DISPOSAL CONCEPTS FOR WASTE IN UNDERGROUND SINGLE-SHELL STORAGE TANKS AT THE HANFORD SITE

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ABSTRACT

The Hanford Site is preparing for the future remediation of single-shell underground storage tanks. A significant portion of that task is the disposal of radioactive and hazardous components of the wastes that are in the tanks, the tanks themselves, and any residual contamination (e.g., soil) resulting from tank operations. This report discusses disposal concepts being studied for application on waste stored in the single-shell tanks.

INTRODUCTION

Defense production activities since the early 1940's have resulted in the generation of large quantities of liquid waste. The waste was either disposed of or placed in tank storage to await treatment and disposal. The underground storage tanks (UST) for waste are of two general varieties: (1) a single-shell tank (SST) design containing the older (pre-1980) wastes, and (2) a double-shell tank (DST) design containing the newer liquid wastes as well as some of the liquid from the SSTs. The present defense waste treatment and disposal plan is to process the liquid waste from the DSTs into high-level waste (HLW) and low-level waste (LLW) fractions. The HLW fraction is scheduled to be immobilized into glass canisters in the Hanford Waste Vitrification Plant (HWVP) presently in the design stage. The resulting glass canisters would be transported to the federal HLW repository for disposal. The LLW fraction is scheduled to be immobilized with grout in the Grout Treatment Facility. Disposal of the grout in onsite subsurface concrete vaults is planned. A plan for the treatment and disposal of the SST wastes has not been prepared yet (1,2). Programs to look at processing and disposal options for the SSTs are underway. A program described in this report is designed to study waste disposal options for the SSTs and other offsite U.S. Department of Energy (DOE) USTs for which a treatment and disposal plan is not yet in place. This report will primarily address disposal concepts for Hanford Site SSTs.

The scope is to evaluate and carry out waste technology development work that will result in the demonstration of disposal methods that can be applied to the remediation of USTs.

LOW-LEVEL WASTE IMMOBILIZATION AND WASTE FORM DEVELOPMENT TECHNOLOGY

The bulk of the SST waste is categorized as low-level. This means that the largest volume of the SST waste should be directly disposable if radionuclides and hazardous waste components can be appropriately immobilized. The present emphasis of the program is development or adaptation of LLW immobilization/waste form technology to tank waste. As a part of this work, waste disposal technology was reviewed for potential application. Waste immobilization technologies and waste forms that can be considered are given in Fig. 1.

DISPOSAL TECHNOLOGY CRITERIA AND ISSUES

The LLW waste forms and packages must meet the following criteria for present disposal.

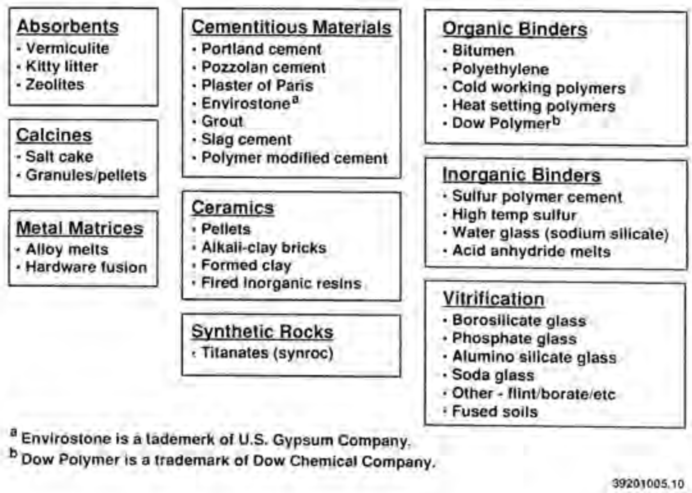

Fig. 1. Waste immobilization and waste form options listing.

- All wastes and waste residues must be converted to a waste solid or solids.
- Waste forms and packages must meet the Hanford Site solid waste disposal criteria given in WHC-EP-0063-3 (3) or a Washington State/U.S. Environmental Protection Agency (EPA) Treatment/Disposal Permit must be obtained.
- For direct disposal, mixed waste (MW) forms must pass the EPA Toxic Characteristic Leach Procedure (TCLP) leach test. (Tank wastes are considered MW because they have hazardous waste components in addition to radionuclides.)

