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VITRIFICATION OF HIGH LEVEL NUCLEAR WASTE INSIDE AMBIENT TEMPERATURE DISPOSAL CONTAINERS USING INDUCTIVE HEATING: THE SMILE SYSTEM*

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ABSTRACT

A new approach, termed SMILE (Small Module Inductively Loaded Energy), for the vitrification of high level nuclear wastes (HLW) is described. Present vitrification systems liquefy the HLW solids and associated frit material in large high temperature melters. The molten mix is then poured into small (~1 m^3) disposal canisters, where it solidifies and cools. SMILE eliminates the separate, large high temperature melter. Instead, the HLW solids and frit melt inside the final disposal containers, using inductive heating. The contents then solidify and cool in place. The SMILE modules and the inductive heating process are designed so that the outer stainless can of the module remains at near ambient temperature during the process cycle.

Module dimensions are similar to those of present disposal containers. The can is thermally insulated from the high temperature inner container by a thin layer of refractory alumina firebricks. The inner container is a graphite crucible lined with a dense alumina refractory that holds the HLW and frit materials. After the SMILE module is loaded with a slurry of HLW and frit solids, an external multi-turn coil is energized with 30-cycle AC current. The enclosing external coil is the primary of a power transformer, with the graphite crucible acting as a single turn "secondary." The induced current in the "secondary" heats the graphite, which in turn heats the HLW and frit materials. The first stage of the heating process is carried out at an intermediate temperature to drive off remnant liquid water and water of hydration, which takes about 1 day. The small fill/vent tube to the module is then sealed off and the interior temperature raised to the vitrification range, i.e., ~1200°C. Material volatized (e.g., small amounts of cesium) during the liquefaction process is retained by an internal lower temperature "cold trap" inside the module. Liquefaction is complete after approximately 1 day. The inductive heating then ceases and the module slowly loses heat to the environment, allowing the molten material to solidify and cool down to ambient temperature.

The process cycle requires approximately one week, with most (60%) of the time used for cool-down. During the process cycle, the outer steel can is maintained at near ambient temperature by air cooling to remove heat generated by parasitic induced currents and conductive heat transfer through the insulating firebrick layer. These parasitic effects increase the amount

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and cost of energy required to process module; however, the cost is still very low. For example, a SMILE module containing 1.25 m^3 of vitrified HLW, (equivalent to the baseline HLW disposal module for the Hanford TWRS program) will require approximately 10 mWh. At a cost of 10 cents per kWh, this corresponds to only $1200, which is negligible compared to the repository fee of several hundred thousand dollars per module. Heat transfer analyses for the SMILE process are described, together with projected glass compositions based on Hanford HLW feeds. Using appropriate frit materials, the composition of SMILE-Hanford HLW glass can be made very similar to that projected for Savannah River waste. Because of its modularity and the elimination of long-term material problems, there do not appear to be major technical issues for the SMILE concept.

DESCRIPTION OF THE SMILE CONCEPT

The present vitrification approach is based on large, high cost, centralized high temperature glass vitrification facilities that melt the solid waste in large furnaces and then pour the molten glass into the final steel disposal containers. In SMILE, the waste solids would be vitrified in-situ inside closed, individual containers that serve as the final geologic disposal containers. The outer SMILE container is maintained at a relatively low temperature while its inner contents are heated to high temperature by an external low frequency (e.g., hertz) inductive heating coil. Thermal insulation between the inner contents and outer container minimizes heat leakage from the hot interior. The SMILE concept appears to offer substantial cost savings over the baseline approach. It also appears to be more reliable and maintainable, is much less demanding on materials, and essentially eliminates the problems of radioactive emissions from hot melters. Besides its advantages of lower costs and simpler operation, SMILE has the unique capability that the vitrified waste inside disposal containers can be reheated to high temperature without degrading canister containment. If long-term radiation damage to the vitrified waste were to prove unacceptable, the inner contents of SMILE containers could be annealed when necessary.

