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VERIFICATION, VALIDATION, AND RELIABILITY OF PREDICTIONS

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ABSTRACT

The objective of predicting long-term performance should be to make reliable determinations of whether the prediction falls within the criteria for acceptable performance. Establishing reliable predictions of long-term performance of a waste repository requires emphasis on valid theories to predict performance. The validation process must establish the validity of the theory, the parameters used in applying the theory, the arithmetic of calculations, and the interpretation of results; but validation of such performance predictions is not possible unless there are clear criteria for acceptable performance. Validation programs should emphasize identification of the substantive issues of prediction that need to be resolved. Examples relevant to waste package performance are predicting the life of waste containers and the time distribution of container failures, establishing the criteria for defining container failure, validating theories for time-dependent waste dissolution that depend on details of the repository environment, and determining the extent of congruent dissolution of radionuclides in the UO_2 matrix of spent fuel. Prediction and validation should go hand in hand and should be done and reviewed frequently, as essential tools for the programs to design and develop repositories.

1. INTRODUCTION

We are here concerned with verification and validation of long-term predictions made in performance assessment of waste repositories. We must make suitably reliable predictions of how a repository will perform, when radionuclides in waste packages will dissolve in ground water, how fast they dissolve and move into the host rock, and when and in what concentrations and amount they reach the environment. There are two quite different interpretations of verification/validation of these predictions.

1.1 The research approach

One approach is to view the objectives of long-term prediction as the same as those of scientific research: to find the truth of the prediction, to establish what will be the actual values of the predicted results, within some allowable measure of uncertainty. These are desirable goals, but repositories and their surrounding media are complex. Predictions must

necessarily look tens and hundreds of thousands of years into the future. It is doubtful that the truthful and accurate predictions sought by the research approach will be attainable in reasonable time and from reasonable effort. Yet, the research approach to performance predictions is characteristic of many of the efforts now underway.

We recognize the considerable research, testing, and analysis necessary to attain adequate predictive capability. But, we speak here of the end goal of predictions. Seeking the most accurate and detailed prediction of how a repository will actually perform in the long-term future is labeled here as "the research approach".

1.2 The design approach

Our objective is to design a repository that performs sufficiently well when measured against criteria for satisfactory long-term protection of public health and safety. The design approach is to develop a reliable prediction that the performance will not fall outside the criteria for acceptability. The need is to establish sufficiently reliable design predictions, not to predict all details of repository performance.

1.3 Predictive reliability

Predictive reliability in design depends on:

- o clear and reliable criteria for acceptable performance,
- o a theory that can reliably predict that performance does not fall outside the criteria for acceptable performance,
- o reliable parameters to apply the theory.

To illustrate, in designing structures we seldom attempt to predict when and how the structure will fail. Instead, we seek a design that we are reasonably sure will not fail. We design to a conservative prefailure criterion, such as a conservatively specified limit for elastic deformation and creep. We adopt conservative parameters to allow for material inhomogeneities, uncertainties, etc., and use simple well-established theory that we know may not be a detailed representation of the complex mechanisms leading to failure. By conservatively confining our design to material conditions well removed from incipient failure, we can adopt a theory for prediction based on clear principles of physics and mathematics, verifiable for this regime of material behavior. Knowing from such a conservative but reliable prediction that the structure will not fail is a

sufficient and satisfactory result, an attainable result that is far more meaningful for decision purposes than the more difficult and uncertain results from attempts to predict the detailed response of a structure stressed to its failure limit.

2 VALIDATING DESIGN PREDICTIONS

Predicting that the system being designed will not fail, that it will not fall outside the criteria for acceptability, is the goal of design. Verifying and validating that such a design prediction is reliable is a far more realistic objective than attempting to verify and validate predictions of actual performance.

Distinctions between the design and research approaches to prediction and validation are too easily obscured. The U.S. Nuclear Regulatory Commission states [22]:

"Validation - Assurance that a model as embodied in a computer code is a correct representation of the process or system for which it is intended."

