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Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

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Characterization of Hanford Tank Waste and the Role of Historic Modeling

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1.0 ABSTRACT

The tank waste characterization process is an integral part of the overall effort to identify, quantify and control the hazards associated with radioactive wastes stored in underground tanks at the Hanford Reservation. Characterization of the current waste tank contents through the use of waste sampling is only partly effective. The historic records must be exploited as much as possible. A model generates an estimate of the current contents of each tank, built up from the estimated volumes of each of the defined waste components. The model combines the best estimate of the waste stream composition for each of the major waste generating processes. All available waste transfer records were compiled and integrated to track waste tank fill history. The behavior of the waste materials in the tanks was modeled, based on general scientific principles augmented with specific measurement data. Sample analysis results were not used directly to generate any of the tank contents estimates, but were used to determine the values of variable parameters such as the solubility. By considering all available information first (including historical model estimates, surveillance data, and past sample analysis results), future sampling resources and other characterization efforts can best be spent on tanks that will provide the largest returns of information.

2.0 INTRODUCTION

The Hanford Reservation system of underground storage tanks contains approximately 230 million liters of waste material distributed among 177 tanks. The waste in these tanks was produced as a part of the nuclear weapons materials processing mission that occupied the Hanford Site for the first 40 years of its existence. Waste tanks (149 single-shell and 28 double-shell tanks) contain a wide variety of waste compositions generated by three distinct chemical separations processes, several waste management/waste volume reduction operations and two tank waste reprocessing flowsheets. Characterization of the tank wastes is required to maintain the safe storage of the wastes, and to guide retrieval, processing, and disposal technology development.

3.0 MOTIVATION FOR CHARACTERIZATION

The tank waste characterization process is an integral part of the overall effort to identify, quantify and control the hazards associated with radioactive wastes stored in underground tanks at the Hanford Reservation. Knowledge of the physical, chemical and radiological properties of the wastes is prerequisite to operations to store, retrieve, process and dispose of the wastes safely.

The Tank Waste Characterization Project currently addresses the information needs identified for the Tank Waste Remediation System (TWRS) through the Data Quality Objectives (DQO) process (EPA 1994).

3.1 Safe Storage of the Waste

Characterization for safe storage of the waste includes the screening of all tanks for key parameters that may indicate potential safety issues. The areas of concern are:

- The presence of energetic compounds in the condensed phase of the waste in a configuration that could support a propagating exothermic reaction (i.e. dry, in the presence of an oxidizer, located where an initiating factor is feasible).

- The presence of flammable gases in the dome space above the waste surface at sufficient concentrations that combustion is feasible.

- The presence of fissile materials concentrated in the absence of significant neutron absorbers so that a criticality event is feasible.

In all areas of concern, several factors must interact to give rise to an actual safety issue. The presence of specific waste components, addressable by characterization, is only one of the contributing factors. Characterization can be used to identify and quantify hazards and develop the most appropriate responses for dealing with them. A combination of active and passive controls can provide multiple safeguards that prevent an event from occuring.

3.2 Safe Operation of the Waste Tank Facilities

Although many of the tanks are currently stabilized and undergo no operations, several operations are ongoing in a subset of tanks. A waste evaporator unit concentrates liquid waste to provide additional storage space in the existing tanks. Removal of pumpable liquid from single shell tanks is performed to prevent leakage into the soil. Transfers of waste between operating double shell tanks is necessary to support these operations. Other specific operations may occur, such as the addition of caustic to a tank to maintain operating specifications.

All operations must be performed in accordance with all applicable regulations. Intrusive operations must be carried out in a manner that ensures that none of the safety issues identified above (see 3.1) are generated during operations. This requires the review of existing characterization information prior to initiation of the operation, and may require acquisition of new information.

In addition, other information on waste characteristics may be required to support physical operations. For example, prior to transfer of waste material, the physical parameters of the waste must be well enough understood to ensure that the material will not solidify during transfer, clogging the transfer lines.

3.3 Resolution of Safety Issues

The above areas address characterization needed to ensure that safety issues are recognized so that they may be corrected, are not created during waste management and disposal operations. Separately, a thorough understanding of the mechanism behind the safety issues is needed in order to select the appropriate response. For example, one needs to understand how flammable gas is generated in the waste material, and what causes it to be retained or released at a given rate. This understanding ensures that the symptoms of a potential problem are correctly identified, that the correct controls or treatment for the problem are applied, and that sufficient foresight is applied in the future to prevent occurences during waste management and disposal operations. The information to resolve safety issues is obtained from many sources, including theoretical work, laboratory experimentation, and in some cases characterization data.