Figure 1 shows the proposed SMILE concept. The empty container consists of a graphite cylinder with an inner alumina liner. The graphite is inductively heated to high temperature by an outer solenoidal coil that is connected to an external power source. The solenoidal coil operates at a low frequency (e.g., 30 hertz), and acts as the primary of a transformer, with the secondary being the graphite cylinder. The graphite cylinder is enclosed in a stainless steel jacket (not shown) with a layer of thermally insulating refractory ceramic, e.g., alumina fire brick (also not shown), between them. The graphite can operate at high temperature, e.g., 1200°C or higher, while the enclosing steel jacket is maintained at a relatively low temperature [200°C, for example] by auxiliary air cooling. The stainless steel jacket completely enclosed the graphite cylinder and its contents except at the top of the container, where the fill/vent tube is initially open.

Place Figure 1 Here

The SMILE version in Figure 1 uses graphite as the conducting secepter that is heated by the inductive heating coil. Other versions are possible. For example, the graphite cylinder could be replaced by an iron container, which would couple even better to the inductive coil, making the heating process more efficient. The iron secepter version of SMILE would have a somewhat lower temperature capability but would still be high enough (i.e., ~1150°C) to vitrify HLW into borosilicate glass. It also is possible to have multiple tubes or cylinders (graphite or steel) at several points in the HLW solids to enable faster and more uniform heating. Here, however, we only analyze SMILE designs with a single graphite secepter that contains all of the HLW solids to be vitrified.

The external fill/vent tube at the top of the container allows the waste slurry (solids plus water) to be introduced and also allows the gases from the drying phase (e.g., steam) to leave. The central internal porous ceramic vent tube collects the gases generated inside the annular porous bed which then allows them to flow to the external vent. The steam travels at most about 12 inches (average about 6 inches) through the porous solid zone to reach the central vent tube, compared to a distance of 15 feet if there were no tube.

Figure 1A illustrates the drying operation. The container is filled with a wet slurry of HLW solids and frit material. Excess water is pumped out through the central vent and external fill/vent tube, leaving behind a settled bed of wet solids. The contents are then sequentially inductively heated to a sufficiently high final temperature (e.g., ~800°C) to first drive off all liquid water, then the water of hydration, and finally any residual decomposition gases. The remaining solids then contain only dry refractory waste oxides and frit.

The external vent/fill tube is then sealed off and the contents inductively heated to a higher temperature (i.e., ~1200°C) to vitrify the HLW (Figure 1B). No radioactive gases, e.g., volatized cesium, are released during vitrification, since the fill/vent tube has been sealed. This minimizes radioactive release and contamination of the vitrification facility. After the HLW solids have been vitrified inside the sealed container, it cools to near ambient temperature, and is discharged to a temporary storage facility. Eventually it would be shipped to an off-site geologic repository.

If an additional protective barrier is desired, the container can be overpacked with an outer steel container. The process is illustrated in Figure 2A. The open outer steel container would be positioned below the inner container and inductive heating coil during the drying and vitrification phase of the process.

Place Figure 2 Here

After vitrification is complete and the inner container has cooled to near ambient temperature, the outer container is raised to enclose the inner container (The outer container fits inside the heating coil). The outer container would then be lowered with the enclosed inner container, and removed through an exit transfer lock at the bottom of the pit. At the beginning of the sequence, both the inner and outer steel container come in through an entrance transfer lock at the bottom of the pit. The inner container enters first, is raised up by the hydraulic lift and then held in place by the center and adjustable side supports. The hydraulic lift is then lowered and the outer container brought in through the transfer lock. The treatment sequence is then initiated. The inner container is filled with wet HLW solids (#1); the water is removed by moderate heating (#2); the vent/fill tube of the inner container sealed (#3); and the HLW solids vitrified at high temperature (#4). In contrast to previous HLW vitrification processes, the SMILE process will not result in any significant release of radioactive volatiles. In the drying step (#2), the only volatiles are steam and decomposition gases (e.g., from residual nitrates) which are trapped externally; in the vitrification step (#4) all volatiles are fully retained inside the inner container.

