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PNL-3245
UC-78

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Olympic Engineering

January 1980

Prepared for the U.S. Department of Energy
under Contract EY-76-C-06-1830

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute

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PNL-3245

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
Under Contract EY-76-C-06-1830

Printed in the United States of America
Available from
National Technical Information Service
United States Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151

Price: Printed Copy $____*; Microfiche $3.00

*Pages	NTIS Selling Price
001-025	$4.00
026-050	$4.50
051-075	$5.25
076-100	$6.00
101-125	$6.50
126-150	$7.25
151-175	$8.00
176-200	$9.00
201-225	$9.25
226-250	$9.50
251-275	$10.75
276-300	$11.00

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DEVELOPING AND TESTING A VERTICAL SINTERING FURNACE FOR REMOTE NUCLEAR APPLICATIONS

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SUMMARY

There is an industry need for process equipment that can be remotely operated and maintained to fabricate nuclear fuels. The remote capability is necessary in order to keep exposures to personnel at the lowest level possible while utilizing nuclear fuel material that will deter proliferation attempts.

Horizontal-type furnaces used to sinter fuel pellets on a production basis are large and thus impractical for remote applications. However, research has shown that vertical-type furnaces are adaptable for use and are cheaper to operate and maintain.

In 1979, Pacific Northwest Laboratory,[a] working under the auspices of the Department of Energy's Fuel Refabrication and Development (FRAD) Program, began developing an advanced concept for a remotely operated furnace designed specifically to sinter nuclear fuel pellets.

The FRAD Program at PNL ended before the sintering of nuclear fuels could be completely verified. However, during 1979, PNL performed a sufficient number and variety of tests to establish that nuclear fuel pellets can be sintered in a vertical furnace. However, additional efforts are required to refine and to establish an efficient operation.

(a) Pacific Northwest Laboratory, located in Richland, Washington, is operated by Battelle Memorial Institute for the Department of Energy.

INTRODUCTION

The nuclear industry has renewed its interest in the development of process equipment for the fabrication of nuclear fuels, i.e., equipment that can be remotely operated and maintained. This interest reflects government and industry concern for maintaining very low levels of personnel exposure to radiation while fabricating proliferation-resistant fuels.

One phase of nuclear fuel fabrication that is being examined for remote operation possibilities is the sintering of fuel pellets, which is the process of exposing the pellets to high temperatures in a reducing atmosphere in order to obtain near-theoretical density and the desired oxygen-to-metal (O/M) ratio. Current production-level sintering furnaces are very large and such furnaces are not readily adaptable for remote use. However, veritical-type furnaces offer the potential of smaller size per throughput and do not require as much mechanical handling of fuel pellets. Vertical furnaces also can be operated continuously. This type of furnace should be adaptable to remote operation in a controlled environment and remote maintenance in, and/or removal from, a radioactive area. Such a furnace would be cheaper and easier to build and would be more adaptable to remote operation than would a horizontal-type furnace.

In 1979, Pacific Northwest Laboratory (PNL) instigated efforts on behalf of the Department of Energy's Fuel Refabrication and Development (FRAD) Program to identify an advanced concept for a sintering furnace with these characteristics:

- vertical design
- compact size
- continuous type of operation
- high throughput-to-size ratio
- improved atmosphere-to-pellet contact for control of the oxygen-to-metal ratio, and
- modular construction for remote application.

The initial goals of the furnace development project were to 1) demonstrate the proof-of-principle of the vertical furnace by the end of FY 79 and 2) verify the sintering of nuclear fuels during FY 80.

This report covers the tasks that were completed during the development of this furnace. These include -

- developing a prototype design
- fabricating the furnace
- conducting preliminary and mockup tests, and
- performing tests at both room and elevated temperatures.

DESIGN FOR PROTOTYPE VERTICAL SINTERING FURNACE

In order to test the concept of remotely operating a vertical-type sintering furnace for nuclear fuels applications, PNL designed and built the prototype shown in Figure 1. This vertical sintering furnace (VSF) is capable of remote operation to accommodate highly radioactive nuclear wastes or fuels containing plutonium, uranium-233, or spikants of other radioactive materials. Because of the demands of machine operation and maintenance in a remote environment, criteria that normally would receive little notice become most important.

