report seemed skeptical of the threat. Thomas Pigford commented that the explosion scenarios had to postulate that the neutron poisons would be washed away faster than the nuclear fuel, and that if this happened suddenly with enough moderation by water and rock, that there could be a sudden criticality excursion with the release of enough energy to create a steam explosion. Someone suggested that higher carbonate and bicarbonate concentrations may lead to relative leaching of neutron poisons from fuel (NOTE: All discussions

of criticality were prior to the March 5, 1995 New York Times article).

Recommendations of Workshop - After hearing the presentations, the assembled group tried to sort through the issues and take account of the resources available to recommend the direction of EPRI's modeling efforts for the next phase. I have attached Tables 1 and 2, provided by John Kessler after the meeting, which summarize the issues addressed and the priorities decided for the EPRI Phase 3 source term.

Conclusions

The EPRI source term workshop was very informative and worthwhile, as was the previous workshop on flow and transport modeling. The source term model is perhaps more complicated and uncertain than the flow and transport models. There are fewer similarities to other problems, and less consensus among the participants than in the previous meeting. I intend to follow up on several references and models related to the source term.

I would be happy to brief you further on my experiences.

Attachments:
1. Tables 1 and 2 on Source Term Issues from J. Kessler
2. "EPRI's Interest in Source Term Issues at Yucca Mountain," by J. Kessler
3. "Important Processes for Yucca Mountain Source Term," by J. Walton
4. "Key Waste Package and Engineered Barrier System Uncertainties," by R.W. Andrews
5. "Further Development of a Source Term Model for a Repository in Unsaturated Tuff," by R. Codell and R. Weller

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Table 1: SUMMARY OF SOURCE TERM ISSUES (unprioritized)
February 22-23, 1995

Air/Water Contact/Entry	Container Effects	Release	Transport/Retardation
• How does water enter?	• Localized/limited container failure	• Key nuclides: Cs-135, Zr-93, Sn-126, Ni-59, C-14, Np237, Tc-99, Se-79, I-129, Cl-36, U, Pu, Am • Grow in: Pa-231, Ra-226, Th-230	• D_e in backfill and rock matrix
• When can water wet the package?	• Cladding failure	• Matrix dissolution = f(CO_3)	• Sorption by backfill
• Composition of water in space and time	• Time of failure	• Solubilities	• Limited penetration
• Criticality	• Manufacturing flaws - early failure - substantially complete containment - model how flaw evolves/heals	• "Gap" nuclide inventory • Mobility of gap and grain boundary nuclides	• Add daughter ingrowth? • Colloids
• Model humidity impacts on container	• Characteristic properties of corrosion products	• Comparative release rates of fissile material vs moderators	• Transient vs steady state models
• Rate of O_2/air ingress to container vs O_2 consumption through penetrations	• Redox buffering	• Colloids • Thin film contact/strange chemistry • Time changes in surface area	

SUMMARY OF SOURCE TERM ISSUES
February 22-23, 1995

Table 2 What auxiliary models/studies should be pursued, to put in IMARC?

General group consensus:

Priority

(1) Penetration size/characteristics

A. Control on diffusive/advective process out of container
B. Control of internal chemistries (oxygen supply vs usage)
C. Control of fraction of source available for mobilization (expert judgement only)

(2) Evaporation effects on the formation of very near-field transport paths

- Surface chemistry during cool-down (both on container and waste surfaces)
- Wetting development and transport pathways (steady and unsteady inflow)
- Affects how containers fail and how waste is released

(3) Effect of backfill and canister corrosion products on release rate to the surrounding rock

- Types (e.g., cement, crushed tuff, sand, iron oxides, chemical 'getters')
- Effective diffusion coefficient

(3) Waste surface area, gap inventory, and matrix dissolution rate sensitivity studies

(4) Colloid transport

- Near-field
- Far-field

(5) Criticality: Understand the problem first (Note: this priority given before the 3/5/95 NY Times article appeared)

(5) Manufacturing flaws:

- What kinds go undetected?
- How many might there be?
- How much earlier will failure occur?
- What is impact?
  - early air oxidation of hot waste
  - early release

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Background

- EPRI is funded by utilities to conduct research on their behalf
- Nuclear Power Group within EPRI focuses on members with nuclear plants
- Spent fuel handling, storage, and disposal a significant concern for those utilities
  - of 36 nuclear power research topics at EPRI, execs ranked these areas high in importance
  - #3: HLW repository issue resolution
  - #7: spent fuel storage and transportation

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EPRI's General Goals in Conducting a Research Program for Yucca Mountain

- Develop an independent capability to identify important technical issues at Yucca Mountain and assess their importance
- Selective demonstration of assessment techniques, technical approaches and solutions
- Assist DOE, EPA, and NRC in resolving difficult licensing issues from a technical standpoint

Fuel Reliability, Storage & Disposal

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EPRI's Total System Performance Assessment (TSPA) Effort

EPRI feels that TSPA

- is the best tool for assessing the real importance of technical issues
- can prioritize an efficient site characterization strategy
- will be the centerpiece of DOE's license application to the NRC
- should be used in DOE's Technical Site Suitability (TSS) determination, too

EPRI TSPA Development History

1989:

- EPRI forms small team of experts (11 people) to develop subsystem models
  - infiltration model: Austin Long
  - hydrology and mass transport model: Frank Schwartz
- Event tree approach, rather than Monte Carlo, chosen for probabilistic assessment

1990: model completed, preliminary studies conducted and report issued

1991-1992:

- Model improved using a small group of experts (14 people)
- Focused on 10.000 year EPA release criterion

EPRI / NPG

EPRI TSPA Development History

1993:

- Model extended to 100,000 years
- Dose assessment capability added

1994:

- sorption in the saturated zone added
- SZ porosity adjusted
- key transuranic specie solubilities adjusted

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EPRI / NPG

Regulations Requiring the Use of Source Term Models

Efficiently meeting the regulations drives EPRI's interests

Does each regulation require a different source term modeling approach?

Substantially Complete Containment

- 300-1,000 years of $\leq$1% container "failure"
- Only outer container corrosion model required
  - conservative model/approach?
    - » constant corrosion rate model?
    - » microbiologically-induced corrosion (MIC) important for 1,000 years?
    - » radiolysis effects
  - "best estimate" model/approach?
    - » Stahl, Lamont, Bullen, or other model?
    - » safe to rule out MIC for the first 1,000 years? Not any more, because of lowered thermal loading

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Regulations Requiring the Use of Source Term Models (continued)

- **Substantially Complete Containment (continued)**
- Input parameters (as a function of time) are needed. Will they be available experimentally and/or from auxiliary models?
  - container surface temperature
  - water phase on container surface
  - chemistry
    - » local general geochemistry
    - » general container surface chemistry
    - » chemistry within corrosion pits

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Regulations Requiring the Use of Source Term Models (continued)

Release and Dose Criteria Require a Full Treatment of Source Term

- Container corrosion
  - rate
  - nature of corroded container
    - » pin holes to complete disintegration
    - » importance of corrosion products on local chemistry and radionculide retention
- Cladding corrosion
- Waste form alteration and dissolution
  - surface area
  - congruent or preferential dissolution
  - locally-controlled solubility limits

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Regulations Requiring the Use of Source Term Models (continued)

Groundwater Travel Time

- Source term considerations specifically excluded by NRC
- DOE's analysis of "significant" transport along fast paths requires a source term
  - » a full treatment of source term is probably necessary
  - » is a 'fast path' source term different than the average source term?

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Conservative vs. Best-estimate Source Term Modeling

Conservative

- regulatory "confidence' that reality is less extreme (i.e., models and parameters should NOT be reality)
- modeling should be simpler
- potential examples:
  - » no reduction of container corrosion rate with time
  - » container vanishes upon first penetration
  - » cladding does not exist
  - » neglect benefits of corrosion products, alteration layers

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Conservative vs. Best-estimate Source Term Modeling (contined)

Best-estimate

- try to model reality (requires "validation" if used to meet the regulatory bases)
- models will probably be much more complicated
- potential examples:
  - » decreasing corrosion rate with time
  - » diffusion through pinholes in a partially failed container
  - » presence of cladding
  - » diffusion through partially failed cladding
  - » corrosion products and alteration layers

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General Goals of This Meeting

- Identify the components of the source term model(s) that are *essential* to demonstration of the regulatory goals
  - how complex before you can see what is essential?
  - conservative or best-estimate approach?
  - issue of model "validation" (corroboration)
- Identify any clarification of the regulatory requirements necessary prior to choosing the correct model(s)
- Identify parts of the EPRI source term model requiring improvement and specific recommendations for improving them on a limited budget

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Important Processes for Yucca Mountain Source Term