Interpreting the above as requiring truthful and accurate predictions of actual performance and assurance that such predictions are, in fact, correct, leads to the tunnel of research with no light at the end! If we adopt the design-approach objective of predicting whether performance falls within the criteria of acceptability, validation becomes more realistic and achievable.

To predict what happens in tens of thousands of years in a repository, we must emphasize sound theories of prediction, more so than in conventional engineering design wherein performance can be predicted, validated, and remedied by real-time testing. To validate that predictions of such long-term performance are reliable, the repository programs must:

- o validate the theory and its applicability,
- o validate the parameters,
- o validate the arithmetic, and
- o validate the interpretation of results.

2.1 Validating the theory

Validating that the theory is itself sound and reliable for predicting the future is a vital step that is too often overlooked. The validity and adequacy of the theory must be scrutinized and challenged through peer review, using the tools of logic, science, and mathematics. Semi-empirical correlations

that contain many adjustable parameters adopted to make the correlation fit real-time data are usually of doubtful utility for extrapolating beyond the era of the experiments. If we must rely on such design approaches, considerable factors of safety must be incorporated to allow for uncertainty. Validation of the extrapolation to the long-term future is difficult to assess.

Unfortunately, this seems to be the dilemma of many predictions of canister life, which rely on real-time corrosion experiments.

Too often we overlook the essential validation of predictive theory and substitute the mere verification that a computer program is in fact producing the result that is arithmetically consistent with the programmed equations.

This narrow definition of "verification" has been adopted by the U.S. Nuclear Regulatory Commission [22], so for this paper we deal with validation as the determination of the reliability of predictions, including the processes of verification.

2.2 The importance of bounding predictions

Usually a reliable theory is simple and its application bounding. Rather than attempting to predict the details of actual performance, the theory is used to predict whether performance will fall outside the criteria for acceptable performance. The use of well established and easily validated design theory to establish bounding values of predicted performance must be balanced against the desire to refine the performance prediction for greater realism, as in the research approach, but with necessarily greater uncertainty in the result.

2.3 The Neretnieks theory to predict canister lifetime

An example of soundly based theory is Neretnieks' clever and innovative prediction [15] of the long-term corrosion of a copper waste canister by ground water. Having established from chemical thermodynamics that sulfide in Sweden's granitic ground water is the only likely corrodant of copper, Neretnieks avoided the uncertainty of extrapolating empirical chemical reaction rates and calculated the diffusive-convective mass transfer of sulfide from ambient ground water through compacted bentonite to the copper surface.

He assumed that any sulfide reaching the copper reacts instantaneously, thereby developing a bounding upper-limit prediction of the copper corrosion rate. His steady-state form of the mass-transfer equation is well established. His governing equations demonstrate that the results will not be affected by sulfide sorption in rock or bentonite. The parameters needed to apply the theory can be measured by well-defined experiments. The diffusion coefficient can be measured in a separate

experiment, or a bounding upper-limit value can be adopted from well-established values for diffusion in a water continuum. The theory requires geometric data on the borehole and waste package, as well as backfill porosity, the thickness of fractures that intersect the borehole, and flow rate of ground water in the fractures.

Such a theory can be validated in part by peer review, as has been done by the many international reviews of the KBS analyses of repository performance.

The theory is by no means an exact representation of all possible phenomena that could affect the rate of mass transfer of sulfide. It assumes no surface diffusion, no osmotic effects or thermal diffusion, no flow or channeling cracks in the bentonite, etc. An essential element of validation is to identify such assumptions, as Neretnieks has done, and to subject them to scrutiny, additional analysis, and experiment to determine their validity.

These are all ingredients of a useful and reliable design theory and of a meaningful validation program.

2.4 Analytical predictions of waste-package performance

A few other examples of predictive theory will provide a focus on what does and does not constitute validation. Examples are given for predicting the transport of contaminants from solid waste into surrounding rock. The conclusions herein also apply to validating predictions of far-field transport to the biosphere.

Beginning in 1980 members of the National Research Council's Waste Isolation System Panel [17] reviewed published theories of how to predict the dissolution rate of borosilicate glass waste buried in a geologic repository.

