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DEVELOPMENT OF ACCEPTANCE SPECIFICATIONS FOR LOW-ACTIVITY WASTE FROM THE HANFORD TANKS

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ABSTRACT

Low-activity products will be in the form of solidified waste and optional matrix and filler materials enclosed in sealed metal boxes. Acceptance specifications limit the physical characteristics of the containers, the chemical and physical characteristics of the waste form and other materials that may be in the container, the waste loading, and the radionuclide leaching characteristics of the waste form. The specifications are designed to ensure that low-activity waste products will be compatible with the driving regulatory and operational requirements and with existing production technologies.

INTRODUCTION

The waste materials legacy from past defense reprocessing operations at Hanford includes about 250 MCi of radioactivity dispersed in more than 240,000 MT of process chemicals. These waste materials have been stored underground as slurries, sludge, saltcake, and highly alkaline solutions in 177 tanks, some dating back to 1944. Many of the tanks are nearing the end of their design life, and some are leaking. In 1992, the Tank Waste Remediation System (TWRS) program was established to manage these wastes in a safe, environmentally sound, and cost-effective manner. The management strategy involves removing the waste from the tanks and separating it into a high-activity fraction, which contains most of the radionuclide inventory, and a low-activity fraction, which contains most of the nonradioactive solids. The separated fractions are then converted into forms suitable for subsequent transport, storage, and disposal (TSD). The high-activity fraction is suitable for deep geological disposal, whereas the low-activity fraction is suitable for near-surface land disposal at Hanford.

To implement the TWRS strategy, several waste management functions must be accomplished, including retrieval of the waste from the tanks; pretreatment to separate the

retrieved waste into high- and low-activity fractions; and transport, immobilization, storage, and disposal of the high- and low-activity waste forms. Under the privatization initiative at Hanford, the U.S. Department of Energy (DOE) will purchase waste treatment and immobilization services from private-sector, contractor-owned and -operated facilities under a fixed-price contract. The privatized work scope will be divided into two phases. Phase 1 will process 6-13% of the tank waste and show the technical and economic viability of procuring fixed-price waste treatment and immobilization services for the range of waste compositions in the tanks. Phase 2, the full-scale production phase, will accommodate removal and processing of the remaining waste. DOE will retain responsibility for TSD of immobilized waste products once they have been accepted from the private contractors. To be acceptable, the products must be compatible with the regulatory and operational requirements governing the TSD functions.

This paper addresses the basis for the specifications developed for the immobilized low-activity waste (ILAW) products. It describes and illustrates, with selected examples, the basis used to identify (1) the specifications needed to control ILAW characteristics and (2) the acceptance criteria and testing methods to be used for assessing compliance.

DEVELOPMENT OBJECTIVES

The general objective of this study is to develop specifications that ensure that ILAW products accepted from private-sector contractors are compatible with DOE's responsibilities for safe, efficient, and environmentally benign TSD of these products. This objective requires the specifications to ensure that, as a minimum, the products be compatible with regulatory and operational requirements for the TSD functions. An additional objective is that the acceptance criteria in each of the specifications should be reasonably achievable by using available ILAW production technologies and be quantitative and amenable to direct compliance determination.

APPROACH

The general approach for specifications is based on regulatory and operational requirements that would govern the TSD of the accepted ILAW products. The basis for the ILAW specifications (i.e., the linkage between the driving requirements and the content of the specifications) was developed in two steps. First, the applicable regulatory and operational requirements were identified and linked to the characteristics of the ILAW products that need to be controlled. This step provided the basis for identifying the needed specifications. Second, the extent to which the ILAW characteristics need to be controlled was addressed by developing the acceptance criteria and associated testing methods included in the specifications. The criteria and testing methods were identified by quantitatively analyzing the linkage between the driving requirements and the ILAW characteristics. In addition, practical production constraints were considered, as appropriate, in establishing the acceptance criteria and the testing methods.

Some of the governing requirements apply directly to ILAW products. Others apply to the systems used for the TSD functions. In some cases, the driving regulatory requirements explicitly define characteristics of the ILAW products that must be controlled, the criteria that identify acceptable ranges for these characteristics, and the testing methods and information that are acceptable for establishing compliance. For these requirements, the basis for the content of the specifications is transparent; the specifications simply implement the driving requirements. For the system requirements, the linkage between the driving requirements and the content of the specifications is often more complex. In this case, the linkage may involve allocating performance to the ILAW products, identifying ILAW characteristics that are important for performing the allocated functions, quantifying the linkage to identify acceptance criteria and testing methods, and identifying data sufficient to ensure that the ILAW products can perform the allocated functions.

IDENTIFICATION OF ILAW CHARACTERISTICS THAT NEED TO BE CONTROLLED

The requirements that apply to the TSD of ILAW are too numerous to list here. However, the requirements include those that apply directly to the ILAW products and those that apply to the TSD systems in which the ILAW products are a component part. The requirements can be categorized as follows:

1. Classification of the ILAW as "incidental waste."

2. Limitations on the occupational doses, population doses, risks, and releases from operating storage and disposal facilities.
3. Exclusion of specific types of radioactive waste materials from land disposal.
4. Requirements on the physical and chemical stability of the waste form.
5. Limitations on doses to the public from a disposal facility after the operational phase.
6. Limitations on the contamination of aquifers that are potential sources of drinking water.
7. Limitations on doses that intruders who inadvertently enter a disposal facility may receive after the operational phase.
8. Operational requirements for the handling, lifting, and moving operations associated with the execution of the TSD functions.

