HIGH-LEVEL WASTE TREATMENT SYSTEMS FOR TREATMENT OF ORPHAN WASTES

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INTRODUCTION

The total past, current, and future investment by the United States to manage and solidify high-level waste (HLW) will be many tens of billions of dollars. Large-scale HLW solidification facilities have been built at the Savannah River Site (SRS), which is near Aiken, South Carolina, and large-scale facilities will be built at the Hanford site, which is near Richland, Washington.

There are a large number of other highly radioactive wastes with no good options for treatment and disposal. The materials are very hazardous and will be difficult to dispose of. But the volumes of materials are small. Their disposal is lower in priority than the high volume HLW streams, which are the pressing priority. We define them as orphan wastes because the path forward for treatment and disposal has not yet been defined. There is no existing treatment or disposal facility (home) to go to.

Examples of orphan wastes include some types of excess fissile materials, control rods, sludges, and hot-cell examination wastes. The difficulties in managing these wastes suggest that the United States should consider converting these wastes into forms that allow processing by the HLW treatment systems now being deployed. This option is described herein.

HLW MANAGEMENT SYSTEM AND APPLICABILITY TO OTHER WASTES

During the cold war, the United States generated large quantities of HLW liquids, salts, and sludges from processing spent nuclear fuel (SNF) and targets for the recovery of plutonium and uranium for defense purposes. The wastes consists of alkaline sludges, mostly metal hydroxides, and salt cakes made primarily of sodium nitrate.

Methods to convert these wastes to HLW glass are similar for both SRS and Hanford wastes. The sludges are washed with water or sodium hydroxide solutions to remove soluble sodium salts and excess aluminum, and the remaining sludges are converted to borosilicate HLW glass. The salts are dissolved in water. The aqueous solutions obtained from dissolving salts and washing the sludge are processed to remove the important soluble radionuclides. These radionuclides are also sent to the HLW glass melter. Finally, the remaining liquid is treated as low-level radioactive waste (LLW). Treatment and disposal of LLW are much less expensive than treatment and disposal of HLW. The total inventory[1] of HLW exceeds 300,000 m^3; thus, there are strong economic incentives to partition the HLW into a HLW glass and an LLW.

Three characteristics of the HLW systems make them potentially attractive for treating orphan wastes.

- *Eliminate Deleterious Effects of Certain Chemical Species in the Final Waste Form*. Many orphan wastes have chemical compositions that make it difficult to produce a high-quality waste form. If the small quantities of such wastes are added to the large inventories of HLW sludges, the changes in composition of the sludge and the resultant HLW glass are very small. HLW glass as a waste form has the desirable property that it can accept small quantities of almost any element without impacting its performance in a repository.

- *Quality of Final Waste Form*. HLW glass is a qualified, high-quality waste form that is accepted by the repository.

- *Economics*. The large-scale facilities imply lower treatment costs per unit of volume. Equally important, use of HLW facilities avoids the costs of building new facilities for treating the orphan wastes and qualifying multiple waste forms for the repository—a very expensive process.

The cost to obtain these benefits is that the wastes must be converted into forms acceptable for the HLW tanks and the glass melter. The conversion process can be conducted near the tank farms or offsite, and the wastes can then be shipped to the tank farms and slurried into the HLW tanks.

EXAMPLE WASTES AND PROCESSING OPTIONS

Three candidate wastes for mixing with HLW are described herein to provide an understanding of the potential possibilities. In each case, a process to convert the waste into a form acceptable for addition to a HLW tank is described. All the options take advantage of the capability of the HLW system to produce a high-quality waste form. Many take advantage of other characteristics of the HLW system.

Uranium-233

The United States is considering disposition of potentially excess ^{233}U. Although ^{233}U is not currently a waste, it seems likely that some portion of the ^{233}U inventory will eventually be disposed of. One option under investigation[2] is mixing the excess ^{233}U with HLW sludge and converting the mixture to HLW. This disposition option is potentially attractive for two reasons.