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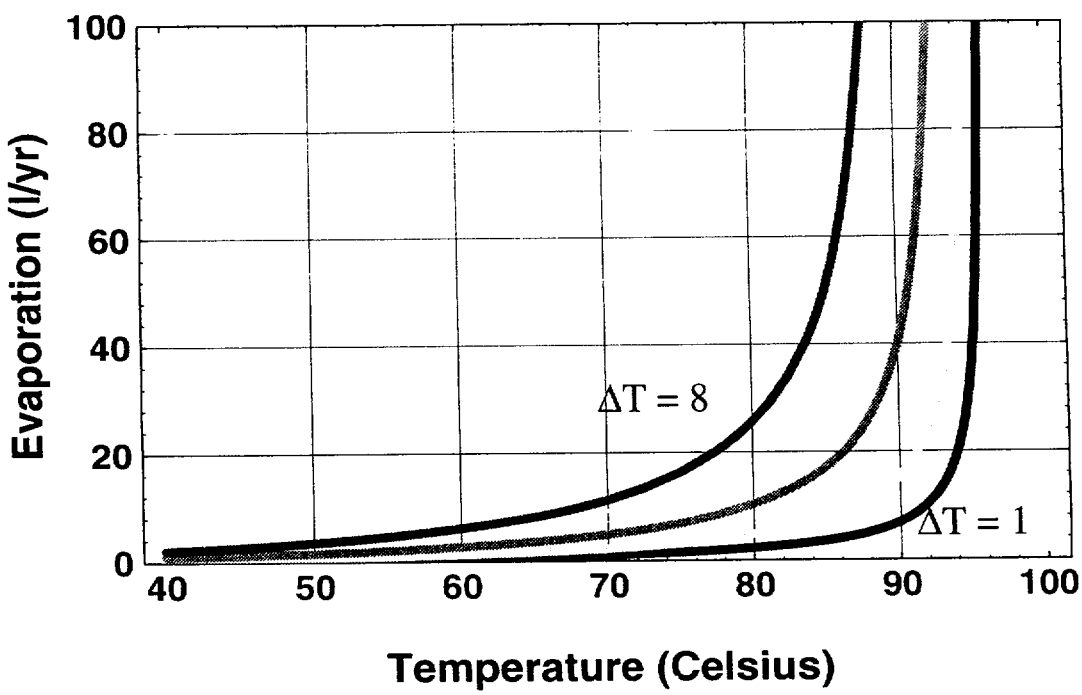
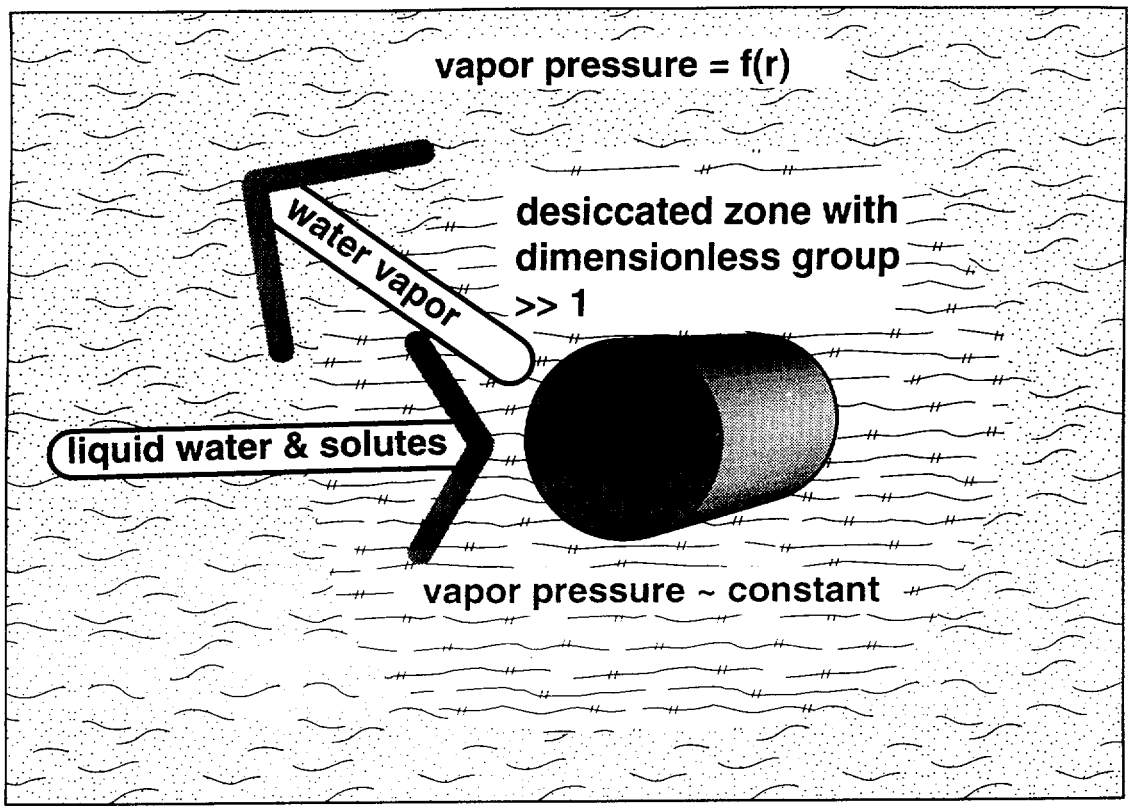
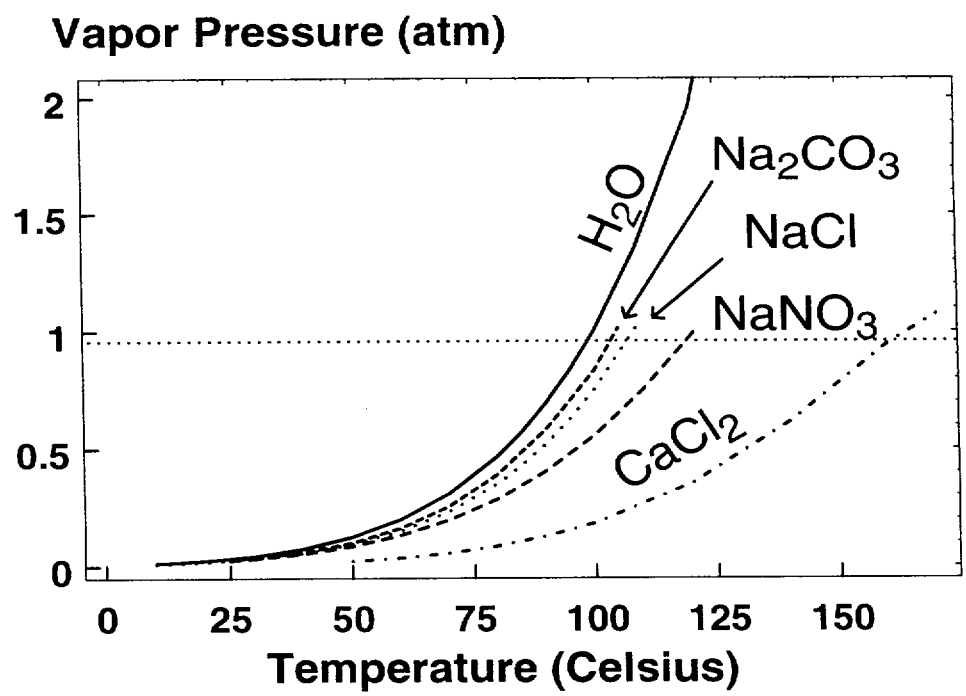


Maximum evaporation rates with rock surrounding the container.

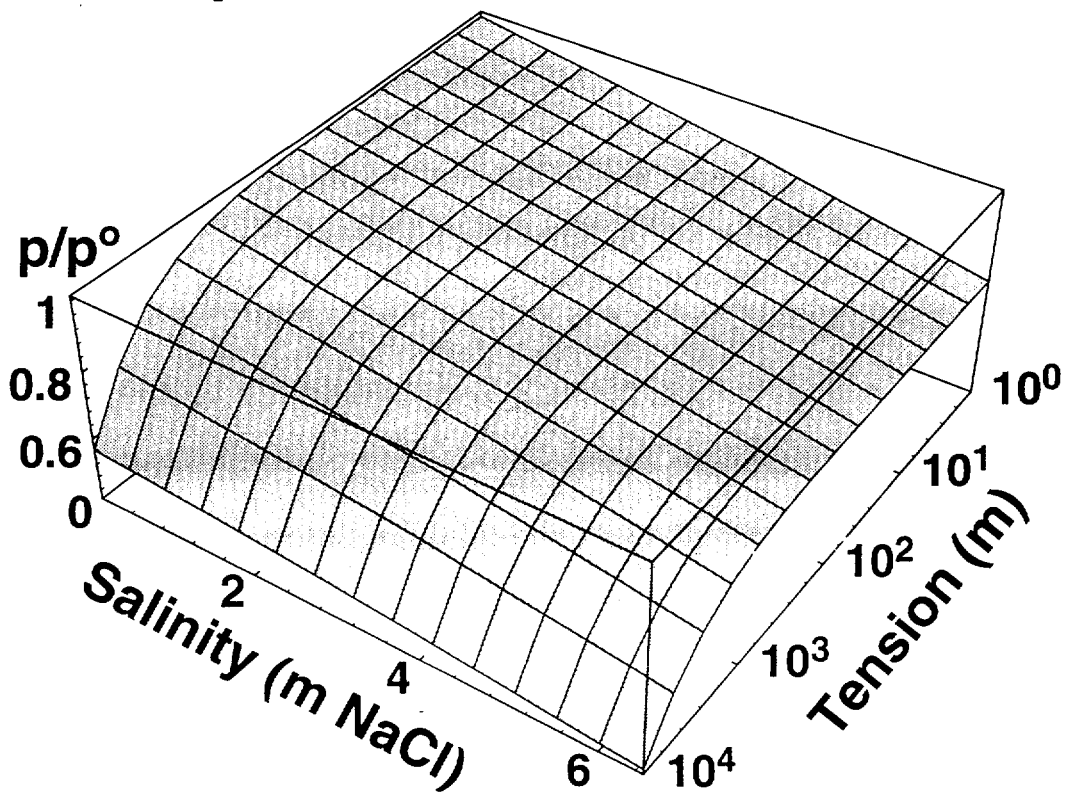
Evaporation Causes a Desiccated Zone





Increased solute concentration in the evaporated solutions can lead to significant vapor pressure reductions & thermodynamic stability of liquid water above the boiling point.

