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APPLICATION OF THE HANFORD THERMITE PROCESS
TO INCREASE IMMOBILIZATION OF IN-TANK SOLIDIFIED WASTES

By

Wallace W. Schulz
Michael J. Kupfer

Chemical Technology Laboratory
Research and Development
Chemical Processing Division

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ABSTRACT

The Hanford Thermite process is a high-temperature process for conversion of high-level, radioactive wastes to insoluble silicates and aluminosilicates. For application of the process to Hanford's In-Tank Solidified (ITS) waste, statistically designed experiments with synthetic waste demonstrate the optimum thermite charge contains 30 to 40 wt% ITS waste, 15 to 35 wt% Fe_2O_3 (or alternatively, 15 to 35 wt% MnO_2), 15 to 20 wt% silicon metal, and 10 to 25 wt% sand. Ignition of such a charge produces a green-black silicate glass (density = $\sim 2.5 \text{ g/cm}^3$; thermal conductivity = $\sim 0.7 \text{ watt/meter } ^\circ\text{C}$) whose leach rate in distilled water at 25°C is in the range 10^{-7} to $10^{-5} \text{ g/cm}^2 \text{ day}$. This glass, judging from initial data, should be an excellent vehicle for immobilization of ITS waste.

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APPLICATION OF THE HANFORD THERMITE PROCESS
TO INCREASE IMMOBILIZATION OF IN-TANK SOLIDIFIED WASTES

INTRODUCTION

Upon completion of the Hanford Waste Management Program^[1] millions of gallons of salt cake generated by evaporation of aged, high-level, alkaline waste liquor will have accumulated in underground storage tanks. This In-Tank Solidified (ITS) waste is essentially a mixture of NaNO_3 , Na_2CO_3 , NaNO_2 , NaOH , and NaAlO_2 ; principal radioisotopes are strontium-90 and cesium-137. Ultimate disposition of this solid, high-level waste has not yet been determined; it may become necessary, however, in the future to convert the salt cake to a more immobile form for long-term storage.

Several high-temperature methods for conversion of ITS and other high-level wastes to immobile silicate and aluminosilicate forms suitable for long-term disposal and/or storage have been proposed recently. These include the Hanford Thermite process,^[2] the Thermalt process,^[2] and the Basalt Melting process.^[2] Candidate feeds, besides ITS waste, to these processes include calcined Purex process waste and the encapsulated $^{137}\text{CsCl}$ and $^{90}\text{SrF}_2$ which will be produced in the Hanford Waste Management Program. Proposed laboratory-scale studies to develop and demonstrate these immobilization processes have been outlined previously.^[2]

This report is the first of a series intended to document progress of the laboratory investigation of the various high-temperature processes. Statistically designed experiments have been performed to define suitable reactant proportions and reaction conditions for operation of the Hanford Thermite process with ITS waste. Results of these experiments are

described here along with other pertinent observations and tests.

SUMMARY AND CONCLUSIONS

Ingredients of the feed charge in the Hanford Thermite process are ITS waste, Fe_2O_3 , Si metal, and Hanford sand.* When ignited, these ingredients combine exothermically to yield a glassy iron silicate in which other silicates and aluminosilicates, including those of various fission products, are dispersed. Typical glasses produced by the Hanford Thermite process leach in distilled water at 25 °C at rates that range from 10^{-7} to 10^{-5} g/cm² day (after 96 hr leaching).

Results of statistically designed experiments with synthetic ITS waste demonstrate optimum proportions of reactants in the initial thermite charge are 30 to 40 wt% ITS waste, 15 to 20 wt% Si metal, 15 to 35 wt% Fe_2O_3 , and 10 to 25 wt% sand. These experiments also show that both the rate of the thermite reaction and the leach rate of the silicate product increase with increasing amount of ITS waste in the thermite charge. Increased dilution of the charge with sand results in decreased reaction rate and decreased density of the silicate product.

Iron metal, produced as a by-product in the Hanford Thermite process, scavenges less than 0.06% of either cesium or strontium from the ITS waste. Substitution of MnO_2 for Fe_2O_3 appears potentially desirable to reduce the amount of metal by-product and to provide a slight decrease in the water leachability of the silicate glass.

* Hanford sand contains, typically, 64% SiO_2 , 12.7% Al_2O_3 , 6.4% FeO , 4.1% CaO , 3.2% Na_2O , 2.0% MgO , 1.8% K_2O , and 1.0% TiO_2 .

PROCESS DESCRIPTION AND FLOWSHEET

Both the Thermite process originally proposed by Spector, Suriani, and Stukenbroecker^[4] and the more recent Hanford version are high-temperature (1500 to 2000 °C) methods for producing insoluble silicates and aluminosilicates of ¹³⁷Cs, ⁹⁰Sr, and various other fission products.* The former uses high-level Purex process waste as feed while the Hanford Thermite process is intended primarily for further immobilization of ITS waste. The principal reaction in these processes is

which serves as a source of heat energy to melt the feed ingredients.

A conceptual flowsheet for application of the Hanford Thermite process to ITS waste is shown in Figure 1. Hanford sand is added to the reaction mixture as a diluent to control the rate of the thermite reaction and to provide additional silicon and aluminum for formation of silicates and aluminosilicates. As shown in Figure 1, the Hanford Thermite process is operable over a wide range of feed compositions and is flexible enough to accommodate any variation in ITS waste compositions. So far this process has been demonstrated only on a batch basis with simulated ITS waste. Both batch and continuous process tests with actual ITS waste are planned for the future.

The final solidified product of the Hanford Thermite process contains fayalite (Fe_2SiO_4) as the primary silicate matrix in which other silicates are homogeneously dispersed.

* It is believed these processes will also produce silicates of plutonium and other actinide elements but this has not yet been established.

FIGURE 1
HANFORD THERMITE PROCESS CONCEPTUAL FLOWSHEET

The typical silicate product (Figures 2a and 2b) is a gray-green to black colored glass whose density is about 2.4 to 2.8 g/cm³ and whose thermal conductivity is about 0.7 watt/meter °C. Its bulk leach rate in distilled water is of the order of 10⁻⁷ to 10⁻⁵ g/cm² day. Judging from these physical and chemical properties, the glass produced by the Hanford Thermite process should be an excellent vehicle further immobilization of ITS waste.

