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ENGINEERING DATA TRANSMITTAL

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TIME/TEMPERATURE TEST RESULTS FOR PFP THERMAL STABILIZATION FURNACES

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Project Hanford Management Contractor for the
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Fluor Hanford Company

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1. INTRODUCTION

The national standard for plutonium storage acceptability (standard DOE-STD-3013-99, generally known as "the 3013 standard") has been revised to clarify the requirement for processes that will produce acceptable storage materials. The 3013 standard (Reference 1) now states that "Oxides shall be stabilized by heating the material in an oxidizing atmosphere to a *Material Temperature* of at least 950°C (1742°F) for ... not less than 2 hours." [italics added for emphasis] The process currently in use for producing stable oxides for storage at the Plutonium Finishing Plant (PFP) heats a furnace atmosphere to 1000°C and holds it there for 2 hours. The temperature of the material being stabilized is not measured directly during this process.

The Plutonium Process Support Laboratories (PPSL) were requested to demonstrate that the process currently in use at PFP is an acceptable method of producing stable plutonium dioxide consistently. A spare furnace identical to the production furnaces was set up and tested under varying conditions with non-radioactive surrogate materials. Reference 2 was issued to guide the testing program.

2. SUMMARY

The process currently in use at the PFP for stabilizing plutonium-bearing powders was shown to heat all the material in the furnace to at least 950°C for at least 2 hours. The current process will work for (1) relatively pure plutonium dioxide, (2) dioxide powders mixed with up to 20 weight percent magnesium oxide, and (3) dioxide powders with up to 11 weight percent magnesium oxide and 20 weight percent magnesium nitrate hexahydrate. Time and temperature data were also consistent with a successful demonstration for a mixture containing 10 weight percent each of sodium and potassium chloride; however, the molten chloride salts destroyed the thermocouples in the powder and temperature data were unavailable for part of that run. These results assume that the current operating limits of no more than 2500 grams per furnace charge and a powder height of no more than 1.5 inches remain in effect, although deeper powder beds (up to 2 inches) also yielded temperatures of greater than 950°C for longer than 2 hours.

3. EQUIPMENT DESCRIPTION

Figure 1 shows the heating and controlling equipment used in these tests. The furnace is a Thermolyne model FA1630-1, which is the same model used in the Thermal Stabilization production areas. The furnace chamber is 17 cm wide, 12.5 cm tall, and 34.8 cm deep (total volume: 7.4 L) with ceramic heating elements on the top, bottom, and both side surfaces. The furnace elements are capable of reaching 1277°C. A ceramic hearth is used on the bottom surface to keep the boat or powder off the bottom element surface per the furnace manufacturer's recommendation. A Hastelloy[1] plate was placed in the rear left corner (as viewed through the furnace door opening) to diffuse the gases flowing through the furnace chamber into the off-gas tubing behind the plate. A ceramic-sheathed thermocouple protruded through the rear of the furnace near the top of the chamber and was used as the temperature input to the furnace heater controller. Another tube penetrated the right side of the furnace rear wall and was used to run thermocouples in through the furnace rear wall to the boat with the powder. This tube was also stuffed with ceramic fiber insulation to hold heat in the furnace chamber and provide friction helping to hold the thermocouples for the powder in position. A ceramic hearth was placed on the bottom heating element's top surface to support the powder boat away from the element. This practice is the same used in the production furnaces at the furnace manufacturer's recommendation.

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[1] Hastelloy is a registered trademark of Haynes International, Inc., Kokomo, Indiana.

FIGURE 1: Furnace and Controlling Equipment

The furnace temperature controller was a Eurotherm model 818, same as used in the Thermal Stabilization production areas and in other applications at the PFP. A load center was installed temporarily in the room to furnish 220-VAC power to the controller and its companion Eurotherm model 831 silicon-controlled rectifier assembly. The controller was programmed the same as the production furnaces with the 4-step heating ramp rate and a 4-hour "dwell" at the operating temperature of 1000°C (only 2 hours at dwell were used).

The furnace boat was a Hastelloy X® open-topped box with a nominal working capacity of 1.42 L. The boat dimensions were 12.9 cm wide by 27.8 cm long with holes at each end for lifting handles that left a working height of 4.0 cm. Total height of the boat was 6.3 cm. A new boat is shown loaded into the furnace in Figure 2. This figure also shows how thermocouples could be run through the edge of the furnace door into the powder or from the furnace rear wall over the back of the boat and into the powder.

The off-gas system is shown in Figure 3. Gases exiting the furnace passed through 1/2-inch stainless steel tubing, then through one of two rotameters before being blown upward to a building exhaust duct at the room ceiling. Normally, the larger rotameter was used and this was an Omega FL-1500A Series with a size 6 glass tube and a glass float. Omega supplied a calibration sheet to allow monitoring the desired 1-1.5 scfm air flow rate on their 0-100 scale. The smaller rotameter was a 0-2 scfh Dwyer Purgerator used for monitoring one run with only a 0.03 scfm (i.e., 2 scfh) air flow rate. A 3-way ball valve at the outlet of each rotameter was used to select the rotameter for each run. A flow control valve (a 1/2-inch ball valve) was left partially open and adjusted to control the rate of flow through the rotameter in use. Motive force for the off-gas flow was supplied by a Gast model R1102 blower with an ultimate flow capacity of 27 scfm. Exhaust from this blower ran through tubing to the top of the room and the entrance to one of the room's E-1 exhaust ducts.