After the heated, sealed inner container cools to near ambient temperature, the hydraulic lift raises the outer steel container around it (Step #5). The outer/inner container combination is then lowered to the bottom of the process pit and removed through the exit transfer lock. The open top of the outer steel container is then covered with a lid, and a welded seal made remotely, completely sealing off the vitrified waste inside a double barrier container (Step #6). The completed inner/outer container combination is shown in Figure 2B. The finished container would be temporarily stored and then sent to an off-site geological repository. The projected time line for processing SMILE containers is given in Table 1. A more detailed description of the SMILE concept is given in the BNL report, "SMILE - A New Approach for the Vitrification of High Level Wastes," by J. Powell, et al.[1]

Place Table 1 Here

ANALYSIS OF THE SMILE CYCLE

Dimensions for a typical SMILE container are given below. These values are based on an initial study of the SMILE concept, and may change somewhat after further, more detailed studies. Outer radius of central vent tube = 5 cm; outer radius of HLW glass region = 32.5 cm; outer radius of alumina cylinder = 33.5 cm; outer radius of graphite cylinder = 41.0 cm; outer radius of thermal insulator = 46.0 cm; outer radius of stainless steel jacket = 47.5 cm; length of canister = 450 cm.

The drying of the wet HLW/frit solids involves a complex conductive/convective transient heat transfer process. Exact analysis of the process is beyond the scope of this paper. However, the transient behavior can be approximately determined from analytic solutions given by Schneider[2] in the Handbook of Heat Transfer. The following thermophysical properties are taken for the bed of HLW/frit solids: $k = 1\times10^{-2}$ watts/cm°K, 70% solids volume fraction in bed, $\rho C_p = 2$ Joules/cm^3 °K [per cm^3 of bed], with a corresponding thermal diffusivity of $\alpha = k/\rho C_p = 5\times10^{-3}$ cm^2/sec. At the beginning of the drying phase, the temperature of the graphite alumina cylinder is raised from its original value, T_o, to T_G and held constant. Heat is conducted/convected radially inwards, with steam and other gases being driven off through the central vent tube. The temperature ratio, T_R, relating the time dependent (Θ = time) temperature at the axis of the HLW/frit bed (i.e., at the central vent tube) to the original temperature T_o, is a function of the Fourier number, F_o,

$$T_R = \frac{[T(O,G)-T_O]}{[T_G-T_o]} = f\,[F_o] = f\left[\alpha\frac{\Theta}{(R_B)^2}\right] \qquad (1)$$

Taking T_G = 1150 K(877°C) and the Fourier number = 0.5, the analytic solution given by Schneider yields a value of T_R = 0.90 (center temperature = 1065K). For the condition of R_B = 32.5 cm and $\alpha = 5\times10^{-3}$ cm^2/sec, the time required is Θ_{DRY} = 29.4 hours. At the end of the drying phase, the temperature of the coolest portion of the bed (i.e., at the central ceramic vent rube) is 792°C, with the average bed temperature being ~800°C. The temperature of the graphite/alumina cylinder is then increased to a higher constant value, T_G*, so that the HLW/frit solids can be vitrified. Fixing a precise value for T_G^* will require experiments but it is expected that T_G^* will be on the order of 1200°C (1473K). Taking temperature ratio, T_R^*, for the vitrification phase is 0.85, the corresponding Fourier number is $F_o^* = 0.4$. The time required for vitrification is Θ_{VIT} = 23.6 hours. The bed temperature at the ceramic vent tube at the conclusion of the vitrification phase is $T^*(0,\Theta_{VIT})$ = 1150°C, based on values of T_G = 1200°C and T_o = 800°C.