Neither the exact amount and composition of the gases given off during thermite immobilization of ITS waste nor their treatment and disposition have been established. Qualitatively, however, some white smoke and nitrogen oxides evolve throughout the reaction; the former is believed to consist of particulate SiO₂ and Na₂O. Limited analyses indicate only 2 to 5% of the cesium and <0.5% of the strontium in the ITS waste volatilize during the thermite reaction.

The volume of immobilized ITS waste obtained by the Hanford Thermite process is about 1.5 to 2 times the volume of the original ITS waste. Methods for reducing the volume of the ITS waste prior to immobilization are currently being investigated.^[2]

Besides fayalite, iron metal is also produced in the Hanford Thermite process. The amount of metal by-product is a function of the amount of Fe₂O₃ in the initial reaction charge. For charges containing 30 to 40% Fe₂O₃ sufficient iron metal is produced to collect as a separate layer at the bottom of the silicate glass (Figure 2a). [Even in this case, however, less than 0.06% of the cesium and strontium in the ITS waste reports to the metal phase.] Considerably less iron is produced with charges containing less than 20% Fe₂O₃ and, in this case, the metal is dispersed throughout the silicate matrix (Figure 2b).

Charge Formulations

	<u>a</u>	<u>b</u>
Wt% ITS-2 waste	30	40
Wt% Si metal	16	18
Wt% Hanford sand	10	27
Wt% Fe ₂ O ₃	44	15

FIGURE 2

APPLICATION OF HANFORD THERMITE PROCESS TO ITS WASTE--
TYPICAL PRODUCTS

Finally, as noted in Figure 1, MnO_2 may be substituted for Fe_2O_3 as the primary oxidant in the Hanford Thermite process. Such substitution offers several potential process advantages. Foremost of these is that little metallic by-product is produced even with charges containing as much as 40 wt% MnO_2 . Leach rates of silicate products made from charges containing MnO_2 are as low as and, in some cases, slightly lower than those of glasses made from charges containing an equivalent amount of Fe_2O_3 .

EXPERIMENTAL

MATERIALS

Simulated ITS Wastes

Table I lists the composition of the two simulated In-Tank Solidified (ITS) types of wastes used in this work. [Both wastes were prepared by thorough mixing of dry ingredients.] These compositions were selected to take into account the variation in NaOH and NaAlO_2 concentrations of actual ITS

TABLE I
COMPOSITION OF SIMULATED ITS WASTE

<u>Component</u>	<u>Composition, wt%</u>	
	<u>ITS-1*</u>	<u>ITS-2**</u>
NaNO_3	34.0	38.0
Na_2CO_3	22.0	13.8
NaNO_2	18.6	19.9
NaOH	12.4	1.8
CsNO_3	4.1	3.6
NaAlO_2	3.4	14.6
$\text{Sr}(\text{NO}_3)_2$	2.1	1.8
SrCO_3	2.1	1.8
NaH_2PO_4	0.7	1.3
Na_2SO_4	0.3	1.6
NaCl	0.3	0.9

* In-Tank Solidified Waste Type #1.

** In-Tank Solidified Waste Type #2.

material. The amounts of inert and radioactive cesium and strontium in actual ITS wastes are very low. To facilitate determination of leach rates additional strontium and cesium were added to the synthetic ITS wastes.

Other Reagents

Technical grade (55% Fe) Fe_2O_3 and technical grade (98% Si) silicon metal powder were both obtained from City Chemical Corporation. Reagent grade Fe_2O_3 used in a few tests reacted more vigorously with silicon metal than did the technical grade material. In a few experiments technical grade (57% Mn) MnO_2 (City Chemical Corporation) was substituted for the Fe_2O_3 .

So-called "Hanford sand" was obtained from the J. A. Jones Company Batch Cement plant located near the 200 West Area. This material, produced by simple screening of gravel, is used as a source of sand in making cement. The Hanford sand, consisting primarily of 10 to 100 mesh size particles, was used as received from the Batch Cement plant.

PROCEDURES

Thermite Charge Preparation and Ignition

Thermite process charges were prepared by thorough mixing of the desired proportions of the dry ingredients--ITS waste, Hanford sand, silicon metal, and, in most cases, Fe_2O_3 . [The Fe_2O_3 was omitted in a few tests, and MnO_2 was substituted for it in several others.] The mixed charge (2.5 kg in each experiment) was poured into a Dixon No. 14 graphite-clay crucible (Dixon Crucible Company).

Thermite mixtures were all ignited outside the 222-S

Building with the lower seven inches of the 9-inch tall crucible buried in sand. An insulated piece of No. 14 nichrome wire connected to a variable transformer was inserted into the mixture (Figure 3). About one gram of Thermet Starting Mixture (a proprietary formulation of the Thermex Metallurgical Corporation) was sprinkled around the wire. This starting mixture ignited when enough current was applied to the wire to turn it bright yellow. The heat released from combustion of the Thermet Starting Mixture ignited the thermite charge. Burning began at the top of the mixture and proceeded downward until all the charge had reacted; complete reaction required 1.5 to 6 minutes depending on the ratio of ingredients. Typical views of the reacting mixture are shown in Figure 4.

The solidified product, after cooling to about 25 °C, was easily removed from the graphite crucible by simply inverting the crucible. [The crucibles appeared to be unaffected by the high temperature of the thermite reaction and were each used many times.] In some cases the volume of the bulk product was determined by water displacement. In all cases the product was broken into chunks to facilitate physical separation of silicate material from large pieces of iron metal.

Off-Gas Composition

Exact composition and amount of the gas evolved when the Hanford Thermite process is applied to ITS waste have not yet been determined. In all cases discussed here gray-white smoke evolved throughout the thermite reaction (Figure 4); this smoke is thought to consist of particulate SiO_2 and Na_2O . Nitrogen oxides were also given off during the reaction.