Other considerations or issues:

- Waste forms should be the following:
  - Solid monoliths
  - Non-leachable or leach resistant including matrix
  - Compatible with packaging
  - Non-flammable and stable to moderate temperature rises
  - Compatible with nitrates/nitrites.
- Waste disposal technology should be the following:
  - Simple and practical
  - Exhibit waste minimization and practical processing costs.

The candidate waste forms included in Fig. 1 are being considered against the foregoing LLW disposal criteria and other considerations. Immobilization methods or waste form candidates generally will be screened by a TCLP test and general waste form characteristics and will then be tested for leach resistance to evaluate potential performance. The disposal options and relative processing complexity are shown in Fig. 2.

SELECTION OF LOW-LEVEL WASTE TECHNOLOGIES AND WASTE FORMS FOR TESTING AND EVALUATION

What are the top waste form technologies presently used or considered for use? The alternatives document, ERDA-76-43 (4), indicated that probably the top four waste technologies are vitrification, metal matrices, inorganic binders, and cementitious materials. Vitrification and metal matrix technologies are expensive, although they provide excellent product quality. The latter two technologies (inorganic binder and cementitious materials) are economically practical waste forms, but the quality is less than probably required for direct disposal of MW materials. It is doubtful that most cementitious forms or inorganic bound waste could meet the TCLP tests. However, polymer modified cements and/or enhanced inorganic binders are expected to pass the test. Therefore, the preliminary selection of technologies and waste forms to test was from vitrification, cementitious, and inorganic binder candidates; the metal matrices at present were excluded because of complexity and cost. Some of the program work has been initiated with grout (cementitious material), soil glass (vitrification), glass beads in sulfur polymer cement, cement, and polymer modified cement. In addition, work is planned with organic binders, primarily water repellent plastic materials. These selections were made based on an engineering report addressing waste form technology by Weimers, et al. (5). Also, if funding permits, a laboratory screening of miscellaneous waste forms and technologies such as ceramics, glass marbles, sulfur polymer cement (SPC), and others will be conducted. Polyethylene, a good representative of organic polymer technology, has been and is continuing to be studied by Brookhaven Laboratory for use as a waste form for UST type liquid wastes (6).

SELECTED WASTE FORM CONCEPTS

The selected waste forms and a brief concept of their potential uses are given as follows:

- Grout
- Soil glass (in-place immobilization)
- Glass beads in an SPC matrix
- Cement and polymer modified cement
- Organic binders
- Others - lab screen of other potential candidates such as ceramics, glass marbles, SPC, and low temperature glasses.

Grout

Work on grout is being done by Pacific Northwest Laboratory (PNL). The concept for grout is essentially the same as that presently being developed for the DSTs. However, the

