

A Method to Evaluate Additional Waste Forms to Optimize Performance of the HLW Repository

D. Gombert, L. Lauerhass
Battelle Energy Alliance
Idaho National Laboratory
2525 Fremont Ave, Idaho Falls, ID 83415-3710
USA

ABSTRACT

The DOE high-level waste (HLW) disposal system is based on decisions made in the 1970s. The de facto Yucca Mountain WAC for HLW, contained in the *Waste Acceptance System Requirements Document* (WASRD), and the DOE-EM *Waste Acceptance Product Specification for Vitrified High Level Waste Forms* (WAPS) tentatively describes waste forms to be interred in the repository, and limits them to borosilicate glass (BSG).

It is known that many developed waste forms are as durable as or better than environmental assessment or "EA"-glass. Among them are the salt-ceramic and metallic waste forms developed at Argonne National Laboratory (ANL). Also, iron phosphate glasses developed at University of Missouri show promise in stabilizing the most refractory materials in Hanford HLW. However, for any of this science to contribute, the current Total System Performance Assessment model must be able to evaluate the additional waste form to determine potential impacts on repository performance. The results can then support the technical bases required in the repository license application.

A methodology is proposed to use existing analysis models to evaluate potential additional waste forms for disposal without gathering costly material specific degradation data. The concept is to analyze the potential impacts of waste form chemical makeup on repository performance assuming instantaneous waste matrix dissolution. This assumption obviates the need for material specific degradation models because it assumes the entire matrix is soluble. The existing analysis models, with appropriate data modifications, are used to evaluate geochemical interactions and material transport through the repository. This methodology would support early screening of proposed waste forms through simplified evaluation of disposal performance, and would provide preliminary guidance for repository license amendment in the future.

INTRODUCTION

The HLW disposal system consists of the Yucca Mountain Facility (YMF) and waste product (e.g. glass) generation facilities, either currently located or planned, at the Savannah River, Hanford, and Idaho sites. This system is based on decisions made in the 1970s, and the DOE has tentatively described waste forms to be interred in the WASRD, as well as the DOE-EM WAPS. It is known that the current system is far from optimal for disposal of the diverse HLW streams, and proven alternatives are available to reduce costs by billions of dollars. These changes are also necessary to meet schedule commitments the DOE has made to host states. Current plants must go forward, but retrofit studies are needed now. Preliminary studies in proprietary proposals show \$2B can be saved in Idaho, and another \$1B in South Carolina at the Defense Waste Processing Plant (DWPF) using current technology. However, much greater benefit is expected at Hanford due to the chemistry of HLW there and the relatively low solubility of some constituents in BSG, which allows significant benefit by using a matrix that would accept a greater waste loading.

Responsibility for management of the HLW disposal system is shared between the DOE Office of Civilian Radioactive Waste Management (DOE-RW) and DOE Office of Environmental Management (DOE-EM). The DOE-RW license application must include technical bases that document the acceptability of waste forms against regulatory performance standards. To facilitate progress on immobilization of HLW, the DOE has described the planned BSG waste form and package requirements in the WAPS and the WASRD. These documents actually serve two purposes: 1) they describe the envelope for acceptable HLW products as part of the technical bases in the license application, and 2) they provide tentative waste acceptance criteria (WAC) for products from existing and planned vitrification facilities. The WAC is only tentative, because the final WAC must be written to conform to the licensing specifications issued by the Nuclear Regulatory Commission (NRC) in the actual license. This governing basis for HLW disposal should be reassessed to consider extensive waste form and process technology research and development (R&D) efforts, which have been conducted by DOE-EM, international agencies (i.e. Australian Nuclear Science and Technology Organization (ANSTO), Commissariat à l'Energie Atomique (CEA)), and the private sector. Note that DOE-RW and DOE-EM have established their baseline as disposal of HLW in a BSG waste form. Investigating other wastefoms is out of their current scope, and direct funding is not available to initiate reevaluation. The work reported here was a preliminary sensitivity study to develop a basis for detailed optimization of the HLW disposal system, which could be done pending direct funding. A previously completed calcine dispositions study was used to evaluate sensitivity of repository performance to waste form durability. This will provide for accelerated HLW disposition, more efficient utilization of the YMF, and overall system cost reduction. The ultimate program will require close collaboration among DOE-EM, DOE-RW, and a team that includes DOE national laboratories, international agencies, and the private sector, undertaking an integrated effort to meet the following goals:

1. Reevaluating and revising the assumptions in the DOE-EM and DOE-RW documents that control the waste form/package/disposal program.
2. Qualifying additional protective, but more efficient, waste matrices by matching the matrix to the waste instead of forcing all wastes into BSG. This does *not* include developing new waste forms, only qualifying existing matrices to demonstrate protection equal to or better than the baseline (Environmental Assessment [EA]) glass.
3. Preparing the technical bases to support inclusion of additional acceptable waste forms for HLW disposal in the final application for the License to Receive and Possess.