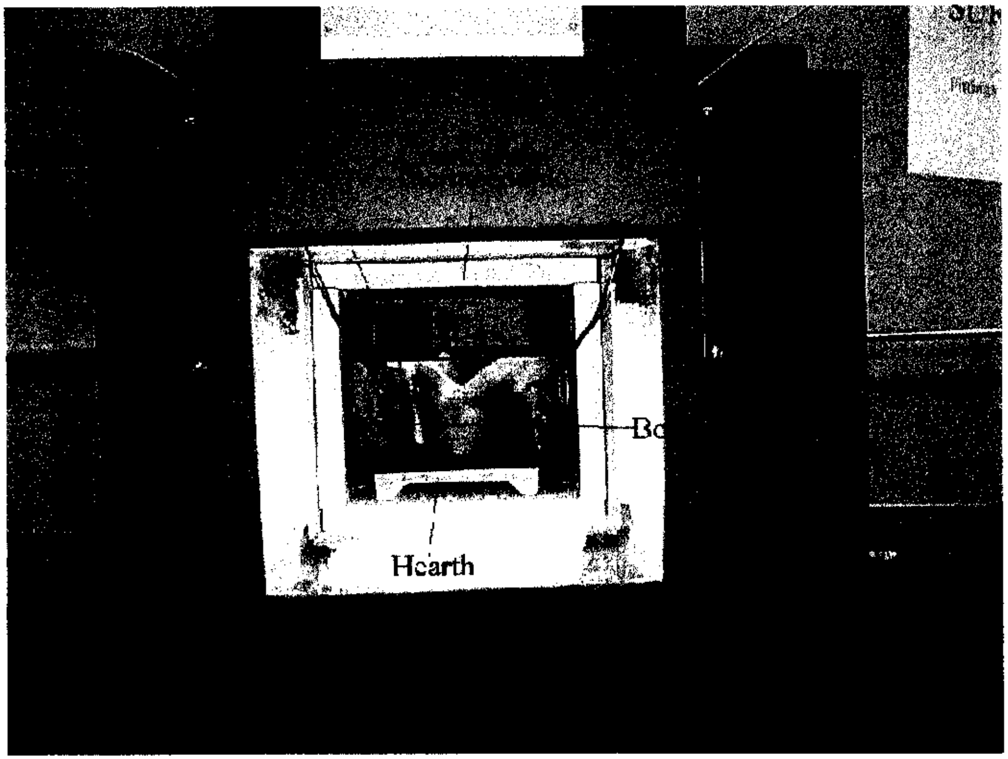

FIGURE 2: Boat Installed in Furnace

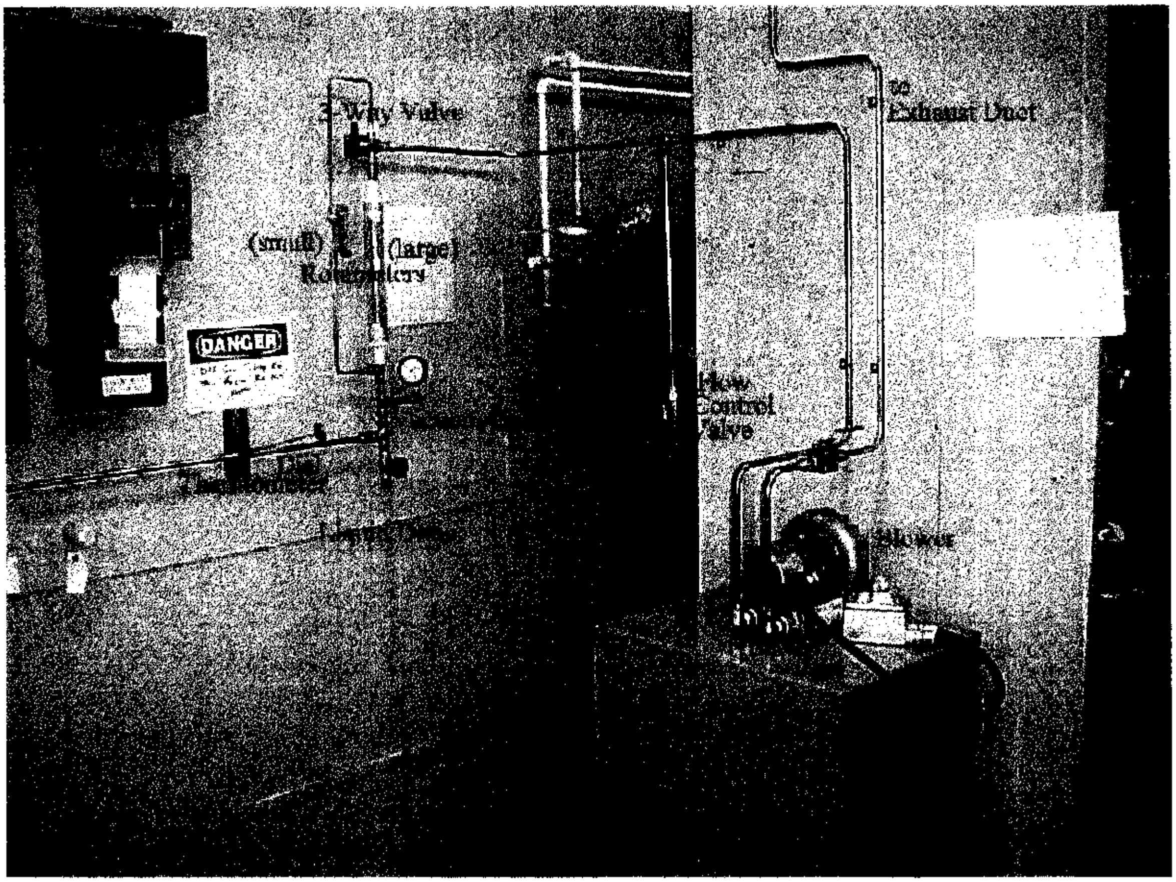

FIGURE 3: Off-Gas System Installation

Temperatures were monitored two places in the off-gas system to inform workers of the off-gas line temperatures as the runs progressed. One type K thermocouple was connected to the temperature indicator and measured the temperature of the line at a spot about 60 cm downstream of the furnace exterior wall. The second indication was a dial thermometer that measured the temperature about 53 cm upstream of the rotameter entrance (about 2 m downstream from the thermocouple). The dial thermometer also confirmed that no temperature corrections needed to be made to the rotameter indications.

A 0-60 inches water vacuum gauge was installed at the base of the rotameters to confirm that no pressure corrections would need to be made to the rotameter indications. The vacuum gauge indicated a vacuum level exceeding 5 inches water only one time when the powder in the boat contained 20 % chloride salts. In that run, the condensed and recrystallized salts were plugging the off-gas line, causing the high vacuum indication.

The powder temperatures in the boat were measured via 1/16-inch-diameter type K thermocouples that were ordered with 24-inch-long Inconel[2] 600 sheaths. This diameter was the smallest available from Omega Engineering, Inc., and the small diameter was needed for maximum flexibility in positioning the thermocouples. The 24-inch lengths were needed to allow penetration from outside the furnace and maximum front-back movement inside the furnace while positioning the thermocouples. Inconel® sheaths were ordered for maximum temperature and corrosion resistance. These thermocouples were connected to an Omega model DP116-KC1-A-MDSS display meter with a manual 10-position selector switch. Three thermocouples were connected for measuring powder temperatures and each run used either 2 or all 3 of those thermocouples. A fifth thermocouple was also attached for measuring the off-gas line temperature as explained two paragraphs above. A calibrated thermocouple signal generator was used to confirm the accuracy of the temperature display at the start of testing.