GENERAL DESIGN CRITERIA

Some of the most important considerations for a remote sintering furnace are as follows:

- Size - The furnace must be as small as possible for a given production capacity for reduced cost, ease of installation, and remote removal for maintenance and/or burial at end of life.
- Operability - The furnace must be simple to operate to avoid excessive maintenance in an environment that requires remote control and maintenance.
- Service - The furnace must be constructed for a minimum lifetime of five years and modular replacement of parts.
- Safety - The furnace must be capable of sintering a product to a controlled oxygen-to-metal ratio (O/M) in a $<5\%$ hydrogen atmosphere in order to avoid all possibilities of fire or explosion.

The prototype VSF does not meet all these criteria, but its mode of operation is the same. Therefore, it can be used as a basis to prove the principle of this sintering method.

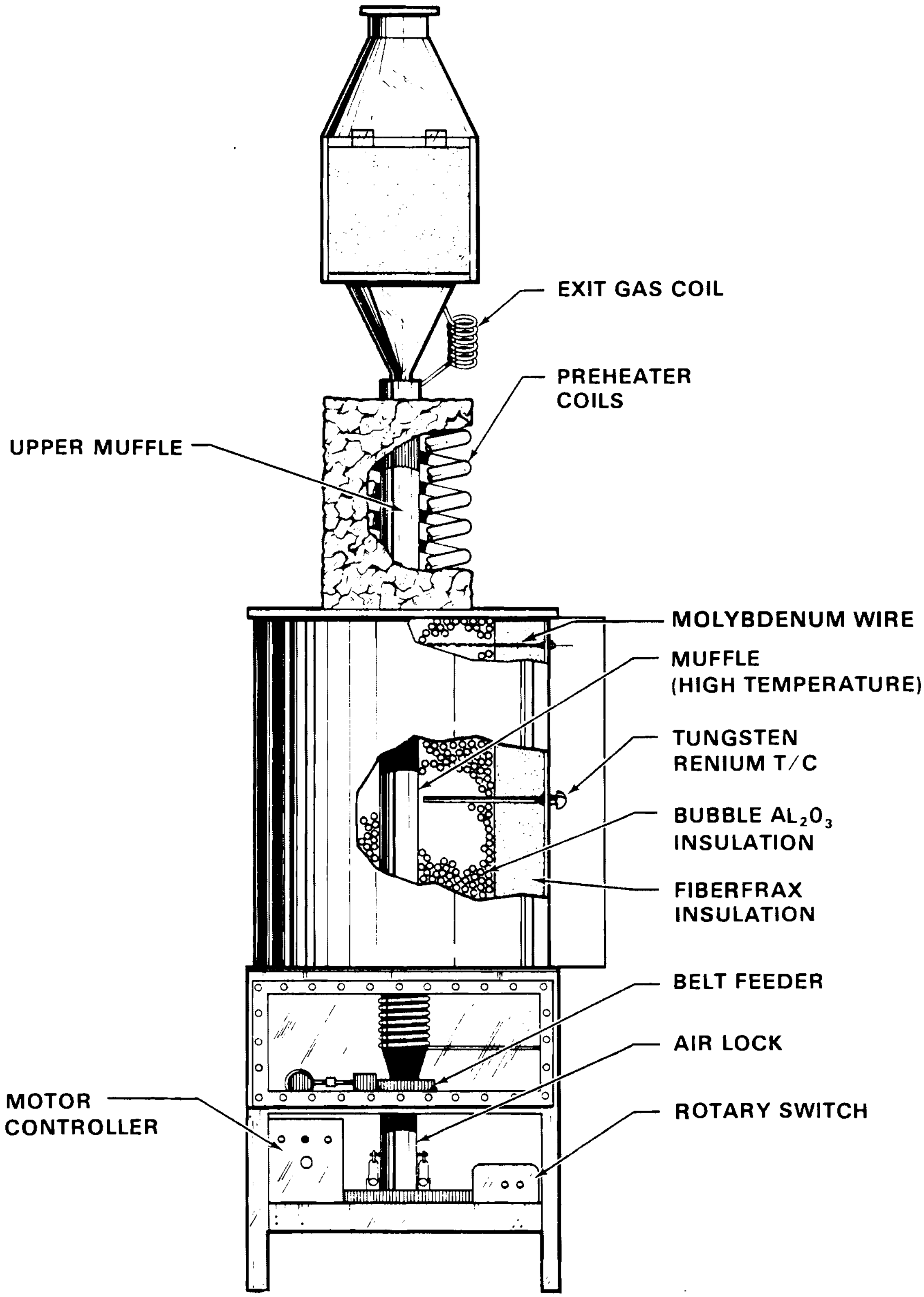

FIGURE 1. Cutaway Drawing of Vertical Sintering Furnace

FURNACE DESIGN

The furnace has a cylindrical muffle that contains the material to be sintered, and is mounted in a vertical position. Gravity is thus utilized to move the product through the sintering cycle. In place of molybdenum boats, or boxes that are required to contain pellets in a horizontal-type furnace, a granular "carrier" protects the pellets from damage during movement through the furnace and can be any free-flowing material that is compatible with and inert to the product at the high sintering temperatures. In the case of mixed oxides of plutonium-uranium, candidates for carrier material are alumina (Al_2O_3), urania (UO_2), and thoria (ThO_2).

The muffle, or central portion of the furnace, is divided into three parts: 1) The entrance, or top portion of the muffle, (the preheat section) is constructed from stainless steel. It includes a hopper or receptacle for receiving the pellets and carrier and an exit for gases. 2) The central portion (the high temperature zone) is made of high purity alumina ceramic wound with molybdenum wire for even heating and is surrounded by insulation to reduce loss of heat from the furnace. 3) The lower portion (the cooling zone) is made from stainless steel. The cooling apparatus surrounding this portion is either coils of water-filled copper tubing or some form of water jacket.