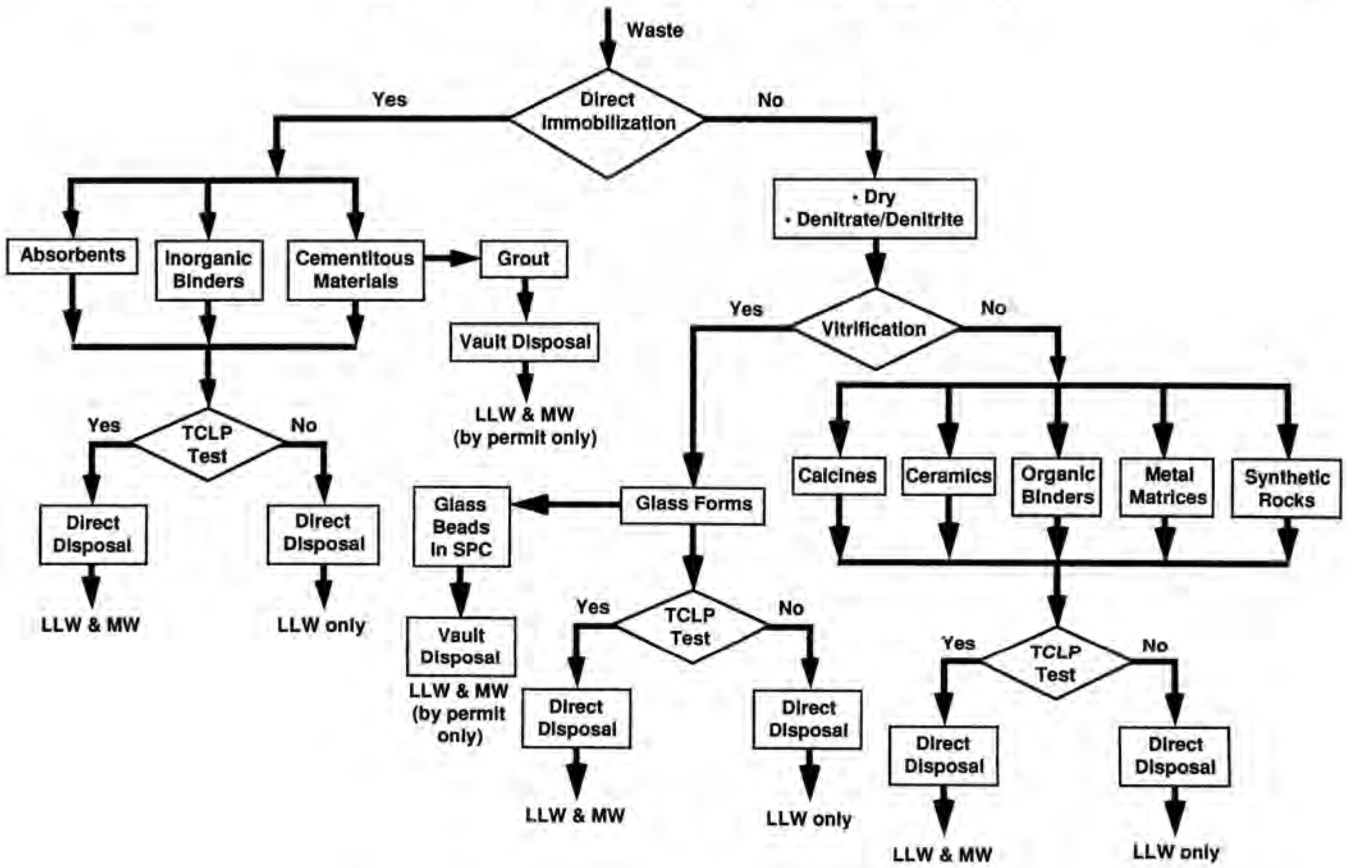

Fig. 2. Low-level waste immobilization (disposal options).

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chemical compositions of the SST wastes are significantly different from the DST wastes. This means that new grout formulations will be required, and therefore the effects of the new formulations will have to be studied and analyzed. In addition, there are substantial chemical variations within the SSTs so that either a number of different formulations will have to be developed or a variable type formulation will have to be derived that will accommodate composition changes. A grout schematic is shown in Fig. 3.

Soil Glass

Some of the soil near and around certain SSTs has been contaminated by tank leaks or other operations. This contaminated soil would typically be a LLW type waste. This soil could be processed or vitrified to form a glass product termed here as soil glass. The process used could be an in situ vitrification or similar process in which the soil was converted in-place into a glass, or it optionally could be removed and converted. The potential composition and use of soil glass is being studied by PNL. Preparation and testing of soil glass prepared from several different Hanford Site soils is presently being done as a part of this work. A schematic of the concept is shown in Fig. 4.

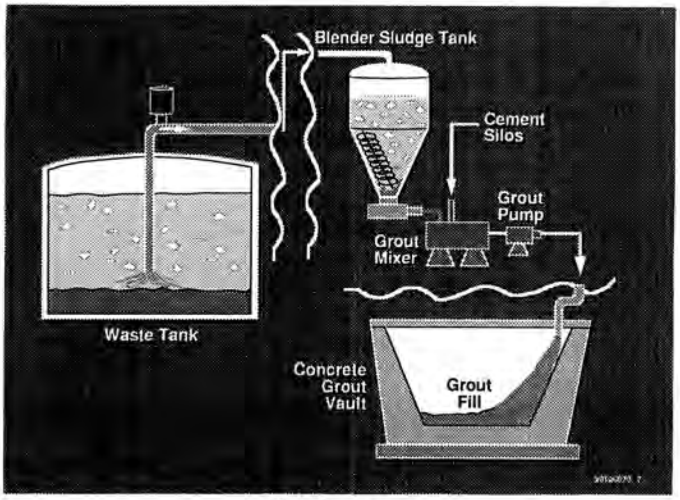

Fig. 3. Grout.