We learned that laboratory leach data were empirically correlated with the ratio S/V of the sample surface area to the volume of simulated ground water in the reacting container. Reasoning that there is indeed a well-defined volume of water in contact with waste samples in such laboratory experiments, with or without periodic replacement of the water, and that internal convection results in near-complete mixing of the laboratory leachant, we questioned the logic of the proposed theory that would seek some equivalent well-mixed volume of water to associate with a waste solid in a repository. Those questions [17,20,21] have not been answered. Such review, challenge, and response are essential parts of the validation process. Confronted with the dilemma of predicting waste performance in a repository, our Berkeley group developed analytical design predictions of the time-dependent rate of dissolution of a waste solid into surrounding porous rock by solving analytically the governing equations for

diffusive-convective mass transfer in a saturated porous medium surrounding the waste. In the first published form of the theory [2,3,17], the concentration of a dissolved species at the surface of the waste solid was assumed to be the saturation concentration, i.e., the effective solubility of the stable solid phase present when saturation is reached. Adopting an upper limit to the diffusion coefficient that neglects tortuosity, it was shown that this bounding upper-limit prediction of the dissolution rate for borosilicate glass in a repository was orders of magnitude less than the rate then deduced from laboratory leach experiments. In the repository porous rock can support a concentration gradient in the ground water and does not allow the complete mixing that occurs in the laboratory leach experiments. The limited diffusive-convective pathways in the rock are not present in the leach experiments.

This design-approach mass-transfer analysis has since been extended to include backfill between the waste solid and rock, rapidly dissolving species that may be present in the fuel-cladding gap and grain boundaries of spent fuel, effects of transient heating of the repository, simultaneous mass transfer of radioactive-decay chains, and effects of flow in the backfill.

A consequence of the mass-transfer analysis of dissolution is to refocus the issues of how to validate performance predictions for waste packages. For a repository in which the waste solid can be in contact with moist porous rock, the focus is no longer on rate of chemical reaction of ground water with the waste but on the properties that govern the rate of mass transfer in the medium surrounding the waste. There are different theories to be challenged and verified, and different confirming experiments to be performed.

Review, challenge, and acceptance of the predictive theory are essential parts of the validation process but are not sufficient. Our exact mathematical solutions still depend on assumptions of the governing processes. Most are defended by more detailed analysis and by experience and data from other fields, but confirming experiments are desirable. We know of only one such validating experiment. McGrail et al. [14] measured the steady-state dissolution rate of a single-component solid surrounded by a bed of spherical particles through which water flowed. The experimental results were to be compared with mass-transfer predictions of actual performance, so the actual solubility and tortuosity-affected diffusion coefficient were measured in separate experiments. The experimental dissolution rate checked the theory within a fraction of a percent, expectedly confirming the prediction. There are many other facets of the theory that need validation, including predictions of transient dissolution rates.

2.5 Validating bounding predictions

Whether the validation experiment should faithfully reproduce the details of the waste solid in the repository depends on what is to be validated. The theory for solubility-limited mass transfer would not be validated by real-time experiments with borosilicate glass surrounded by backfill and rock to simulate repository conditions. Our more detailed analyses of mass-transfer rate using the empirical reaction rates between water and solid as a boundary condition [29] predict that during the first few years the dissolution rate would be governed by chemical reaction rate, but thereafter by the exterior-field diffusion rate. A delay of a few years for the exterior-field diffusion to control dissolution is unimportant in a geologic repository, but it is crucial in real-time laboratory experiments. This seems to be true also for the dissolution of the UO_2 matrix in spent fuel. As in the development of the predictive theory itself, validation experiments must be devised to challenge and confirm the theory in its intended application.