The shape of the cylindrical ceramic muffle, the outside of which is wound in molybdenum wire, concentrates electrical power in a specific area and permits excellent heat transfer through the muffle to the product. The low heat losses resulting from the use of this insulated muffle contribute to the high throughput capacity that is possible. The small size of the furnace, its type of construction, and its simplicity of operation combine to make it adaptable to the semiremote or remote operation that is necessary for nuclear fuels and other highly radioactive materials.

Special Design Considerations

To remove the fire or explosion potential, less than 5% hydrogen in the furnace gas is required. This criterion permits a safe furnace operation but introduces the problem of insufficient reduction of highly radioactive fuel made from plutonium or uranium-233. In typical horizontal furnaces that use boats, the mixed oxide pellets must be kept to a single layer or at most two to three layers in order to assure even, thorough penetration of the low hydrogen content gas. The fuel load is thus reduced by a factor of four to six, and a diffusion distance of only two to three layers can still cause a gradient in the pellet O/M ratio, which may require costly rework and adjustment. The horizontal furnaces are therefore excessively large and inefficient. The vertical furnace is being developed to sinter bulk quantities of highly radioactive fuels[a] using the safe, low hydrogen content furnace gas while meeting necessary reducing conditions.

DESIGN CHANGES

Results of tests performed on the VSF can be found in the section, Preliminary and Mockup Tests. Suggestions for changes in the furnace design that resulted from these tests are included here. These changes fall into two categories: changes to be made to the prototype and features to be incorporated in the design of the next version of the VSF.

Changes in the Prototype

Tests conducted to date indicate that the following changes in the prototype furnace should be made prior to performing a substantial number of further tests:

- Taper the inside of the high-temperature muffle by gradually increasing its diameter at least 0.075 in./ft.
- Reduce space between the preheat and high-temperature muffle windings.
- Alter spacing on high-temperature windings to compensate for downward flow of material.
- Eliminate restriction at base of inlet funnel.
- Incorporate thermocouple installation that will provide a more representative reading of the temperature in the hot zone.
- Increase capacity of exit gas coil.

