A COMPARATIVE STUDY OF ALTERNATIVE HIGH-LEVEL WASTE SOLIDIFICATION PROCESSES

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INTRODUCTION

Numerous processes have been proposed for solidifying high-level radioactive wastes. These processes must be evaluated to determine which one is best suited to a particular waste. An evaluation of the solidification processes should be based on a thorough understanding of the equipment requirements for each process. Unfortunately, determining equipment requirements is very difficult because most of the proposed processes are at such preliminary stages of development. There are still uncertainties about how the product and process must perform. For example, should each waste form be sampled and analyzed? Should each waste form be overpacked? Should the process be capable of recycling and solidifying all of its scrap and by-products, or will other processes be permitted to solidify these wastes?

In spite of limited knowledge of equipment requirements for high-level waste solidification processes, we at Pacific Northwest Laboratory attempted to compare the basic requirements for nine of the better known processes. These processes are:

1. in-can glass melting process (ICGM)
2. joule-heated glass melting process (JHGM)
3. glass-ceramic process (GC)
4. marbles-in-lead matrix process (MLM)
5. coated supercalcine pellets-in-lead matrix process (CSPLM)

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6. SYNROC process
7. titanate process
8. concrete process
9. cermet process.

PROCESS DEFINITION

For the purpose of comparing the nine solidification processes, we selected bases that seemed to us to be reasonable for operating a high-level waste solidification plant. The more important bases are listed below:

- Each process begins with the receipt of high-level liquid waste and ends with the production of a sealed and decontaminated canister of solidified waste.
- The process solidifies liquid waste generated when 2000 MTHM in spent, light-water reactor fuel rods are processed during a year.
- The process operates 24 h/d for 300 d/yr.
- The process is designed to operate at 1.5 times the nominal rate.
- Only high-level waste is solidified. Intermediate- and low-level wastes are solidified in other processes.
- The waste contains 34 W/kg on an oxide basis. This is equivalent to the typical heat loading of high-level waste five years after removal from a reactor.
- At the time of solidification, a maximum of 3 kW are permitted per canister of waste.
- Each waste canister is limited to the maximum dimensions of 0.3 m (2 ft) in dia and 3.1 m (10 ft) in height.
- The solidification process produces no off-standard waste forms and no scrap except where experience has indicated the production of significant volumes of scrap.
- Each process uses essentially the same off-gas and canister processing equipment. Canister overpacking is not required.

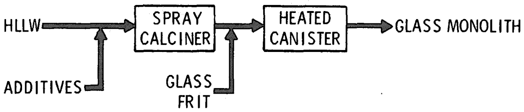
- Each process is assumed to be available for operation at least 67% of the time. At this rate of availability, the process can operate at design rate and still meet production requirements.

Using these bases and data provided by advocates of the various processes, we proceeded to define the basic equipment requirements for each process. We found that additional data were needed, especially pertaining to the properties of the waste between its liquid state and its final, solidified form. Therefore, we made assumptions about the properties of the waste, such as its density, thermal conductivity, specific heat, volatility, hygroscopicity, ability to flow, and ability to sinter. We also found that several options exist for producing a particular waste form. Many judgements were made in narrowing the number of options to a single basic process for producing a particular waste form.

PROCESS DESCRIPTIONS

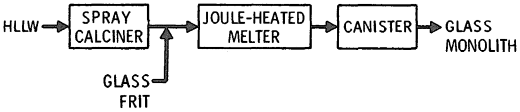
The nine basic processes are described below in very simple terms.

In-Can Glass Melting Process



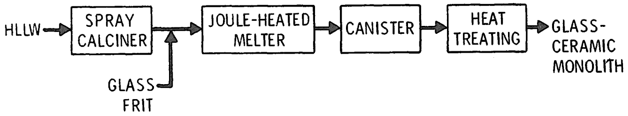
HLLW is dried in a spray calciner. The dry powder, or calcine, and glass frit fall into a heated canister where they melt together to form a glass monolith.