4. EXPERIMENTAL METHOD

First, the desired blend of powder was selected for addition to the furnace boat. The chemicals were weighed and their amounts recorded in the laboratory notebook. The chemicals were then mixed by hand with a spatula until the mixture appeared uniform to the unaided eye. During the chloride salts run, additional sodium and potassium chloride aliquots were kept separate for use around the thermocouple tips. The powder mixture was then poured into the furnace boat and left with an unsmoothed top surface. The powders poured into the boats in the production process are not smoothed out to reduce radiation exposure.

Next, the boat was loaded into the furnace and the thermocouples were placed into the powder. This step was fairly simple for one worker if the thermocouples were run into the furnace at the front door, either at the top or bottom. This method, though, caused a visible crack at the doorway and allowed additional air in through the furnace front. The additional air here cooled the boat front more than in the production furnaces, so the temperatures in the boat front were lower than they would be in production. Usually, the thermocouples were run through an insulation-stuffed tube in the back of the furnace so that the furnace door would be fully closed, as the production furnace would be, and the temperatures would represent production conditions. Running the thermocouples in at the furnace back required at least a second worker so that the thermocouples away from the front of the boat could be moved back along with the boat as they were positioned and the boat pushed back all the way into the furnace. Modified forceps or similar tools were then used to move the powder around to form mounds above the thermocouple locations and to make certain that the top surface of the powder was far from smooth, as it normally is in the production furnaces.

The furnace heater controller was then started by depressing its power button and tapping the Run/Hold button on the controller face. The furnace was allowed to proceed through its pre-programmed cycle. The programmed cycle warmed the furnace in 4 separate ramping steps to 1000°C and held it at 1000°C for four hours. The 4 separate ramping steps were heating to 500° at a rate of 300°C/hr, then to 700°C at 200°C/hr, then to 900°C at

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[2] Inconel is a registered trademark of Huntington Alloys, Inc., Huntington, West Virginia.

125°C/hr, and finally to 1000°C at 75°C/hr. Although the controller was programmed to hold the furnace at 1000°C for 4 hours, the power to the furnace heaters was always shut off after only 2 hours at 1000°C as this time duration was adequate to show that the powder would be above 950°C for at least those 2 hours (in one run ,the furnace ran about half an hour longer due to an inability to get back to the room at the desired time from another assignment). As soon as the heaters were heating the furnace, the air flow through the furnace was started and a ball valve was adjusted to achieve the desired flow rate of air through the furnace.

After cooling overnight or longer, the furnace was opened to confirm the positions of the thermocouples in the powder. At this time, the boat could be removed from the furnace or a new run could be started with the same powder and thermocouple arrangement. The boat might be removed to (a) change the powder completely, (b) add or subtract some powder, (c) reposition the thermocouples, or (d) any combination of (a)-(c). The boat and thermocouples might be left alone to (1) repeat all the same conditions or (2) test a different air flow rate with the same powder and thermocouple arrangement.

Powders removed from the boat were either kept and stored for reuse or were transferred to the PFP Analytical laboratory for future use in a Loss On Ignition reference powder. After confirming that no future use existed for any kept powders, the PFP Solid Waste Operations organization was contacted to coordinate disposal.

5. EXPERIMENTAL DESIGN

The tests were performed as closely as possible to the current Thermal Stabilization production process except that nonradioactive powders were substituted for plutonium dioxide. The heat-up rates and time spent at maximum operating temperature were the same as used in production. Tests were performed for four combinations of feed materials and impurities. At least one third of the runs were repeated. The powder mixtures used in these tests were:

- Pure ceric oxide (CeO_2 - purity as purchased) to represent pure product and mixed oxides.
- Ceric oxide with 10-20% MgO to match expected magnesium hydroxide precipitation process product and some RMC Line scrap compositions.
- Ceric oxide with 10% NaCl and 10% KCl to simulate Rocky Flats oxide and oxide scrap with volatile salt impurities that will have endothermic reactions during processing.
- Ceric oxide with 10-20% metal nitrates to simulate other scrap feeds with exothermically decomposing salts.

Initial plans to add carbon fines to some powders were dropped after the success of the runs with the other powder mixtures. The carbon fines would only have generated additional heat that might have raised temperatures higher than the already acceptable temperatures, depending on how quickly all the carbon fines oxidized. Most likely, the carbon fines would have oxidized long before the furnace reached normal operating temperature and their oxidation would have had no effect on the time the powders spent above 950°C.

6. ADEQUACY OF SURROGATE POWDERS

These tests were conducted in a non-radioactive laboratory to allow faster setup and easier chemical handling. A non-radioactive surrogate had to be used for the PuO_2 that is stabilized in the Thermal Stabilization furnaces. Ceric oxide [cerium(IV) oxide or CeO_2] was chosen due to its similarity to PuO_2 in properties and behavior as shown in past studies (References 3 and 4). Exact similarity was not necessary for these tests because (1) the powders processed through the Thermal Stabilization furnaces will vary in properties and because (2) "worst case" conditions are preferable in experiments to be certain that the test results will apply in all possible production conditions. The ideal surrogate needs to have a heat capacity at least as high as that of plutonium dioxide and a thermal conductivity at least as low in order to make the surrogate as difficult or more difficult to heat throughout the powder bed; however, the surrogate might still be adequate in these tests if the surrogate heat capacity is greater than that of plutonium dioxide.

The heat capacity of the respective powders was the property most needing to be matched in these experiments. The first requirement for getting a chemical up to a desired temperature is to make certain the required amount of heat reaches the chemical. Conducting this heat to all parts of the chemical is also important, but the slow heating rates used in the Thermal Stabilization furnaces and the relatively thin depth of chemicals to be heated in this boat arrangement make conduction through the powder bed less of a problem here. Perry's Chemical Engineers' Handbook (Reference 5, Table 2-194) supplied the formulae for calculating the heat capacities of the non-radioactive oxide powders considered as surrogates in terms of calories per mole per °C. The molecular weights were then factored into these heat capacities to recalculate them on a weight basis (cal/gm/°C). The bulk densities were then measured in the laboratory for conversion to a volumetric basis (cal/cc/°C). The heat capacities were needed on both weight and volumetric bases because there are both mass and volume loading limits for the Thermal Stabilization production boats. The Handbook of Chemistry and Physics (Reference 6, pages D-64 through D-66) was used for determining the molar heat capacities of radioactive oxides. The calculated heat capacities for all three bases are listed below in Tables 1-3.