- *Radiation Control*. Some of this uranium requires heavy shielding because it contains a ^{232}U impurity and its decay product ^{208}Tl. Thallium-208 emits a 2.6-MeV gamma ray. The HLW system is one of the few systems capable of accepting wastes with high radiation levels.

- *Criticality and Safeguards*. Some HLW tanks contain HLW sludges with large quantities of depleted uranium (DU), which can be used to isotopically dilute the ^{233}U and eliminate safeguards and nuclear criticality concerns.

The ^{233}U can be prepared for addition to the HLW tanks by dissolving it in nitric acid, adding gadolinium for operational criticality control, and then neutralizing the solution with sodium hydroxide. This produces a sludge similar to the existing HLW that can be mixed with DU-containing HLW sludge before conversion to HLW glass. There are several other processing options to prepare a feed to the tanks.

Europium Control Blades

Several research reactors use aluminum control blades containing europium-oxide particles dispersed in the aluminum[3]. The expended blades pose a potential radiological hazard[4] because of (1) the high radioactivity levels and (2) the long-lived europium isotopes ^{152}Eu ($T_{1/2}$ = 13.48 years) and ^{154}Eu ($T_{1/2}$ = 8.59 years).

Aluminum corrodes rapidly in sodium hydroxide solutions such as those found in HLW tanks. The control blades could be dissolved in the existing HLW. The resultant HLW would be processed into an LLW stream that contains the aluminum and a HLW stream that is converted to HLW glass. As a consequence, most of the mass of the control rods (the aluminum) is disposed of as LLW at relatively low costs with only the small-volume, highly radioactive europium being converted to HLW glass.

Sludges

The decontamination of processing facilities generates highly radioactive, high-sodium, transuranic, acidic liquids requiring treatment and disposal. A recent U.S. National Academy of Sciences report described[5] the difficulty in treating and disposing of some of these wastes at the Idaho National Engineering and Environmental Laboratory. An option is neutralization of the liquid with sodium hydroxide, evaporation to dryness, and shipment to HLW tanks. The transportable solid product is chemically similar to HLW sludges and can be mixed with the HLW sludge. The resultant HLW would be processed into an LLW stream that contains most of the sodium in the initial liquid and a HLW stream that is converted to glass. Only the highly-radioactive impurities in the initial liquid go into the HLW glass.

REQUIREMENTS FOR TREATMENT BY THE HLW SYSTEM

There are multiple requirements that must be met to process wastes using an HLW system. These requirements are associated with: (1) transport, (2) criticality control, (3) chemical form, (4) physical form, (5) radionuclide content, and (6) institutional issues. Some of these requirements—such as over the road transport—are reasonably well defined. Other requirements are less well defined. For example, for very-radioactive materials such as curium, there have been concerns that the high radiation levels might change the behavior of certain separation operations associated with HLW system operations.

ECONOMICS

Disposal of HLW is expensive, so disposal of orphan wastes as HLW waste is attractive only for streams that cannot be economically disposed of by other means. The process for disposing of a waste needs to be designed carefully to produce as little HLW glass as possible. The incremental cost to produce and dispose of an HLW glass log is ~$500,000. A glass log can accept ~500 kg of waste. The characteristics of the HLW system imply small costs for addition of aluminum, sodium and other alkali metals, boron, or silicon in various chemical forms to the HLW tanks. These elements (1) are separated from the HLW into the LLW stream with low disposal costs or (2) are glass components, which must be added to make HLW glass. Most other elements become part of the waste matrix of HLW glass and have high associated costs.

CONCLUSIONS

Safety concerns about some of the older HLW tanks and political concerns imply that the near-term priorities will be to solidify the existing waste. However, as progress is made, there will be strong economic incentives to broaden use of HLW systems to treat other wastes and a general recognition that the HLW systems can reduce the risks associated with treatment and storage of other wastes. This suggests that the United States should begin to consider how to use the HLW systems to properly treat for final disposal the large number of small-volume, difficult-to-treat, highly-radioactive wastes.

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