Vapor Pressure Lowering

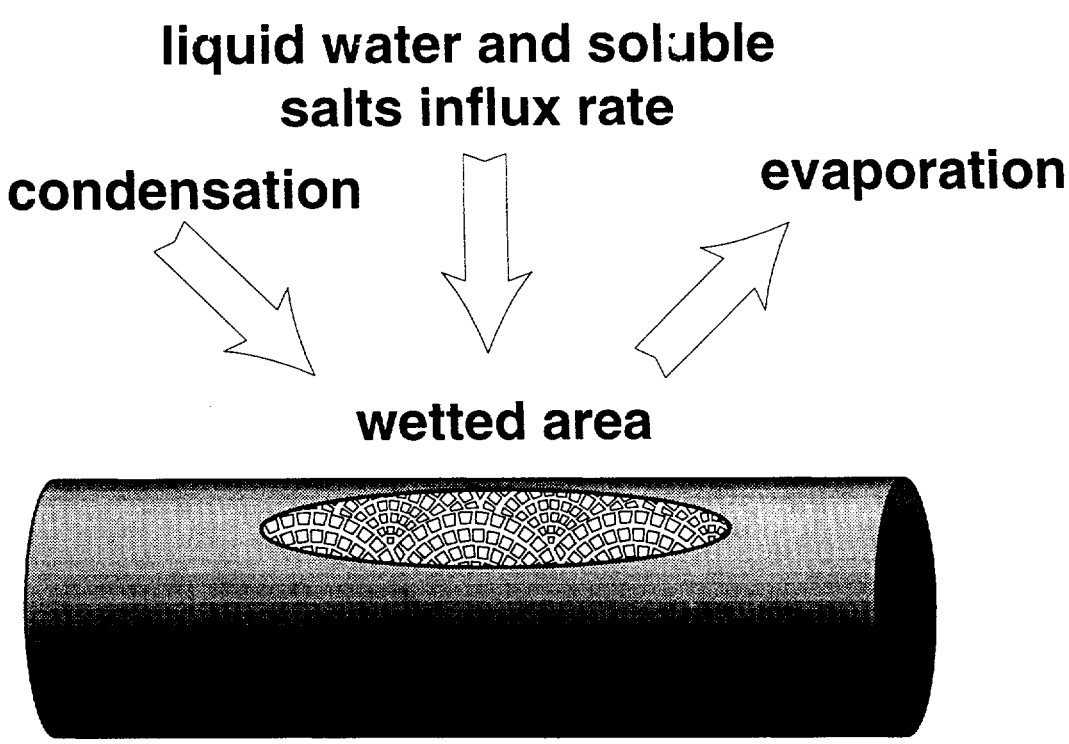


Environment Overview

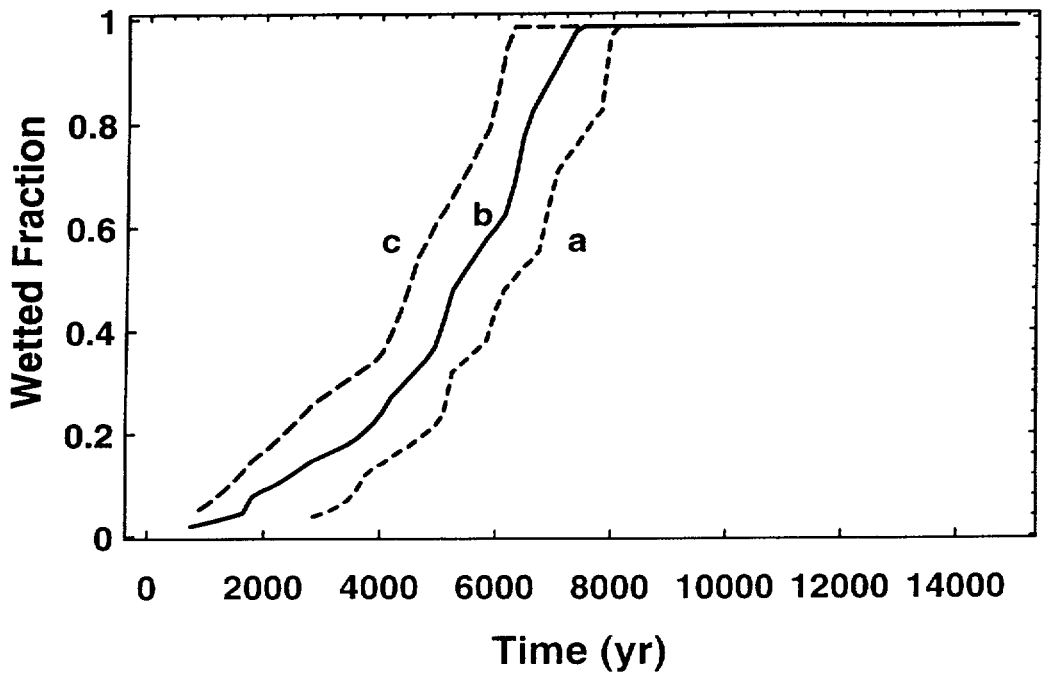
- Thermal – hydrologic – chemical coupling
- Water present as thin films
- Vapor pressure of water remaining in desiccated zone is reduced by surface tension in small pores or from solute concentration
- High spatial and temporal variability of water chemistry
- Small temperature changes result in large changes in water chemistry
- Violates key assumptions in current PA's, especially mass transport approach
- Most experimental data not relevant to anticipated environment
- OK, but give me some example calculations . . .

Calculations (SCCEX)

- Conduction only heat transfer with 3 dimensional Green's function solution and convolution integral with time variant heat generation rate
- Quasi-steady state evaporation based upon binary diffusion of air and water vapor
- Combined solution of set of ordinary differential equations representing analytical solutions to partial differential equations
- Vapor pressure calculated as a function of salinity and T
- Water chemistry simplistically reduced to sodium chloride concentration (salinity)
- Constant water drip rate of 1 liter per year, J-13 water from "magic source"