This project is focused on only the technical evaluation necessary to support the first bullet above. Performance of the repository system is modeled by the Total System Performance Assessment (TSPA), a collection of mathematical constructs designed to evaluate the performance of the whole system. The objective of this project was to evaluate the current models to develop an approach by which sensitivity analyses could be used to determine which waste form parameters significantly affect repository performance as measured by impacts to human health and the environment.

BACKGROUND

Borosilicate Glass as a Mandated Wasteform

The YMF will be a long-term repository for spent nuclear fuel (SNF) and stabilized HLW from fuel reprocessing. The combined performance of the waste matrix, the engineered waste package(s) and facility design, and the effects of the hydrogeologic interaction at the site govern the repository performance as a system for sequestering radionuclides from the biosphere. This system has been the focus of technical analysis for nearly three decades, with emphasis on system performance in protecting the public from radiological hazards. However, the system has not been optimized for HLW disposal.

While DOE HLW/SNF inventory represents about 10% of the repository space, and only about 5% of the activity is in the HLW, there is potential for significant cost-savings in optimizing the HLW processing strategy. HLW processing is costing billions of dollars, thus the potential savings from even small-percentage cost savings are significant. A key limitation on a comprehensive engineering analysis to optimize HLW disposal is the DOE self-imposed requirement in the WAPS that all HLW be converted to BSG. The technical community within the DOE complex knows this to be an inefficient approach for many wastes, but institutional barriers limit action to change the system.

While the United States Environmental Protection Agency (EPA) has identified vitrification as the “Best Demonstrated Available Technology” (BDAT) for HLW, it does not designate BSG as the only acceptable formulation; hence no regulatory driver exists to restrict the acceptable waste form for HLW to BSG. Borosilicate glass was chosen because it is a very stable material, capable of hosting a wide variety of elements in its amorphous matrix. Both DOE-RW and DOE-EM have adopted BSG as the only qualified waste form for HLW disposal, though other forms are used for plutonium and SNF. Although other waste forms are not specifically precluded, BSG is specifically defined in the key documents that govern HLW disposal at YMF, including the WAPS [1], and the WASRD [2]

The WAPS and WASRD restrict the envelope of acceptable waste forms for HLW even further by specifying an outdated version of the Product Consistency Test (PCT), as delineated in American Society for Testing and Materials ASTM C-1285-94 that cannot be obtained through ASTM since it has been superseded by ASTM C-1285-02. PCT measures individual constituents dissolved from the glass into the solution, e.g. boron, sodium, and lithium. The current version of PCT has been expanded to include glass and glass ceramics and provides the protocol determining the appropriate constituents to be measured. A similar protocol should be established for waste forms that do not contain a glassy phase. Though it is commonly stated that HLW forms need not be glass, DOE-RW and DOE-EM regulations and guidance form the de facto WAC understood by the sites immobilizing HLW, and these documents are written around BSG glass. By specifying an outdated version of the PCT standard, the documents do not support testing alternative matrices.

The benefits and limitations of BSG are well known, well researched and widely documented. There are, however, alternatives including iron-phosphate glasses, glass-ceramics, metallic forms, and hydroceramics (waste forms). The knowledge gained from over 30 years of R&D within the DOE complex, international agencies and the private sector is largely being disregarded, even though independent evaluations funded by DOE-EM have recommended these alternatives be considered.

Waste forms must not only incorporate the wide variety of HLW streams at Hanford and Savannah River, but also high activity waste streams generated during accelerated closure activities, and sodium bearing waste and calcine waste at Idaho, which are dramatically different in chemical composition and physical form. It must also be expected to ultimately include wastes that will be generated in meeting the nation's future energy needs. It is known today that waste forms other than BSG are acceptable choices to immobilize key radioactive and hazardous components in existing and future waste streams (e.g., through a technical down selection process a *ceramic* waste form was selected to immobilize plutonium). As large volume waste streams are addressed like calcine, it is imperative to match the waste form to the waste stream characteristics. As an example, INL calcine contains components that are difficult to incorporate in a BSG waste form. It has been demonstrated that waste loadings exceeding 50% can be achieved using a cold crucible induction melter to generate an iron-phosphate glass-ceramic waste form.[3] If calcine is mandated to be immobilized in a BSG, the resulting waste form is neither economical nor an optimally performing waste form. Requiring a single host matrix for a variety of wastes, some of which are nearly insoluble in the BSG matrix (e.g. phosphorous and sulfur) results in lower waste loading that leads to greater waste volume, and, in turn, higher processing and operations costs at both the treatment and disposal facilities. These overall HLW disposal system inefficiencies cost

more money and take more time. Higher waste processing and disposal efficiency can be realized by performing the engineering analyses and trade-studies necessary to select the most efficient methods for processing the full spectrum of wastes across the DOE complex.