These categories of requirements provide a framework for discussing the characteristics of the ILAW that need to be controlled. The ILAW specifications that implement the driving requirements in each of the above categories are identified in Table 1. To illustrate how the implementing specifications were identified, the following paragraphs summarize the requirements for classifying the ILAW as incidental waste and how these requirements are linked to implementation of controls on radionuclide concentrations in the ILAW products.

The waste currently stored in the Hanford tanks is managed as high-level waste. It is important that the ILAW products satisfy requirements for classification as incidental waste to allow it to be managed under low-level waste (LLW) regulations. The U.S. Nuclear Regulatory Commission (NRC) has identified three conditions that must be satisfied to classify the separated low-activity waste as incidental waste[1]:

1. The wastes will be processed to remove *key* radionuclides to the maximum extent that is technically and economically practical.
2. The wastes will be incorporated in a solid physical form at a concentration not exceeding the applicable concentration limits for Class C LLW, as defined in 10 CFR Part 61.

3. The wastes will be managed pursuant to the Atomic Energy Act, so that safety requirements comparable to the performance objectives defined in 10 CFR Part 61 are satisfied; i.e., incidental waste shall meet the NRC low-level waste disposal requirements.

Table 1: Summary of the Driving Requirements and the Specifications that Implement these Requirements

Driving Requirements	Implementing Specifications
- Requirements to classify ILAW as incidental waste - Requirements to limit dose to inadvertent intruders	- Radionuclide concentration limitations
- Control of occupational and population doses from operating facility	- Surface dose rate limitations - Surface contamination limitations - Radionuclide release rate airborne - Closure and sealing
- Exclusion of certain materials from disposal	- Free liquids - Pyrophoricity or explosivity - Explosive or toxic gases
- Requirements on the waste form and the optional matrix and filler materials	- Compressive strength - Thermal degradation - Stability in radiation field - Biodegradation - Immersion stability - Leach testing - Dangerous waste limitations
- Requirements for public and groundwater protection after disposal	- Radionuclide release rate - Void space
- Operational requirements	- Package handling - External temperature - Mass - Compression testing - Waste loading - Size and configuration - Container material degradation - Labeling and manifesting

Condition 1 is somewhat ill defined because the NRC did not define the phrase "to the maximum extent that is technically and economically practical" and is, therefore, open to different interpretations. It suggests that only a "small fraction" of the radionuclide inventory can be processed into the ILAW if it is to be classified as incidental waste. In the ILAW specifications, concentration constraints, together with waste loading limitations, are designed to meet the intent of the NRC requirements. Specifically, the concentrations of ^{137}Cs and ^{90}Sr are limited because these radionuclides dominate the radioactivity concentrations of the waste and are, therefore, considered to be the *key* radionuclides for compliance. In addition, ^{99}Tc is considered to be a *key* radionuclide because of its importance for compliance with the disposal system requirements. Condition 2 is implemented through the requirements that the waste form be a durable solid and that the concentrations of all radionuclides in the waste form not exceed Class C limits. Condition 3 is implemented by including the NRC 10 CFR Part 61 requirements and the associated NRC guidance (e.g., *Technical Position in Waste Form*[2]) in the set of driving requirements for the ILAW specifications.

RATIONALE FOR THE ACCEPTANCE CRITERIA AND TESTING METHODS

The content of this section illustrates the development of the acceptance criteria and the corresponding testing methods. It focuses on the basis for the acceptance criteria and testing method included in the radionuclide release rate specification (i.e., the specification that implements the requirements in Row 5, Table 1). The approach is organized into three general steps (Table 2). Each of these steps is described briefly in the following paragraphs.

Analyses have been conducted to examine the performance of engineered and natural systems for near-surface disposal of glass waste forms at Hanford.[3,4] These analyses show that the groundwater protection requirements impose the most restrictive limitations on the radionuclide release rate. The key radionuclide that may contribute to the calculated drinking water dose for the first 10,000 years is ^{99}Tc; therefore, it is the focus of the following discussion. The available analysis results indicate that a fractional release rate from the engineered disposal system of about one part per million per year of the estimated inventory (2.4 x 10^{-4} Ci for ^{99}Tc) was needed to meet the maximum concentration limit beta/gamma standard. Based on these results, the allowed release rate of ^{99}Tc from the ILAW disposal system was estimated to be 2.6×10^{-2} Ci/yr (31 Bq/s).