Glass Beads in Sulfur Polymer Cement

This is a pumpable glass waste form proposed by Boomer (7) and re-evaluated by Weimers (5). This concept is thought to be a possible option to grout as a pumpable slurry that could be disposed of in the vault barriers. The glass beads themselves should pass the TCLP test. The SPC matrix acts as an additional leach barrier and a carrier after it melts at 119°C (8). The intent is to use the SPC matrix as a fluidizing liquid to carry the glass beads through transfer pumps and pipes. The SPC matrix is being worked on by PNL, who has investigated optional glass compositions with simulated SST liquid waste. A schematic of the concept is shown in Fig. 5.

Cement and Polymer Modified Cement

Cement is a currently available waste form technology that appears to meet all the required LLW criteria with the possible exception of being able to pass the TCLP test. Modifying the portland cement formulation with a latex or other appropriate organic polymer material to make the cement product impervious to water should result in a product able to pass the TCLP test. Westinghouse Hanford Company (WHC) is performing the work on cement and modified cement. A parametric study of cement, water, and SST interstitial waste liquid has been completed. The resulting triangular phase diagram is given in Fig. 6. Compositions selected from this phase diagram will be used for preparing polymer modified cement test specimens. A schematic of the concept is shown in Fig. 7.

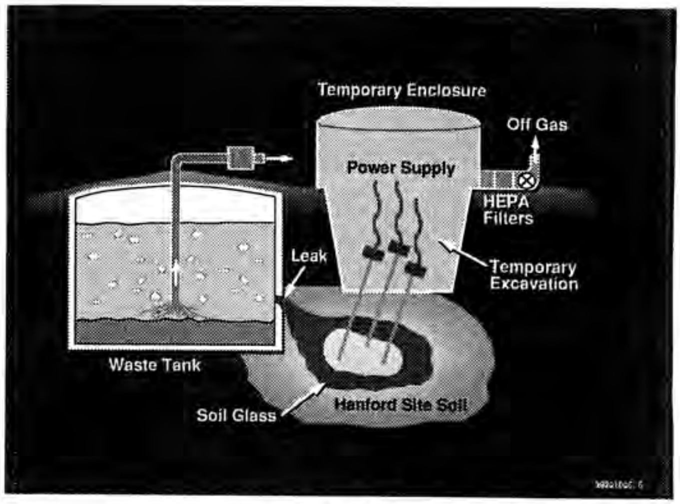

Fig. 4. Soil glass.

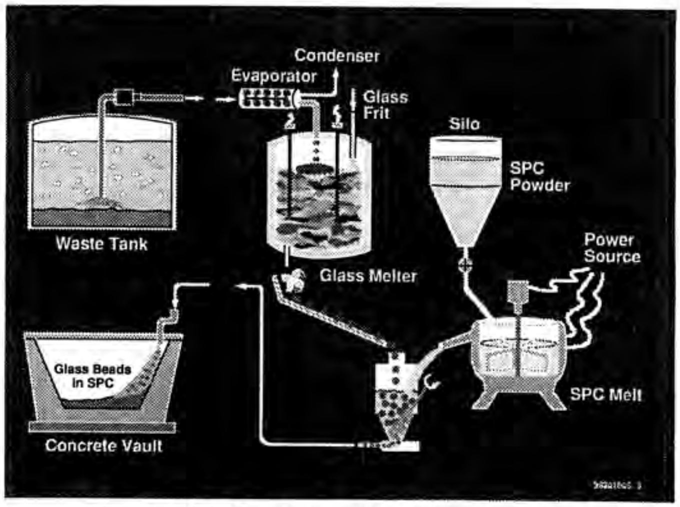

Fig. 5. Glass beads in sulfur polymer cement.