Matching an Optimal Matrix to the Waste Chemistry

The potential benefits of matching waste chemistry to the host matrix and using currently available technologies have been documented in several studies [4,5,6,7,8,9] conducted by DOE over the past several years. Matrix chemistry may include multiple phase BSG, non-BSG (i.e. aluminosilicate glass, iron phosphate glass), glass-ceramics, and ceramics, as long as standards are met for durability and stability. If a collaborative effort between DOE-EM and DOE-RW were initiated based on performance requirements, near term benefits could be realized by DOE operations such as the Defense Waste Processing Facility (DWPF) at the Savannah River Site, as well as the Waste Treatment Plant, currently under construction at Hanford by using materials such as iron phosphate or ceramics that can doubly contain higher waste loadings [1,2].

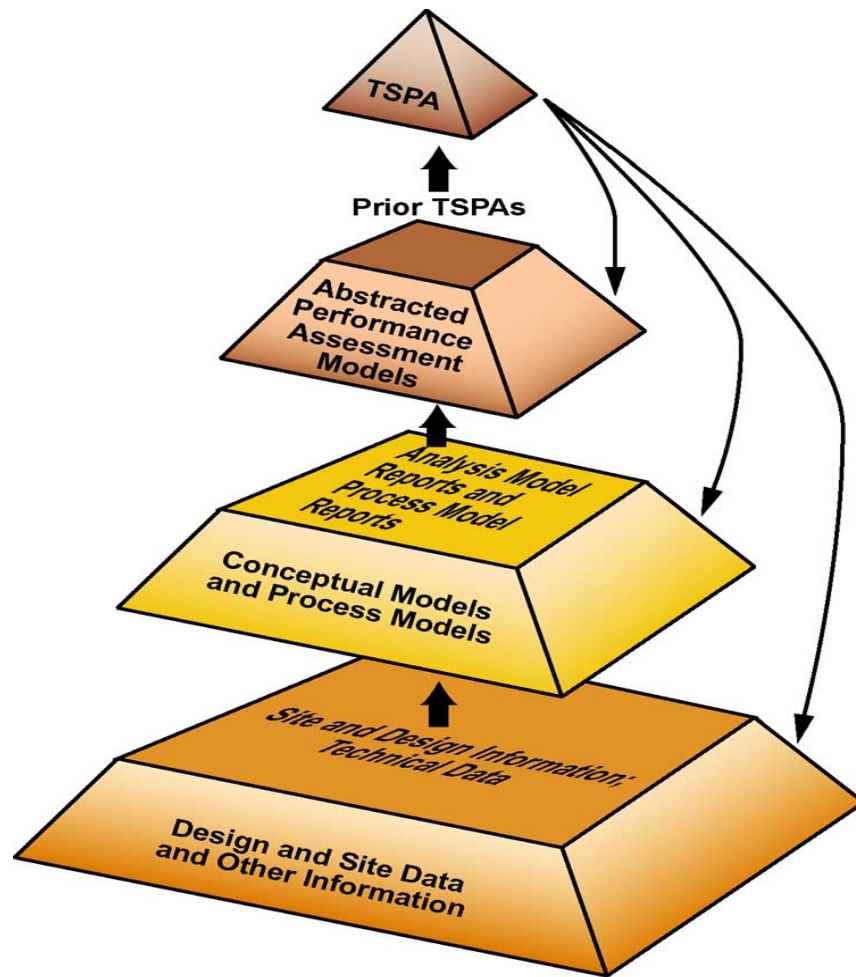
INVESTIGATION OF TSPA SUB-MODEL INTERACTIONS/REPRESENTATIONS

The YMP developed the Total System Performance Assessment (TSPA) Model to analyze the ability of natural barriers and engineered barrier systems (EBS) of the repository to isolate nuclear waste for the regulatory period, currently 10,000-years following repository closure. Performance assessments and related supplemental analyses of the Yucca Mountain repository have been iteratively conducted following the publication of the Nuclear Waste Policy Amendments Act of 1987, Public Law No. 100-203. The iterative assessments incorporate improved understanding of the processes affecting repository performance and, through additional field observations and laboratory analyses, better identification and quantification of the values of the parameters used in the TSPA

The general TSPA approach is illustrated by the pyramid in Fig. 1. Starting with a basic design concept, information is collected about the features, events and processes which characterize the site and associated engineered barriers. This information is used to develop models of the relevant processes, first at the conceptual level and then at the detailed or process level and finally at a simplified or abstracted level suitable for inclusion in the probabilistic TSPA model. Throughout this process, uncertainty in the understanding is quantified to the extent possible, or appropriately bounded in cases of high complexity. The role of the performance assessment is then to integrate these processes and to evaluate the significance of the uncertainty with respect to the ability of the system to meet regulatory objectives.

Release of material from a repository waste package is a complex process, dependent on many features of the repository design. These features are modeled in the TSPA[10]. Among these are the waste package materials, the presence of drip shields in the repository, and the temperature of the repository (which is a function of radionuclide decay) that control the rate of water access to the disposed waste. Fig. 2 illustrates the interconnection and flow of information between the process component models of the TSPA that embody these features.