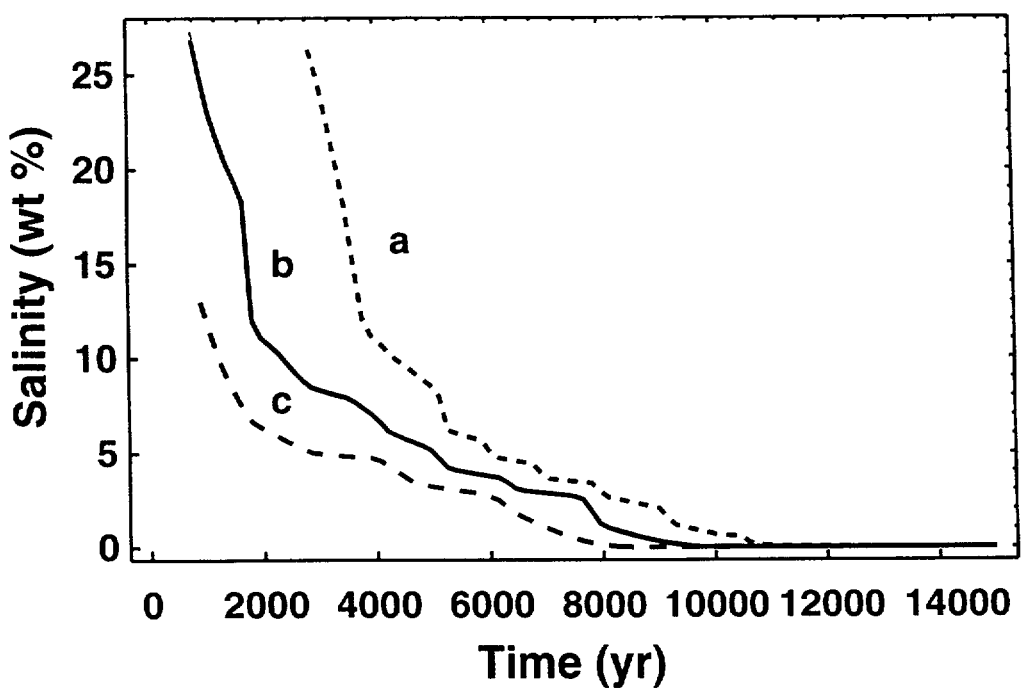


Container Wetting



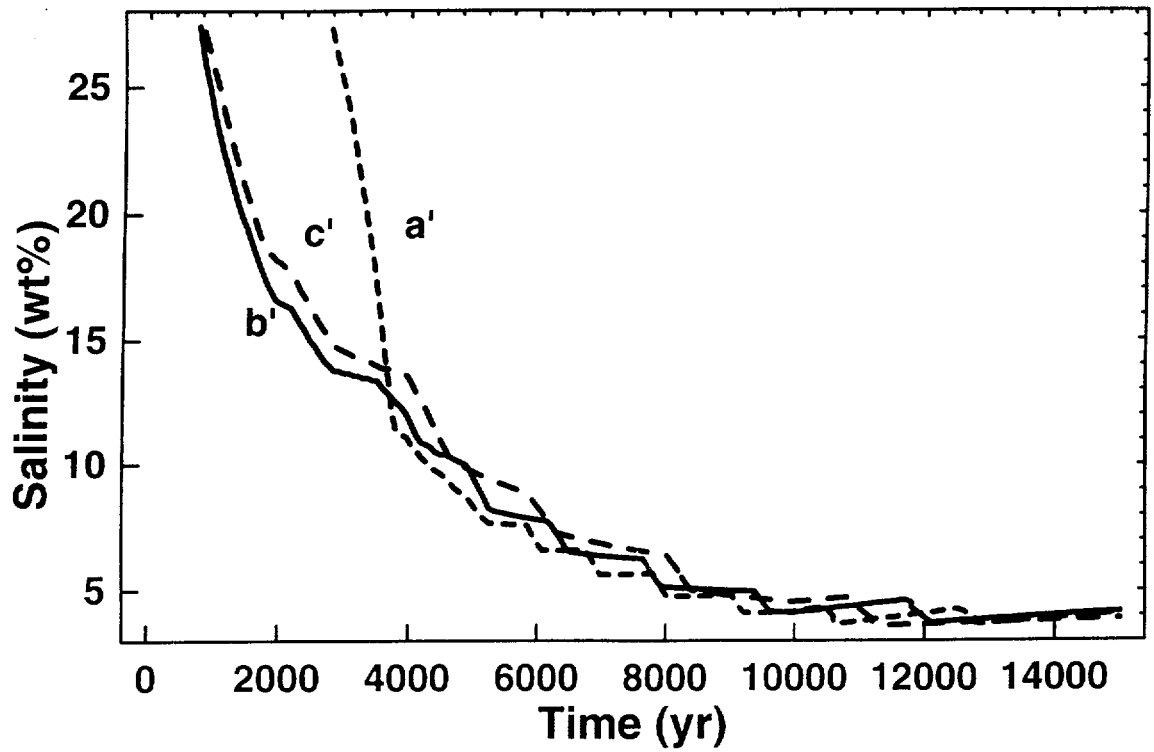
a, b, c = hot, warm, cold repository

Salinity



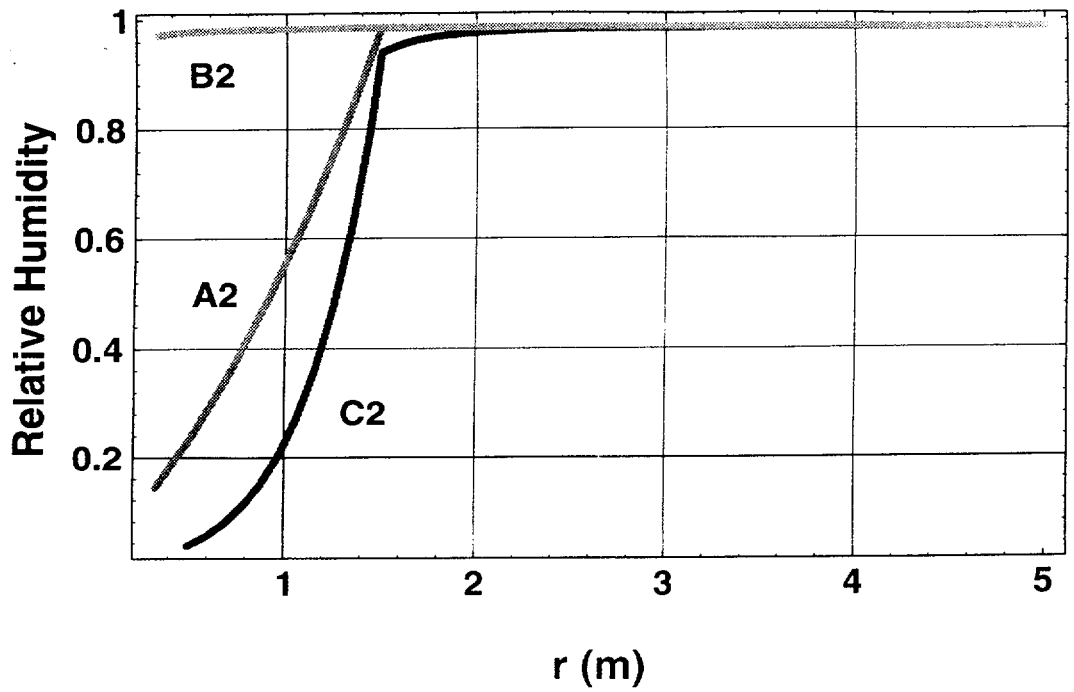
a, b, c = hot, warm, cold repository

No Drip – Salinity

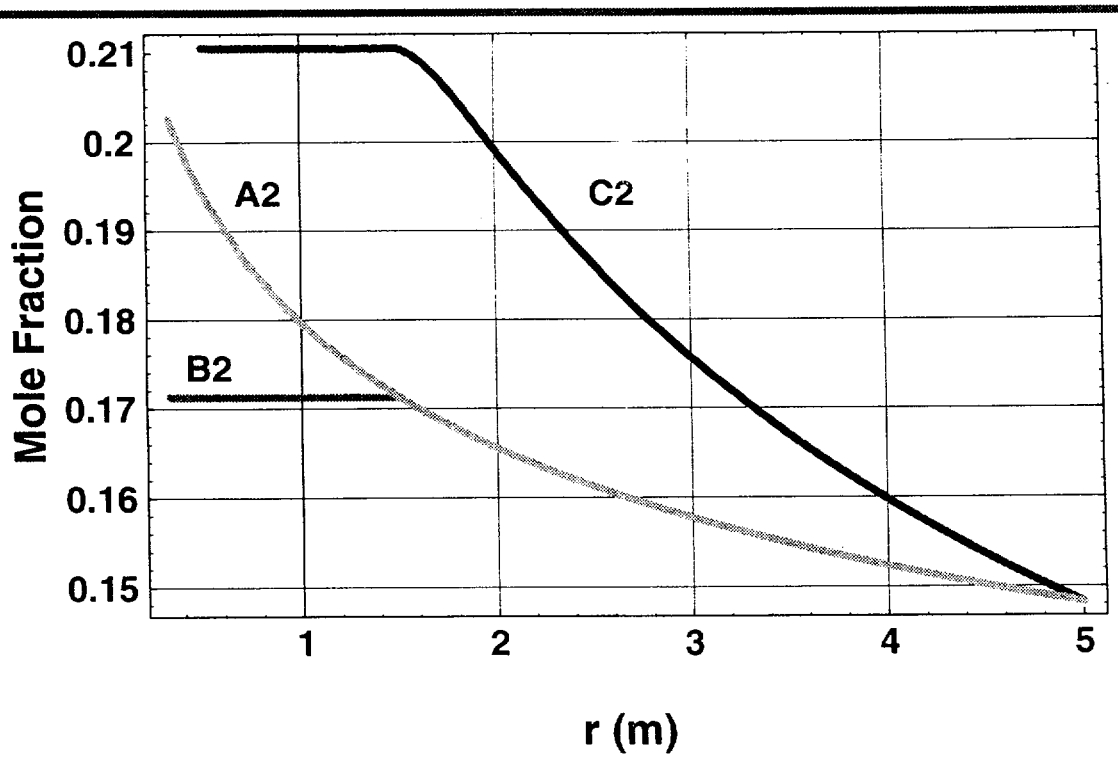


Analytical Solution

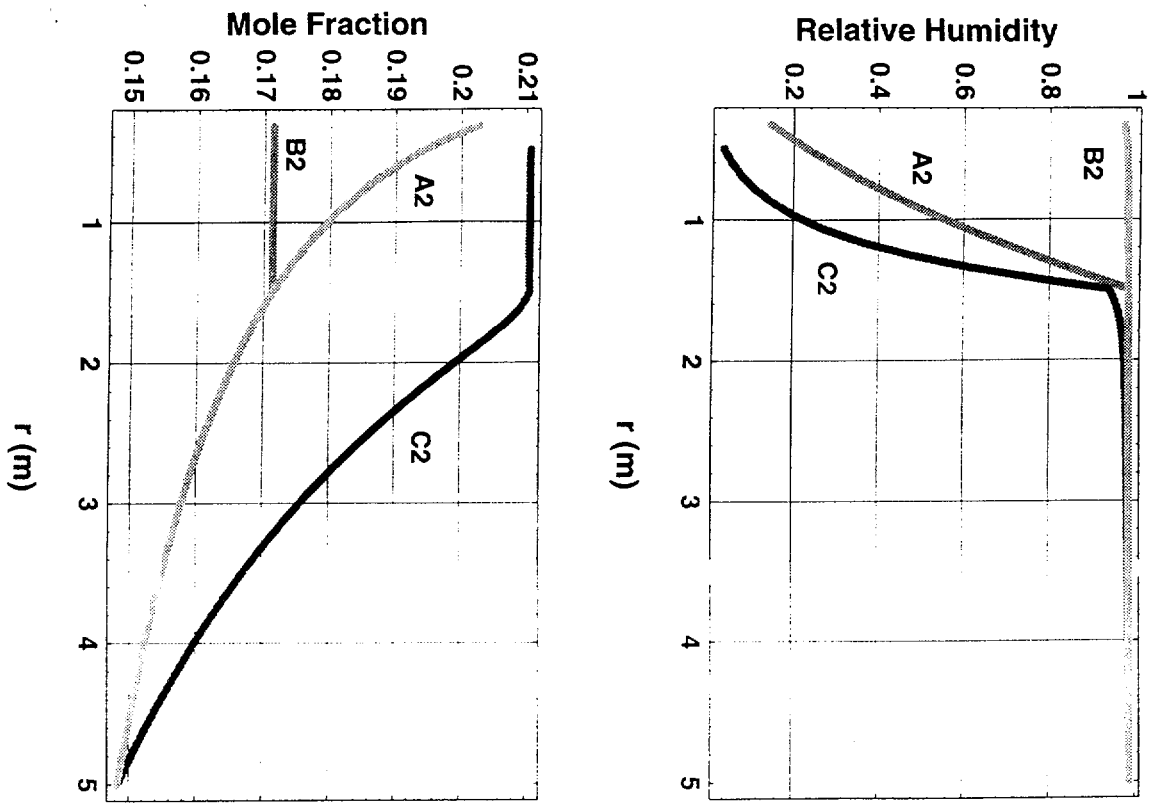
- Quasi-steady state, heterogeneous media
- Equate liquid and vapor fluxes at any point (radius)
- Liquid flux with Darcy's Law & van Genuchten parameters
- Vapor with simplified Dusty Gas Model
- Limiting case reduction to constant vapor pressure of water (i.e., no leachate generation) based upon dimensionless variables
- For limiting case, precise hydraulic properties are no longer required to predict performance
- Granular backfill materials promote limiting case situation and reduced relative humidity



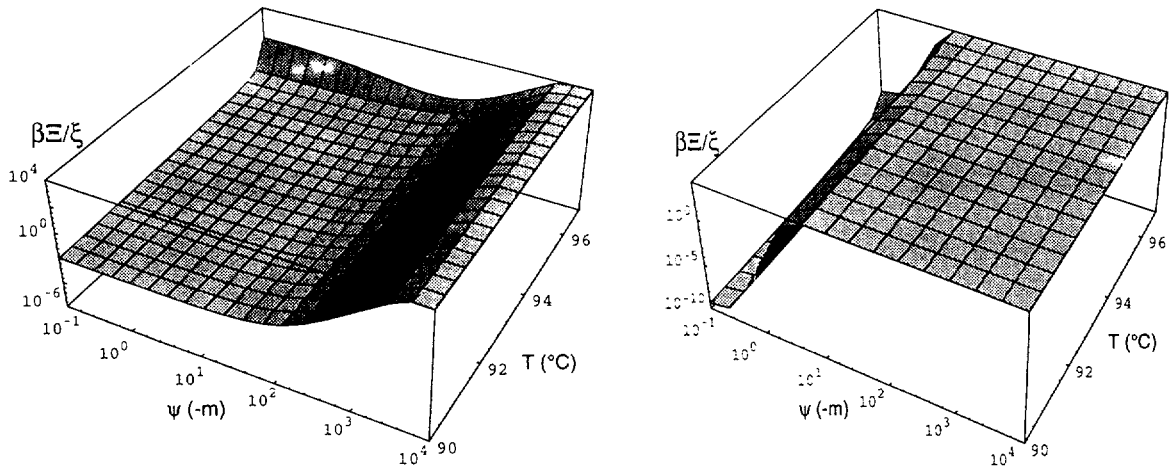
1,000 years, 28 kW/acre repository; A2 = SCP, 1 m backfill; B2 = SCP, no backfill; C2 = MPC, 1 m backfill



