A PROPOSED ASTM STANDARD PRACTICE FOR THE PREDICTION OF LONG-TERM BEHAVIOR OF WASTE PACKAGE MATERIALS IN A GEOLOGIC REPOSITORY

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ABSTRACT

Subcommittee C26.13, "Repository Waste Package Materials Testing," of the American Society for Testing Materials (ASTM) has been investigating the problem of making long term predictions of the behavior of materials that will be used in a geologic repository for high-level nuclear waste. Since radionuclides in the waste forms (currently spent nuclear fuel and borosilicate glass) must be contained for thousands of years, predictions of materials behavior are needed over the same extended periods of time. Such predictions present unique problems to scientists. The ASTM provides an appropriate forum for reaching a consensus solution to this problem since nuclear waste producers and receivers, licensing agencies, and general interest scientific groups are represented. The Subcommittee is now nearing completion of a Standard Practice on this subject. The purpose of this paper is to introduce the scientific community to the proposed Standard Practice. The Standard is based on development of a model for the alteration of the material in repository service. As much as possible, the model is based on a mechanistic understanding of the alteration modes. The model is then used to predict the long-term behavior of the material.

INTRODUCTION

In the USA, the Nuclear Waste Policy Act of 1982 (NWPA)(1) mandated disposal of high-level nuclear waste and spent nuclear fuel in a deep geologic repository. Responsibilities for carrying out this mandate were divided among three Federal agencies: The Department of Energy (DOE) for operational aspects, The Environmental Protection Agency (EPA) for standards for protection of the general environment, and The Nuclear Regulatory Commission (NRC) for criteria for licensing aspects of construction, operation, closure and decommissioning of such repositories. The requirements specify that containment of radionuclides shall be substantially complete for a period to be determined, but to be not less than 300 years. They further specify limits on controlled release for a period of up to 10,000 years. Borosilicate glass has been selected in the US and elsewhere as an appropriate wasteform for achieving the necessary immobilization of radionuclides from high-level liquid waste; spent nuclear fuel is presently the only other wasteform.

Evaluation of the behavior of materials over these extraordinarily long periods of time poses particular problems. However, such evaluations are necessary both in the material selection process and to provide reasonable assurance that the selected materials will fulfill their intended functions and, therefore, that the system as a whole will meet the above mentioned requirements. The problems stem from the fact that there is little, if any, data on material behavior pertaining to periods of thousands of years on exactly the materials of concern in high-level nuclear waste repository. Thus, methods are required for predicting the long-term behavior of such materials. Such predictions should draw on all existing knowledge, a carefully planned testing program that makes optimum use of the time (and funding) that is available, and any long-time data on natural or historical materials which, while not identical to the materials of interest, may be reasonably considered to be analogous to them in some relevant respect. This paper outlines a consensus approach to this problem which seeks to capture the essential elements of such a program together with the logical interrelationships between them.

THE ASTM

The American Society for Testing and Materials (ASTM) provides an appropriate forum for reaching a consensus solution to the above problem. Typically, producers, users, and general interest viewpoints are represented in the final consensus. An ASTM Standard Practice (SP) on the subject is now nearing completion. The objective of this paper is to publicize the proposed SP and the major conclusions reached by ASTM Committee C26.13 (Repository Materials) which is preparing that document. Such a consensus view should provide a valuable guide to ensure that the data obtained by DOE to support its license applications are both relevant and adequate. Members of C26.13 (in addition to the present authors) and others who are actively participating in preparing the SP are:

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DISCUSSION OF THE STANDARD PRACTICE

The overall logic of the Standard Practice is diagramed in Fig. 1. This figure formalizes the intent of C26.13 that all the elements shown should be present in the materials evaluation program for the repository project and illustrates the essential interrelationships between the various activities. The five stages indicated on Fig. 1, Problem Definition, Testing, Modeling, Prediction, and Confirmation, are discussed below. An application of the Standard Practice will be illustrated by using the high-level waste glass development program as an example in which a large number of the activities in Figure 1 are already underway.