From an alternative waste form evaluation perspective, the focus of all the models depicted in Fig. 2 is the Waste Form Degradation Model (WFDM) whose components and how they interact with repository features, events, and processes (FEPs) are shown in greater detail in Fig. 3. The WFDM includes sub-models for radionuclide inventory, in-package chemistry, cladding failure, waste form degradation, dissolved concentration limits of radioactive elements, and colloid formation and stability. From the standpoint of alternative waste form considerations, the waste glass degradation, in-package chemistry and dissolved concentration limits of radioactive elements sub-models are of particular interest as described in the following sections.



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Fig. 1. Total System Performance Assessment pyramid

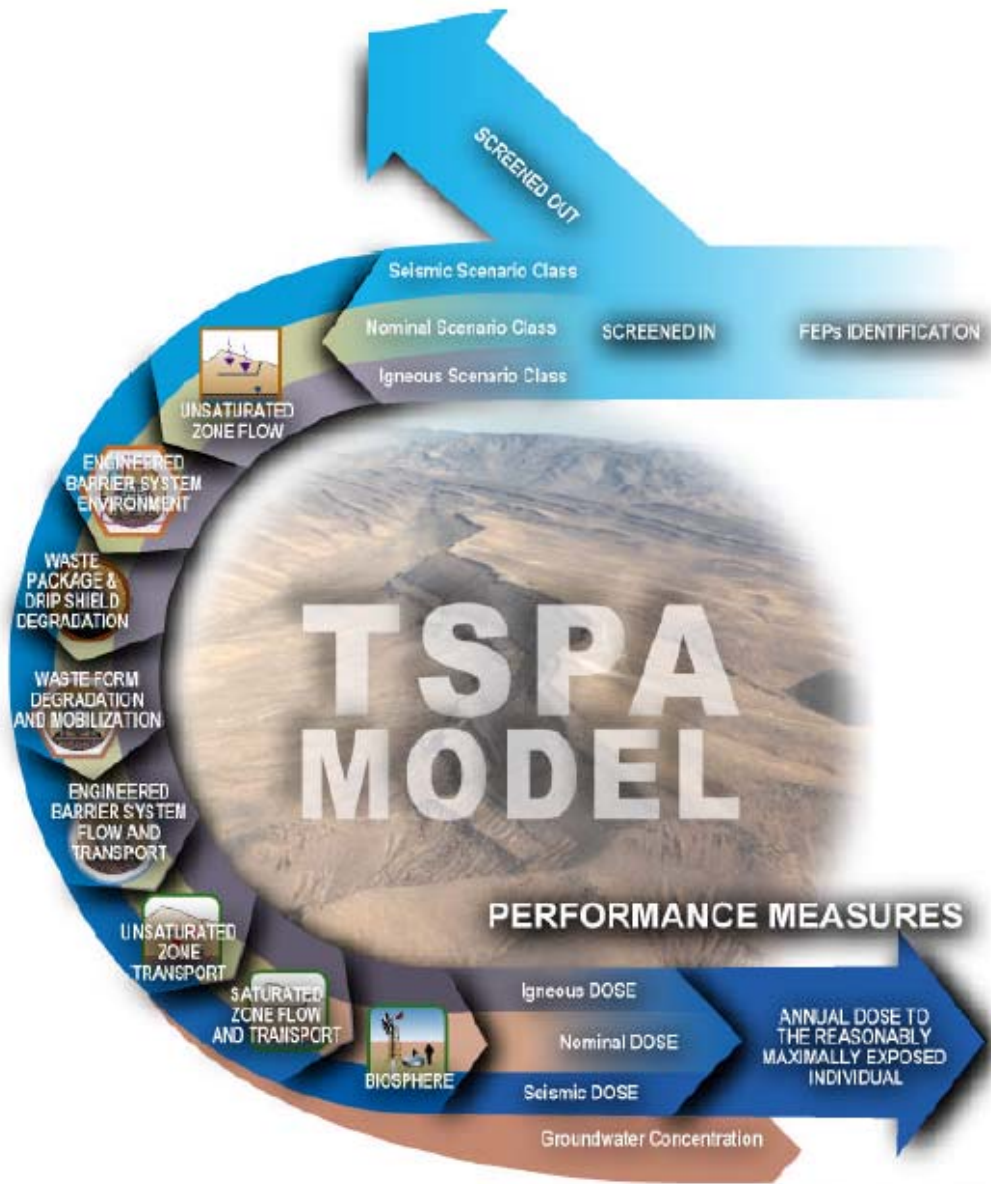


Fig. 2. Process models incorporated in the TSPA Model

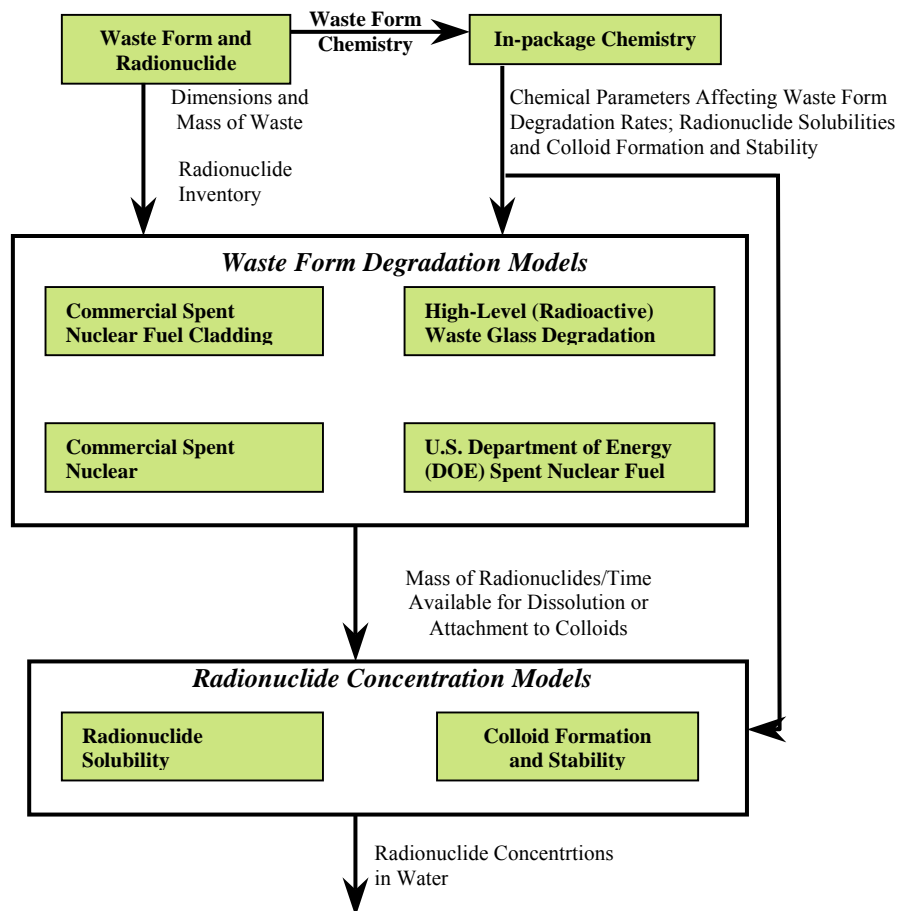


Fig. 3. Components of the Waste Form Degradation Model

Waste Glass Degradation

The Waste form degradation is modeled within the TSPA using empirical degradation rate formulas developed for the three different waste form types; Commercial SNF, Defense SNF and HLW as BSG. Degradation includes transformation of the solid glass to another solid phase as well as the chemical dissolution of the glass. Defense SNF and HLW glass are combined and disposed of in Co-Disposal Waste Packages. The HLW degradation model derived by YMP based on years of study of glass degradation data[11], is formed around the use of BSG as the HLW form, and is a phenomenological Arrhenius-type rate equation model that is only a function of the key repository variables pH and temperature.

The release rate of radionuclides (into the waste package) resulting from the degradation of HLW glass is calculated as the product of three terms. These are:

$$Ri = \text{rate}_G \times S \times Ii \quad (\text{Eq. 1})$$

Where

- Ri = release rate of radionuclide i from HLW glass (g i /day)
- rate_G = specific degradation rate of the glass (g glass/(m² day))
- S = surface area of glass contacted by water (m²)
- Ii = inventory of radionuclide i in the glass (g i /g glass).

The radionuclide release rate given in Equation 1 is used in the TSPA model to estimate the mass of a particular radionuclide i available for dissolution into water in a waste package during a particular period of time. The concentration of a given radionuclide i in water exiting the waste form is equal to the dissolved concentration of that radionuclide in solution plus the concentration of that radionuclide attached to or embedded in colloids.

The specific degradation rate of the glass is given as:

$$\text{rate}_G = k_E \times 10^{\eta \text{pH}} \times \exp^{(-E_a/RT)} \quad (\text{Eq. 2})$$

Where

k_E = glass degradation rate coefficient (g glass/(m²•day))

η = the pH dependence coefficient

pH = $-\log[\text{H}^+]$

E_a = the effective activation energy (kJ/mol)

R = the gas constant (8.314 J/mol•K)

T = the temperature (K)

The value of k_E accounts for the effects of glass composition, including heterogeneity of the waste inventory, as well as the effects of solution composition (except pH).

In-Package Chemistry

The in-package chemistry model simulates chemical interactions of influx water with the dissolved waste package materials and waste forms for waste packages under different physical, hydrologic, and chemical conditions. When run in the process model mode, this model utilizes the geochemical reaction-path code EQ3/6[12,13] which calculates thermodynamic solution equilibria within a waste package at a given time with consideration for precipitated secondary mineral phases as waste package components slowly dissolve.