TABLE 1: Calculated Oxide Molar Heat Capacities

(heat capacities in calories per mole per degree C)

Temperature (°C)	MgO	CaO	Al_2O_3	CeO_2	ThO_2	UO_2	PuO_2
500	11.4	13.6	28.3	16.9	17.9	19.8	19.3
700	11.8	14.6	29.6	17.4	18.5	20.4	20.1
900	12.1	15.6	30.7	17.9	19.0	20.8	20.9
950	12.2	15.9	30.9	18.1	19.2	20.9	21.1
1000	12.3	16.1	31.2	18.2	19.3	21.0	21.3
1100	12.4	16.6	31.7	18.4	19.6	21.2	21.6

TABLE 2: Calculated Oxide Mass Heat Capacities

(heat capacities in calories per gram per degree C)

Temperature (°C)	MgO (MW = 40.3)	CaO (MW = 56.1)	Al_2O_3 (MW = 102)	CeO_2 (MW = 172)	ThO_2 (MW = 264)	UO_2 (MW = 270)	PuO_2 (MW = 271)
500	0.284	0.242	0.277	0.098	0.0678	0.0733	0.0712
700	0.293	0.260	0.290	0.101	0.0701	0.0754	0.0743
900	0.300	0.278	0.301	0.104	0.0721	0.0771	0.0771
950	0.302	0.283	0.303	0.105	0.0726	0.0775	0.0778
1000	0.304	0.287	0.306	0.106	0.0731	0.0778	0.0785
1100	0.307	0.296	0.311	0.107	0.0741	0.0786	0.0798

TABLE 3: Calculated Oxide Bulk Volumetric Heat Capacities

(heat capacities in calories per cubic centimeter per degree C)

Temperature (°C)	MgO (sp. gr. = 0.34)	Al_2O_3 (sp. gr. = 1.01)	CeO_2 (sp. gr. = 1.84)	PuO_2 (sp. gr. = 2.3*)
500	0.0966	0.280	0.180	0.164
700	0.0996	0.293	0.186	0.171
900	0.102	0.304	0.191	0.177
950	0.103	0.306	0.193	0.179
1000	0.103	0.309	0.195	0.181
1100	0.104	0.314	0.196	0.184

* traditional bulk specific gravity for oxides produced in most precipitation processes

Tables 2 and 3, in particular, show that the ceric oxide used in these tests requires more heat per gram or cc to reach 950°C and beyond than required by plutonium dioxide. Ceric oxide requires just over 4/3 as much heat per gram as plutonium dioxide and 6.5-10 percent more heat per cc. As far as heat capacities of the powders are concerned, the ceric oxide clearly required more heat during the tests than plutonium dioxide would have required. Temperatures in a boat with plutonium dioxide, then, should be no lower than the temperatures measured in these experiments and might be slightly higher.

The thermal conductivity of the powder is important because it is the property that determines if the heat energy can be transferred throughout the powder before the heating cycle is completed. A few data were found for ceric oxide (References 7 and 8) and magnesium oxide thermal conductivities (Reference 5, Table 2-374); however, only one datum pertaining directly to the thermal conductivity of plutonium dioxide could be located (Reference 9). Table 4 compares the thermal conductivity data available for these dioxides. The data for ceric oxide are not in perfect agreement between the 2 references, but they show that ceric oxide is a much better conductor than plutonium dioxide for the temperatures listed. Both conductivities are expected to continue decreasing with increasing temperatures, and in relatively similar patterns, so the thermal conductivity of plutonium dioxide is expected to remain much lower than that of ceric oxide throughout the temperature range of 0-1200°C. Ceric oxide is, then, a questionable surrogate as far as thermal conductivities are concerned. The effect of thermal conductivity, though, may be less than that of heat capacity because of the long heating cycle with slow heating rates used, the heating from 4 sides, and the relatively thin layer of powder in the boat. Regardless of how poorly the plutonium dioxide might conduct heat into the interior of the powder, the heaters would still respond the same way to the controlling thermocouple. The incoming heat to the powder would accumulate at/near the top surface if not conducted quickly enough into the powder interior. As the surface temperature rose from this accumulated heat, the temperature difference between surface and interior would drive more of the heat into the interior, still raising the interior's temperature. Reference 10 also includes test results from processing sludges in the Thermal Stabilization production furnaces in 1995. In those process tests, the temperatures measured at the thermocouples placed into the powder were reaching their peak temperatures at the same time the furnace started its 2-hour dwell at 1000°C. Adding magnesium oxide to the powder in the boat is expected to affect the thermal conductivities proportionately with either ceric oxide or plutonium dioxide.

TABLE 4: Thermal Conductivities of Ceric and Plutonium Dioxides

	Thermal Conductivities (W/m/°K)			
Temperature (°C)	CeO_2 (Ref. 7)	CeO_2 (Re. 8)	MgO	PuO_2
27			48	0.174*
38		11.5		
47	11.7			
93		10.9		
106	10.0			
131	6.3			
149		9.00		
227			27	
300				
527			13	
727			10	
927			8	
1019	1.7			
1127			7	

* = for 2.0 gm/cc; temperature assumed (not stated in reference)

The non-radioactivity of the ceric oxide also makes it more difficult to reach high temperatures than plutonium dioxide. No attempt was made to predict how much the PuO_2 temperatures would be raised by the radioactive decay heat, but the internal heat generation within the powder can only raise the powder temperature, however slightly, not lower it.

7. EXPERIMENTAL RESULTS

7.1 Temperature Indication Check

The accuracy of the temperature indicator used for the thermocouples in the powder needed to be known so that temperature recordings could be made with confidence. A check of the indicator was performed prior to starting the first test using a Transmation model 1604P Mini-Temp Calibrator to send a calibrated thermocouple input. This check was witnessed by Quality Control. The calibrated temperatures sent to the indicator and the resulting temperature indications are in Table 5.

TABLE 5: Temperature Indication Check

Known Temperature, °C	Indicated Temperature, °C
0	-1
900	899
950	948
1200	1200

Desired accuracy was +/- 3°C, so the indicator gave acceptably accurate temperatures throughout the range of interest (900-1200°C). The temperature indications that disagreed with the input temperature signal always indicated lower than the real temperature; thus, any temperature indications showing the powder to be at or above 950°C were actually showing that the powder was above 951-952°C. During the tests, the temperatures recorded as data were the actual indications unadjusted for this slight inaccuracy. This report uses the indicated temperatures for all powder temperatures; the real powder temperatures should be a degree or two warmer for all but the highest temperatures recorded.

7.2 "Status Quo" Tests

The first tests to heat powder in the test furnace were labeled as "Status Quo" tests because they represented the current type of feed to the production furnaces and were intended to demonstrate that the current product batches were reaching 950°C for at least 2 hours. The four Status Quo runs were attempted with pure ceric oxide (as purchased) because the production furnaces are currently processing reasonably pure plutonium dioxide. Three of the runs were completed. The first of the 4 runs had to be discontinued when power was lost to the heating system due to a failure in the ground fault circuit interrupter most of the way through the warmup portion of the heating cycle. These tests used a powder bed of very near 2500 grams, which is the mass limit for criticality prevention in the Thermal Stabilization production furnaces. Table 6 lists summary data for these runs.