Problem Definition

The basis for any materials study is formed by information generated in the first two interrelated boxes in Fig. 1: the identification of candidate waste package concepts and materials, and the definition of a credible range on environmental conditions. Information gained from an assessment of these factors combined with literature data then allows one to identify possible modes of alteration for the particular material under consideration. At this point one is in position to begin the test planning and modeling activities in Stages II and III of Fig. 1.

It is important to obtain the best possible information at the beginning of the investigation since the goals of the study are not only to extrapolate site relevant data to long time periods, but also to provide an understanding of alteration modes over a wide range of conditions. In this way the testing program can be focused and the need to perform large numbers of experiments over the same wide range is reduced. For example, the spent fuel to be stored in the repository will come from many different sources, and thus will have been exposed to varying reactor operating conditions. It is very likely that no two sources will provide spent fuel having the same characteristics. Because of the resources and time involved in testing spent fuel performance, only a limited number of tests will be performed. Thus, the performance of the untested spent fuel will have to be projected by using a model. The information generated in Stage I of Fig. 1 is critical in providing confidence that the model will be able to make the predictions required.

For the particular material in question exposed to the range of environmental conditions identified, possible historical or natural analogs should be sought. Such materials provide a unique source of alteration data at very long times and should therefore be fully exploited. It is recognized, however, that satisfactory analogs may not always be available as is likely in the case, for example, of the stainless steels.

Testing

In order to achieve the goal of providing a test database to support reliable long-time predictive behavior models testing is required to:

a) Provide data to establish the possible alteration modes and mechanisms for materials of interest over the expected range of conditions

b) Provide data for predictive behavior models

c) Simulate in a short period of time material conditions or aspects which could occur over long periods of time (i.e., artificially "age" materials) in the repository environment

d) Provide an adequate data base for model validation and reliability and uncertainty analyses

e) Provide data for confirmation, to the extent possible in the preclosure period of the repository, of material behavior model predictions

f) Establish a basic materials property data base for materials for those intrinsic properties necessary for the implementation of the behavior models.

It has proved useful to divide tests by the function they fulfill in the logic for the development of predictive models shown in Fig. 1. Broadly, attribute tests, characterization tests, accelerated tests, and service condition tests play various roles in providing data for model development and validation. Any or all of these tests may also be performed on candidate analog materials. Confirmations tests, on the other hand, are intended to provide data in the period after model validation, and presumed license application, but prior to repository closure to provide further confirmation of materials behavior predictions based on the model. More specifically: attribute tests are conducted to determine intrinsic properties of materials such as density, radionuclide content, etc; characterization tests are conducted principally to furnish information for a mechanistic understanding of alteration; accelerated tests are conducted with variables set at values that will result in increased rates of alteration compared to those expected in service (with due consideration to possible changes of mechanism); service