Where one can locate some integral experimental data on conditions that seem to simulate the real material in the repository, exact or close confirmation with the predictions are not necessarily expected. For example, even if one were to find buried copper surrounded by a well-characterized diffusive-convective transport medium as analyzed by Neretnieks, the corrosion rate inferred from the measurements might be much lower than predicted because of a slow chemical reaction rate. The validating result would be that Neretnieks predicts corrosion rates greater than measured, confirming that his predictions are bounding. The actual data could be useful, however, if they yielded enough information about the chemical reaction rate to be included in reliable long-term predictions of repository performance.

Using geologic analogues to validate predictive theory is a worthwhile objective, but to date has achieved little success, in some cases because of incorrect application of theory [4,18]. Using the predictive theory to confirm observations of results of long-term dissolution and transport of a mineral requires knowledge of the controlling parameters and their change over the long time when the dissolution process has occurred. Such attempts may be worthwhile, however, as will be any exercise that questions and challenges the validity of the theory.

2.6 Application to spent fuel

Our mass-transfer analyses predict very slow solubility-limited dissolution of the UO_2 matrix of spent fuel, with fractional dissolution rates of the order of $10^{-12}/\text{yr}$ to $10^{-8}/\text{yr}$, depending on whether the environment is reducing or oxidizing. These are bounding estimates and neglect expected reduction of the liquid

diffusion coefficient by tortuosity. Similar fractional dissolution rates of the other radioelements in the UO_2 matrix are predicted if these radioelements dissolve congruently with the UO_2 .

The assumption of congruent dissolution is reasonable but requires experimental validation. Present experiments [5,6,8,9,27,28] indicate congruent dissolution for the actinides and some of the fission products. Data for other fission products, such as Cs-135, in the matrix are difficult to obtain, because of interference from the more rapid short-term release from grain boundaries and from the fuel-cladding gap. Validation of congruency should include tests of possible continuing releases of radioelements accompanying the possible restructuring of the spent fuel matrix to a more stable solid form of uranium when the solution is saturated with uranium, as well as tests and further analyses of the effect of alpha radiolysis to create a moving redox front of uranium precipitation [16].

Our mass-transfer analyses predict that during the first few hundred years of exposure of spent-fuel to ground water release rate into rock will be dominated by the mass-transfer of readily soluble species present in grain boundaries and in the fuel-cladding gap, assuming about one percent of the total inventory of these radioelements can be readily dissolved in water [12]. Validation must include careful measurements of the inventory of radionuclides available for such rapid dissolution and release.

Our mass-transfer analyses have been extended to predict the effect of repository heating on the transient release of radionuclides into the rock. Reliable prediction requires valid determination of the effects of temperature on solubilities, the diffusion coefficient, and the sorption distribution coefficients.

2.7 Validation of waste-package dissolution rate in tuff

Validation of waste-package performance sometimes has little to do with the waste package. For example, the design for a waste repository in unsaturated tuff provides an air gap between each waste package and the surrounding rock. Infiltration water can drip on the waste package. The tuff project [24] estimates upper limits to waste-package release rates from a waste package by multiplying saturation concentrations of waste constituents by the volume flow rate of ground water contacting each waste package. The simple predictive theory seems reliable and requires only data on flow rate and saturation concentrations. It also requires validation that the porous rock does not contact the waste packages. Otherwise, diffusion pathways from dissolved species on the waste surface into the rock can result in transient diffusive mass-transfer rates as much as three orders

of magnitude greater than the bulk-flow solubility-limited release rate at the low flow rates predicted for the tuff repository. Therefore, key issues of validating waste-package release rate predictions include the long-term integrity of the air gap and the diffusion coefficient for dissolved species in tuff.

The tuff project also presents [24] an alternative estimate of the release rate from a waste package in contact with moist tuff, based on a mass-transfer equation [10] from a boundary-layer approximation. Here validation must question the theory, which agrees with our exact solution [3] at high flow rates but considerably underestimates the mass-transfer rate at the low water infiltration rates (ca. 0.003 to 1 mm/y) estimated for the tuff project.