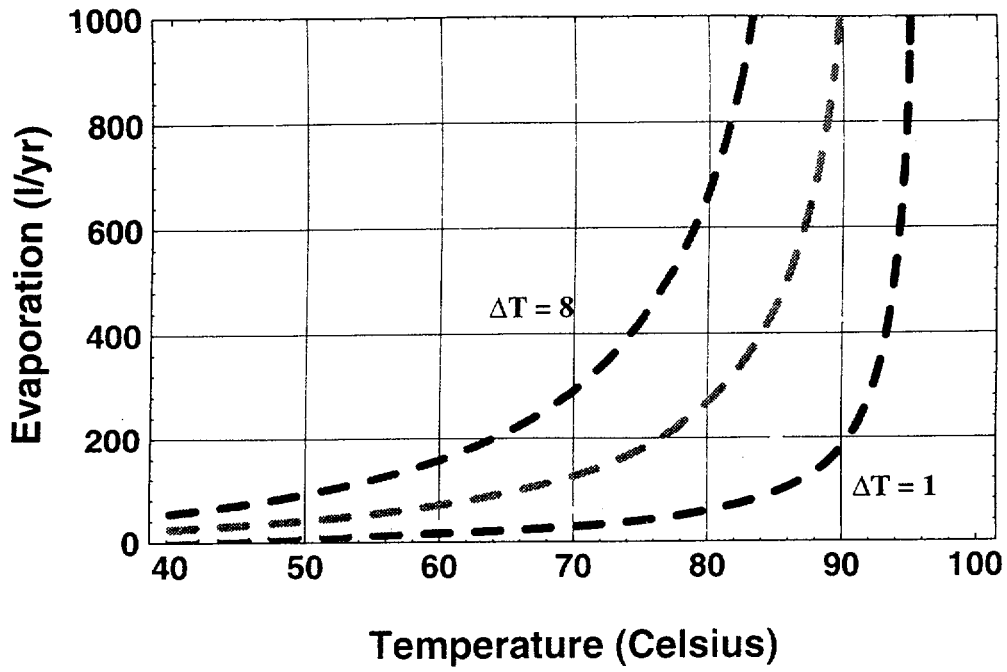
Limiting case present in backfill leads to constant vapor pressure and lower relative humidity



Dimensionless Group



Behavior of dimensionless group is different for rock (left) and granular backfill materials such as sand (right). Granular backfill promotes limiting case solution and dry waste.

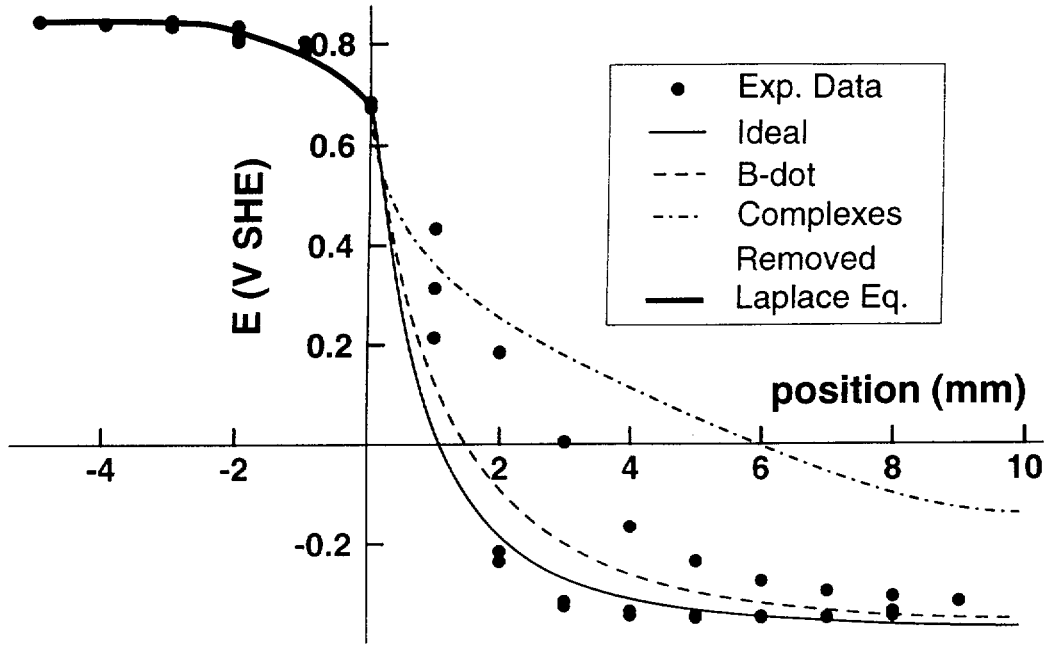


Maximum evaporation rates with sand backfill. Note that ΔT also increases with backfill.

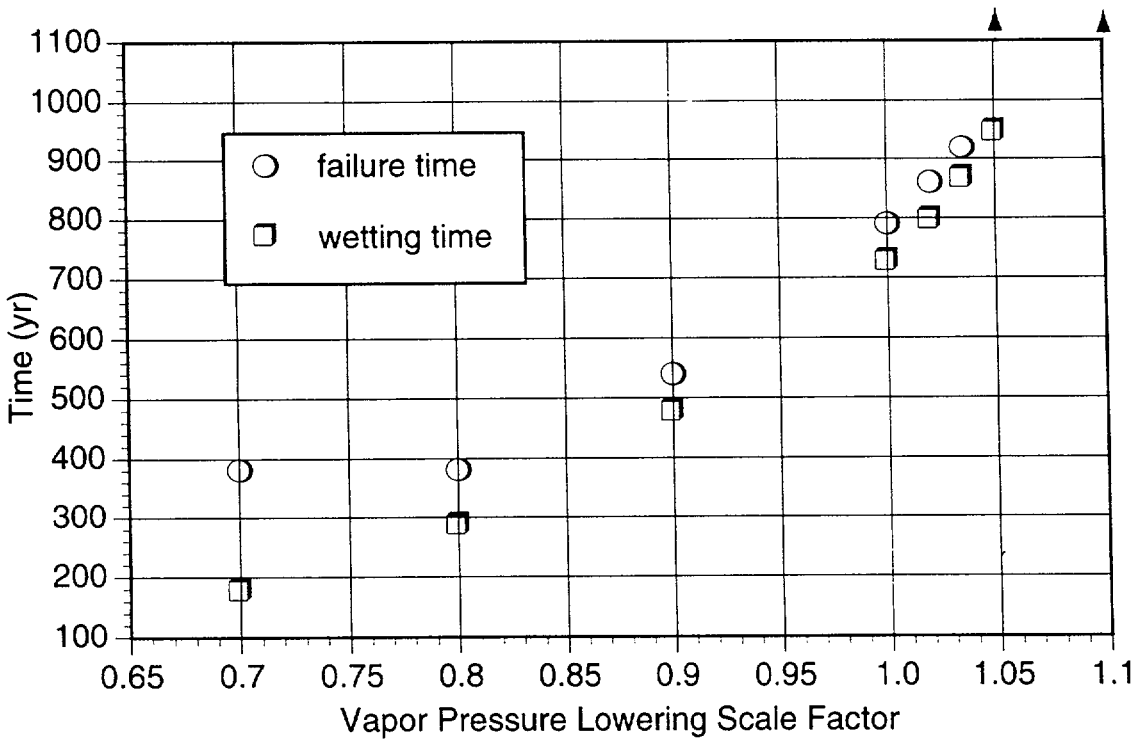
Waste Package Corrosion

- Corrosion potential is unifying concept – valid for any metal
- Corrosion potential versus pitting & repassivation potentials
- Obtain critical potentials from experiments combined with more detailed mechanistic modeling (TWITCH)
- SCCEX calculated corrosion potential considering scale formation, salinity, oxygen solubility, diffusion through scale layer, and water reduction reaction
- Water chemistry is initially saturated brine with trend toward dilute solutions
- High chloride leads to pitting failure soon after wetting – although data is not adequate for firm conclusions

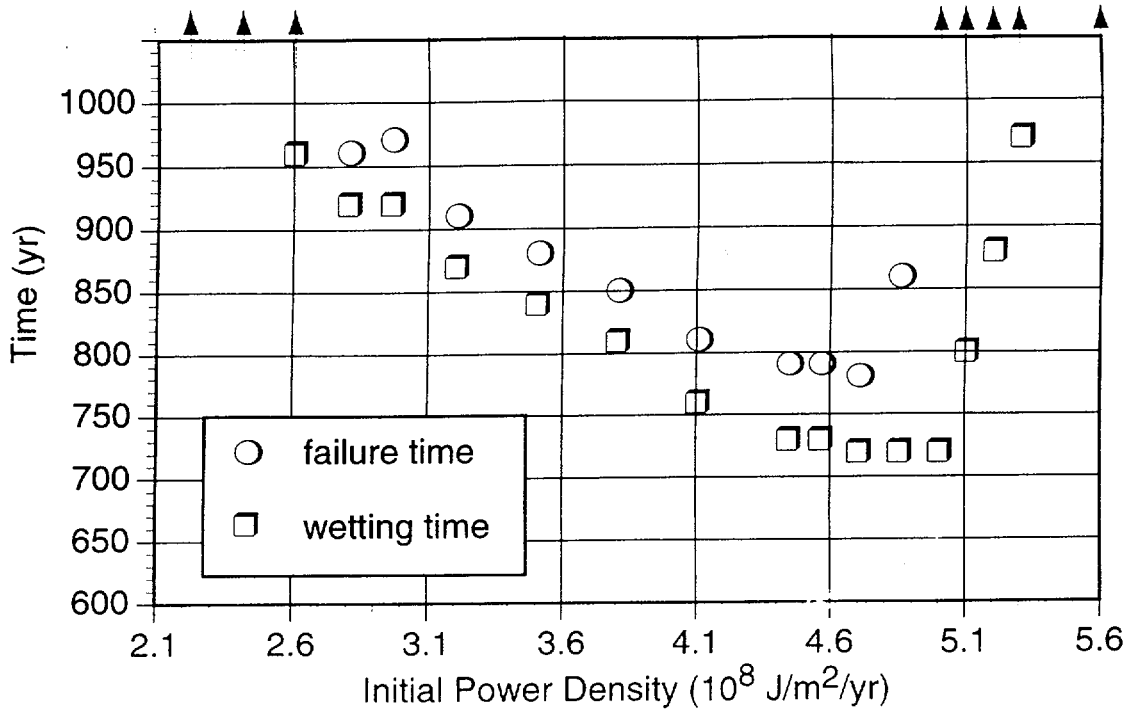
Waste Package Corrosion (TWITCH)



Comparison of measured and modeled potentials in a crevice with TWITCH code



Influence of maximum salt on container failure



Influence of thermal loading on container failure.