The total energy input to the bed for drying and vitrification supplied by the graphite/alumina heater is $Q_{TB} = Q_{DRY} + Q_{VIT} = 1.16\times10^9$ Joules/meter, based on the change in enthalpies of the original components. Two additional energy inputs are supplied by the graphite/alumina heater: 1) energy to raise the heater from a cold state to its final operating temperature, and 2) energy lost to the cool outside steel container by conduction through the insulating alumina brick layer during the drying and vitrification phases. The first term equals 0.75×10^9 Joules/meter, the increase in enthalpy of the graphite cylinder and alumina liner. The second term is considerably greater, 2.42×10^9 Joules/meter. The temperature, T_{SS}, of the outer steel container is maintained at a relatively low value, e.g., 500K (227°C) by air cooling. The thermal conductivity of the insulation is taken as equal to that given by Marks[3] for #16 insulating brick refractory. The total energy input from the heater then equals $Q_{TOT} = Q_{DRY} + Q_{VIT} + Q_{HEATER} + Q_{INSUL} = 4.33\times10^9$ Joules/meter. This corresponds to a heater energy input that is approximately 3 times greater than that required to dry and vitrify the HLW/frit solids themselves. The total energy input for a 5 meter long container is 2.16×10^{10} Joules or ~6000 kWh. At an electrical energy cost of 10 cents per kWh, this corresponds to a cost of ~$600 per container. The average electrical power input during the drying and vitrification phases is 113 kW(e).

There is an additional energy input and cooling load associated with parasitic I^2R losses generated in the outer stainless steel container by the inductive heating coil. The SMILE container is equivalent to a single turn secondary of a transformer, with the primary being the inductive heating coil. The large size of the container, plus the low heating rate enables the use of low AC power frequencies, i.e., 30 hertz, in the inductive heating coil. The alternating magnetic field produced by the inductive heating coil will induce currents in the outer stainless steel container as well as in the graphite cylinder, however, and energy losses to the steel must be included as part of the overall energy losses. They will not affect the internal temperature distribution, but do affect the cooling load on the outer steel container. Analyses of the inductive heating process find that the parasitic currents in the steel container approximately double the required input power, from 113 kW(e) to a total of 230 kW(e).

COMPOSITION OF HLW GLASS PRODUCED BY SMILE

SMILE can be used with all processing alternatives, including enhanced sludge washing and acid dissolution (e.g., TRUEX). As an example, HLW from the TRUEX D process proposed for the Hanford TWRS program can be combined with suitable frit material to yield a final borosilicate glass composition that closely approximates Savannah River HLW glass.

Table 2 gives the weight percent of the various solid oxides (after heating) in the TRUEX D waste[4] and compares it to the weight percent distribution for Savannah River waste.[5] There are substantial differences, and some components fall outside of the quoted possible range. However, by addition of appropriate frit material, the combination of Hanford TRUEX D waste and frit can closely match the composition of Savannah River waste glass. Table 3 shows the resultant composition produced by adding frit in the ratio of 3 parts frit by weight to 1 part of TRUEX D waste. The resultant composition matches that of the Savannah River glass for all components except those marked with an asterisk. For those that are different, the discrepancies are relatively small, with the principal differences being a higher amount of Al_2O_3 (6.9 vs. 2.8 wt.%) and a somewhat lower amount of Fe_2O_3 (10.1 vs. 14.5 wt.%). These differences are not expected to change glass properties significantly, however. These relatively small differences could be further reduced by minor adjustments in the concentrations of the other components.

Place Tables 2 and 3 Here

SMILE DEVELOPMENT

There do not appear to be any major technical feasibility issues for SMILE. The inductive heating coil is low tech and thermal insulation techniques are well established. The handling and lifting equipment appears straightforward. Material behavior during the vitrification phase should be confirmed; however, it appears molten glass can be contained without problems in both alumina and graphite. Because the SMILE concept involves small modules, and it does not require the high temperature components to operate for long periods, it should be possible to readily demonstrate the concept at full scale, and show that containers can be reliably produced. Technology modifications can be made quickly and easily, since the time to process a container is only a few days. This eliminates waiting for many months to detect problems, and then waiting for many more months to see if the required modifications are successful.

Borosilicate glass can be produced in alumina, steel, or graphite containers. Issues relating to heat transfer and energy cost do not appear to be significant. The issue of how much gas is released inside a SMILE container during the vitrification phase, and whether it would result in an objectionable pressure increase, can be resolved at an early stage by laboratory tests on simulated wastes. As discussed earlier, if the release amount is considered excessive, the container could be vented during vitrification with an attached cold trap to catch whatever cesium would volatize.