FIGURE 3
EXPERIMENT ARRANGEMENT FOR IGNITION OF THERMITE PROCESS CHARGES

FIGURE 4

APPLICATION OF HANFORD THERMITE PROCESS TO ITS WASTE--
TYPICAL REACTIONS

In a few crude measurements the evolved gas was passed through an 8-foot long section of 4 3/4-inch diameter 304-L stainless steel pipe containing plugs of glass wool at the 4, 6, and 8-foot marks. Subsequently, the pipe and glass wool filters were leached with water and 1M HNO₃ and the leachates analyzed (by atomic absorption methods) for cesium and strontium.

Product Quality Tests

Several properties of the thermite reactions and products were measured to evaluate effects of changes in reactant proportions. These included:

1. Reaction Rate. Reaction rate, estimated by personal observation, was recorded on a scale of 1 to 5 corresponding to reactions which were judged to be, respectively: violent, vigorous, moderate, slow, and quite slow.
2. Time to Complete Reaction. The time necessary for the reaction to complete itself, another indication of reaction rate, was recorded. For a 2.5 kg charge, a violent reaction (Type 5) generally took less than 2 minutes to complete, while the slowest reaction (Type 1) took 7 minutes or more.
3. Appearance of Silicate Product. By inspection the silicate product was judged as either glassy or crystalline; X-ray diffraction was used to decide doubtful cases. Porosity of the product was also estimated qualitatively.
4. Density of Silicate Product. Product density was calculated from the volume of water displaced by a weighed sample.

5. Leach Rates. A jaw-type rock crusher was used to break Hanford Thermite process silicate products into small pieces. The crushed material was screened, and the 14 to 20 mesh (U.S. Standard Sieve Series) fraction taken for leach tests. This fraction was rinsed with distilled water and then dried in air. Of particular significance is the fact that no effort was made to remove particles of iron metal from the silicate product either before or after crushing. Hence, we believe that the crushed material used in the leach tests was representative of the massive silicate product produced in the thermite reaction. Pictured in Figure 5 are a lump of typical silicate product before crushing and the 14 to 20 mesh particles used in leach tests.

Leach tests were performed with 15 to 25 grams of the dried 14 to 20 mesh material which was assumed to consist of 0.11-cm diameter spheres. [The value of 0.11 cm is the average of the openings (0.14 and 0.084 cm, respectively) of 14 and 20 mesh screens]. Total surface area of the weighed leach samples was estimated from the weight and surface area of a counted number of the 0.11-cm diameter pieces.

Pyrex vessels (Figure 6), similar to those described by Paige,^[6] were used in leach tests; the test material was supported on a stainless steel screen. Airlift circulators were used to circulate 200 ml of distilled and deionized water over the sample pieces. Test samples were leached initially for 24 hours at 25 °C and, then after changing out the leach liquor, for 96 more hours at 25 °C. Final leach solutions were analyzed (atomic absorption) for Fe, Na, Si, Ca, Mg, Al, Sr, and Cs. Bulk leach rates

Top: Lump of silicate product produced by Hanford
Thermite process.
Bottom: 14 to 20 mesh particles of silicate product used
in leach tests (10X).

FIGURE 5
IMMOBILIZED ITS WASTE SPECIMENS

FIGURE 6
LEACH TEST APPARATUS

were calculated from the total weight of elements leached into solution per day. Specific leach rates were also determined for cesium and strontium.

The procedure just described provided a rapid and convenient way of determining and comparing leach rates of the various Thermite process product. However, leach rates determined by this procedure are uncertain to the extent of the errors involved in estimating the surface area of the test pieces. The assumption that the test pieces are spherical leads to calculation of minimum surface area; hence, leach rates quoted in this paper are believed maximum values.

6. Thermal Conductivity. The "hot wire" procedure described by Underwood and McTaggart^[6] was used to measure the thermal conductivity of representative Hanford Thermite process products at 25, 110, and 300 °C. Bulk products were sectioned (diamond saw) into two pieces; a rectangular groove (1/8-inch wide by 1/8-inch deep) was sawed in one face to contain the hot wire. The two halves, held together by copper wire, were wrapped in glass wool, placed in an appropriate heating mantle, and heated to the desired temperature.

Statistically Designed Experiments

Statistically designed experiments were performed to establish effects of the various Hanford Thermite process ingredients on product properties and to determine optimum proportions of the reaction ingredients.

Following preliminary experimentation, the following levels were chosen for the three independent variables in the statistically designed experiments with ITS-1 waste:

Silicon metal: 13, 16, and 19 wt%
Hanford sand: 0, 10, and 20 wt%
ITS-1 waste: 12, 20, 28, and 40 wt%

The balance of the mixture in each test was Fe_2O_3 . For a full factorial design (2 variables at 3 levels and 1 variable at 4 levels) a total of ($3^2 \cdot 4$) or 36 experiments were performed. A few additional tests were also performed to establish experimental error.

Levels of the independent variables in the statistically designed experiment with ITS-2 waste were as follows:

Hanford sand: 0, 10, and 20 wt%
ITS-2 waste: 14, 22, 30, and 40 wt%

In all tests the amount of silicon metal was held constant at 16 wt% and Fe_2O_3 made up the balance of the charge. A total of 12 experiments corresponding to a full factorial design ($3 \cdot 4$) were executed.

For each test the response variables were reaction violence, reaction time, product density, and product (both bulk and specific ion) leach rates.

RESULTS AND DISCUSSION

STATISTICALLY DESIGNED EXPERIMENTS

Primary Findings

Conditions and principal results of the statistically designed experiments performed with ITS-2 and ITS-1 wastes are listed in Tables II and III, respectively.* Standard regression analysis techniques were used to interpret these data and to generate the equation of the response curve for each of the

* Leach rate data for cesium and strontium for these experiments are listed in Tables A-1 and A-2 in the Appendix.