Wasteform Alteration and Release

- Radionuclide adsorption on corrosion products
- Complexity of environment:
  - T, salinity, maybe pH and oxidation vary dramatically over time and space & all dependent upon partially failed geometry, pore size distributions and continuity of the "porous medium"
  - can no longer assume a "lumped system," many solubilities and many alteration rates varying on small scales
- Wasteform alteration and corrosion begin long before there is any leachate generation & this can lead to pulse releases
- Galvanic corrosion cells may modify environment (steel, cladding, fuel) and alteration rate
- Much interesting work on dissolution & mass transport but many years away from being defensible in a PA

Conclusions

- Granular backfill materials and dimensionless variables can be used to greatly simplify PA
- The solution does not appear to be converging – new information on waste package environment suggests PA calculations & assumptions worse than originally thought
- The old standby mass transport approach is not well suited to Yucca Mt. & should be abandoned as centerpiece of waste package PA
- No leach approach with bounding calculations elsewhere is most promising – no leach period can be extended >> 10,000 years with proper design
- DOE is risk averse. Perhaps, rather than simply serving as a independent check, EPRI could take a leading role in defining new approaches?

rock can get dried out below residual wetness.
becomes sponge for next deluge

Key Waste Package and Engineered Barrier System Uncertainties

R.W. Andrews
Civilian Radioactive Waste Management System
Management and Operating Contractor

Presented at
Electric Power Research Institute
February 22, 1995

Key Waste Package/EBS Uncertainties

- Near Field Environments
  - Drift-scale thermo-hydrologic
  - Drift-scale thermo-chemical-hydrologic
  - Waste-package scale thermo-hydrologic
- Waste Package Degradation
  - Criteria for corrosion initiation
  - Degradation model
    - » Localized corrosion
    - » Stress-corrosion cracking
    - » Microbiologically-influenced corrosion
    - » Cathodic protection
  - Corrosion rates for corrosion resistant materials
    - » Stochastic pit growth model parameters
  - Percent of waste package surface "failed" vs time

Key Waste Package/EBS Uncertainties

- Radionuclide Mobilization
  - Cladding degradation models
    - » Percent of waste form surface exposed vs time
  - Accessibility of radionuclides (gap; grain boundary)
  - Waste form surface area vs time
  - Water contact with waste form surface area
    - » Percent of waste form surface in contact with water
  - Radionuclide solubilities from undersaturation
  - Colloid formation, stability and effect
  - Effect of man-made materials

Key Waste Package/EBS Uncertainties

- Radionuclide Release
  - In-drift advective flux
  - Effective diffusion coefficient in backfill/packing material
  - Retardation in backfill/packing
  - Mechanical and hydrolgic stability of backfill/packing
  - Colloid mobility

FURTHER DEVELOPMENT OF A SOURCE TERM MODEL FOR A REPOSITORY IN UNSATURATED TUFF

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ABSTRACT

Performance assessments for a potential HLW site at Yucca Mountain should consider the unique characteristics of burying nuclear waste in unsaturated rock. Source term models have been a key part of NRC's first two performance assessments, but have relied on simplified concepts with large uncertainties. NRC is improving its preliminary models for the source term in several important areas, including: (1) incorporation of the latest designs for waste packages; (2) heat transfer near the waste; (3) contact of waste by liquid water, especially thermally induced flows; (4) mechanisms for waste package corrosion; (5) incorporation of spatial and temporal variability at several scales; (6) release of gaseous ^{14}C from the waste; (7) modes of waste package failure other than corrosion; (8) dissolution of radionuclides from the waste form; (9) waste forms other than spent nuclear fuel; (10) abstraction of complex models for use in total system performance assessments; and (11) identification of anticipated and unanticipated events and processes for subsystem performance assessment of the engineered barriers.

I. INTRODUCTION

The potential HLW repository at Yucca Mountain, Nevada would contain waste packages of spent nuclear fuel and other high-level radioactive waste emplaced in partially saturated rock. Excavation and operation of the mined repository will affect the properties of the rock. The repository environment will vary with time as a result of waste heat and radiation that are anticipated to promote significant changes in the chemical and hydrologic environment surrounding the waste packages. In addition, natural and human-induced disruptive events and processes may change the environmental conditions or configuration of the repository.

While many of the problems of the near field environment would be shared with repository concepts for other national programs[1], the prospective Yucca Mountain repository has a set of characteristics in its design that are unlike most other repositories: (1) The waste will be buried in a deep unsaturated rock layer; (2) the environment will be oxidizing because of the presence of air; and (3) The unsaturated nature of the rock would permit waste package design and emplacement concepts which minimize the potential for contact with liquid water and corresponding corrosion (e.g., waste package surrounded by an air gap instead of backfill), although this consideration depends on the final repository design adopted. Furthermore, most of the waste buried in the U.S. repository will be unreprocessed spent fuel, while most other national programs appear to favor vitrified waste from reprocessed fuel. Spent fuel will provide a more sustained heat source and a different mix of radionuclides than reprocessed fuel.

The NRC's regulatory needs for source term modeling has two aspects:

(1) Total system performance assessment - The NRC staff's first two Iterative Performance Assessments (IPA)[2,3] focussed on assessing the performance of the total geologic repository system. The source term models were developed to take into consideration both anticipated and unanticipated processes and events likely to affect repository performance.

(2) Subsystem performance requirements - In developing the standards for disposal of HLW in geologic repositories (10CFR60), the NRC adopted a multiple barrier approach to waste isolation and established minimum performance standards for each of the major elements or subsystems of the geologic repository. The waste packages and underground facility together constitute the "engineered barrier system" (EBS). 10CFR60 established subsystem performance requirements for "Substantially Complete Containment" of radionuclides by

the waste packages for a period of 300 to 1000 years and controlled release from the EBS thereafter. Unlike total repository system performance, subsystem performance needs only to consider anticipated processes and events.

As a regulator, NRC's needs for developing these kinds of models and using them in evaluations such as IPA are somewhat different from those of DOE. The overall objective of IPA is to develop, maintain and enhance the NRC staff capability to review effectively performance assessments developed by DOE, evaluate the DOE site characterization program, and provide input to regulatory guidance and ongoing evaluation of federal regulations for a HLW repository, primarily 40CFR191 and 10CFR60. NRC does not have to demonstrate the performance of either the total repository or its subsystems.

Development of models for repository performance assessment, which support methods for assessing compliance with the regulations, will be an ongoing activity supporting the development of Compliance Determination Methods (CDM) within the framework of NRC's Systematic Regulatory Analysis process[4]. Iterative development of these methods will reflect progress in DOE's site characterization and repository design activities. These CDMs will be incorporated eventually in the NRC's License Application Review Plan (LARP). The development of the source term models used at NRC will be the joint responsibility of the NRC Division of High-Level Waste Management NMSS, NRC Office of Nuclear Regulatory Research and the Center for Nuclear Waste Regulatory Analyses (CNWRA). This paper will describe the directions for improving the present source term models that will be used in further Iterative Performance Assessments (IPA) and technical analyses for the Yucca Mountain repository.

Refinement of models for the source term must rely on the collection of experimental data on all of the areas that will be covered in this paper. While NRC has experimental programs in most areas, the budget for such research is limited, and we expect most of the necessary data to come from DOE and its contractors. NRC does not currently have any experimental work on irradiated nuclear fuel.

II. MODEL ABSTRACTION

While realism and accuracy are important, the computer code describing the highly complicated phenomena for release of radionuclides from waste packages will need to be run repeatedly as part of the probabilistic analysis in IPA, and should therefore be highly efficient as well. Even though the current NRC analysis of the release of radionuclides to the geosphere from the waste form was highly simplified, the source term represented a large proportion of the overall complexity of the assessment[5]. The recommended improvements discussed in this paper will add further to the complexity of the models. A substantial effort will be required to develop models that take into account the many complications of the source term, yet are simplified enough to be practical for IPA.

Three principles useful for developing models suitable for IPA are (1) abstraction - results of complex models that are too computationally intensive to include directly in IPA can be abstracted to faster, more efficient models that contain the most important information, (2) conservatism - a model may not have to be an accurate reflection of the prototype, so long as its results are demonstrably conservative, and (3) parsimony - only phenomena important to performance need to be modeled, and only to the degree needed by the compliance demonstration strategy. Development of models for use in the IPA source term will progress on two levels. Many of the more detailed analyses, codes and models will be developed under the Engineered Barrier System Performance Assessment Code (*EBSPAC*) project. *EBSPAC* is being developed by a cooperative effort of the NRC and CNWRA, mainly to support determination of compliance of the proposed engineered barrier system with NRC performance requirements in 10CFR60.113. In addition, codes abstracted from *EBSPAC* will be incorporated in the SOurce TErm Code *SOTEC*[5]. *SOTEC* has been used in the NRC's Phase 2 IPA to provide rates of release of dissolved and gaseous radionuclides from the EBS to the geosphere, and will be described in an accompanying paper[6]. Among the models to be considered for further development under *EBSPAC* are two-phase flow of liquid and gas in the near field, geochemical effects at elevated temperatures, various corrosion processes, waste-form dissolution, radiolysis, mass transport, mechanical stress, and the performance of partially failed waste packages.