The time-dependency of the in-package chemistry is simulated taking into account the following information at each time step assuming a fixed seepage dripping water rate in a given run:

- inflow of water seepage into the package
- inventory of dissolved waste components remaining in the waste package from the prior time step
- additional dissolved waste components from the current time step based on the kinetics of waste form degradation
- thermodynamic solution equilibria as calculated by EQ3/6
- outflow of equilibrated solution from the waste package with volume equivalent to water inflow

Fluid composition predictions from the in-package chemistry model for a single co-disposal waste package example is shown in Fig. 4. Note that over the 100,000 year time period simulated, the concentration of any one specie can vary by 7 or 8 orders of magnitude. This example is just one case of many that has been examined with different assumptions regarding high/low glass dissolution rate, high/low canister steel degradation rate, or fluid flux rate.[14]

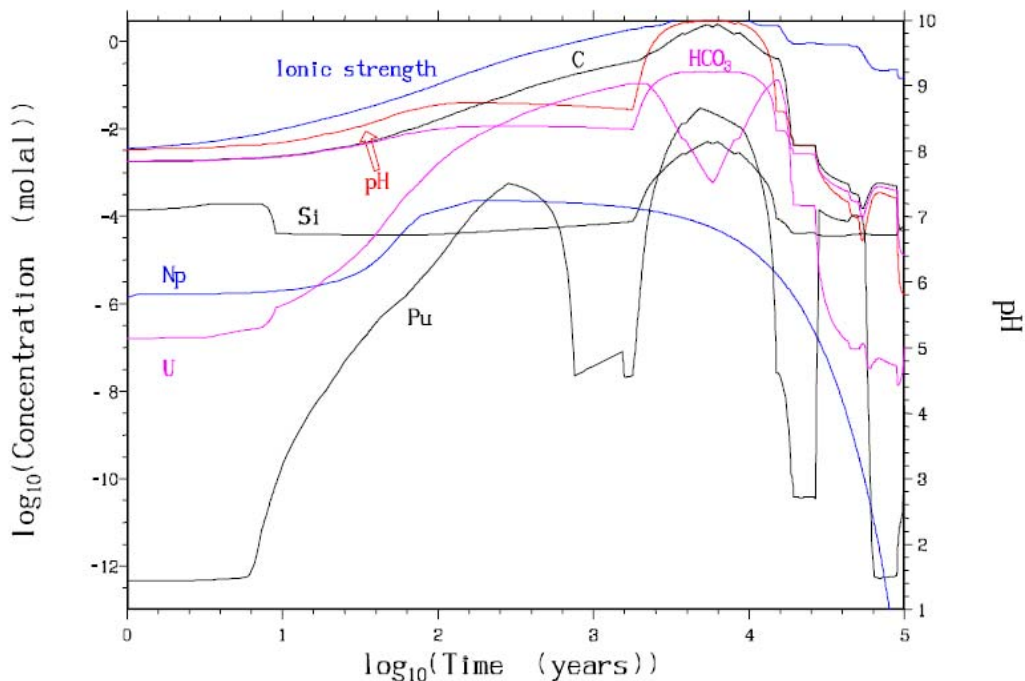


Fig. 4. Predicted changes in fluid composition for case with high glass dissolution; low steel degradation; fluid flux – $0.0015\text{m}^3/\text{yr}$

The outputs of the in-package chemistry process model (pH, ionic strength, redox potential, total carbonate, chloride and fluoride concentrations) are used, either directly or indirectly, as inputs to the models that evaluate dissolved concentrations of radionuclides, commercial SNF matrix degradation, HLW glass degradation, and colloid stability in the TSPA calculations.

While the in-package chemistry process model simulates in-package chemistry in detail, the in-package chemistry model abstraction utilizes simplified expressions, either parameter distributions or regression equations, of the process model results for use in TSPA calculations. An example utilizing such an abstraction is presented in the next section discussing dissolved radioactive element concentration limits. The probabilistic mode employed by the TSPA allows for a sampling of these abstracted parameters distributions, with numerous TSPA runs defining a range of potential repository responses.

Dissolved Concentration Limits for Radionuclides

For the TSPA, fourteen elements have been identified as important to total dose[15]: actinium, americium, carbon, cesium, curium, iodine, neptunium, plutonium, protactinium, radium, strontium, technetium, thorium and uranium. When water comes into contact with spent fuel and waste glass, these radionuclides will dissolve into solution up to a solubility limit that will depend on the particular radionuclide, solution chemistry, and the secondary phases likely to form in equilibrium with the fluid. Through a series of calculations using the speciation-solubility code associated with EQ3/6, it was established that the solubility for these radionuclides were primarily a function of pH, CO_2 fugacity and fluoride concentration.