2.8 The effect of the performance criterion

The above illustration for the tuff project also points out that the validation issues depend on the performance criteria. In those countries where the performance criterion is stated as a maximum allowable concentration of dissolved species in ground water reaching the environment, calculation of bulk-flow solubility-limited release rate, based on total water flow through the repository, would be a bounding value of the source-term for far-field calculations. However, the U.S. Nuclear Regulatory Commission requires that the release rates of radionuclides from waste packages into the surrounding rock be no greater than 10^{-5} /yr of the 1000-year inventories. At the low flow rates predicted for the U.S. repositories, the transient rates of molecular diffusion of dissolved species into the surrounding rock dominate the release rate into the rock if diffusion pathways are present, and the estimate of bulk-flow solubility limited release is not bounding.

2.9 Validation of waste-package dissolution rate in salt

To demonstrate expected compliance with NRC's release-rate criterion, the U.S. salt repository project adopted a simple bounding estimate by calculating the rate of migration of brine into the cavity between the waste-package and bore hole and multiplying by the solubility of each elemental species in the waste solid [25]. The theory is conceptually simple, but its application requires a valid theory to predict brine migration in salt. Unfortunately, the earlier concept that brine migrates through polycrystalline salt according to laws derived for thermally induced migration within individual crystals is now known to be wrong [13]. After leaving a salt crystal, brine migrates along grain boundaries and is driven largely by pressure gradients. Predicting the space-time-dependent pressure gradients in heated salt and predicting the resulting brine migration is a formidable challenge.

Here a more mechanistic theory [19] of release rate into salt seems more reliable and may be easier to validate. We expect that a borehole cavity to receive migrating brine will not persist for many years. Bradshaw [1] has calculated that within a few years after waste is emplaced the heated salt will consolidate against the waste package. Thereafter, no brine can accumulate in the bore hole, and pressure gradients are in the direction to cause some migration of brine back into the rock. If our present estimates are correct that brine migration must cease after a few years, the rates of release of dissolved species into the salt can be solved by the applying the analytical solutions of mass transfer by molecular diffusion in the grain-boundary liquid. The new theory for release rate in salt involves entirely different phenomena and parameters, and requires a different validation program than does the borehole-accumulation theory used by the project.

2.10 Validation of the time of container failure

The U.S. Nuclear Regulatory Commission requires substantially complete containment of the radioactive waste for 300 to 1000 years. The iron-alloy containers planned for the U.S. projects are not amenable to the mass-transfer predictions of container life adopted for the KBS-3 copper containers. Even though the designed life for the U.S. containers is much shorter than for the KBS-3 containers, developing and validating a theory for predicting the container life may be a more formidable problem.

Here the validation of container-life predictions is clouded by the lack of a clear criterion for acceptable performance. What constitutes "substantially complete containment"? It is tempting to adopt the simple concept of uniform corrosion and assume that the container fails when all of the metal corrodes away. However, fabricated enclosures usually fail first by localized cracks and penetrations. The usual theories of design are difficult to apply. If water penetrates a partly failed container, release of dissolved radionuclides might be small because of tortuous pathways through partly failed outer layers and corrosion products and because of solid phases of low-solubility corrosion products. The protective features of these phenomena should be taken into account, where possible. However, there must be a compromise between the increased detail for realism as contrasted with the loss of predictive reliability, when the greater detail invokes additional physical parameters and requires more data and validation than may be possible within available resources and time.

Our analytical solution [11] for diffusion through well-separated holes shows that for small holes in a thinned container the area proportionality assumed by some waste package codes in predicting release rates is not obeyed. For example, if the Zircaloy

cladding of a single fuel rod contains a sufficient number of 2-mm holes so that the total hole area is about 0.3 percent of the container area, the rate of diffusive transport through the holes is over 40-fold greater than predicted on the basis of hole-area proportionality. This is a consequence of the large concentration gradients and large diffusive fluxes near the hole edges, and it may explain observations by Johnson et al. [7] of large releases of cesium through apertures in Zircaloy cladding. Of course, the holes could become plugged with corrosion products, or the failure phenomena may be such that the fuel cladding is penetrated by only a few openings, so that the net release rate could be appreciably lower than that of an uncontained waste solid. Obtaining sufficient data to reliably predict the effect of partial failure of waste containers on release rate is a challenge to experiment and theory. We will not be able to predict and validate container life until a meaningful criterion for container failure is developed and until a reliable theory to predict acceptable performance towards that criterion is developed.