Joule-Heated Glass Melting Process



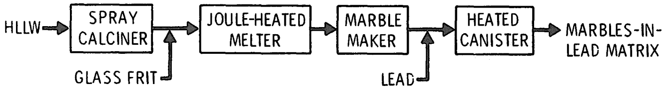
Calcine and glass frit fall into a ceramic-lined melter. The calcine and frit are melted together by heat that is generated by the resistance to the passage of electrical current through molten glass. The resulting glass is poured into canisters to form glass monoliths.

Glass-Ceramic Process



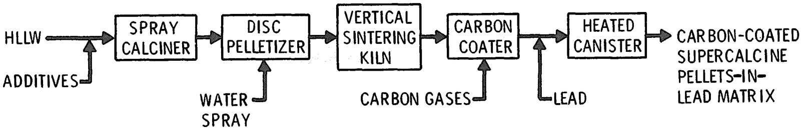
This process is identical to the joule-heated glass melting process described above except that the canisters of glass are specially heat-treated to promote crystalline growth within the glass matrix. The result is a glass-ceramic monolith.

Marbles-in-Lead Matrix Process



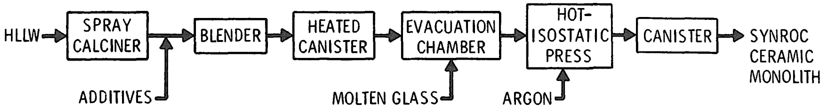
Glass is produced in a joule-heated glass melter as in the joule-heated glass melting process. The glass is continuously poured into a marble-making device. The resulting marbles are loaded into a canister, heated, and cast in a lead alloy.

Coated Supercalcine Pellets-in-Lead Matrix Process



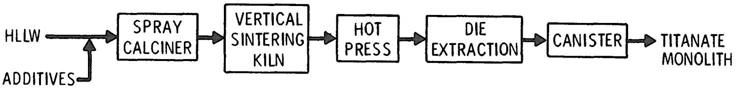
High-level waste is calcined in a spray calciner and is then pelletized in a disc pelletizer. The resulting pellets are sintered in a vertical, sintering kiln and are then coated with a thin layer of pyrolytic carbon. The coated pellets are loaded into a canister, heated, and cast in a lead alloy.

SYNROC Process



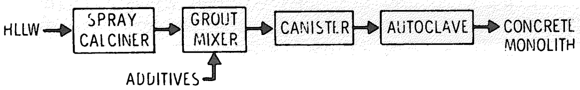
Calcined waste is blended with SYNROC additives that are sized to yield a maximum, dry bulk density. The blend is loaded into a heated alumina vessel where it is sintered into a semi-dense billet. While the alumina vessel and its contents are hot, gases are evacuated from the vessel. Then a charge of viscous glass is added to submerge the sintered SYNROC billet in glass. The billet is then hot-isostatically pressed using molten glass as the pressing medium. The alumina vessel with its glass and SYNROC contents is then loaded into a metal canister.

Titanate Process



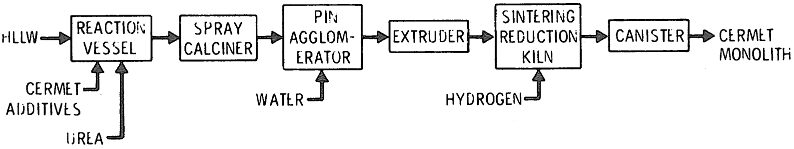
High-level liquid wastes and special calcia- and titania-bearing additives are dried in a spray calciner. The resulting product is calcined in a vertical, sintering kiln. This material is loaded into a graphite die and is then uniaxially hot-pressed. The resulting ceramic block is removed from the die and loaded into a metal canister.