The effects of varying pH and CO_2 fugacity are directly considered and are provided to the TSPA as lookup tables for a range of pH and CO_2 fugacity values. The effects of different fluoride concentrations

are secondary in nature and are addressed through the addition of a probabilistic uncertainty to the table lookup values.

As an example of radionuclide solubility abstraction, consider implementation of radium solubility in the TSPA. Radium is one element whose solubility is a function of pH and no other variables. Radium solubility is represented as a piece-wise linear function of pH as described in Table I.

Table I. Radium Solubility Values[15]

pH Range	Radium Solubility (mg/L)
3.0 to 7.75	0.02
7.75 to 9.75	1.2
> 9.75	No Limit

Previously Completed Studies for Calcine Disposition

A previous repository analysis for the Idaho Cleanup Project (ICP) Calcine Disposition Program involved calculating geochemical effects on the waste package[16] and radionuclide release from the placement of untreated calcine in the repository[17,18]. In this prior study, the degradation model was set, through a software switch in the geochemistry analysis and the TSPA, to make all material from the calcine matrix available for transport through the repository immediately upon breach of the waste package [19,20]. For the studies conducted in this analysis, only the release of material from the calcine matrix was assumed to be instantaneous. Transport and other chemical reaction of the regulated materials released from the calcine matrix through the repository were not modified. This investigation, supported by laboratory dissolution data that showed very rapid calcine dissolution rates (instantaneous on the geologic time scale of the analysis), indicated no release of materials from the repository in excess of defined regulatory limits.

It has previously been tacitly assumed that implementation of a different repository waste form in the repository analysis would require years of effort to establish a technically defensible basis for new degradation models or parameters for that waste form. The calcine results indicate that extensive data defining waste form degradation may not be required to demonstrate preliminary regulatory compliance for disposal. This is because, as the calcine analysis showed, the waste form degradation may not be controlling for material release of a specific waste form from the repository. Key to the proposed approach is that HLW-form-specific degradation mechanisms and empirical models derived from extensive material testing may not be necessary to model repository performance.

There are many other factors involved, such as the presence of the engineered barrier systems EBS in the repository, the material source term available for release from the waste form, and the transport of the materials of interest once they are released from the waste form (which is influenced by, among others, the geochemistry of the disposal system, itself partially a function of the chemical constituents of the materials in the waste form). If one assumes that waste form degradation is not a controlling mechanism, chemical interaction between the waste form and the repository becomes a major influence. Evaluation of the changes in repository analysis parameters due to changes in the chemical makeup of the waste form may become controlling as was partially evaluated for calcine. This assumption, however, is extremely dependent on the specific chemistry of the waste form, including the projected source term of regulated materials.

As other waste form combinations are examined, the following situations and motivations might lead to relaxation of the instantaneous dissolution assumption:

- Instability in the EQ3/6 calculation due to the rapid release of chemical inventory
- Non-acceptable releases of radionuclides that significantly compromise repository performance
- Verification of the “conservative” nature of the rapid dissolution assumption

Analysis of the system chemistry may indicate that major changes to the structure of the TSPA or sub-programs are required to evaluate the new waste form in the repository system. Potential developments which may require model modifications include:

- The predominant specie in an alternative waste form (e.g. phosphate in iron phosphate glasses) could form compounds that are absent from or require modification to the EQ3/6 database in order to be properly represented
- The predominant specie in an alternative waste form may have a dramatic effect on key radionuclide solubilities that need to be included in model abstractions in order to properly simulate repository performance. This would require enhancement of radionuclide solubility models and expansion of the lookup tables supporting the radionuclide solubility abstraction.

In these cases, a technically defensible mechanism to modify the models would be needed, and investigated under future work. However, in the absence of any modification requirements, the use of the instantaneous release assumption will be used as the starting point of the proposed investigation.

RECOMMENDED RESEARCH DESIGN AND METHODS FOR FUTURE ACTIVITIES

The research methodology proposed for assessing alternative waste forms relative to BSG is based on the strategy outlined above and is depicted in Fig. 5. To demonstrate the methodology it is recommended that initial research focuses on a single waste stream and a potential waste form matrix such as iron-phosphate glass. The following key tasks are envisioned based on the activities shown in Fig. 5:

1. Identify a waste stream and potential waste form for consideration based on potential benefit to site or repository.
2. Derive required information for the waste form. This includes items such as the radionuclide inventory, quantities of materials, and matrix chemistry).
3. Define waste form proposed packaging configuration. This information is required since all modeling activities are based on the waste package configuration.
4. Perform the in-package chemistry analysis using the appropriate computer tools for the analysis, notably the EQ3/6 package used by YMP. Compute relevant chemical parameters, such as waste package pH, radionuclide solubility, colloid stability within the waste package, etc based on instantaneous material release (modifying the generic release rate equation shown in the flowchart). Compare this information against existing model bases to determine compatibility for TSPA analysis.
5. Modify parameters within the TSPA model to incorporate the physical and chemical information defined in steps 1-4 and perform TSPA analyses. Perform parametric geochemical and TSPA analyses to examine the impact of different waste package failure rates, changes in radionuclide inventory, number of waste packages, and other parameters. The merits of the alternative waste form can be assessed by comparing these analysis results with those of a base case utilizing BSG rather than the alternative waste form for immobilization of the waste stream of interest.