TABLE II
RESULTS OF THERMITE REACTIONS
ITS-2 WASTE

Sl	Ingredients (wt%)			Reaction Rate* 1-5	Reaction Time min	Product Density g/cm ³	General Appearance	Bulk Leach Rate	
	Waste	Sand	Fe ₂ O ₃					g/cm ² day	
								24 hr	96 hr
16	22	10	52	3	4.8	2.6	Glassy, nonporous	1.3 x 10 ⁻⁶	1.4 x 10 ⁻⁷
16	14	0	70	3	3.7	3.2	Crystalline and glassy mix	3.4 x 10 ⁻⁶	8.5 x 10 ⁻⁷
16	30	0	54	4	3.9	2.8	Glassy, small pores	7.3 x 10 ⁻⁶	1.2 x 10 ⁻⁶
16	30	20	34	3	4.5	1.8	Glassy, small pores	5.2 x 10 ⁻⁶	1.6 x 10 ⁻⁶
16	14	20	50	1	6.5	1.9	Glassy, porous	5.2 x 10 ⁻⁶	3.7 x 10 ⁻⁷
16	22	20	42	2	5.7	1.7	Glassy, small pores	4.7 x 10 ⁻⁶	2.2 x 10 ⁻⁶
16	22	0	62	4	4.0	2.8	Glassy, nonporous	6.4 x 10 ⁻⁶	1.1 x 10 ⁻⁶
16	30	10	44	3	3.0	2.6	Glassy, nonporous	3.6 x 10 ⁻⁶	8.5 x 10 ⁻⁷
16	14	10	60	2	4.6	2.5	Glassy, nonporous	2.0 x 10 ⁻⁶	4.0 x 10 ⁻⁷
16	40	0	44	5	2.2	2.7	Glassy, nonporous	6.9 x 10 ⁻⁶	1.6 x 10 ⁻⁶
16	40	10	34	4	3.9	2.4	Glassy, some pores	2.3 x 10 ⁻⁶	7.1 x 10 ⁻⁷
16	40	20	24	3	4.7	1.9	Glassy, porous	1.5 x 10 ⁻⁵	3.8 x 10 ⁻⁶

* Reaction rate defined on page 12.

TABLE III
RESULTS OF THERMITE REACTIONS
ITS-1 WASTE

Si	Ingredients (wt%)			Reaction Rate* 1-5	Reaction Time min	Product Density g/cm ³	General Appearance	Bulk Leach Rate g/cm ² day	
	Waste	Sand	Fe ₂ O ₃					24 hr	96 hr
19	28	10	43	4	2.7	2.7	Glassy, nonporous	1.1 x 10 ⁻⁵	1.6 x 10 ⁻⁶
13	20	0	67	2	5.3	2.9	Glassy, nonporous	2.0 x 10 ⁻⁶	3.0 x 10 ⁻⁷
16	20	10	54	3	4.4	2.8	Glassy, nonporous	3.7 x 10 ⁻⁶	1.5 x 10 ⁻⁶
				3	4.2	2.6	Glassy, nonporous	7.3 x 10 ⁻⁶	8.8 x 10 ⁻⁷
19	12	0	69	4	2.8	3.0	Glassy, porous	3.3 x 10 ⁻⁵	5.4 x 10 ⁻⁶
13	28	0	59	3	5.0	3.0	Glassy, nonporous	2.2 x 10 ⁻⁵	9.4 x 10 ⁻⁶
19	12	20	49	2	4.8	1.9	Glassy, small pores	7.0 x 10 ⁻⁷	1.5 x 10 ⁻⁷
19	12	10	59	4	3.2	1.7	Glassy, small pores	1.2 x 10 ⁻⁶	1.6 x 10 ⁻⁷
13	28	20	39	2	6.7	2.8	Glassy, nonporous	9.9 x 10 ⁻⁶	2.2 x 10 ⁻⁶
16	20	0	64	4	3.3	3.1	Glassy, nonporous	4.0 x 10 ⁻⁶	2.4 x 10 ⁻⁶
				4	3.6	3.0	Glassy, nonporous	5.8 x 10 ⁻⁶	2.7 x 10 ⁻⁶
13	12	10	65	2	5.7	3.0	Glassy, some pores	6.8 x 10 ⁻⁶	2.7 x 10 ⁻⁶
13	20	10	57	2	4.5	2.9	Glassy, nonporous	1.2 x 10 ⁻⁵	3.3 x 10 ⁻⁶
16	12	0	72	3	2.6	3.1	Crystalline, nonporous	2.0 x 10 ⁻⁶	4.6 x 10 ⁻⁷
19	20	10	51	4	2.1	2.7	Glassy, nonporous	1.4 x 10 ⁻⁶	4.0 x 10 ⁻⁷
19	28	0	53	5	1.6	3.1	Glassy, nonporous	2.0 x 10 ⁻⁵	6.2 x 10 ⁻⁶
16	20	20	44	3	3.6	2.4	Glassy, small pores	1.2 x 10 ⁻⁶	1.3 x 10 ⁻⁷
13	12	20	55	1	5.0	2.9	Glassy, porous	5.0 x 10 ⁻⁶	9.5 x 10 ⁻⁶
16	12	20	52	2	5.0	2.2	Glassy, nonporous	1.6 x 10 ⁻⁶	1.3 x 10 ⁻⁷
16	28	0	56	4	3.2	3.2	Glassy, nonporous	3.7 x 10 ⁻⁵	1.1 x 10 ⁻⁵
13	20	20	47	2	5.7	2.9	Glassy, nonporous	4.3 x 10 ⁻⁶	1.2 x 10 ⁻⁶
19	28	20	33	4	2.3	1.9	Glassy, small pores	2.3 x 10 ⁻⁵	4.8 x 10 ⁻⁶
19	20	20	41	3	4.8	1.7	Glassy, small pores	3.3 x 10 ⁻⁶	8.4 x 10 ⁻⁷
16	28	20	36	3	4.8	2.4	Glassy, nonporous	4.4 x 10 ⁻⁶	1.2 x 10 ⁻⁶
16	12	10	62	2	4.5	2.9	Glassy, nonporous	2.3 x 10 ⁻⁶	3.8 x 10 ⁻⁷
13	12	0	75	3	4.3	3.1	Crystalline, some pores	1.4 x 10 ⁻⁵	2.7 x 10 ⁻⁶
16	28	10	46	4	4.0	2.6	Glassy, nonporous	1.4 x 10 ⁻⁵	2.0 x 10 ⁻⁶
13	28	10	49	3	6.0	3.0	Glassy, some pores	3.2 x 10 ⁻⁵	5.8 x 10 ⁻⁶
19	20	0	61	4	2.8	2.9	Glassy, nonporous	2.5 x 10 ⁻⁵	1.7 x 10 ⁻⁶
13	40	0	47	3	4.5	2.6	Crystalline, glass mix, porous	3.3 x 10 ⁻⁴	1.0 x 10 ⁻⁴
13	40	10	37	4	4.5	2.4	Glassy, nonporous	3.2 x 10 ⁻⁵	5.8 x 10 ⁻⁶
13	40	20	27	4	4.5	2.7	Glassy, nonporous	2.4 x 10 ⁻⁵	7.0 x 10 ⁻⁶
16	40	0	44	5	2.6	2.7	Dull glass, some pores	5.4 x 10 ⁻⁵	8.3 x 10 ⁻⁶
16	40	10	34	4	2.9	2.6	Glassy, nonporous	2.3 x 10 ⁻⁵	1.1 x 10 ⁻⁵
16	40	20	24	4	3.6	2.3	Glassy, some pores	4.4 x 10 ⁻⁵	1.8 x 10 ⁻⁵
19	40	20	21	4	3.7	2.4	Glassy, some pores	7.1 x 10 ⁻⁵	2.3 x 10 ⁻⁵