(a) Other nuclear materials such as calcined waste can be sintered in this type of remotely operated furnace that is fired in either a controlled environment or air. See section, "Other Applications for the Vertical Sintering Furnace."

Changes in the Furnace Design

The following changes or new design features should be incorporated into the design of the vertical furnace for future versions of the VSF.

General

- additional thermocouple(s) to indicate position and length of hot zone
- improved transition from round cooling zone to rectangular belt feeder base
- design changes for modular construction, remote operations, and remote maintenance
- larger exit gas coil capacity
- ancilliary equipment such as separators and elevators.

Muffle

- replacement of two-section (3-foot ceramic plus 2-foot stainless steel) muffle with five-foot long ceramic muffle
- first portion of the ceramic muffle wound specifically for the lower temperature preheat section
- at least a 3/8 inch ID increase over the entire length of muffle
- entrance for the control thermocouple drilled in muffle
- larger six-inch diameter muffle tested or length of hot zone increased for demonstration of the potential of higher process rates.

Insulation

The temperature of $<200^{\circ}F$ on the skin and flanges of the prototype furnace indicate a 24-inch diameter should allow sufficient insulation for a 4-inch muffle furnace. A 30-inch diameter would probably be necessary for a furnace with a 6-inch or larger muffle.

Belt Feeder

- a two and one-half to three-inch belt width
- an endless belt to avoid problems, such as differential thickness, gaps, etc., encountered with current crudely spliced belts

- a fine mesh wire belt with rubber-coated drive pulley
- a drive and bearings that are capable of performing in a radioactive environment.

FURNACE FABRICATION, STARTUP AND OPERATION

This section contains information on a few aspects of furnace fabrication. It also includes information on furnace startup and its actual operation as background for the tests that were performed.

FURNACE FABRICATION

The VSF as it appeared after fabrication is shown in Figure 2. Note the foil-wrapped preheater coils around the upper muffle.