3.4 Preparation for Waste Disposal

Safe storage and operation of the waste tanks is an interim step until facilities are available for the retrieval, pretreatment, processing and final disposal of the material. The equipment and facilities for these steps are still in the design phase. Characterization of the waste requires that the physical and chemical properties be adequately quantified, both in terms of average values and of bounding values, to support design. In addition, physical samples of waste material are required for small and large scale testing of various treatment processes.

4.0 TANK WASTE CHARACTERIZATION PROCESS

Characterization of the current waste tank contents through the use of waste sampling is only partly effective. The waste tanks contain very few access ports, limiting the number of samples obtainable. The access port locations do not necessarily support obtaining representative samples, particularly in tanks where the waste material is heterogeneous. Sampling is expensive and complex because of the radioactive and chemically hazardous nature of the waste. The historic process records must be exploited as much as possible to improve overall waste characterization. By considering all available information first (including historical model estimates, surveillance data, and past sample analysis results), future sampling resources and other characterization efforts can best be spent on tanks that will provide the largest returns of information (Brown et al., 1995).

4.1 The Overall Process

The approach to characterization of each tank is to compile all available information about each tank to provide an estimate of the contents. Information sources include models of waste contents developed from historic records of processes and waste transfers, surveillance and monitoring data, sampling and analysis results, and models of chemical behavior. The estimates developed from these sources are improved by sampling or other measurement to provide additional data. Grouping of similar tanks is employed in developing the estimates (Hill et al, 1995; Remund et al., 1995a).

Based on the amount and quality of data extant, an estimate of tank contents and the identified needs, a plan is developed to obtain additional data (Dove et al., 1995; Homi and Dodd, 1995). The development of a defensible plan requires an understanding of the quality of existing data and the capabilities and limitations of the tools which may obtain new data.

4.2 Role of Historic Modeling

The waste tanks contain a wide variety of waste compositions, principally generated by three distinct chemical separation processes, several waste volume reduction operations and two tank waste reprocessing flowsheets. All of the processes contributing to the waste generation underwent significant evolution over time. Extensive (albeit incomplete) records were kept describing the initial placement of waste in specific tanks and the later transfers of waste between tanks. The complete reconstruction of current tank contents through use of the records is complicated by several factors:

- Although the flowsheets for the waste-generating processes are well known at the start of a process, the evolution of the process during plant operation is not well documented. In addition, process vessel corrosion and impurities in process chemicals can dramatically affect the nature of the waste stream.

- Active concentration of tank waste supernatants in six different evaporator campaigns spanning the fifty years of processing have not been thoroughly documented. There are many uncertainties in the tank transaction histories for these campaigns. The waste heating during evaporator campaigns may also have accelerated chemical reactions, changing waste properties from those described in the flowsheets or in early sample analysis.

- The waste transactions associated with the removal of tank waste for the two major tank waste reprocessing campaigns, the Uranium Recovery campaign in the 1950's and the Cesium/Strontium removal campaign in the 1960-70's, are incomplete. These processes were based on assumed waste characteristics, not actual waste characteristics. Significant changes were performed as the processes evolved during the course of the reprocessing campaigns. In many cases these changes were not well documented.

The construction of a model of tank contents combines the best estimate of the waste stream composition for each of the major waste generating processes and several minor processes. Altogether, 48 distinct waste types are modeled, refered to as the Hanford Defined Wastes (Agnew, 1994a). All available waste transfer records were compiled and integrated to track waste tank fill history (Agnew, 1994b, 1994c, 1994d, 1994e). The behavior of the waste materials in the tanks is modeled, based on general scientific principles augmented with specific measurement data (Agnew, 1995f, 1994g). Although sample analysis results are not used directly to generate any of the tank contents estimates, sample analysis is used to determine the values of variable parameters such as the solubility and precipitation rates of specific analytes in specific waste materials.

The model generates an estimate of the current contents of each tank, built up from the estimated volumes of each of the defined waste components (Brevick et al., 1995a, 1995b, 1995c, 1995d). The quality of these waste content estimates is now being reviewed and quantified through several parallel activities. A Monte Carlo simulation has been performed to quantify the uncertainty of the model estimates for several tanks, based on the variability of key model parameters such as limiting solubility. This intial variance or uncertainty estimate has been used in the systematic comparison of tank contents estimates with actual sample data (Remund et al., 1995b). Initial results are promising in most cases (i.e., relatively good agreement between model estimates and sampling estimates). However, some results indicate that there are significant differences between the model estimates and sampling results. This observation could indicate:

- incomplete modeling of the major error sources in the Monte Carlo simulation,
- overly optimistic estimates of variance based on sampling results,
- systematic errors in the model that require correction,

or some combination of the above factors.