* Reaction rate defined on page 12.

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several test variables. [Typical response curves are plotted in the Appendix.] Significant findings are highlighted here; additional details of these experiments are discussed subsequently.

1. Optimum Reaction Mix. Most efficient immobilization of ITS waste by the Hanford Thermite process is realized when the initial reaction charge contains 30 to 40 wt% ITS waste, 15 to 20 wt% Si, 20 to 35 wt% Fe_2O_3^* , and 10 to 25 wt% Hanford sand. These conditions permit a controlled reaction and yield a dense product of minimum leachability.

2. Silicate Product Leach Rates. Leach rates of silicate products made from ITS-1 waste are lower than those of silicates produced from ITS-2 waste. In both cases, leach rates increase as the amount of waste in the initial thermite charge increases. Also for ITS-1 waste, leach rates decrease as the amount of either silicon metal or sand in the charge increases.

3. Reaction Rate. The rate of the thermite reaction increases when the amount of either silicon metal or ITS waste in the charge increases. Conversely, the reaction becomes less rapid as the charge is diluted with Hanford sand.

4. Reaction Time. The time to complete the thermite reaction increases as more Hanford sand is added to the charge but decreases as the amount of silicon metal in the charge increases. Hence, reaction time is essentially the converse of reaction rate.

* Other results (see Page 26) indicate satisfactory silicate products are obtained with charges containing as little as 15 wt% Fe_2O_3 .

5. Product Density. Increased dilution of the thermite charge with Hanford sand results in decreased density of the silicate product.

Detailed Results

Leach Rates. For the conditions listed in Tables II and III bulk leach rates (96 hr) of immobilized ITS-1 and ITS-2 waste range from 1.3×10^{-7} to 10^{-4} and from 1.4×10^{-7} to 3.8×10^{-6} g/cm² day, respectively. Specific leach rates for cesium and strontium vary proportionately with bulk leach rates and for immobilized ITS-1 and ITS-2 waste range from $<10^{-9}$ to 1.4×10^{-6} and from $<2.0 \times 10^{-9}$ to 3.6×10^{-5} g/cm² day, respectively. Because steady-state may not have been reached after only 96 hr leaching, it is expected that the leach rate values listed in Tables II and III may be somewhat higher than equilibrium values. Products obtained with the flowsheet conditions shown in Figure 1 are expected to have a bulk leach rate of about 10^{-7} to 10^{-5} g/cm² day. In comparison, solidified high-level Purex waste made by various calcination procedures or by the Phosphate Glass process leaches at rates of 10^{-6} to 1.0 g/cm² day.^[7]

Bulk leach rates of both immobilized ITS-1 and ITS-2 waste increase as the amount of waste in the initial thermite charge increases. However, as evident from response curves plotted in the Appendix (Figures A-1 and A-2), the effect is much more pronounced for ITS-1 waste than for ITS-2 material. Compared with ITS-2 waste, ITS-1 waste contains about 7 times as much NaOH and only about one-fourth as much NaAlO₂. Although exact mechanisms are not understood, this difference in composition is sufficient, apparently, to increase water solubility of immobilized ITS-1 waste. In any event, flowsheet conditions given in Figure 1 are

flexible enough to immobilize satisfactorily ITS wastes of widely varying compositions.

Bulk and specific leach rates for both immobilized ITS-1 and ITS-2 waste determined after only 24 hours' leaching were higher than those measured after 96 hours' leaching. This behavior is consistent with the experience of other investigators who observed leach rates of various solidified Purex waste forms to decrease with leaching time.^[8,9]

Appearance of Silicate Product. With both ITS-1 and ITS-2 wastes, crystalline products were produced only with reaction charges containing 0 to 10% Hanford sand and 12 to 14% waste. [In accordance with Reaction 1 the crystalline products were identified as fayalite by X-ray diffraction analysis.] Glassy products obtained in all other cases were gray-green to black in color; thin sections of the black glass were translucent and appeared emerald-green in bright light.

Except for the upper crust which was slightly porous most silicate products were quite dense and nonporous. For charges containing either low amounts of waste or low amounts of silicon combined with a large amount of Hanford sand, product porosity increased with decreased thermite reaction rate. Generally, products of acceptable low porosity were produced from moderately vigorous (Type 3) reactions.

Metallic Iron By-Product. Iron metal is a by-product of the Hanford thermite process when Fe_2O_3 is used as the oxidizer. The iron metal produced in many of the tests with ITS waste was dispersed as tiny spheres in the fayalite matrix. In other tests, particularly the more vigorous

reactions, the iron coalesced and collected at the bottom of the silicate glass as a large, massive slug. The weight of iron produced varied considerably from test to test depending upon the amount of Fe_2O_3 used.