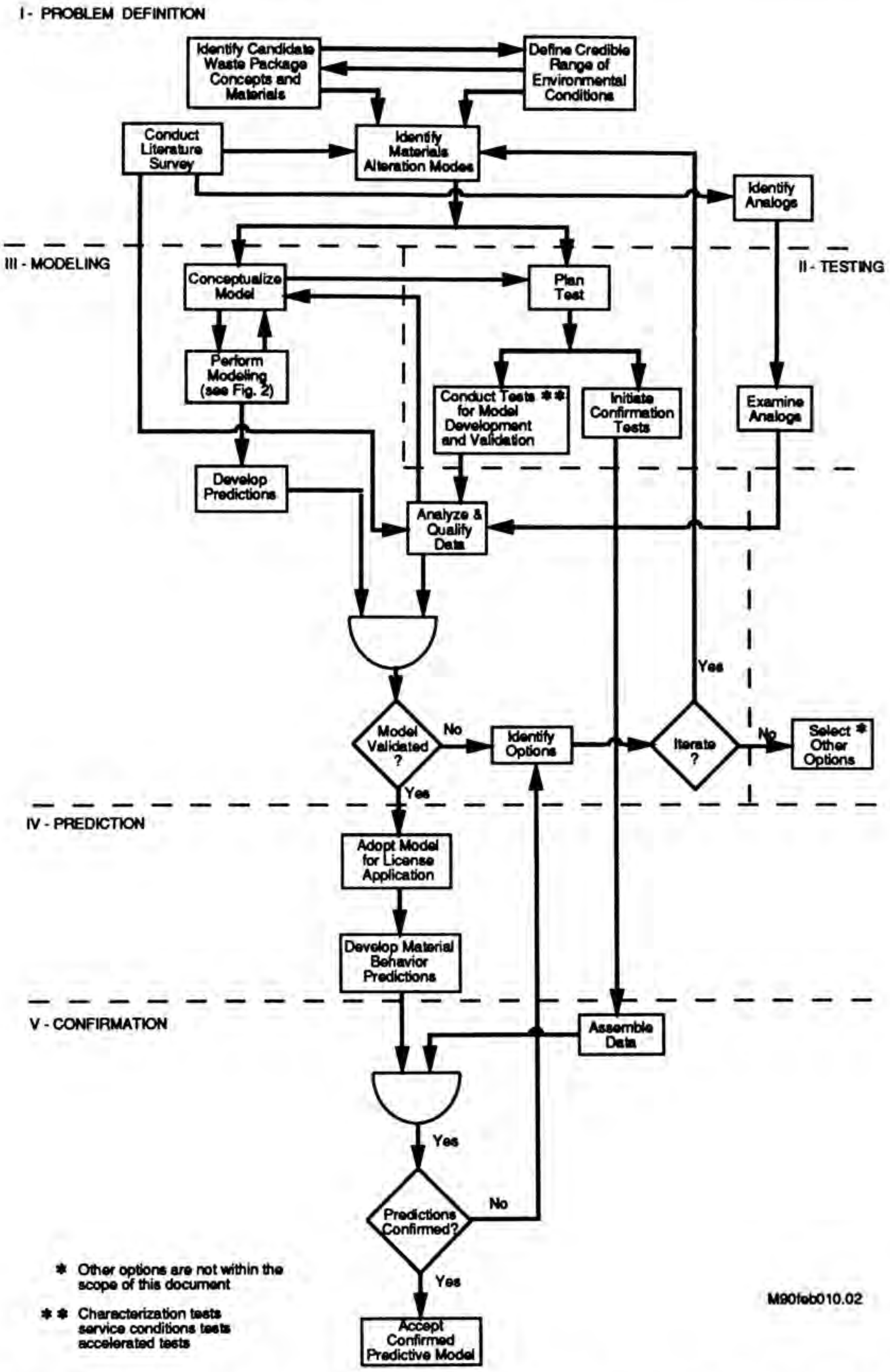

Fig. 1. Logic for the Development of Predictive Models for the Post-Closure Behavior of Waste Package Materials.

condition tests are conducted with variables set within the *ranges* expected in actual service.

Modeling

Model conceptualization will likely begin at an early stage based on findings from Stage I and more formalized modeling activities will develop as test data becomes available. Figure 1 reflects the basic objective of obtaining a validated model which yields predictions that compare satisfactorily with available data. This is achieved by an iterative process of model refinement, data generation, and comparison. However, even if model predictions compare well with data this result does not in itself ensure that the model will yield reliable predictions over a repository-relevant period of time. For this reason some forms of model, e.g. those incorporating a large component of mechanistic understanding of the alteration processes involved, are to be preferred over others, e.g. purely empirical models. In fact, the committee consensus was that while purely mechanistic models are most probably, unattainable, purely empirical models are unacceptable, as is reflected in Figure 2.Acceptable alternatives to purely mechanistic models are:

1) Semi-Empirical Models in which one or more of the parameters of the model are empirically determined. For any empirically determined parameter, the need for the use of such a parameter must be explained, including why a mechanistic understanding is lacking. A full description of what effects could invalidate the use of the parameter for conditions that extend beyond the time used to obtain the empirical value, should be included. Additionally, a description of how the empirically derived parameter interacts with other mechanistically determined parameters must be given, and some method of accelerating the process which the empirical parameter describes must be presented to demonstrate that effects that may occur with increased reaction time do not adversely affect the choice of the empirical parameter. The number of empirically derived parameters must be the minimum possible.
2) Bounding Models which are predominantly mechanistic in nature but refer specifically to the bounding conditions expected in the repository. In this manner the complexity of modeling a process where one or more physical parameters have variable values is eliminated. For example, in the repository the near-field temperature will decrease with time. If the bounding temperature is chosen as the maximum temperature, then the variability of the process with temperature can be eliminated. This option is applicable only if the value used for the bounding parameter can be shown, based on a mechanistic interpretation of the process, to be that value which causes the maximum degree of degradation for the expected range of repository conditions. A thorough evaluation of the bounding parameter chosen, and the effect of that parameter on the reaction process must be documented prior to the use of the bounding condition.

Prediction

Predictions are made at several stages in the logic of Fig. 1. It is useful to differentiate between the two distinct purposes of these predictions. "Model-Testing Predictions" are made in the Modeling (III) and Confirmation (V) Stages and involve comparison with existing experimental data to generate feed-back for model development, validation, and confirmation. However, the primary concern in Stage IV is with the particular problems that arise in using the model to generate justifiable "Repository Service Predictions". These are predictions, made after model validation, of the behavior of each material under the expected service conditions over the extended periods of time that are necessary for assessment of repository performance in connection with license application.

It is recognized that the time variable is unique in the sense that while test data can be acquired over the expected range of service conditions, it is unlikely that data will extend to repository relevant times. Thus, in many cases predictions will be made by extrapolation of available data using behavior models. Where appropriate analogs are available, however, the extrapolation may become an interpolation, affording further confidence in the predictions. Since perfect matches of composition are unlikely it should be noted that models will also be required to interpolate or extrapolate in composition in order to fully utilize available analog data.

Since repository system performance requirements(2) divide the controlled storage time into two periods, one of substantially complete containment and one of gradual release, the critical period over which the materials behavior predictions must be made will depend on the function of the particular materials in the engineered barrier system. For example, the onus of the substantially complete containment requirement may rest mainly on the container materials. Behavior predictions for these materials would therefore be most critical in the containment period, which might be as long as 1000 years after closure of the repository. Spent fuel and glass, however, play central roles in the gradual release phase and therefore considerably longer term predictions are *important for these* materials.

Environmental conditions to which materials will be exposed in the repository may change with time after emplacement and consequently a number of possible progressions of conditions with time, or "repository scenarios", each with some associated probability of occurrence, will have to be considered in making materials behavior predictions.