Concrete Process



Calcine is mixed with water and cermet-forming additives and is then cast in a canister. The canister is placed in an autoclave, where elevated temperatures and pressures accelerate the setting and curing processes. The concrete monolith is placed in air storage where dewatering slowly occurs. The canister is later sealed.

Cermet Process



High-level liquid waste and cermet-forming additives are dissolved in molten urea. The solution is then spray-calcined. The resulting calcine is agglomerated into small, dense granules and is then extruded. The extruded rod is sintered and reduced under an argon-hydrogen atmoshpere. This yields a dense cermet rod containing ceramic and metallic phases. The cermet rod is then loaded into a metal canister.

PROCESS COMPARISONS

For each of the solidification processes we determined the number of processing components, the number of motorized parts, and the number of mechanical steps required to produce a single canister of solidified waste. We considered only the minimum in-cell requirements to operate the mainline process at design capacity. Therefore, we did not include backup equipment that would be part of a well designed process. We also did not include services or equipment located outside the cell. Finally, in-cell service equipment, which is not part of the mainline process, was not included. Examples of in-cell service equipment are vacuum cleaning systems, sampling systems and decontamination systems.

We first determined the number of processing components for each process. A processing component was defined as a unit of equipment that alters the physical or chemical form of the waste and/or stores it. Included in this definition are tanks, hoppers, furnaces, coolers, mixers, off-gas process vessels, and canister processes, such as seal welding and decontamination. For each of the processes, we found that the 3-kW limitation per canister was so restrictive that 6.5 canisters per day were produced regardless of the process. Therefore, the number of canister processing components was the same for each process. The number of off-gas processing components was also found to be basically the same for each process in spite of considerable variability in off-gas volumes and flow rates.

The total number of processing components for each process is shown in Fig. 1. The in-can glass melting process, for example,

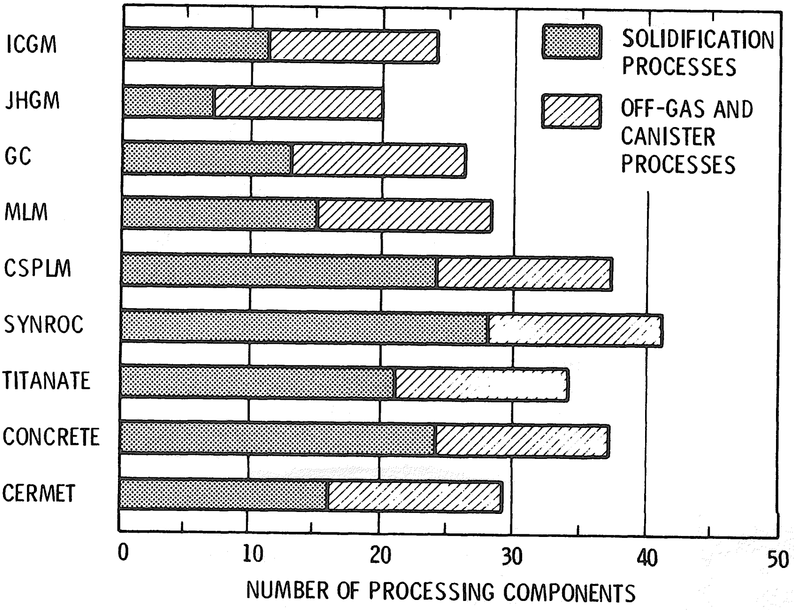


Fig. 1. A Comparison of the Number of Processing Components in Each of the Solidification Processes

consists of 24 processing components. The different processing components for the in-can glass melting process and the number of each are listed below:

HLLW holding tanks (3)
HLLW feed tank (1)
spray calciners (2)
seal pot (1)
in-can glass melters (4)
venturi scrubber (1)
knockout pot (1)
NO_x recovery unit (1)
condenser (1)
ruthenium removal bed (1)
iodine removal bed (1)
condensate evaporators (2)
forced-air cooling station (1)
seal-welding station (1)
leak-check station (1)
decontamination stations (2).