Judging from several analyses of the iron by-product from typical Thermite process tests, 0.05% or less of the strontium and cesium in the ITS waste charge report to the metal phase. Tests with actual ITS waste will enable accurate determination of the distribution of ^{137}Cs , ^{90}Sr , and other radionuclides, *e.g.*, ^{106}Ru and ^{239}Pu , between metal and silicate phases.

Product Density. Density of silicate products containing ITS-1 waste was influenced by a strong interaction between contents of silicon metal and Hanford sand in the thermite charge. High-Si-low-sand mixes produced dense products while low-density silicates were obtained with high-Si-high-sand charges. Similar effects were noted for the density of products made from ITS-2 waste. With this waste, high-sand mixes produced silicates of uniformly low density with no apparent effect from the amount of waste in the charge.

Reaction Rate and Time. That reaction rate should vary directly with the amount of silicon and/or ITS waste in the thermite charge follows from the Law of Mass Action. [The decrease in reaction rate associated with dilution of the charge with sand is also accounted for on the same basis.] Nitrate in the ITS waste, along with the added Fe_2O_3 , oxidizes the silicon metal fuel.

OTHER EXPERIMENTS AND OBSERVATIONS

In addition to the principal concern with the statistically designed experiments miscellaneous other tests and

observations related to application of the Hanford Thermite process to ITS waste have been made. Data from these latter tests are discussed here.

Thermal Conductivity of Silicate Products

The thermal conductivity of actual ITS waste at 25 to 30 °C is typically 0.7 to 0.8 watts/meter °C.^[10] Immobilized ITS waste produced by the Hanford Thermite process has a thermal conductivity in about the same range (Table IV). For both materials thermal conductivity increases with temperature. As noted in Figure 1, immobilization of ITS waste by the Hanford Thermite process does not change its volume.

TABLE IV
THERMAL CONDUCTIVITY OF SILICATE PRODUCT

Waste	Composition of Initial Charge, wt%			Thermal Conductivity watts/meter °C at		
	Hanford			25 °C	110 °C	300 °C
	Sand	Fe ₂ O ₃	Si			
28 ^a	20	38	16	0.74	0.76	-
22 ^b	20	42	16	0.47	0.64	0.76
20 ^b	10	44	16	0.61	0.72	0.80

^a ITS-1; *cf.*, Table I.

^b ITS-2; *cf.*, Table I.

Thermal conductivity data for solidified Purex process wastes, resulting from the Pot Calcination, Fluidized Bed Calcination, Spray Melt, and Phosphate Glass processes have been cited by Schneider.^[11] Thermal conductivity of immobilized ITS waste exceeds that of solids produced by the former two methods and is at the lower limit of the conductivity of waste resulting from the Spray Melt and Phosphate Glass processes.

Volatilization of Cesium and Strontium

According to limited off-gas analyses (see page 12) only 2 to 5% of the cesium and <0.5% of the strontium in the ITS waste volatilize during the thermite reaction. Verification of these results is provided by analyses of the final silicate products which account for 95 to 98% of the cesium in the charge of ITS waste. Definitive information on the amounts of ^{137}Cs and other radionuclides, including ^{106}Ru , in the off-gas from the Hanford Thermite processes will be provided from tests now in progress with actual ITS waste.

Substitution of MnO_2 for Fe_2O_3

Judging from limited testing, substitution of MnO_2 for Fe_2O_3 in the thermite charge offers several process advantages. Foremost of these is that little metallic by-product is produced even with high amounts of MnO_2 in the charge. Leach rates of silicate products made from charges containing MnO_2 are also slightly lower than those of glasses made from charges containing an equivalent amount of Fe_2O_3 (Table V). Technical grade MnO_2 is also less expensive than technical grade Fe_2O_3 (\$0.35/pound *vice* \$0.57/pound).

Hanford Thermite process products resulting from charges containing 15 to 40 wt% MnO_2 are very similar in appearance to those made from charges containing 15 wt% Fe_2O_3 (compare Figures 7a and 2b). Comparison of Figures 7a and 2b with Figure 2a illustrates how much more metallic iron is produced from charges containing greater than 20 wt% Fe_2O_3 .

Data in Table V also show that leach rates of glasses made from ITS-1 waste are higher than those of material made from ITS-2 waste. This phenomenon and its explanation were commented on earlier (page 21).

FIGURE 7
HANFORD THERMITE PROCESS PRODUCTS

TABLE V
SUBSTITUTION OF MnO_2 FOR Fe_2O_3 IN HANFORD
THERMITE PROCESS

Charge Composition, wt%					Product Leach Rates, g/cm ² /day					
Sl	ITS				Bulk		Strontium		Cesium	
	Waste	MnO	Fe ₂ O ₃	Sand	24 hr	96 hr	24 hr	96 hr	24 hr	96 hr
18	40 ^a	0	15	27	6.3×10^{-4}	2.7×10^{-4}	1.1×10^{-4}	5.0×10^{-5}	2.7×10^{-4}	1.4×10^{-4}
18	40 ^b	15	0	27	2.8×10^{-4}	8.9×10^{-5}	9.0×10^{-5}	2.7×10^{-5}	1.4×10^{-4}	3.5×10^{-5}
18	40	0	15	27	1.4×10^{-4}	3.5×10^{-5}	5.8×10^{-5}	1.4×10^{-5}	3.3×10^{-5}	8.3×10^{-6}
18	40 ^c	15	0	27	1.1×10^{-4}	2.5×10^{-5}	2.8×10^{-5}	7.0×10^{-6}	6.1×10^{-5}	1.1×10^{-5}
16	25	25	0	34	4.0×10^{-4}	9.5×10^{-5}	3.0×10^{-5}	1.0×10^{-5}	2.9×10^{-5}	1.0×10^{-5}
16	30 ^d	20	0	34	7.5×10^{-4}	1.3×10^{-4}	3.4×10^{-5}	3.3×10^{-6}	1.3×10^{-5}	6.0×10^{-6}
19	40 ^e	18	0	23	1.8×10^{-4}	4.0×10^{-5}	ND ^f	ND ^f	ND ^f	ND ^f

^a ITS-1; see Table III.