The Status Quo runs confirmed that the powder was heating acceptably throughout the bed. The initial prediction was that "near dead center" should be the coldest spot in the powder. The temperatures there remained above 950°C for over 3 hours and peaked at about 1020°C. The entire bed of powder was declared to be above 950°C for just over 2-3/4 hours, but that time duration is based on two assumptions. The first assumption is that the powder surface is warmer than the interior while the powder is still heating. Once the powder in the bed interior shows that 950°C has been reached, the powder on the top surface should be well above 950°C. The second assumption is that the top surface of the powder will cool faster than the interior due to the air flow rate after heater power is shut off. The time that the controlling thermocouple dropped to 950°C is assigned as the last instant that the powder surface is guaranteed to be at or above 950°C. This method for determining the time the entire bed spent above 950°C was used in all sections of the testing.

The correct "coldest spot in the powder" was later determined to be at the front bottom corners of the powder in the boat (see Section 7.3 of this document). The coldest powder temperatures were also known by then to occur with the lowest air flow rates. Having determined that, Run SQ-04 was set up with a thermocouple in one front bottom corner and with an air flow rate of 1.0 scfm, which is the lowest flow rate allowed in the Thermal

TABLE 6: "Status Quo" Tests Summary Data

Run # and Date (Year 2000)	Bed Comp.	Bed Amt. (gm)	Air Flow Rate (scfm)	TC 1				TC 2				TC 3				Whole Bed Time @ > 950 C (hr)
				Place	Entry Route	T_{max} (°C)	Time @ > 950°C (hr)	Place	Entry Route	T_{max} (°C)	Time @ > 950°C (hr)	Place	Entry Route	T_{max} (°C)	Time @ > 950°C (hr)	
SQ-1 4-5	Pure CeO_2	2495	1.3	3-D Center	Rear	818 and still heating when power lost	N/A	Not used	N/A	N/A	N/A	Not used	N/A	N/A	N/A	N/A
SQ-2 4-6	Pure CeO_2	2495	1.3	3-D Center	Rear	1022	3.08	4" back, 1" down, just L of center	Front	1014	3.00	Not used	N/A	N/A	N/A	2.78
SQ-3 4-7	Pure CeO_2	2495	1.3	3-D Center	Rear	1019	3.05	4" back, in air?	Front	995	2.76	8" back, near center, 1/2" up from bottom	Front	1052	3.53	2.77
SQ-4 5-18	Pure CeO_2	2495	1.3	Rt front corner, bottom	Rear	1041	3.6	Center, 1/8" off bottom	Rear	1169	5.0	Not used	N/A	N/A	N/A	3.4

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Stabilization production furnaces. The bottom front corner exceeded 950°C for 3.6 hours in this run and peaked at 1041°C. The entire bed remained at or above 950°C for 3.4 hours in this run. The two time durations are different because the controlling thermocouple cooled to 950°C eleven minutes sooner than the thermocouple in the bottom front corner after power to the heaters was shut off.

Powder temperatures were consistently higher than the 1000°C temperature at the controlling thermocouple during these runs and the ensuing runs from the other phases of this study. The higher powder temperatures are caused by the combination of radiant heating and the air flow rate through the furnace chamber. The controlling thermocouple juts out into the air space of the furnace chamber where the air flow attempts to cool it. The heater controller then requires the heaters to send sufficient heat to maintain 1000°C there. The same heat output rate is radiated to the powder surface and the boat walls. The boat is a much larger target and receives far more radiant heat than the controlling thermocouple. The powder has less contact with the flowing air in terms of specific surface (surface area per unit of mass); thus, a higher fraction of the heat stays in the powder. The powder continues to warm until a steady-state temperature is reached that allows the incoming radiant heat transfer and the exiting convective heat transfer to equalize. In most runs, this top powder surface temperature, while unmeasurable, was so high that the interior of the powder reached 1000°C before the controlling thermocouple did and stayed well above that temperature. Eventually, 5 of the 18 total runs had at least one thermocouple indicate temperatures above 1100°C; two runs had peaks of 1169°C and 1177°C.

7.3 "Sensitivity" Tests

Eleven "Sensitivity" tests were performed to determine how sensitive the recorded temperatures were to thermocouple location, powder composition, and air flow rate. Powder mass became an accidental 4th parameter when a mixed powder of nearly 2468 gm was used and yielded temperatures that did not reach 950°C in the powder. That amount of the mixed powder overfilled the boat and the other criticality prevention limit of a maximum 1.5-inch powder bed depth was used (and deliberately exceeded to maintain "worst case" conditions for testing). Thermocouple entry location became an accidental 5th parameter as testing proceeded.

Four of the 11 Sensitivity runs used pure ceric oxide as the powder; the other 7 runs used a mixture of 13.8 weight percent magnesium oxide in ceric oxide. Magnesium oxide was added because it is a known future component in furnace feeds from the Magnesium Hydroxide Precipitation Process that will be used for stabilizing plutonium solutions.

The powder bed exceeded 950°C for at least 2 hours in seven of the 11 Sensitivity runs as shown in Table 7. The first 2 runs with the mixed CeO_2/MgO powder bed had trouble exceeding 800°C at the front of the boat, although the center temperatures exceeded 950°C for over 2.5 hours and reached over 1000°C at peak temperature. The large amount of powder used (2468 gm) was the primary cause for the low temperatures in this bed. The MgO sand is very light and its 13.8 weight percent was about 50 volume percent of the powder (eyeball estimate). The powder had to be piled high in the center to keep it from falling out through the holes in the boat front for the removal handle. The powder center was higher than the boat wall and may have approached a 3-inch depth. The average depth was probably well over 2 inches, but the removal handle's holes made it impossible to spread out the powder and measure it. The amount of powder for the remaining CeO_2/MgO runs was reduced to 1884 grams, which still exceeded the 1.5-inch depth limit for the production furnaces, but kept the highest mounds in the powder bed below or at least near the top of the boat wall.