2.11 Validation of the time distribution of container failures

Prediction and validation of the time distribution of container failures is important to some of the repository projects. An estimated distribution of container failure over many hundreds of thousands of years was found to be beneficial to the predictions of satisfactory performance in Sweden's KBS-3 analysis. The U.S. projects are dealing with steel containers designed to much shorter lifetimes. If the U.S. Nuclear Regulatory Commission's release-rate criterion can be met by statistically averaging the failures of waste-package containers, and if container failures are sufficiently widely distributed over time, the large early mass-transfer rates predicted for long-lived constituents in individual waste packages can be averaged out.

However, if the mean time to failure is as short as a few hundred years and if enough failures occur earlier, the U.S. criterion for release rate maybe difficult to meet for shorter-lived radionuclides of initially high inventory, such as Cs-137 and Sr-90. Because of uncertain theory for predicting life of the steel containers, uncertainty in extrapolating laboratory corrosion data, and uncertainty of what constitutes container failure, we are a long way from even describing a means of validating the time distribution of container failures. This questions the justification for extensive work on developing elaborate calculational techniques that incorporate yet-unspecified statistical distribution of container failures. It is easy to verify the code arithmetic for predictions with assumed distributions of container failure, but predicting and validating failure distribution seems beyond our present capability.

3. WHEN TO MAKE AND VALIDATE PERFORMANCE PREDICTIONS

Projects for geologic waste disposal are complex, challenging, and embody many disciplines of science and engineering. There is a tendency to concentrate now on developing predictive methodology, on compiling scientific data for a full understanding of the geologic system, on developing formalism and procedures for carrying out validation. It is largely only in the mandated Environmental Assessments that we find comprehensive attempts to actually predict the long-term performance of the U.S. repositories, and even these predictions are sometimes treated as being only rudimentary exercises, to be set aside with the expectation of more refined and elaborate predictions that may emerge later. If so, the real value of these early predictions is being lost!

The best way to assess the adequacy of data and theory is to apply them towards the final product of long-term prediction and subject the predictions and predictive techniques to open scrutiny, challenge, and peer review. This crucial element of the validation process is most valuable when done in the early stages of a project, when we have the greatest to learn from the validation challenges and time to adjust our program to make more reliable predictions. Frequent and periodic tests of our ability to predict long-term performance and validation of those predictions are essential tools to focus the programs of design, research, development, and testing and should be priority programs for each project [13].

We need less concentration on the methodologies and formalism of performance assessment and validation and more on doing performance assessment and validation, on defining the issues of reliable prediction, and on resolving those issues.

4. SUMMARY

The goal of performance assessment for geologic waste disposal is to make reliable predictions that the long-term performance will not fall outside the criteria for acceptability. Predictive reliability depends on:

- o clear and reliable criteria for acceptable performance,
- o a theory that can reliably predict that performance does not fall outside the criteria for acceptable performance,
- o reliable parameters to apply the theory.

To validate that predictions of long-range performance are reliable, the repository programs must:

- o validate the theory and its applicability,
- o validate the parameters,
- o validate the arithmetic, and
- o validate the interpretation of the results.

Validation must emphasize questioning, challenging, and testing the theories used for prediction. It must include identification of the substantive issues of prediction that need to be resolved. Validation and prediction should go hand in hand and should be done early and periodically in the repository projects.

Theories of mass transfer by diffusion and convection provide a theoretical foundation for predicting the rate of transport of dissolved species through backfill and into surrounding rock. They can predict bounding values of the rate of corrosion of copper containers.

Reliable prediction of the lifetime for substantially complete containment of steel containers is difficult because of uncertainty in the predictive theory and lack of definition of container failure. Predicting the time distribution of container failure is even more uncertain. A program for adequate validation of such predictions cannot yet be fully defined.

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