^b ITS-2; see Table II.

^c ND = not determined.

Elimination of Fe_2O_3 and MnO_2 from Thermite Charge

In several process tests neither Fe_2O_3 nor MnO_2 were added to the thermite charge; charges for these tests contained 50 wt% ITS waste, 34 wt% Hanford sand, and 16 wt% silicon metal. The resulting silicate product in each case was typically a nonporous, dense (2.4 g/cm^3), gray-colored glass (Figure 7b) containing little, if any, metallic components. Unfortunately, the bulk leach rate (2.6 to $5.8 \times 10^{-4} \text{ g/cm}^2 \text{ day}$) of this glass was 10 to 1000 times higher than that of silicates obtained with Fe_2O_3 or MnO_2 in the reaction mixtures.

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APPENDIX

APPENDIX

Standard regression analysis techniques were used to fit an equation of the type

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_1^2 + b_5X_1^2 + b_6X_3^2 + b_7X_1X_2 + b_8X_1X_3 + b_9X_2X_3 + b_{10}X_1X_2X_3$$

for each of the response variables (*e.g.*, leach rate, density, etc.) of the statistically designed experiments described in Tables II and III. In these equations the "b's" are numerical coefficients, while X_1 , X_2 , and X_3 , are, respectively, the percentages of silicon, ITS waste, and Hanford sand in the various thermite mixes. Plots of these equations of response curves are shown in Figures A-1 through A-8.

To facilitate their handling, bulk leach rates and specific cesium and strontium leach rates (both 24- and 96-hour data) were expressed in natural logarithmic form. [In each case the exponential term was discarded prior to taking the logarithm.] The average of the logarithms of the six leach rates available at each thermite charge composition is called the average $\ln$ leach rate and is shown as the response variable in Figures A-1 and A-5.

Tables A-1 and A-2 list leach rates for cesium and strontium from silicate products produced in the statistically designed experiments with ITS-1 and ITS-2 wastes, respectively.

TABLE A-1
STRONTIUM AND CESIUM LEACH RATES
ITS-2 WASTE

Si	Waste	Sand	Fe ₂ O ₃	Strontium Leach Rate		Cesium Leach Rate	
				g/cm ² day		g/cm ² day	
				24 hr	96 hr	24 hr	96 hr
16	22	10	52	1.6 x 10 ⁻⁸	<4.3 x 10 ⁻⁹	<1.6 x 10 ⁻⁸	<4.3 x 10 ⁻⁹
16	14	0	70	3.2 x 10 ⁻⁸	<5.9 x 10 ⁻⁹	3.2 x 10 ⁻⁸	<4.9 x 10 ⁻⁹
16	30	0	54	8.9 x 10 ⁻⁸	1.9 x 10 ⁻⁸	3.9 x 10 ⁻⁸	7.1 x 10 ⁻⁹
16	30	20	34	1.3 x 10 ⁻⁸	3.3 x 10 ⁻⁹	1.3 x 10 ⁻⁸	<3.3 x 10 ⁻⁹
16	14	20	50	<9.5 x 10 ⁻⁹	<2.4 x 10 ⁻⁹	9.5 x 10 ⁻⁹	2.4 x 10 ⁻⁹
16	22	20	42	2.5 x 10 ⁻⁸	4.9 x 10 ⁻⁹	9.0 x 10 ⁻⁹	4.8 x 10 ⁻⁹
16	22	0	62	3.6 x 10 ⁻⁸	4.5 x 10 ⁻⁹	3.9 x 10 ⁻⁸	1.4 x 10 ⁻⁸
16	30	10	44	<1.1 x 10 ⁻⁸	<2.8 x 10 ⁻⁹	1.1 x 10 ⁻⁸	<2.8 x 10 ⁻⁹
16	14	10	60	1.3 x 10 ⁻⁸	<3.3 x 10 ⁻⁹	1.3 x 10 ⁻⁸	<3.3 x 10 ⁻⁹
16	40	0	44	2.4 x 10 ⁻⁸	6.0 x 10 ⁻⁹	<2.4 x 10 ⁻⁹	<6.0 x 10 ⁻⁹
16	40	10	34	1.9 x 10 ⁻⁸	3.3 x 10 ⁻⁹	1.9 x 10 ⁻⁸	<3.3 x 10 ⁻⁹
16	40	20	24	2.1 x 10 ⁻⁸	5.3 x 10 ⁻⁹	<2.1 x 10 ⁻⁸	7.5 x 10 ⁻⁹