Organic binders, including polymers, are generally leach resistant, but they are not compatible with nitrates and nitrites forming flammable, combustible, or chemically unstable products. If the nitrates and nitrites can be destroyed cost effectively via heat, chemical addition, etc., then polymers might be acceptable. Possible polymer candidates include both cold forming materials, such as vinyl esters and specialized epoxy systems, and heat forming plastics such as

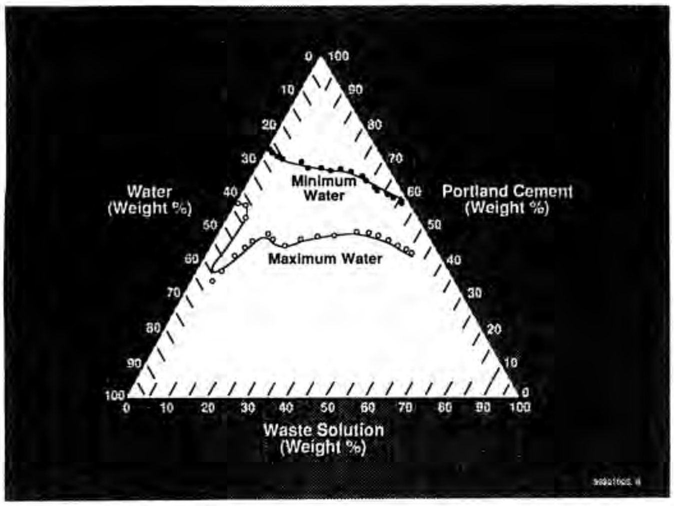

Fig. 6. Phase diagram for cement - water - tank waste.

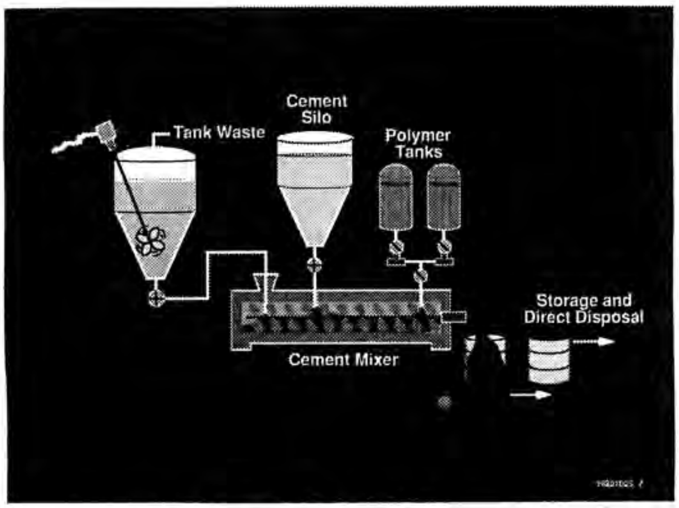

Fig. 7. Polymer modified cement.

polypropylene, acrylates, phenolic resins, etc. A schematic of the concept is shown in Fig. 8.

Others Category--Laboratory Screen of Other Potential Candidates

Other potential LLW form candidates selected from the chart in Fig. 1 include ceramics, organic polymers, SPC and other melts, low temperature glasses, etc. Ceramics are more leachable than glasses but also have a higher waste packing efficiency and can be easier and less expensive to prepare. If a relatively nonleachable ceramic waste form can be developed that would pass the TCLP test, then it could be considered for additional testing and evaluation.

Glass Marbles - The preparation of glass marbles is a current relatively inexpensive technology that should be adaptable to LLW disposal. Black glass marbles have been made from simulated HLW by PNL, but the concept was never utilized, possibly because the marble form is not as high

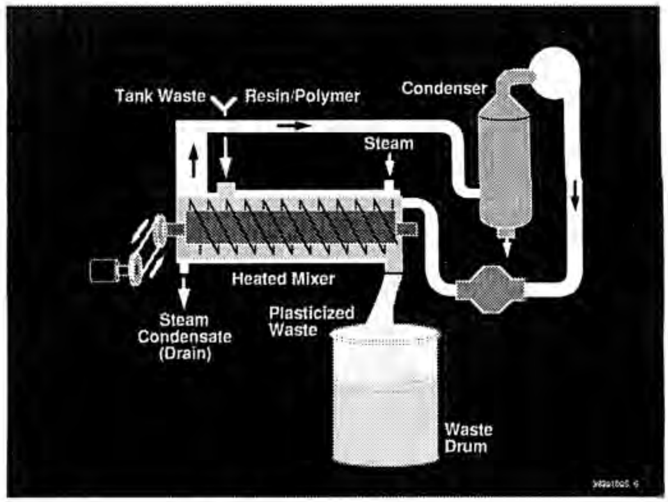

Fig. 8. Organic binders.