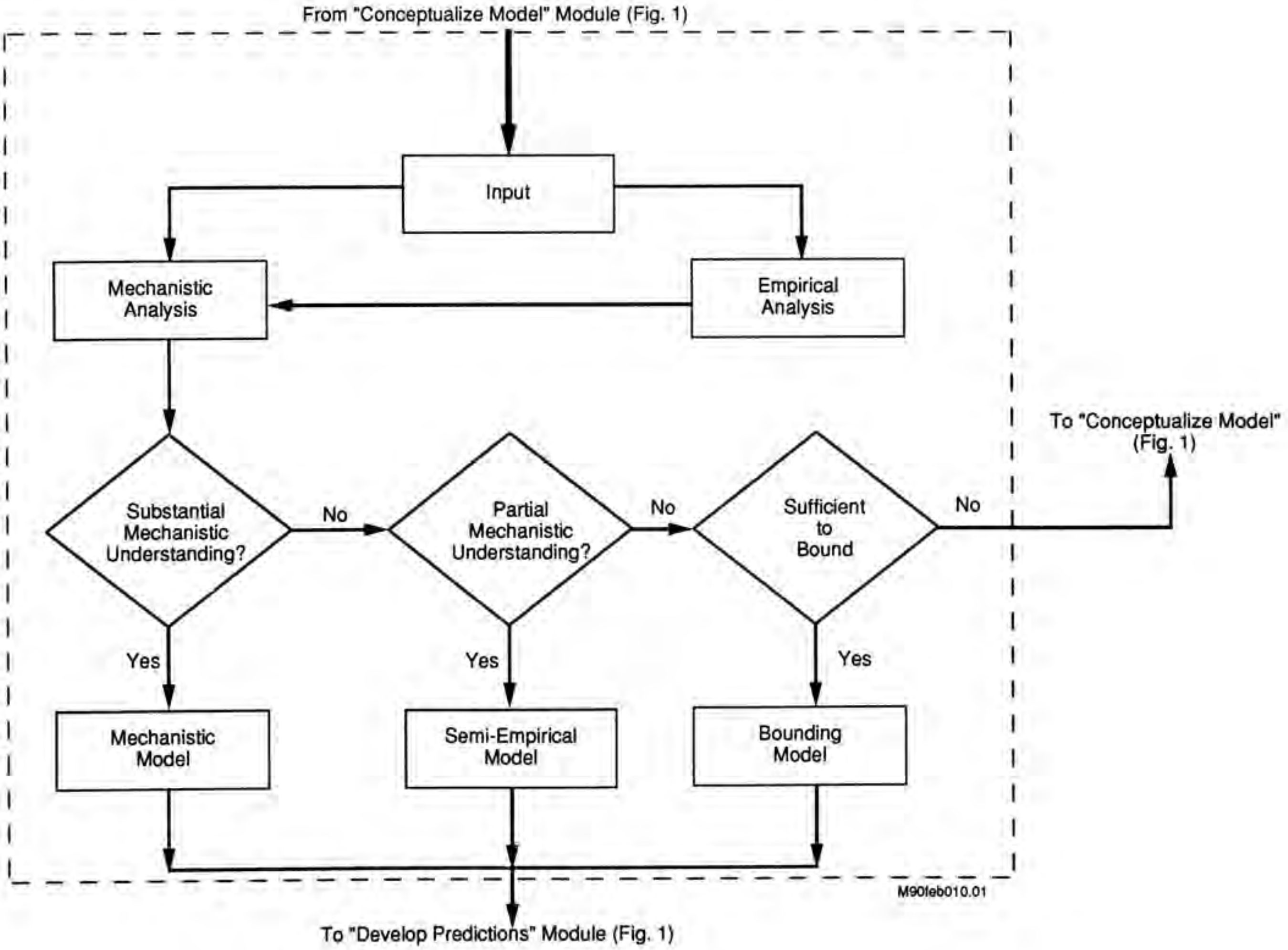

Fig. 2. Detail of "Perform Modeling" Module in Fig. 1.

(Methods for combining scenarios are considered performance assessment and are therefore outside the scope of the SP).

The uncertainty in environmental conditions, and in their time dependence, will contribute to the overall uncertainty in the final materials behavior predictions. Since it is essential to estimate the confidence that can be attached to the predictions, a proper analysis of all sources of uncertainties is necessary. Uncertainties in the predicted long-term behavior will derive from two other principal sources: normal experimental uncertainties in the database which are propagated into the predictions--these can be handled by appropriate statistical procedures; uncertainties in the form of the model that is used, essentially, for the purpose of extrapolation to long-times--these will depend on the confidence with which known or conceivable mechanisms can be excluded for particular materials. The latter are perhaps the most difficult to quantify, but emphasis on mechanistic understanding in model development will increase the likelihood that the true form of the dominant alteration processes is captured, or at least closely approximated. There remains, at least in principle, a possible contribution from inconceivable mechanisms the effects of which, ipso facto, cannot be quantified. However, to this level at least, the predictions should represent the "state of the art."