III. RECOMMENDATIONS FOR IMPROVING THE NRC SOURCE TERM MODEL

The development of the following improvements to *EBSPAC* and *SOTEC* are suggested from NRC's experiences with IPA[3,4]:

A. Develop Models for Current Waste Package Designs

IPA Phase 2 was based on the waste package concept described in the Yucca Mountain Site Characterization Plan[7] of single-walled packages placed vertically in boreholes, with an air gap between the container and the surrounding rock. However, since the issuance of the SCP, the DOE has developed a significant interest in more robust waste package concepts for both borehole and drift emplacement, including consideration of overpacked multi-purpose canisters for spent fuel. As part of its ongoing waste package Advanced Conceptual Design (ACD)[8], the DOE has identified various concepts for evaluation: (1) metallic multi-barrier containers, (2) metallic shielded containers, (3)small metallic multi-barrier container, (4) non-metallic multi-barrier container, (5) overpacked multi-purpose canister, (6) Universal Cask, (7) SCP single container, and (8) Defense High-Level Waste Container. Concurrent with the waste package ACD program, DOE is assessing the merits of various repository thermal loading strategies (i.e., cold, intermediate and hot). Any decision regarding repository thermal loading should be integrated with the waste package concept. The behavior of the waste packages in the environment of the geologic repository will depend markedly on the concept finally adopted, the mode of emplacement (e.g., borehole or drift), and whether backfill or an air gap will be employed in the balance of the engineered barrier system design. The current models in *EBSPAC* and *SOTEC* will be modified as DOE progresses in site characterization and makes decisions about its thermal loading strategy, waste package design, waste package materials, and additional engineered barriers.

B. Improve Models for the Effects of Heat

The present temperature model in *SOTEC* uses a semianalytical approach for heat transfer by conduction-only. More realistic models which take heat and mass transfer in two-phase flow into account to better estimate the temperature in the near field and the transfer of liquid water and water vapor[9] could be incorporated in *EBSPAC* and abstracted for *SOTEC*. The more realistic models are needed to better predict the onset of corrosion, subsequent failure of the waste package and the interaction of liquid water with the waste form.

C. Improve Model for Modes of Water Contact

IPA Phase 2 analyses employed simplified treatments for the thermal, hydrological and geochemical environments, based on the SCP concept[7]. Failure of the waste packages by corrosion and transport of dissolved radionuclides to the geosphere are expected to require contact with liquid water. In IPA Phase 2, waste packages were considered to remain dry until their surface temperature dropped below the boiling point of water, and were then assumed to come into contact with liquid water from dripping fractures and wet rock. Improved models should consider plausible rewetting mechanisms for dry rock, such as; (1) the possible influx of liquid water from dripping fractures, (2) condensation of water vapor on container surfaces because of capillary and solution effects[10], (3) rise in the water table, and (4) water reflux driven by repository or geothermal heat[9]. Additionally, alterations to the hydrologic environment from climate change or hydrothermal processes, along with man-made changes need to be factored into models for water influx. Models to determine accurately the contact of waste with liquid water will be highly design-specific to the repository concept finally adopted. Much of this work is expected to stem from confirmatory lab-scale and field heater tests used to validate mathematical models of two-phase heat and mass transfer[11]. Since the experimental data must be necessarily of short duration and small scale relative to those of the repository, reliable mathematical models may be the only way to extrapolate results to greater times and distances. In this regard, the basis for the development of these models will rely on a mechanistic understanding of the processes and events related to waste package interaction with its environment.

D. Develop More Mechanistic Models for Waste Package Corrosion

Corrosion will be affected by a number of coupled processes that include heat and mass transfer, production of metal ions within the crevices and pits and hydrolysis. While the IPA Phase 2 models assumed corrosion to begin when the temperature dropped below the boiling point of water, episodic evaporation and condensation of water on the surface of waste packages may result in high concentrations of aggressive ions well above that temperature[10].

The present version of *SOTEC* considered simplified models for corrosion. General corrosion is assumed to begin when repository temperatures drop below a specified (input) temperature. Once corrosion begins, the corrosion rate is described by a generic power law equation[12]. General corrosion is assumed to represent a passivated metal and is independent of the corrosion potential.

Models of crevice (and pitting) corrosion in *SOTEC* assume a temperature and radiation dependent corrosion potential, and a constant corrosion propagation rate. Episodic evaporation and condensation of water on the surface of waste packages might result in high concentrations of aggressive ions, making them susceptible to localized corrosion. Crevice corrosion is affected by a number of coupled processes, including mass transfer, production of metal ions within the crevice and hydrolysis. While mechanistic models for crevice corrosion exist[13], such models are too complex to implement directly in a total system performance assessment. Therefore, a strategy to develop a simple parametric equation for crevice corrosion for use in *SOTEC* was adopted.

The model for crevice corrosion in *SOTEC* has two characteristics, (1) critical potential for initiation and repassivation and (2) propagation rate. Initiation time is dependent upon the corrosion potential of the metal exceeding the critical potential for crevice corrosion initiation. Subsequent to initiation, crevice corrosion was assumed to penetrate the waste package at a constant empirical rate.

Mechanistically, pitting corrosion is similar to crevice corrosion and it is possible to develop detailed models[14] for predicting critical pitting potentials or electrochemical conditions within the pit. Alternatively, heuristic stochastic models may also be developed[15]. For predictive purposes, critical potential for the nucleation of pits and a critical age for stable pits must be assumed for these stochastic models[16]. These parameters can not be derived from short-term laboratory experiments without the understanding afforded by physics-based deterministic models that take into account more fundamental concepts. Such deterministic models are not suitable for total system performance assessment because of their complexity. The development of the *SOTEC* code followed a strategy similar to that for crevice corrosion with a constant propagation rate assumed for pitting. Empirical models based on experimental data and other models presented in the literature have been developed recently at CNWRA as part of the *EBSPAC* activities[17].

A needed improvement to the *EBSPAC* program and *SOTEC* is a model for initiation and propagation of stress corrosion cracking or other localized corrosion, taking into account the chemistry near the waste packages and mechanical stresses. Prototypes of such models exist[17], but are not yet developed to a point of being reliable predictors of experimental results. Furthermore, they are unsuitable for IPA because of their long run times. Under the *EBSPAC* program, the NRC and CNWRA staffs will select and further develop a mechanistic model for initiation and propagation of stress corrosion cracking. This model will use the chemistry in the vicinity of the waste packages and the stress levels developed from seismic displacement and other loads. This work will emphasize the evaluation of the critical potential concept through a combination of modeling and experimental work. Models of these phenomena will be abstracted for use in *SOTEC*.

E. Include Spatial and Temporal Variability and Parameter Distributions

For the sake of computational speed, IPA Phase 2[4] used an approach in which the entire repository was represented by only seven zones, within which all waste packages were of the same design and experienced the same environment. Future IPAs should deal with the difficult problem of spatial and temporal variability of material properties and external driving forces. If highly simplified models are required, then the IPAs should be able to demonstrate how spatial and temporal variabilities propagate through the system. IPA Phase 2 began to explore appropriate ensemble averages of the temporally and spatially varying input parameters for the simplified models.

An issue related to spatial and temporal variability is the distributed nature of some parameters. The distinction here is that the models might be able to account explicitly for known variations of parameters in space (e.g., temperature) and time (e.g., water flux). The distributed parameters cannot be completely characterized spatially or temporally, but should nevertheless be taken into account in the models. For example, *SOTEC* assumes that the oxidizing fuel can be characterized by a single set of parameters. The spent fuel however is highly heterogeneous, consisting of grains of varying size and material properties[18]. Improved source term models should take the variability of the properties of the fuel into account either explicitly or by defining effective input parameters that capture the variability without making the models too complex for total system performance assessments.

F. Improve Model for the Dissolution of Radionuclides from the Waste Form

The chemistry within the waste package was treated in a highly simplified manner in IPA Phase 2. Upon waste package failure, water was assumed to infiltrate the waste package and come into intimate contact with the fuel. Radionuclides were released from the waste form to the inside of the waste package at a rate determined by the alteration rate of UO_2, and instantaneously from crud, gap, grain boundaries, and cladding. Transport out of the waste packages was limited by solubility of the nuclides. The models did not consider colloids explicitly; colloids were considered to be part of the dissolved inventory. The model could be improved by taking into account the formation and subsequent transport of colloids. Reflective of this interest, the NRC staff is presently conducting a literature survey of the role of colloids in the release and transport of radionuclides from vitrified waste forms and spent fuel. Improvements in the model for waste form dissolution should also consider speciation of the elements released to the water, the contribution of minerals from the groundwater and corrosion products from the waste package, the changing temperature, and other factors such as ionizing radiation. The *EBSPAC* program will also investigate models of spent fuel dissolution based on electrochemical theories[19].

G. Improve Model for Transport from the Waste Package

Mass transfer out of the waste package by flowing water and diffusion was included in IPA Phase 2, based on the SCP conceptual waste package. These transport mechanisms are highly specific to both waste package and overall repository design. Plausible mechanisms for transport from failed waste packages will need to take into account the design finally adopted by DOE. Both the NRC and DOE iterative performance assessments considered rather simplistic idealizations for transport from the waste packages such as bathtub or moist continuous cases[5]. However in future assessments, the NRC staff plans to develop a more mechanistic model which predicts the mass transport of radionuclides from the waste packages. The transport model, in conjunction with the waste form dissolution model, should consider the rates that water contacts and enters the waste package, interacts with the waste form, and transports radionuclides from the waste package by both advection and diffusion. The model should recognize that the suite of waste packages will represent a broad range of varying stages of degradation, with some completely intact and others significantly degraded from both anticipated and unanticipated processes and events. These conditions are progressive over the 10,000 year period of interest. Although conservatively neglected in *SOTEC*, the model could include recognition that degraded waste packages, including failed fuel (e.g., defective cladding), can still contribute to the isolation or controlled release of radionuclides, as demonstrated in static dissolution tests[20].