an integrity waste form as canister contained glass monoliths. However, for LLW, the integrity of the glass marble is expected to be sufficient and able to pass the TCLP test. The glass marble is highly mobile and can be easily transferred much like a liquid using either displacement pumps, vacuum transfers, or gravity feed. The glass marble might offer a low-cost, glass option that is easy to load automatically or remotely. A schematic of the concept is shown in Fig. 9.

Sulfur Polymer Cement - Sulfur polymer cement is an inorganic polymer (about 90% sulfur) mixed with a heat setting organic polymer to add additional water and acid resistance to the material. The material is formed at modest temperatures and can be used to encapsulate waste residues into a concrete like material. Use of other low or moderate temperature melts might be desirable.

Low Temperature Glasses - Low temperature glasses are an inexpensive immobilization agent using boric acid or phosphoric acids whose anhydride forms a low temperature glassy material. The low temperature glasses are generally slightly

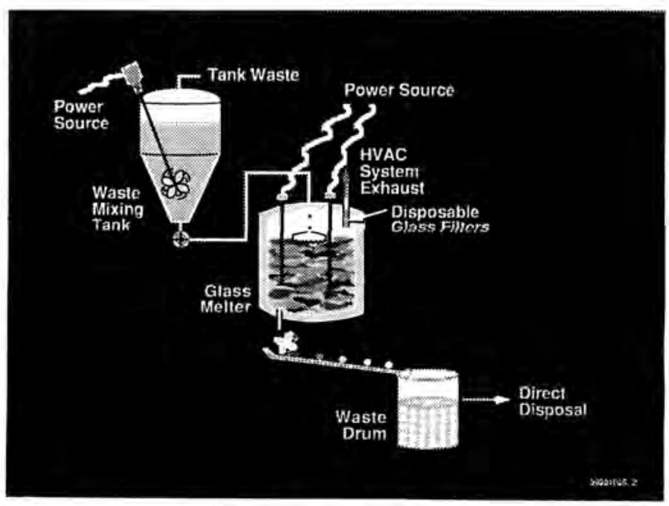

Fig. 9. Glass marbles.

soluble in water and must be made leach resistant in order to be used for this application. Other waste forms that could be considered are metal matrices and synthetic rock (synroc) type forms, but they are generally expensive and are more appropriately considered for transuranic (TRU) or HLW waste forms.

TRANSURANIC AND HIGH-LEVEL WASTE FORMS (FUTURE WORK)

The TRU waste category is included here even though it is not listed for DST waste processing. The DST plan is to fractionate the waste between LLW and HLW. Unfortunately, any low activity material with TRU radionuclides then becomes HLW. Cost differences between LLW and HLW are approximately a factor of 20 or more. Cost differences between LLW and TRU are a factor of five or less. Therefore, there appears to be a cost incentive to include a TRU waste plan.

It is expected that the best LLW form candidates will also be applicable to TRU waste forms. One exception might be the metal matrix waste form. This waste form is particularly suited for TRU waste. Several extractant type metal alloys can concentrate TRU radionuclides and form very stable, non-leachable solids. This form has the additional advantage that it can be readily recovered and the waste form reprocessed to recover the TRU material if desired at some future date. The higher cost of the metal matrix form for TRU material is offset by the above listed advantages, and it also generally makes substantial volume reduction possible. In any event, the TRU waste forms must meet Waste Isolation Pilot Plant (WIPP) Waste Acceptance Criteria (WAC).