A second reason for the low temperatures near the front of the boat was the thermocouple entry point for the front thermocouples. The two front thermocouples were about 1 inch back from the front wall and different depths (0.5 inch down and 1.5 inches down, each about 1 each out from the boat centerline). Positioning the thermocouples was much easier if the they entered through the edge of the furnace door, which is in the front of the furnace. This entry point, though, left a triangular crack on each side of the door and a rectangular crack of 1/16-inch width (the diameter of the thermocouple sheath) at either the top or bottom of the door, depending on which surface was

TABLE 7: "Sensitivity" Tests Summary Data

Run # and Date (Year 2000)	Bed Comp.	Bed Amount (gm)	Air Flow Rate (scfm)	TC 1				TC 2				TC 3				Whole Bed Time @ > 950°C (hr)
				Place	Entry Route	T max (°C)	Time @ > 950°C (hr)	Place	Entry Route	T max (°C)	Time @ > 950°C (hr)	Place	Entry Route	T max (°C)	Time @ > 950°C (hr)	
Sens-01 4-11	CeO_2	2495	1.3	Btwn Boat & Side Htr	Rear	1065	N/A (not in powder)	3-D Center	Front	1016	2.98	Center, 8.5" Back, 1.5" Down	Front	1074	3.85	2.77
Sens-02 4-12	CeO_2	2495	1.5	Btwn Boat & Side Htr	Rear	1068	N/A (not in powder)	3-D Center	Front	1020	3.08	Center, 8.5" Back, 1.5" Down	Front	1079	3.90	2.83
Sens-03 4-13	CeO_2	2495	1.0	Btwn Boat & Side Htr	Rear	1073	N/A (not in powder)	3-D Center	Front	1006	2.95	Center, 8.5" Back, 1.5" Down	Front	1071	2.92	2.80
Sens-04 4-14	CeO_2	2495	1.5	Btwn Boat & Side Htr	Rear	1078	N/A (not in powder)	3-D Center	Front	1012	2.97	Center, 8.5" Back, 1.5" Down	Front	1077	3.90	2.78
Sens-05 4-17	13.8% MgO in CeO_2	2468	1.0	3-D Center (1.5" Down in 3.125" Powder	Rear	1006	2.6	1" Back, 1/4" Right, 1.5" Down	Front Bottom	797	N/A	1" Back 1/4" Left, 0.5" Down	Front Bottom	844	N/A	N/A
Sens-06	13.8% MgO	2468	1.5	3-D Center	Rear	1018	3.0	1" Back,	Front Bottom	814	N/A	1" Back,	Front Bottom	860	N/A	N/A

4-18	in CeO$_2$							1/4" Right, 1.5" Down				1/4" Left, 0.5" Down				
Sens-07 4-20	13.8% MgO in CeO$_2$	1884	1.0	Rear, Center, 3/4" down	Rear	1023	3.1	3-D Center	Front Top	1106	4.2	Front, Rt of Center, 3/4" down	Front Top	964	2.2	2.2
Sens-08 4-21	13.8% MgO in CeO$_2$	1884	0.03	Rear, Center, 3/4" down	Rear	1028	3.3	3-D Center	Front Top	1070	4.0	Front, Rt of Center, 3/4" down	Front Top	979	2.5	2.5
Sens-09 4-24	13.8% MgO in CeO$_2$	1884	1.0	Rt Rear Corner, Bottom		1063	3.8	Rt Front Corner, Bottom	Front Top	923	N/A	Left front Corner, Bottom	Front Top	924	N/A	N/A
Sens-10 4-25	13.8% MgO in CeO$_2$	1884	1.5	Rt Rear Corner, Bottom		1069	3.85	Rt Front Corner, Bottom	Front Top	932	N/A	Left Front Corner, Bottom	Front Top	933	N/A	N/A
Sens-11 4-27	13.8% MgO in CeO$_2$	1884	1.0	Front Right Bottom, 2" Back		1072	3.85	Rt Front Corner, Bottom	Rear	990	2.6	Off-Gas Line, just out of furnace	N/A (not in powder)	262 (not in powder)	N/A	2.6

used. Nearly all the air inleakage used to maintain the oxidizing atmosphere came through these cracks and the front of the boat was constantly cooled by air that had barely been warmed. The effect was more pronounced if the thermocouples entered at the door bottom and flowed directly up and across the boat front. Even with the powder mass decreased to 1884 gm, the front of the boat did not always reach 950°C (Runs Sens-09 and Sens-10) because of the air flow through the edge of the door. When all the thermocouples entered through an unused tube at the back of the furnace, the furnace door rested against the wall surface, just as it does in Thermal Stabilization production use. While still not sealed, less air passed through the door edge and more air entered via other leak paths. The air that still entered at the door edge was also warmed first as it passed through a very narrow opening between two very hot surfaces. This air cooled the boat front less, allowing the powder temperature in front to exceed 950°C early enough to stay there for at least 2 hours.

Run Sens-11 illustrates how the front of the boat was cooled more even after all the thermocouple entries were moved to the rear of the furnace. This run had 2 thermocouples imbedded in the powder. One was in the right front corner at the bottom surface. The other thermocouple was also at the bottom, but two inches behind the first and also at the right edge of the boat. The front thermocouple exceeded 950°C for 2.6 hours and peaked at 990°C. The thermocouple 2 inches behind there exceeded 950°C for 3.85 hours and peaked at 1072°C.

The powder composition's effect on temperatures in the bed could not be determined. The discovery of the effects of thermocouple entry point, the great difference in powder volume for the same mass of the mixed CeO_2/MgO powder, and time constraints prevented getting a match of run conditions that might reveal a direct effect of powder composition on the powder temperature.

Air flow rate affected the powder temperatures distinctly, if slightly. Runs Sens-01 through Sens-04 used the same powder bed, thermocouple locations, and thermocouple entry points. Their only difference was the air flow rate through the furnace. The temperatures for each of the 2 thermocouples in the powder are highest for the 2 runs at an air flow rate of 1.5 scfm, slightly lower at 1.3 scfm, and lowest at 1.0 scfm. The differences between the temperatures at the differing air flow rates are only a few degrees, but the pattern is consistent for each thermocouple. Runs Sens-05 and -06 are identical except for the air flow rate and they also show higher powder temperatures with higher air flow rates. Runs Sens-07 and -08 differ from this pattern but that might be because the comparisons are between 1.0 scfm, which is the lowest air flow rate allowed currently in production, and 0.03 scfm (2 scfh) which was tested to determine if a really air flow rate might have an effect. finally, Runs Sens-09 and -10 show the same pattern with the temperatures at 1.5 scfm 6-9°C higher than those at 1.0 scfm.

Repeatability was difficult to obtain due to the accidental discoveries of thermocouple entry route and powder depth as accidental test parameters. Only one pair of runs had the same conditions for air flow rate, thermocouple positions, thermocouple entry points, powder composition, and powder mass. Those runs were Sens-02 and -04. In those runs, the thermocouple at the 3-dimensional center of the bed stayed above 950°C for 3.08 and 2.97 hrs, respectively, with peak temperatures of 1020°C and 1012°C, respectively. The other thermocouple was about 3 inches behind the centered thermocouple and about 1 inch deep in the powder. This thermocouple stayed above 950°C for 3.90 hours in each of the runs with peaks of 1079°C and 1077°C, respectively.