There is a substantial information base on the properties of the various materials that would be used for SMILE, and the thermal and electrical designs of the SMILE container and process facility appear to be straightforward. However, SMILE will require the normal engineering development associated with any new process, which involves going from bench scale to pilot plant to full scale.

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3. Marks Standard Handbook for Engineers, Eighth Ed., p. 6-172/3, T. Baumeister, E. Avalene, and T. Baumeister III (1978).
4. K. BOOMER, "Tank Waste Technical Options Report," WHC-EP-0616 (1993).
5. M. PRODINEC, G. WICKS, and N. BIBLER, "Borosilicate Glass as a Matrix for the Immobilization of Savannah River Plant Waste," p. 336, DOE/TIC 462 (Vol. 2), P. Hofmann, ed. (1982).

Table I
TIME LINE FOR PROCESSING SMILE CONTAINERS

Process Step		Description	Approximate Time Required
1A.	Position empty inner container inside heating coil	The inner container is introduced through the entrance transfer lock, raised by the hydraulic lift inside heating coil, and held in place by engaging the central support rod and emplacing movable side supports.	2 Hours
1B.	Introduce HLW slurry into inner container & pump out water	A slurry of HLW solids and water is pumped into the inner container through the external fill/vent tube. A separate small tube (not shown) inside the central porous vent tube sucks up water that drips out of the packed annular bed. The HLW solids remaining will then be like a bed of wet sand.	2 Hours
2.	Inductively heat the graphite/alumina inner container to dry out the HLW solids	The inductive heating coil heats the graphite/alumina container. Heat is thermally conducted into the wet HLW solids bed, vaporizing and driving off liquid water, water of hydration, and decomposition gases. The steam exits through the central porous ceramic vent tube to the top of the inner container where it is collected by a transfer line that connects to an external condenser. The coupling between the container and transfer line is remotely engaged.	29 Hours
3.	Seal external vent/fill tube	Disconnect transfer line to vent/fill tube, insert plug, and seal weld shut.	2 Hours
4A.	Inductively melt HLW waste	Increase temperature of graphite/alumina container - continue heating until HLW waste is vitrified.	24 Hours
4B.	Cool Inner Container	The melted HLW waste solidifies and the inner contents cool off to ~ 200° C.	100 Hours
5.	Enclose Inner Container in Outer Steel Container	The outer container is lifted to enclose the cooled inner container. The combination is then lowered and removed through the exit transfer lock.	2 Hours
6.	Seal Outer Container	A cap is put in the outer container and seal welded shut.	2 Hours
			163 Hours

Table II
COMPARISON OF TRUEX SOLID OXIDE WASTE COMPOSITION
WITH SAVANNAH RIVER WASTE

Component	Solid Oxide Waste %		
	Savannah River [DOE/TC-14621 (V.2) M. Plodine, et al., p. 336]		Hanford TRUEX D [WHC-EP-0616 Appendix G]
	Avg.	Possible Range	Avg.
Fe_2O_3	48	15-56	7.9
MnO_2	12	4-15	0.4
Other solids (Fission products + others)	11	0-12	18.5
Al_2O_3	9	1-51	27.6
NiO	6	2-10	2.6
U_3O_8	5	0-13	5.0
CaO	4	1-6	0.6
SiO_2	3	0-10	22.8
Na_2O	2	1-7	14.6

Table III
COMPARISON OF SAVANNAH RIVER & SMILE TRUEX GLASS WASTE

Component	Weight %		
	Savannah River Glass	SMILE-TRUEX Glass	Frit for SMILE Glass
SiO_2	41.1	40.0*	45.9
Fe_2O_3	14.5	10.1*	10.9
Na_2O	13.0	13.0	12.5
B_2O_3	10.4	10.4	14.0
Li_2O	4.0	4.0	5.4
MnO_2	4.0	4.0	5.3
Other solids	3.0	4.6*	---
Al_2O_3	2.8	6.9*	---
NiO	1.8	1.8	1.5
MgO	1.4	1.4	1.9
U_3O_8	1.4	1.2*	---
CaO	1.1	1.1	1.5
TiO	0.7	0.7	0.9
ZrO_2	0.4	0.4	---
La_2O_3	0.4	0.4	0.2
Total	100.0	100.0	100.0

*Different composition from SRP glass.