We also determined the number of motorized parts. A motorized part was defined as any in-cell component of the mainline process that rotates, translates or elevates. We did not include transfer devices that have no moving parts, such as steam jets and air lifts. In Fig. 2 the total number of motorized parts for each process is shown. The in-can glass melting process, for example, uses 42 motorized parts. The motorized parts for the in-can glass melting process and the number of each are listed below:

cranes (2)
manipulators (2)
tank agitators (4)
feed pump motor (1)
frit addition valve rotators (4)
spray calciner vibrators (6)
off-gas damper positions (2)
diverter valve actuators (2)
cone valve elevators (4)
ICGM furnace translators (4)
wire brusher motors (rotator and elevator) (2)
welder rotator (1)
canister revolving air-lock motor (1)
canister translator (1)
decontamination vessel elevator (1)

power lead clamping motor (1)
vacuum pump motor (1)
off-gas system pump motors (3).

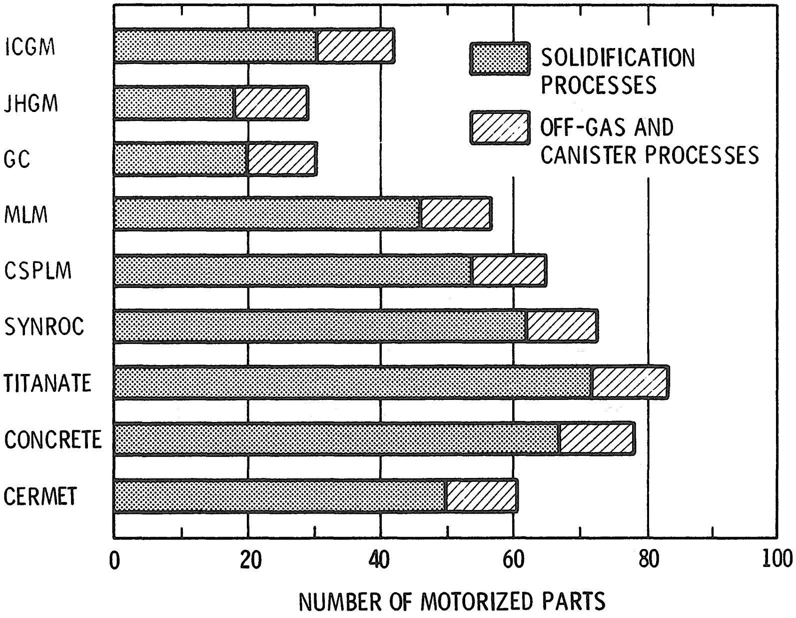


Fig. 2. A Comparison of the Number of Motorized Parts in Each of the Solidification Processes

We also determined the number of mechanical steps required to produce a canister of waste that is ready for shipping. A mechanical step was defined as any intermittent, motorized action that results in moving a charge of waste or a canister or that results in readying a processing component for operation. Examples include all crane and manipulator actions, vibration-induced material transfers, canister rotations and translations, and in-cell valving operations. Each translation step, such as sliding a canister under a loading spout, requires a repositioning step. In this case, it is sliding the canister out from under the spout after loading. Operations that are more or less continuous, such as agitation and pumping of HLLW, were not included as mechanical steps. The total number of mechanical steps per canister of waste produced are shown in Fig. 3 for each of the nine processes.