7.4 Mixed CeO_2 - Chloride Salts Test

One run was performed with a mixture of 80 weight % CeO_2 and 10 % each of NaCl and KCl. The total powder weight was 2542 grams, which included separate piles of pure NaCl (13.4 gm) in the front bottom corner of the boat and pure KCl (10.6 gm) in the bottom edge of the boat halfway back. These locations were also the spots where the 2 thermocouples were placed into the powder. The thermocouples were placed directly into these pure salts to determine the effect of the salts melting during heating.

This run is best described as one that "failed right after it succeeded." The temperature in the coldest spot of the powder, the front bottom corner of the boat, was at an indicated 949°C when the furnace reached its controlled dwell temperature of 1000°C. This thermocouple reached the minimum 950°C seconds later and peaked at 960°C within 30 minutes. Fifty minutes after reaching 950°C, the thermocouple began sending erratic signals to the indicator, usually in the range of 956-958°C but covering a 951-960°C span. Over the next hour, the thermocouple sent increasingly erratic signals and then failed entirely. The second thermocouple in the powder also began sending erratic signals about 75 minutes after the furnace began its 2-hour dwell at 1000°C. It, too got increasingly erratic after that time, but did not fail completely. The thermocouple controlling the furnace temperature kept sending fairly steady signals, but the temperatures varied with changes made to the off-gas flow rate. This run failed to show that the powder temperature would be at or above 950°C for at least 2 hours due to the thermocouple failure in the front corner of the boat and the erratic signals from the thermocouple halfway back in the boat. The results of all the other runs and this run's temperatures recorded over time imply, however, that the powder temperatures would have been above 950°C for at least 2.1 hours had the thermocouple sheaths not corroded, allowing the thermocouple failure.

The off-gas flow rate had to be adjusted frequently after the furnace reached 1000°C. The chloride salts were evaporating more quickly than expected by this time and recrystallizing in the off-gas line. As the off-gas line became more coated with the chloride salts, the flow rate would have to be increased at the control valve. The changes in air flow altered the cooling on the controlling thermocouple, so the temperature would change until the controller reacted to bring the temperature back to 1000°C. By that time, the flow rate would change again and steady temperatures in the furnace were not maintained. The recrystallized salts eventually coated the entire off-gas line, including the entire inside surface of the rotameter tube. The entire off-gas line had to be removed and each piece cleaned separately following this run.

The molten and vaporized salts damaged more than just the thermocouples. The boat with the powder was very difficult to remove from the furnace after this run. A long spatula had to be forced down each side of the boat in order to allow its removal. The boat wall and the heating element on one side had fused together over a small area, somehow, and the fused point had to be broken by the spatula before the boat could be removed. This break left a small hole (about 1.5 cm diameter) in the side wall of the boat near the back corner and a matching "crater" in the heating element. Two small globs of resolidified metal were found on the furnace bottom after the boat was removed. The reason for the boat fusing to the heating element is not certain; however, one theory is that the chloride salt vapor penetrated the porous heating element surface and allowed the electrical current to arc to the boat wall, melting the boat wall at that point. There were no signs of any salt or powder splattering anywhere in the furnace during or after the run. The boat was replaced before continuing with the next series of runs for nitrate salts.

The powder in the boat solidified into a fairly cohesive "brick" as it cooled after the run. The cohesion within the powder is most likely a result of the chloride salts melting during the run and holding the powder together after the chloride salts resolidified. The brick crumbled slightly around the edges unless handled very carefully and could also be ground up with only light pressure from a spatula. The brick could also be picked up carefully and held intact for photographs in any orientation. When placed upside down in a pan, the top surface was not ground down even though it was placed repeatedly in this position and is normally kept in that orientation. Figure 4 shows the bottom surface of this brick while resting upside down in a pan after removal from the boat. Note that the thermocouple tips are still in place in the holes where the pure chloride salt samples melted during the run.

7.5 Mixed CeO_2 - Nitrate Salts Tests

Three runs were performed with a powder matrix of ceric oxide, magnesium oxide, and hydrated magnesium nitrate. These runs were performed to determine the effects of hydrated nitrate salts left in Magnesium Hydroxide Precipitation Process product that had not been heated adequately on that process' hot plates. A portion of the remaining powder from the last Sensitivity Run was used as part of this powder bed. A weighed portion of magnesium nitrate hexahydrate was then added to complete the bed composition. In each succeeding run, additional hydrated nitrate salt was added to the remaining powder from the previous run; thus, the concentration of magnesium oxide kept increasing while the ceric oxide and magnesium nitrate hexahydrate concentrations

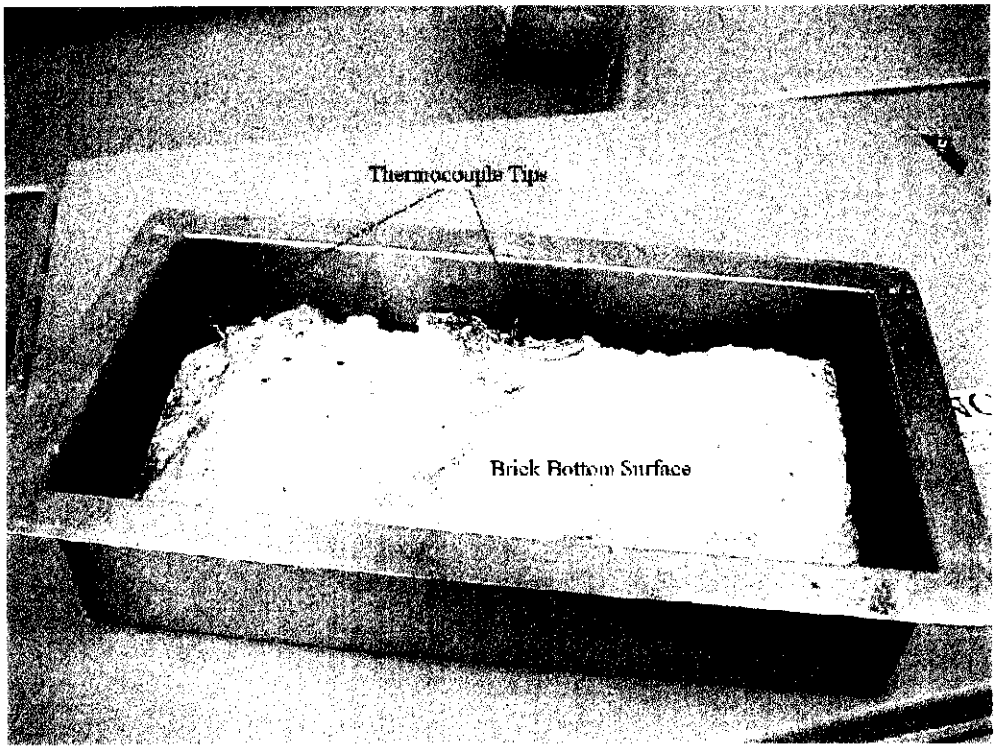

FIGURE 4: Powder Bottom Surface from Chloride Salts Run

decreased slightly. Table 8 shows that the powder exceeded 950°C for at least 3.4 hours in the coldest spot (front bottom corner) in each run. The coldest peak temperature recorded was 1033°C.