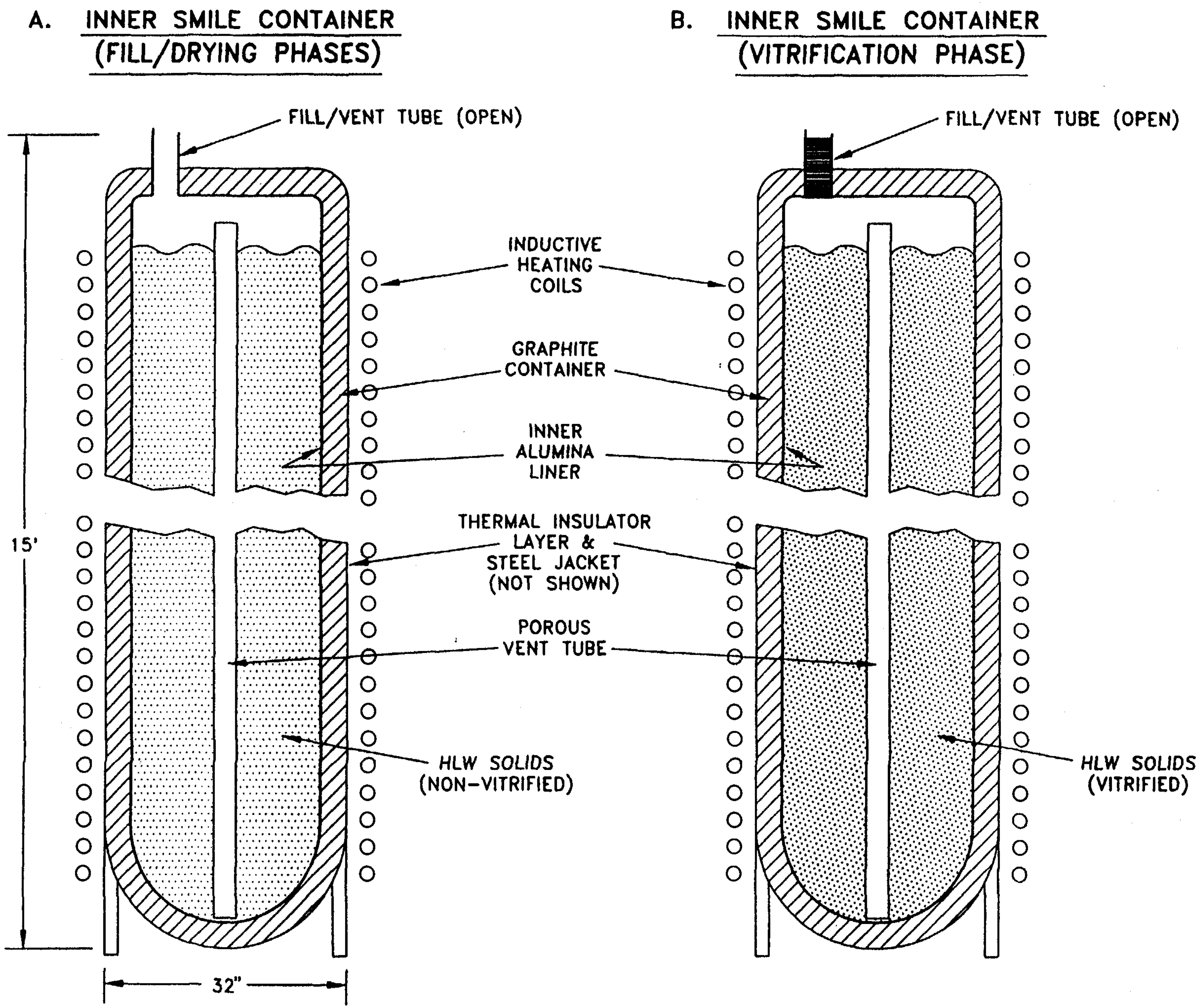

FIG4

Figure 1. The SMILE (Small Module Inductively Loaded Energy) Concept