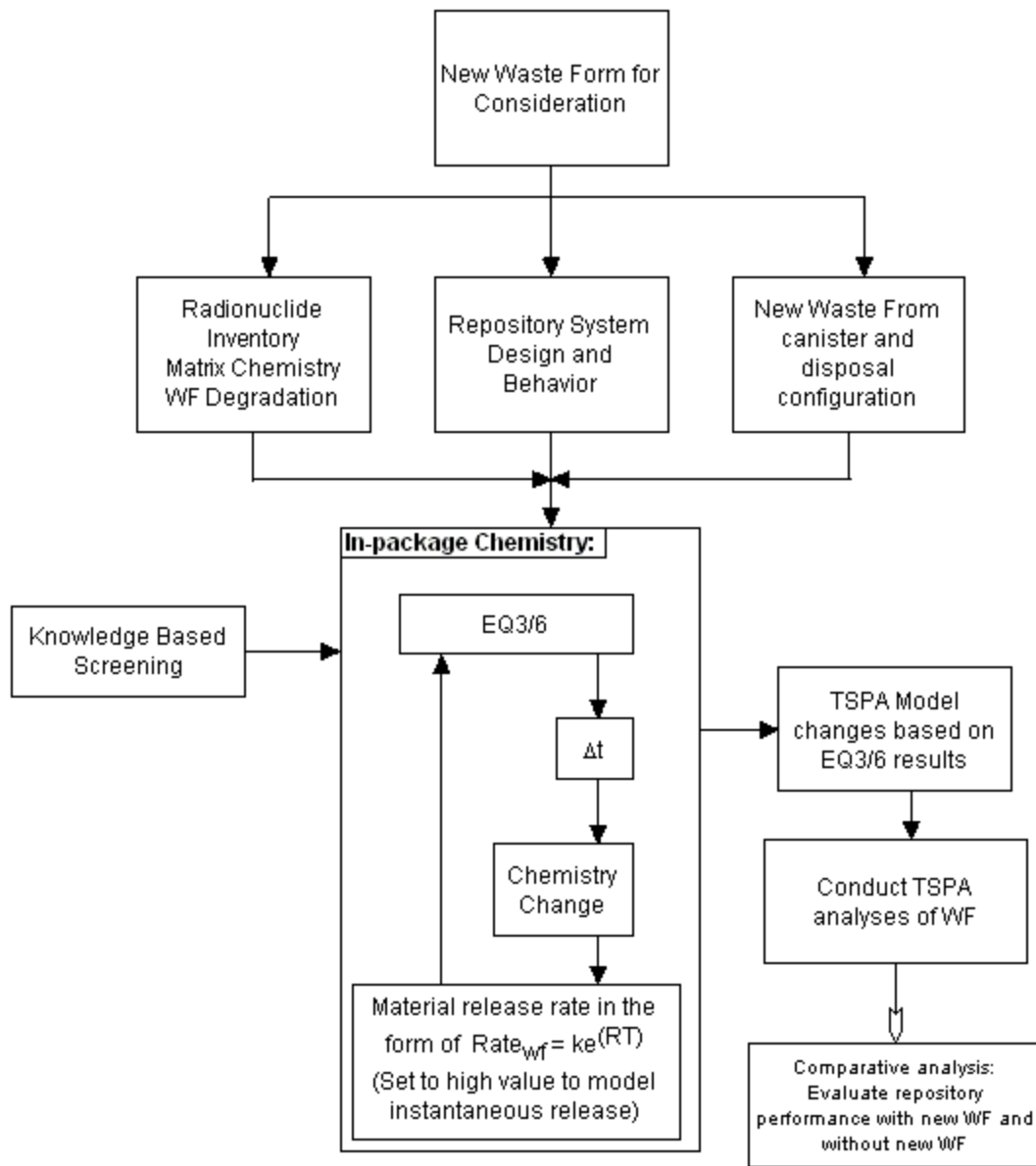


Fig. 5. Proposed research methodology

CONCLUSIONS

The calcine disposition study showed that at least for this waste stream, waste form durability has no significant impact on repository performance. The preliminary sensitivity study using the results of the calcine study indicates that the proposed methodology assuming zero waste form performance could shorten the time required to establish the technical bases for accepting additional waste forms for disposal in a geologic repository. The proposed research will not lead to a complete answer, but will provide additional basis for the methodology and will point toward needed data to establish complete technical bases without the need to start the process used to characterize BSG again. This approach should be a cost-effective strategy to match the waste form to the waste chemistry making the HLW disposal system more efficient.

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