The hydrated nitrate salts presented other problems, but these problems were gone long before the furnace reached 1000°C. The water in the hydrated nitrate salt condensed in the off-gas line and was suspended on top of the rotameter, making monitoring of the air flow rate a bit more difficult. The NO_X gases from the decomposition of the nitrates was partially absorbed by the condensed water, forming what was quickly dubbed "acid rain" in the off-gas line. This liquid was drained periodically by shutting off the air blower, then opening a drain valve. The draining operation was usually over within 30 seconds. The Thermal Stabilization production furnaces might not have the capability to drain the liquids, so any liquids formed could be drawn into the process vacuum pump. Acidic liquids in that pump's seal water could affect the seal water pH and require compensatory actions at that point to prevent corrosive damage to the pump.

Once the water and nitrates were gone from the powder bed, the bed heated quickly and easily reached 950°C at the coldest spot at least 70 minutes before the controlling thermocouple began its 2-hour-long dwell at 1000°C.

8. COMPARISON TO EARLIER STUDY

Similar process tests were performed in the Thermal Stabilization production furnaces while processing sludges in June, 1995 (Reference 10). Powder temperatures in those process tests were significantly lower than the temperatures reported here and they seldom exceeded 950°C. Two reasons account for the lower temperatures in these process tests.

First, the sludges being processed in the production furnaces had varying compositions from run to run and were distinctly different from the dry powders used in the tests reported here. Because of their nature, the sludges could easily have had significant amounts of water or other liquids, plus corrosion products and possibly other miscellaneous materials. All of the powders reached a steady-state temperature at the same time the furnace reached its steady-state dwell at 1000°C, but the steady-state powder temperatures varied from 618-952°C.

TABLE 8: Nitrate Salts Tests Summary Data

Run # and Date (Year 2000)	Powder Comp.	Powder Amount (gm)	Air Flow Rate (scfm)	TC 1				TC 2				Whole Bed Time @ > 950°C
				Place	Entry Route	T_{max} (°C)	Time @ > 950°C	Place	Entry Route	T_{max} (°C)	Time @ > 950°C	
Nitr-1 5-23	69% CeO_2, 11% each MgO and $Mg(NO_3)_2$, 8% H_2O	2039	1.0	Rt Front Corner, Bottom	Rear	1047	3.67	2-D Center about 1/2" Down	Rear	1130	4.67	3.5
Nitr-2 5-31	66% CeO_2, 14% MgO, 11% $Mg(NO_3)_2$, 8% H_2O, 1% Corrosion	2107	1.5	Rt Front Corner, Bottom	Rear	1062	>3.5	1/2" in front of 2-D Center, 1/2-way Down	Rear	1126	4.67	3.7
Nitr-3 6-2	67% CeO_2, 17% MgO, 8.5% $Mg(NO_3)_2$, 6% H_2O, 1% Corrosion	2067	1.0	Rt Front Corner, Bottom	Rear	1033	3.45	1/2" in front of 2-D Center, Bottom	Rear	1177	5.1	3.33

Endothermic reactions occurring in the sludges are assumed to have caused the lower steady-state temperatures. The feed items categorized as "sludges" have now been completely processed.

A second contributor to the lower powder temperatures in those process tests could be the thermocouple entry routing. The thermocouples placed in the powder had to be routed through the front door of the furnace. As explained in section 7.3 of this document, this routing would have allowed a larger fraction of the air to flow in through the front door of the furnace and give that air more of an opportunity to cool the front areas of the boat. This thermocouple entry method also made it more difficult to be certain of where the thermocouple tips were located after the furnace door closed. Bending the thermocouple sheaths in the surrogate tests reported here was much easier because the apparatus was in an open room, not in a glovebox, and bare hands could be used to position them such that closing the furnace door had little or no effect on their final test location.

9. CONCLUSIONS & RECOMMENDATIONS

1. The thermal stabilization method currently in use in the production furnaces was shown to exceed the requirement that all of the feed material be heated to at least 950°C and held there for at least two hours. This process works for the pure oxides currently being processed and for the future Magnesium Hydroxide Precipitation Process product that will be processed. The thermal stabilization process works even if the feed still contains appreciable water and nitrate salts. The process works for all amounts of feed up to the mass limit of 2500 gm and up to the depth limit of 1.5 inches. The process also worked in the test furnace for powder depths in the 1.5-2.0-inch range. The thermal stabilization process currently in use should be kept in use unchanged except for potential changes known to increase the powder temperature (slower heating rate, longer time at 1000°C, steady-state temperatures higher than 1000°C, etc.).

2. Appreciable quantities of chloride salts are very damaging to the furnace and associated apparatus. Chloride salts should not be processed in these furnaces unless permission is received to process those feeds at temperatures low enough to avoid melting the chloride salts. If lower temperatures may not be used, then the feeds should have the chloride salts removed before the feeds enter the furnaces.

3. Hydrated salts and nitrate salts do not prevent the thermal stabilization product from heating to above 950°C for at least 2 hours, but they cause condensates that might cause other problems in the off-gas system. Nitrate salts and wet solids should be avoided in the furnaces by proper use of the hot plates in the Magnesium Hydroxide Precipitation Process. Hot plates might also be useful keeping other types of feed contaminants out of the furnaces and off-gas system.

4. The temperatures in the powder get higher with increasing air flow rate through the furnace chamber, at least within the current bounds of 1.0-1.5 scfm. This air flow rate should be maintained near the 1.5 scfm upper limit as much as possible so that the product will be a few degrees warmer and the corresponding amount less reactive on removal from the furnaces.

5. Running thermocouples through the furnace front door causes lower temperatures at the front of the boat, frequently too low to meet the temperature/time requirement. If additional thermocouples must be used inside the furnace, a way should be found to have them enter at the rear of the furnace or, at a minimum, enter the front in a manner that keeps the door surface flush with the front/side wall surfaces during operation.

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