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TABLE A-II
STRONTIUM AND CESIUM LEACH RATES
ITS-1 WASTE

Si	Ingredient (wt%)			Strontium Leach Rate		Cesium Leach Rate	
	Waste	Sand	Fe ₂ O ₃	g/cm ² day		g/cm ² day	
				24 hr	96 hr	24 hr	96 hr
19	28	10	43	4.0 x 10 ⁻⁸	1.0 x 10 ⁻⁸	2.3 x 10 ⁻⁸	<6.0 x 10 ⁻⁹
13	20	0	67	<6.0 x 10 ⁻⁸	<1.3 x 10 ⁻⁸	<3.0 x 10 ⁻⁸	9.0 x 10 ⁻⁹
16	20	10	54	1.3 x 10 ⁻⁸	3.0 x 10 ⁻⁹	3.2 x 10 ⁻⁸	1.3 x 10 ⁻⁸
				2.0 x 10 ⁻⁸	7.2 x 10 ⁻⁹	3.2 x 10 ⁻⁸	6.5 x 10 ⁻⁹
19	12	0	69	5.3 x 10 ⁻⁸	1.1 x 10 ⁻⁸	8.9 x 10 ⁻⁸	8.0 x 10 ⁻⁸
13	28	0	59	1.4 x 10 ⁻⁷	6.6 x 10 ⁻⁸	2.5 x 10 ⁻⁷	1.5 x 10 ⁻⁷
19	12	20	49	1.0 x 10 ⁻⁸	3.0 x 10 ⁻⁹	<1.0 x 10 ⁻⁸	<3.0 x 10 ⁻⁹
19	12	10	59	2.0 x 10 ⁻⁸	2.9 x 10 ⁻⁹	1.2 x 10 ⁻⁸	<3.0 x 10 ⁻⁹
13	28	20	39	1.3 x 10 ⁻⁷	2.4 x 10 ⁻⁸	3.2 x 10 ⁻⁸	<4.0 x 10 ⁻⁹
16	20	0	64	3.9 x 10 ⁻⁸	2.7 x 10 ⁻⁸	5.5 x 10 ⁻⁸	2.9 x 10 ⁻⁸
				2.8 x 10 ⁻⁸	2.5 x 10 ⁻⁸	4.9 x 10 ⁻⁸	3.0 x 10 ⁻⁸
13	12	10	65	3.4 x 10 ⁻⁸	7.2 x 10 ⁻⁹	1.4 x 10 ⁻⁸	3.5 x 10 ⁻⁹
13	20	10	57	5.9 x 10 ⁻⁸	1.7 x 10 ⁻⁸	7.2 x 10 ⁻⁸	2.2 x 10 ⁻⁸
16	12	0	72	<2.0 x 10 ⁻⁸	<5.0 x 10 ⁻⁹	<1.0 x 10 ⁻⁸	<3.0 x 10 ⁻⁹
19	20	10	51	<1.5 x 10 ⁻⁸	<4.0 x 10 ⁻⁹	<9.0 x 10 ⁻⁹	<3.0 x 10 ⁻⁹
19	28	0	53	6.0 x 10 ⁻⁸	1.3 x 10 ⁻⁸	2.0 x 10 ⁻⁷	7.8 x 10 ⁻⁸
16	20	20	44	<1.3 x 10 ⁻⁸	<3.0 x 10 ⁻⁹	<1.3 x 10 ⁻⁸	<3.0 x 10 ⁻⁹
13	12	20	55	9.6 x 10 ⁻⁹	<2.0 x 10 ⁻⁹	1.1 x 10 ⁻⁸	8.8 x 10 ⁻⁹
16	12	20	52	<1.3 x 10 ⁻⁸	<3.0 x 10 ⁻⁹	<1.3 x 10 ⁻⁸	<3.0 x 10 ⁻⁹
16	28	0	56	9.1 x 10 ⁻⁸	3.7 x 10 ⁻⁸	1.3 x 10 ⁻⁷	6.4 x 10 ⁻⁸
13	20	20	47	1.6 x 10 ⁻⁸	2.0 x 10 ⁻⁸	<1.6 x 10 ⁻⁸	<4.0 x 10 ⁻⁹
19	28	20	33	1.9 x 10 ⁻⁸	4.7 x 10 ⁻⁹	6.2 x 10 ⁻⁸	1.5 x 10 ⁻⁸
19	20	20	41	2.3 x 10 ⁻⁸	<2.4 x 10 ⁻⁹	1.1 x 10 ⁻⁸	<2.8 x 10 ⁻⁹
16	28	20	36	1.9 x 10 ⁻⁸	<2.2 x 10 ⁻⁹	2.9 x 10 ⁻⁸	2.5 x 10 ⁻⁹
16	12	10	62	1.0 x 10 ⁻⁸	2.5 x 10 ⁻⁹	1.2 x 10 ⁻⁸	<2.1 x 10 ⁻⁹
13	12	0	75	8.0 x 10 ⁻⁸	2.0 x 10 ⁻⁸	9.8 x 10 ⁻⁸	3.4 x 10 ⁻⁸
16	28	10	46	5.3 x 10 ⁻⁸	5.8 x 10 ⁻⁹	5.3 x 10 ⁻⁸	1.3 x 10 ⁻⁸
13	28	10	49	1.1 x 10 ⁻⁷	2.1 x 10 ⁻⁸	1.8 x 10 ⁻⁷	6.1 x 10 ⁻⁸
19	20	0	61	4.4 x 10 ⁻⁷	<3.9 x 10 ⁻⁹	2.3 x 10 ⁻⁷	5.8 x 10 ⁻⁹
13	40	0	47	6.2 x 10 ⁻⁷	3.6 x 10 ⁻⁷	3.5 x 10 ⁻⁶	1.4 x 10 ⁻⁶
13	40	10	37	4.7 x 10 ⁻⁸	4.4 x 10 ⁻⁸	2.5 x 10 ⁻⁷	6.3 x 10 ⁻⁸
13	40	20	27	5.0 x 10 ⁻⁸	5.0 x 10 ⁻⁹	2.3 x 10 ⁻⁷	3.2 x 10 ⁻⁸
16	40	0	44	4.7 x 10 ⁻⁸	1.3 x 10 ⁻⁸	1.5 x 10 ⁻⁷	3.1 x 10 ⁻⁸
16	40	10	34	2.4 x 10 ⁻⁸	9.7 x 10 ⁻⁹	6.9 x 10 ⁻⁸	1.5 x 10 ⁻⁸
16	40	20	24	6.1 x 10 ⁻⁸	3.1 x 10 ⁻⁸	7.9 x 10 ⁻⁸	3.8 x 10 ⁻⁸
19	40	20	21	8.1 x 10 ⁻⁸	3.5 x 10 ⁻⁸	1.0 x 10 ⁻⁷	3.4 x 10 ⁻⁸

FIGURE A-1
ITS-1 WASTE--LEACH RATE RESPONSE CURVE

FIGURE A-2
ITS-1 WASTE--DENSITY RESPONSE CURVES

FIGURE A-3
ITS-1 WASTE--REACTION RATE RESPONSE CURVES

FIGURE A-4
ITS-1 WASTE--REACTION TIME RESPONSE CURVES

FIGURE A-5
ITS-2 WASTE--LEACH RATE RESPONSE CURVES

FIGURE A-6
ITS-2 WASTE--DENSITY RESPONSE CURVES

FIGURE A-7
ITS-2 WASTE--REACTION RATE RESPONSE CURVES

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FIGURE A-8
ITS-2 WASTE--REACTION TIME RESPONSE CURVES

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