Confirmation

The role of confirmation tests was discussed briefly in the Testing Section, above. The intention here is to utilize the expected 70 year period between repository licensing and closure. Confirmation tests will produce data up to closure to provide further confidence in the predictive capabilities of the model. It is also likely that during this period new developments in material science will occur that are relevant to the long-term prediction scheme. Thus, provision should be made for further refinement of the model and long-term predictions in this pre-closure period.

APPLICATION TO THE HLW VITRIFICATION PROGRAM

Problem Definition

The principal source of information towards defining the credible range of environmental conditions is presently the Yucca Mountain Site Characterization Plan (3). This document summarizes the geological and hydrological data that have been accumulated on the Yucca Mountain Site and provides estimates of parameters such as ground water composition and flow rate, atmospheric composition, and rock temperature. These are initial estimates and will be revised as construction and exploration of the actual repository site begins; the present data are therefore unlikely to be definitive. Thus, in the HLW vitrification program many of the activities shown in Fig. 1 are occurring in parallel to site characterization. Activities are occurring in an iterative fashion, rather than purely sequentially; new activities are initiated on the basis of the best presently available data and subsequently refined. Such an approach is favored primarily on the basis of expediency.

Borosilicate glass has been selected as the most appropriate means of solidification and stabilization of accumulated liquid high-level nuclear waste in the US and elsewhere. Advantages include its ease of processing and high durability. Other waste package materials and concepts are being selected for the Yucca Mountain Site to further ensure the safety of storing the radioactive waste. These include an overpack of a metal such as a Cu/Ni alloy. An example of a "non-material" waste package concept is the proposed air gap between the rock and the canister which for low flow rates would provide a surface tension barrier against water contacting the container.

Since the remainder of the logic of Fig. 1 is best illustrated by its application to one particular material we will focus on the glass wasteform for the rest of this discussion. A variety of alteration modes might be of interest, including spontaneous changes such as devitrification (which incidently, can be shown to occur at repository temperatures only over time periods even greater than those required for most radionuclide decay (4,5)) and the effects due to radiation. However, the most important alteration modes will involve interaction with water as either liquid or vapor. With liquid water pertinent alteration modes would include dealkalization through ion exchange, gel layer formation, and matrix dissolution. For vapor, crystals form on the surface as the glass hydrates.

An important activity in the Problem Definition Section is the identification of possible natural or historical analogs. For glass there are a large number of natural and historic analogs(6) (e.g. basalts, obsidians, medieval window glasses) which can provide data to substantiate the continued importance of selected alteration modes over extended periods of time or to suggest new modes. The good agreement between extrapolated short term laboratory test data and field data which has been demonstrated for basalt and granite(7) and microtektite alteration(8) provide examples in which the analog helps substantiate both the test method and the extrapolation technique. Furthermore, correlations which include alteration data on a wide variety of glasses (waste, natural, historic)(9) lend confidence to the premise that similar alteration modes are important for the material of interest and the analogs. *It should be emphasized* however that the value of analogs is inevitably limited by the extent of knowledge of the conditions to which they were exposed and the closeness of their composition to that of the material of interest.

Testing

Test planning will draw upon ideas developed from attempts to identify alteration modes as well as from model concepts. For example, matrix dissolution facilitated by hydroxyl ion is an important alteration mode for glass which would suggest that solution pH is an important variable to consider in leach tests. A considerable amount of testing has already been completed on simulated glass wasteform materials. Attribute tests provide basic data on intrinsic glass properties such as density and thermal conductivity. Tests for model development and validation, which include characterization tests, accelerated tests, and service condition tests have the common purpose of providing data to support the development of material behavior models. The SP recognizes that these tests may be functionally interconnected and this is the case for many of the tests that have been used to investigate the leach behavior of waste glasses. Tests such as MCC1(10), MCC3(10), PCT(11), and pulsed(12) or continuous flow(13) tests, and drip tests(14), function as characterization tests. However, to the extent that variables such as temperature or glass surface area to solution volume ratio (S/V) may be overstressed, such tests can also function as accelerated tests. Similarly, by conducting such tests with relevant variables set to values expected to be found in service, by using realistic values for temperature, S/V, and ground water composition, for instance, the same basic test procedure can provide service condition test data.