Additional samples of key waste types are being acquired and analyzed to allow better definition of the composition of the defined wastes types used in the model (Simpson and McCain, 1995). The evaluation and improvement of the historic model will continue until a quantitative variance estimate can be provided for each of the contents estimates. The availability of the resulting tank contents estimates will allow reduced sampling of many tanks, better grouping of similar tanks, and more effective planning of sampling events.

4.3 The Role of Sampling and Analysis

Analysis of sample material alone will not provide adequate characterization information for the Hanford underground storage tanks. It is not possible to design a sampling scheme that provides a statistically significant number of truly random samples.

Most tanks have very few available access ports or risers (many have no more than two accessible risers). The locations of the risers often leads to sampling of non-representative material. The risers have in some cases been used for dumping of additional waste material. Previous samples may have been taken from the risers, disrupting the solid materials. Intruments may have been introduced into the waste through a riser. Removal of instruments may have required decontamination, introducing water or other solutions into the waste near the area where samples are taken.

Given these constraints, the value of sample analysis may be called into question. However, when combined with the historic model and contemporary surveillance data, and allowing for potential biases and sampling error, a well thought out sampling scheme can provide significant information. The first step in the development of the sampling scheme is to consider the historic information, including the waste type predictions, records of any previous sample activity and photographs of the waste tank contents. After this initial assessment, contemporary surveillance data is considered. Review of the available risers is necessary so that any factors that may make a specific sample different from others in the tank are understood. Review of the photographs can reveal surface heterogeneity and give clues to the relationship between a sample at a specific location and the overall tank content. It must be noted that some tanks may be so heterogeneous that very little information can be gained from any small number of samples. In these cases, an alternative approach (such as adding liquid to turn the waste into a homogeneous slurry and retrieval into an interim storage tank) must be considered.

When a sampling approach is selected, it is also necessary to consider how the acts of acquiring, removing, transporting and analyzing the sample may affect the parameters being measured (Winkelman and Eberlein, 1995). Any systematic biases that are introduced during the sampling process need to be considered when using the results to reconstruct the total tank contents.

Given all the constraints, the most realistic approach to tank waste characterization must consider all available data, start with a model of the waste generated from the historic records, and then use sampling to confirm or refine specific aspects of the model.

4.4 Optimization of the Process

Programs requiring data regarding waste composition document their issues and information needs and identify tanks which must be characterized to meet the needs. The programs identify criteria by which all tanks can be prioritized with respect to each issue. The Characterization Project integrates the information needs and tank priorities to define an overall plan for obtaining new information through sampling. The process for developing the priority list and generating a sampling schedule includes the application of technical and operational constraints. The most effective overall prioritization of tank sampling events ensures that early events provide data that supports characterization of multiple tanks. High priority sampling events are those that provide data to:

- Allow resolution of safety issues affecting multiple tanks.

- Define the appropriate approach to characterization of multiple tanks for safety issue identification and resolution.

- Improve the estimates of waste content for multiple tanks, particularly with regard to important safety and disposal parameters.

It is not yet possible to determine the total number of samples needed to characterize the wastes adequately (either for an inidividual tank or for the entire tank farm system). An initial set of 28 tanks has been identified as high priority to address the above issues (Brown et al., 1995). It is anticipated that the information gained from those samples will support resolution of issues (reducing the characterization needs associated with other tanks) and provide quantitative information about the quality of the tank content estimates developed from historic data. Once the uncertainty associated with the historic models is quantified, it may be possible to use the model estimates alone to make future decisions regarding the operation and disposal of specific tanks. This application of the historic model has the potential to greatly reduce future sampling requirements.

5.0 CONCLUSIONS

Continued evaluation of historical information and modeling techniques is needed to enhance the value of that information and potentially reduce the need for future in-tank sampling operations. As additional knowledge is gained, that knowledge is fed back into the process to help prioritize, guide and define future efforts so that the most important information is obtained as soon as practicable. Continued evaluation and improvement of sampling and measurement methods is required to improve the ability to obtain new information and to understand its meaning and limitations.

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