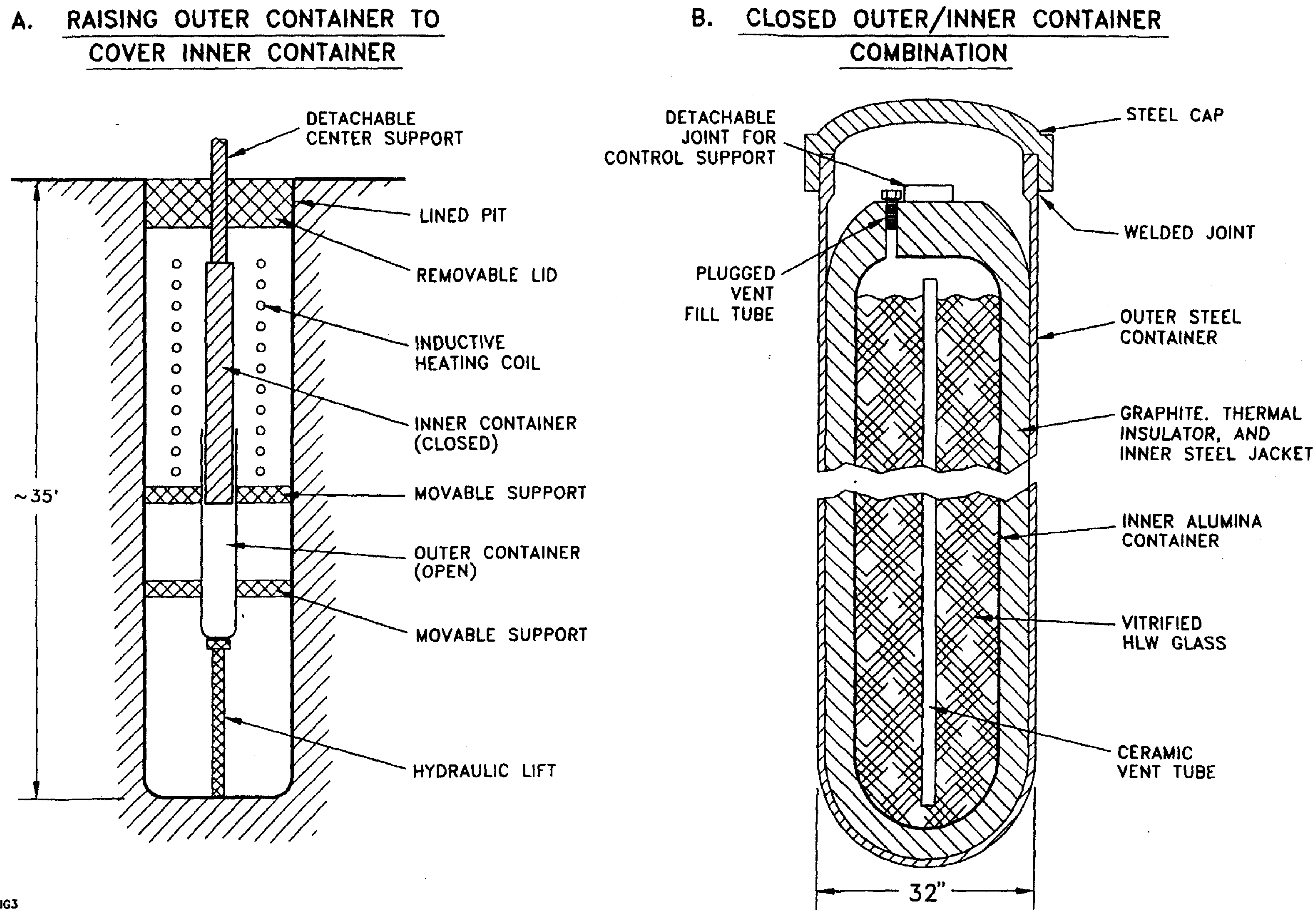

FIG3

Figure 2. SMILE Module Details

MASTER RECORD REPORT

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