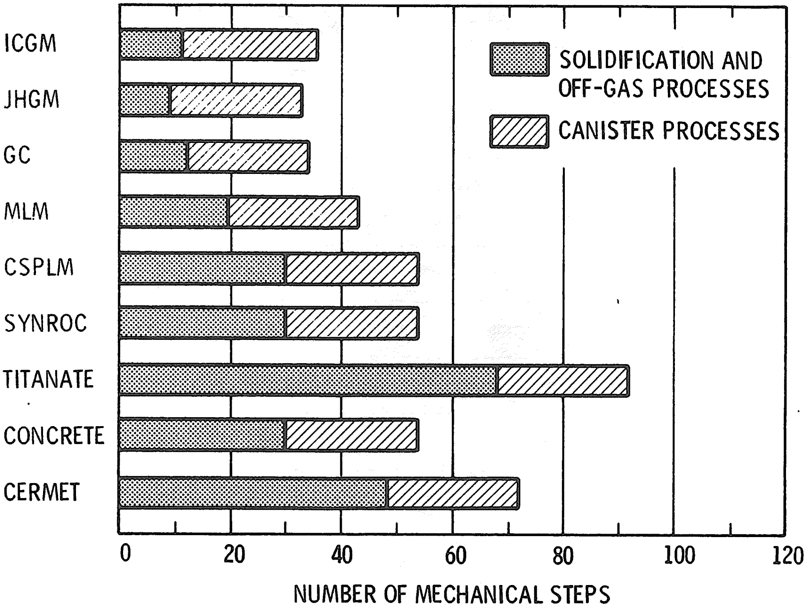


Fig. 3. A Comparison of the Number of Mechanical Steps in Each of the Solidification Processes

The mechanical steps required to produce a canister of waste by the in-can glass melting process are listed below:

1. unload canister from air-lock transfer rack (crane)
2. place insulating cover on canister (crane)
3. load canister in furnace (crane)
4. translate furnace to loading position (motor)
5. lower fill spout/cone valve (motor)

6. switch diverter valve position to canister (motor)
7. when canister is filled, switch diverter valve position to other canister (motor)
8. when canister is cool, elevate fill spout/cone valve (motor)
9. translate furnace to unloading position (motor)
10. move canister to cooling station (crane)
11. remove insulating top (crane)
12. when cool, move canister to welding station (crane)
13. place welding surface preparation system on canister (manipulator)
14. operate perparation system (2 motors)
15. remove preparation system (manipulator)
16. place lid on canister (manipulator)
17. place welder on lid (manipulator)
18. operate welder (motor)
19. replace welder (manipulator)
20. move canister to decontamination station #1 (crane)
21. move canister to submerged, revolving air lock (crane)
22. revolve air lock (motor)
23. move canister to decontamination station #2 (crane)
24. translate canister to power connection point (motor)
25. connect power lead (motor)
26. retract canister translator (motor)
27. elevate decontamination vessel under canister (motor)
28. lower decontamination vessel (motor)
29. translate canister holder in (motor)
30. disconnect power lead (motor)
31. translate canister out to unloading (motor)
32. install helium leak-test head (manipulator)
33. operate vacuum pump (motor)
34. when tested, remove helium leak-test head. (manipulator)
35. move canister to exit air-lock transfer rack (crane).

CONCLUSIONS

In general, and probably as expected, it can be concluded that the processes with the fewest processing components require the fewest motorized parts and the fewest steps to produce a canister of waste. The requirements for the in-can glass melting, joule-heated glass melting and glass-ceramic processes are lower in all categories when compared to any of the remaining six processes. The average difference in requirements between the in-can glass melting, joule-heated glass melting and glass-ceramic processes and the remaining six processes is about a factor of 1.5 to 2. As previously stated, in-cell and out-of-cell service equipment were not included. Equipment requirements are also likely to change as these processes reach a more advanced state of development. Other equipment will likely be required as process and waste form criteria are established.

Although the equipment and process steps identified in this study are of somewhat limited value, they could serve as a basis for more advanced analyses. For example, the reliability of each unit of equipment and each process step identified in this study could be estimated. Repair or replacement times for failed equipment could also be estimated. With reliability and repair or replacement data, it may also be possible to determine the additional equipment requirements, if any, for operating the process at its design rate. With a more complete summary of equipment requirements, other important processing parameters, such as product variability, safety and ease of decontamination, could be estimated.