Several of the test procedures developed for waste glasses, such as MCC1(9) and the pulsed flow test(7), have also been applied to a variety of natural and historical analogs. The application of accelerated test methods to natural glasses has also been used to demonstrate the utility of the test method to accelerate reaction processes without altering the reaction mechanism (15). The combination of such data with field data on analogs is an important component of model validation.

Confirmation tests require special consideration since the intention here is to exploit as fully as possible the period of approximately 70 years between final model validation (and expected license application) and repository closure to acquire confirmation data. Thus, such tests must be specially designed in order to provide data over the considerably longer period of time than that required for tests for model development and validation and should adequately address conditions and synergistic effects expected to occur in the repository environment. Recently a new partial replacement leach test has been developed(16) which addresses the particular problems that arise in providing confirmation leach test data. Also, a laboratory analog test method developed for saturated environments (17) has been modified to also address unsaturated conditions (18). Tests have been ongoing using transuranically doped glasses for several years (18) and the procedure is being applied to glass made from sludge taken from actual high-level waste storage tanks. These test methods should be capable of contributing model development and validation data and continuing over the period necessary to provide confirmation data.

Modeling

While empirical models can be useful tools for assessing data consistency, providing input for further testing, and contributing towards the development of mechanistically based models, the consensus expressed in the SP is that they are not in themselves suitable for the purpose of long-term prediction. Glass leach rates provide an illustration of the statistical basis for this restriction though similar arguments would apply to material alteration rates in general.

It is frequently observed that glass leach rates decrease rapidly with time as solution saturation is approached and at longer times rates become approximately constant or slowly changing with time. Thus, after the initial rapid decrease, leach rate (R) data extending over a time t_1 (a few years) might be well represented empirically by $R = a_0 + a_1t$, with a standard deviation of σ. However, a term a_2t^2 of the order of % would not be detected. The possible contribution of this term at $t_2 = 1000$ yrs, say, is $a_2t_22 = \sigma t_22/t_12$ which for $t_1 = 3$ yrs gives $a_2t^2 = 10^5 \sigma$. Clearly the problem is even worse for higher order terms. This problem dramatically confounds attempts to make empirical extrapolations of data in time by the necessary factors of 10^3 to 10^4. Simply stated, dependencies of alteration rates on high order powers of time must be ruled out by mechanistic arguments, otherwise effects smaller than experimental uncertainties at short times may dominate at long times. If high order terms can be ruled out, the form of the resulting model is then determined and, to the extent the mechanistic analysis is correct, prediction uncertainties are limited to uncertainties in the constants in the model which are derived from test data (neglecting for the moment uncertainties in environmental conditions).

Numerous models have been proposed for the leach rates of waste glasses (19) each with varying degrees of mechanistic and empirical contents and incorporating one or more principal alteration modes. Much of the more recent modeling efforts, however, have centered around a combination of a kinetic model for the glass dissolution process coupled with a geochemical code (e.g. PREEQE(20)) for handling speciation, saturation, and secondary phase formation effects (21-24). Presently this appears to be one of the most promising approaches towards achieving a validated model for alteration of waste glass in contact with liquid (ground) water.

Prediction and Confirmation

While a number of predictions of long-term behavior have been made on the bases of several existing models (9, 21, 22, 24, 25), a fully validated model has not yet been achieved for glass. This is primarily because of the complexity of the leaching process and uncertainties in defining site specific repository conditions. Thus, predictions of the kind envisaged in Section IV of Figure 1 have not yet been made. Similarly, data for the Confirmation Section will only be obtained many years into the future. Continued testing and modeling efforts are therefore required.

CONCLUSION

While each material and its associated set of alteration modes will undoubtedly have its own unique problems, it is expected that the logic expressed in Figs. 1 and 2, and further detailed in the SP, will provide a useful guide in addressing the problem of predicting long-term materials behavior.

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