H. Improve Models for the Release of Gaseous ^{14}C

For several reasons, the release of ^{14}C from the waste was one of the most important radionuclides identified the IPA Phase 2 study: (1) ^{14}C can be released from the waste form as a gas whether or not there is liquid water involved; (2) the estimated inventory of ^{14}C in the spent fuel probably exceeds that which would be allowed under 40 CFR 191; and (3) there may be direct pathways for gaseous flow from the waste to the atmosphere through the unsaturated zone. *SOTEC* considers the release of gaseous $^{14}CO_2$ emanating from the waste, tied to the oxidation of UO_2 from diffusing oxygen and escape of $^{14}CO_2$ through the oxidized film[21]. There is little if any direct evidence of gaseous ^{14}C emanation from spent fuel however, and the ^{14}C releases in *SOTEC* were based largely on speculation on the mechanisms for oxidation of the UO_2, and diffusion of $^{14}CO_2$ through the oxidized film and interstitial spaces.

A better model for gaseous ^{14}C emanations must await more definitive experimental data on spent fuel. Among the questions that await experimental results for the ^{14}C model are[22]; (1) the chemical form of ^{14}C in the spent fuel, (2) the diffusion of the ^{14}C through the spent fuel matrix and the product of its oxidation, and the form of the diffusing substance (e.g., elemental carbon, CO, CO_2),(3) the variation in ^{14}C inventory in the different fuel assemblies because of type of fuel, type of reactor and burn-up, (4) the rate of oxidation of ^{14}C in elemental or compound form; (5) the preferential locations of unoxidized ^{14}C in the fuel (e.g., along grain boundaries, within fuel grains), and (6) the effect of the state of UO_2 fuel oxidation on release; e.g., powdering of U_3O_8 at high temperature[23].

I. Modes of Waste Package Failure Other Than Corrosion

Waste packages might also fail from mechanisms other than corrosion such as seismic shaking, volcanism, and inadvertent human intrusion. While IPA Phase 2 considered failure by drilling, volcanism and seismicity,

the models were highly simplified. Improved models of seismic failure might take into account the range of frequencies of earth motion, and realistic dynamic modes of the waste packages. Models for failure by volcanism might take into account mechanisms of interaction between magma and the waste packages; e.g., corrosive gases and viscous forces. Improved models for human intrusion might consider the site-specific likelihood for drilling, shear forces from drilling fluids, and mechanisms that could bring radioactive material to the surface. These disruptive events could also have a significant effect on the other aspects of the repository performance.

Analytical expressions for buckling are only available for simplified geometries and loading conditions with static loads. A buckling model for a complex geometry and multiple and transient loads would require a complicated and computationally intensive simulation unsuited for IPA. The structural failure considered in IPA Phase 2 was for buckling of a highly simplified waste package weakened by corrosion, with external forces from seismic shaking. Structural failure depends on the engineering design, and the model should be adjusted accordingly for changes. Once the engineering design has been finalized, the structural failure of the waste packages from dynamic and other forces could be analyzed deterministically by numerical and experimental techniques and abstracted for IPA. These analyses would include the possible impact of mechanical fatigue of the waste packages from recurrent, low-intensity seismic activity.

J. Include Models for Other Waste Forms

The NRC staff's first two IPAs focussed on evaluating the performance of waste packages for spent fuel, recognizing that by the year 2030, spent fuel will constitute roughly 97% of the curie inventory of waste expected to be emplaced in a repository[24]. However, this does not mean that the waste form resulting from the planned vitrification of existing defense-related HLW can be neglected as a potential contributor to the overall source term. Accordingly, in future IPAs, the staff will develop a source term model for the expected inventory of glass waste packages with special consideration to the kinetics of glass dissolution, formation of secondary silicate mineral, colloid formation and mass transport of radionuclides. Further, waste forms other than LWR spent fuel and defense-related glass may ultimately need to be considered if they are determined to be potentially significant sources. These may include any transuranic or greater-than-Class-C wastes.

K. Develop Source Term for Subsystem Performance Assessment

Unlike total repository system performance, subsystem performance needs only to consider anticipated processes and events. Accordingly, in order to develop methods to conduct *subsystem* performance assessment (as opposed to *total system* performance), the NRC staff must be able to distinguish those processes and events that are anticipated from those that are unanticipated. 10CFR60 specifies that any processes and events initiated by human activities are unanticipated but the staff has yet to fully characterize natural processes affecting the geologic setting as anticipated or unanticipated. Development of such guidance should accompany source term development for subsystem performance assessments.

IV. CONCLUSIONS

The first two iterations of NRC's performance assessments[2,3] for the Yucca Mountain repository used a set of highly simplified models and a limited set of scenarios to determine the rates of radionuclide release to the geosphere. This paper summarizes the direction for improving models of the source term for IPA and compliance determinations, based on the experiences from these two iterations. Future iterations will consider changes to the design of the waste package concepts, and will rely on data collected under NRC and DOE programs being conducted on thermal hydrology[25], waste package materials[25], and measurements of radionuclide releases from actual and simulated nuclear waste[26].

DISCLAIMER

The views expressed in this paper are the sole opinions of the authors, and do not necessarily reflect the official position of the Nuclear Regulatory Commission.

REFERENCES

1. NEA, *The Status of Near-Field Modeling - Proceedings of a Technical Workshop - Cadarache France, 11-13 May, 1993*, Nuclear Energy Agency, Organization for Economic Cooperation and Development, Paris France, (1993).

2. R.B. Codell, N. Eisenberg, D. Fehringer, W. Ford, T. Margules, T. McCartin, J. Park, J. Randall, *Initial Demonstration of the NRC's Capability to Conduct a Performance Assessment for a High-Level Waste*

Repository, NUREG-1327, U.S. Nuclear Regulatory Commission, (1992).

3. N. Eisenberg, M. Federline, R. Wescott, B. Sagar and M. Silberberg, "IPA Phase 2 and NRC's HLWM Regulatory Program", these proceedings (1994).

4. R.L. Johnson, J.J. Holonich, M.P. Lee, M.S. Delligati, "Nuclear Regulatory Commission Staff Development of the License Application Review Plan for a High-Level Radioactive Waste Repository", *High Level Radioactive Waste Management*, April 26-30, 1993, Las Vegas Nevada, American Nuclear Society, La Grange Park, Illinois, 1243 (1993).

5. B. Sagar, R.B. Codell, J. Walton, R.W. Janetzke, *SOTEC, a Source Term Code for High-Level Nuclear Waste Geologic Repositories*, Center for Nuclear Waste Regulatory Analyses, CNWRA 92-009, (1992).

6. R.B. Codell, J. Walton, B. Sagar, "Models for Source Term, flow, transport and dose assessment in NRC's Iterative Performance Assessment, Phase 2", These Proceedings, (1994).

7. U.S. Department of Energy, *Site Characterization Plan Overview, Yucca Mountain Site, Nevada Research and Development Area, Nevada*, Nevada Operations Office/Yucca Mountain Project Office, Nevada, DOE/RW-0198, (1988).

8. U.S. Department of Energy, *Site Characterization Progress Report, October 1, 1992 - March 31, 1993*, U.S. Department of Energy, DOE/RW-0423, Office of Civilian Radioactive Waste Management (1993).

9. T.A. Buscheck, and J.J. Nitao, "The Analysis of Repository Heat Driven Hydrothermal Flow at Yucca Mountain", *High Level Radioactive Waste Management*, April 26-30, 1993, Las Vegas Nevada, American Nuclear Society, La Grange Park, Illinois, p. 847 (1993).

10. G. Cragnolino, N. Sridhar, J. Walton, R. Janetzke, T. Torng, J. Wu, and P. Nair, *Substantially Complete Containment - Example Analysis of a Reference Container*, Center for Nuclear Waste Regulatory Analyses, CNWRA 94-003 (1994).

11. T.A. Buscheck, D.G. Wilder, and J.J. Nitao, "Large Scale In Situ Heater Tests for Hydrothermal Characterization at Yucca Mountain", *High Level Radioactive Waste Management*, April 26-30, 1993, Las Vegas Nevada, American Nuclear Society, La Grange Park, Illinois, 1854 (1993).

12. R.E. Williford, "Uncertainties in Container Failure Time Predictions", *Material Research Society Symposium Proceedings*, **212:** p. 335 (1991).

13. J.C. Walton, S.K. Kalandros, *Twitch - A Model for Transient Diffusion, Electromigration, and Chemical Reaction in One Dimension*, Center for Nuclear Waste Regulatory Analyses, CNWRA 92-019 (1992).

14. J.C. Farmer, G.E. Gdowski, R.D. McCright, and H.S. Abluwablia, "Corrosion Models for Performance Assessment of High-Level Radioactive Waste Containers", *Nuclear Engineering Design*, **129:** p. 57 (1991).

15. G.A. Henshel, " Stochastic Models for Predicting Pitting Corrosion Damage of HLRW Containers", *Proceeding s of FOCUS'91, American Nuclear Society*, La Grange Park, Illinois: p. 8 (1991).

16. N. Sridhar, *A Preliminary Assessment of Pitting Corrosion Models, Progress Report*, Contract NRC-02-88-005, Center for Nuclear Waste Regulatory Analyses (1992).

17. N. Sridhar, H. Walton, G. Cragnolino, P. Nair, *Engineered Barrier System Performance Assessment Codes (EBSPAC) Progress Report, October 1, 1992 through September 25, 1993*, Center for Nuclear Waste Regulatory Analyses, CNWRA 93-021(1993).

18. R. Stout, E. Kansa, H. Buchanan, R. Einziger, and L. Thomas, "Spent Fuel Waste Form Characteristics: Grain and Fragment Size Statistical Dependence for Oxidation Response", *High Level Radioactive Waste Management*, April 28-May 3, 1991, Las Vegas Nevada, American Nuclear Society, La Grange Park, Illinois, 103 (1991).

19. D.W. Shoesmith and S. Sunder, "The Prediction of Nuclear Fuel (UO_2) Dissolution Rates under Waste Disposal Conditions", *J. Nucl. Mat.* **190:**, p. 20 (1992).