Table 2: General Steps Involved in Identifying the Radionuclide Release Rate Acceptance Criteria in the ILAW Acceptance Specifications	
Step	Description
1	Determine the constraint on the release rate from the disposal system for radionuclides (e.g., ^{99}Tc) that may contribute to groundwater contamination.
2	Determine how the release rate constraint should be apportioned to the ILAW products that result from processing a portion of the total tank inventory.
3	Establish a linkage between the radionuclide release rate requirements and characteristics of the waste form that can be measured by using a practical test method that can be implemented in a production environment.

Because the waste in the tanks will not be processed in one batch or blended to form a uniform feed material, the second step addresses the allowed radionuclide release rate from the ILAW products that result from processing a portion of the waste. The maximum release rate from the ILAW generated from processing a portion of the inventory is obtained by conservatively allocating the release rate performance of the ILAW and apportioning the "allowed" release rate from the disposal system in direct proportion to the fraction of the radionuclide inventory processed; i.e.,

$$R_x < A_x I_x^p / I_x^{tot},$$

where:

R_x is the allowable release rate of component x from the ILAW produced when a portion of the total waste inventory in the tanks is processed (Bq/s);

A_x is the release rate constraint for component x from the disposal facility;

I_x^p is the inventory of component x processed; and

I_x^{tot} is the total inventory for component x estimated to be in the liquid fraction of the tank waste following solid/liquid separation.

Based on the A_x value of 31 Bq/s and an estimated I_x^{tot} value of 1.1×10^{15} Bq, it follows that A_x/I_x^{tot} (s^{-1}) has the value 2.8×10^{-14} (s^{-1}). Substituting these parameter values and rearranging terms makes it possible to express the constraint for technetium release in the following form:

$$R/I^p < 2.8 \times 10^{-14}\ (s^{-1}).$$

The long-term radionuclide release rate from the ILAW packages in their disposal environment is not measurable. To assess compliance of the ILAW products with the above constraints, it is necessary to link them to corresponding criteria that apply to measurable characteristics of the waste form. To examine the constraints on measurable ILAW characteristics that correspond to the radionuclide release rate constraints outlined above, it is instructive to use silicate glass waste forms as a reference waste form.

Assuming, conservatively, that the radionuclide release rate from a silicate glass waste form is constrained only by the rate of corrosion of the glass matrix, the radionuclide release rate constraints can be expressed as follows:

$$R_x < C_g\, S_g\, I_g / \rho\, V_g\ ,$$

where:

R_x is the rate of release of component x (Bq/s);

C_g is the corrosion rate of the glass [kg/(m^{2}s)];

S_g is the glass surface area available for corrosion per package (m^2);

I_g is the radioactive inventory of radionuclide x (Bq) equal to I_x^P, where F is the fraction of the processed inventory solidified in the glass;

ρ is the glass density (kg/m^3); and

V_g is the glass volume (m^3).

Hence, it follows that the constraint on the technetium release rate discussed earlier can be expressed in the following form:

$$\left\{\frac{C_g}{\rho}\right\} \left\{\frac{S_g}{V_g}\right\} F < 2.8x10^{-14}(s^{-1}).$$

(The actual release rate of individual radionuclides may be lower due to other considerations such as solubility constraints and incorporation/sorption onto secondary alteration phases formed in the corrosion process.[5])

To assess compliance with this constraint, each of the parameters on the left side of the above expression must be determined. Testing techniques appropriate for measuring these characteristics for a silicate glass waste form are illustrated by the following discussion of the testing method identified, in the radionuclide release specification, for measuring the glass corrosion rate (i.e., C_g).

Glass corrosion rate is a complex function of many glass composition and environmental factors. Since the long-term rate of corrosion cannot be measured, the general approach adopted was to identify a short-term test method likely to bound the long-term rate. At the outset of the development of the ILAW specifications, a workshop was held to identify suitable testing methods. Static and flow-through testing options were identified. Because data were not available for the testing conditions and the range of waste glass compositions that might be considered, experimental testing programs were conducted to measure the corrosion rates of a range of glass compositions under a variety of testing conditions using flow-through and static testing methods designed to measure the initial corrosion rates (when the feedback effects due to the buildup of silicic acid and other constituents in solution are minimized). The results indicated that the testing methods examined could discriminate between the corrosion behavior of the various glass compositions examined. However, they also showed that for the flow-through test, the results were very sensitive to the testing procedures used. As a result, the decision was made to specify use of the product consistency test (PCT) in the ILAW specifications. The PCT has been developed for high-level borosilicate glass waste form testing and has been standardized.[6] In the ILAW specifications, the value of C_g to be used in evaluating compliance is specified as the value based on the normalized silicon release in a seven-day PCT at 25°C the surface area of the tested glass to be determined as specified in the ASTM Method C1285-94.[6]

CONCLUDING REMARKS

The ILAW specifications were developed by using a systematic approach based on linking the driving requirements to characteristics of the ILAW that need to be controlled. Also, the acceptance criteria and testing methods included in the specifications were based on practical production constraints and comments received from contractors and other interested parties on an early draft of the specifications and on the ILAW specifications included in the draft request for proposals. The completed specifications include the requirements normally applied to the TSD of LLW and, as illustrated in this paper, also include some additional requirements that derive from implementation of the driving requirements for the Hanford ILAW products.

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