The HLW fraction will most likely be incorporated into glass. However, the compositions of the SST waste are different from DST waste, so at least some change in glass formulations will be required. The HWVP will be designed for borosilicate glass melts in a large glass melter. There are a number of different glasses that could be considered as an option to borosilicate glass, such as alumina silicate glass, phosphate glass, a stabilized soda glass, flint glasses, stabilized borate glasses, etc. Also, vitrification processing in something other than a large glass melter might be considered, such as in-can melting that could be installed and operated in existing remote cells. The purpose of this work would be to look at HLW options that could either produce an acceptable glass form at a lower cost, produce a better quality glass form, or develop options that could be installed and operated within present Hanford Site facilities.

DOME FILL (FUTURE)

Dome fill addresses the need to fill the emptied tanks, tank farms, or tank domes with a fill material after the tank wastes have been removed, or after part of the waste has been removed and the residual waste stabilized. An engineering study has been performed by PNL. The study reported some testing on locally available fill material such as crushed basalt that would exhibit good subsidence control and would not be expected to react with any possible waste residues. A number of other candidate materials could be considered: granular clay, zeolite absorbents, minerals such as granular dolomite, coarse graded sand, washed and sized gravel, and cementitious materials. In addition to the inert or the absorbent materials, it appears advantageous to closely study use of the cementitious materials (Fig. 10).

Two such candidates could be considered: a commercial product named Controlled Density Fill® (CDF)*, and high impact strength concrete (without sand or gravel). The CDF material is now used in many places instead of fill dirt or clay and can be formulated to exhibit a specific compaction strength. It has good fluidity and is considered good for filling up void areas. The high impact strength concrete (9) considered here is light weight, pumpable, and sets up into a material that can absorb high impacts with little or no spilling or damage to the bulk solid. Impacts, penetrations, and boring

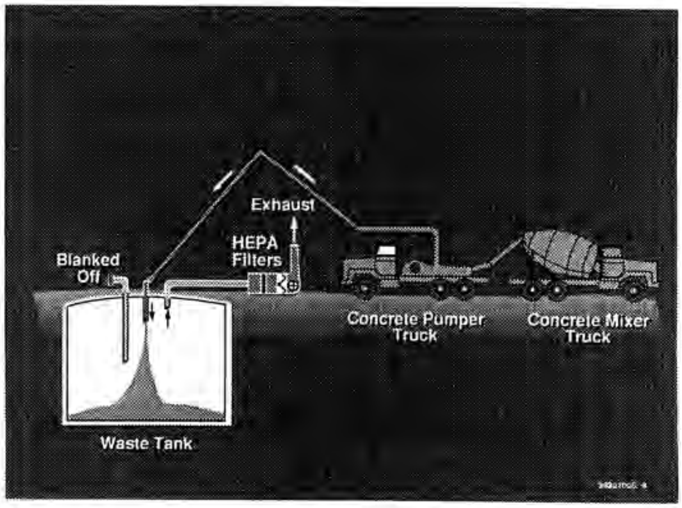

Fig. 10. Dome fill with cementitious materials.

have little detrimental affect upon the total fill. The cost for the cementitious material versus natural fill is probably more than offset by the ease with which cementitious materials can be pumped into the tank voids, etc. Also, cementitious materials offer some additional encapsulating value if any waste residues remain in the tanks.

IN-PLACE IMMOBILIZATION (FUTURE)

The in-place immobilization technology is designed for the tank waste treatment option where a portion or all of the waste is left to be immobilized in-place. Candidate options might include the following:

- In-place vitrification
- In-place grouting
- In-place cementation
- In-place polymerization.

Issues include the following:

- Development of in-place technologies are primitive.

* Controlled Density Fill (CDF) is a product of Pozzolanic.

- The feasibility of meeting in-place safety, regulatory, and quality concerns remains to be demonstrated.
- If in-place methods prove successful, costs should be substantially lower than ex-tank methods.

SUMMARY

The goal of this and related programs is to develop or establish technology that can be used to prepare a cost effective remediation plan for the processing and disposal of SST waste. Technology development has begun for candidate LLW forms and is planned for TRU waste and HLW. Fill technology support for the emptied tanks, stabilized tanks, or dome areas is outlined. Also, in-place immobilization will be investigated as a possible option to the other technologies, which include tank waste removal, waste immobilization, waste disposal, and empty tank backfill.

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