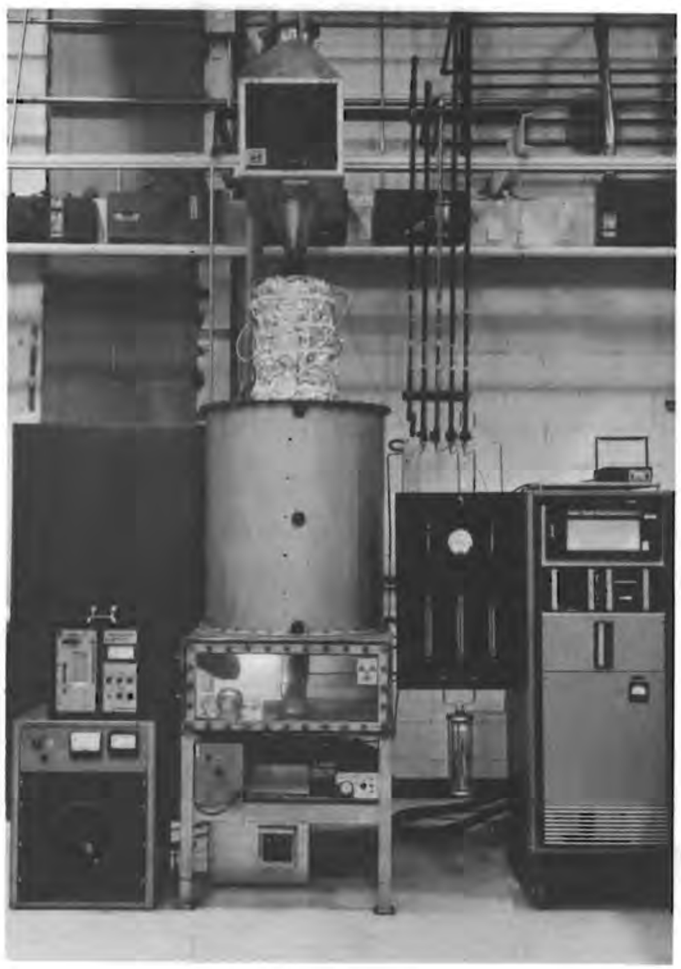

FIGURE 2. The Vertical Sintering Furnace

Furnace Exit Seal

An airlock on the furnace base was required to keep air out of the furnace yet permit passage of the carrier and pellets leaving the reducing gas environment. This barrier was first attempted with a rotary valve air lock. The manufacturer claimed that the airlock would seal against 15 psi differential pressure; however, considerable modifications were necessary before it would handle abrasive grain and pellets without jamming. When the modified valve was checked for air lock sealing, it leaked too much to be used.

An airlock was then constructed of 2 three-inch butterfly valves in a 3-inch ID rubber-lined steel cylinder. The valves were actuated by two small air cylinders operated by four-way solenoid valves and a multiple rotary switch. In actual operation, the space between the valves is purged with nitrogen for three minutes during each cycle to remove any trapped air before opening the inside valve to the furnace environment. With this air lock, it was possible to maintain the level of oxygen in the furnace at <20 ppm during operation and with the carrier moving through it. Commercial butterfly valves were also used in an air lock of similar design for subsequent furnace operation and testing.

Furnace Entrance Seal

Oxygen from the air outside the furnace is prevented from entering the top of the open hopper by the upward movement of furnace gas through the bed of alumina particles and the particles themselves. Tests made with an oxygen monitor have indicated that two inches of carrier is sufficient to maintain the level of oxygen in the furnace to below 5 ppm. Therefore, it was found to be unnecesssary to use any air lock device on the furnace entrance.

Temperature Profile/Control

Carrier was placed in the furnace and the temperature was raised to 1000°C as shown by the control thermocouple. The hot zone of the furnace was probed with both a chromel-alumel and rhenium thermocouples. The maximum temperature readings of these thermocouples closely correlated around 1225°C. The furnace temperature was raised to 1500°C as indicated on the control console and a probe of the internal temperature showed 1830°C in the hottest portion of the furnace, which was about 6 in. below the control thermocouple position. A temperature profile was taken with the maximum overall furnace temperature of 1700°C.

The temperature probes showed the following:

- there is a temperature delta of several hundred degrees between the control thermocouple and actual maximum temperature inside the muffle,
- the heat transferred to the preheat zone by the hot atmospheric gases was negligible compared with movement of the cold carrier,
- the preheat zone remained essentially at room temperature, and
- the sintering or hot zone was shifted downward from the winding and muffle center by the carrier movement.

Because of this temperature information, clam shell heating elements were added around the stainless steel preheat section and connected to a controlled source of power for preheating. With the preheat coil in operation, new temperature profiles were established for different maximum or desirable furnace temperatures.

The new furnace profile indicated that carrier grain entering the central section, or high heat zone of the furnace, is in the range of between 500° and 600°C. This drop from a maximum temperature of 850°C in the preheat zone is caused by the gap between the windings of the separately heated muffles.

FURNACE STARTUP

When the furnace was first brought up to operating temperature, the granular alumina formed into a hard cake. This prevented insertion of a thermocouple probe from the hopper down into the hot zone. When the packed or caked alumina was removed and the temperature raised again to 1700°C, the results were similar. It was postulated that impurities from the new muffle and high temperature cement placed on the windings were condensing out and onto the carrier in the preheat section, then becoming partially sintered when the carrier reached the hot zone. Laboratory analysis of a sample indicated rather large amounts of fluorine, lithium and silica, which may have contributed to the caking.

The carrier suspected of being contaminated was removed, new carrier grain was placed in the furnace, and the maximum furnace temperature was raised to 1700°C. The results were similar to the first tests, but the cake that formed was more fragile, breaking up readily. However, even this fragile cake was sufficient to block the belt feeder. Tests were continued for several days and it was found that the cake would form at temperatures above 1100°. Pure arc-fused granular alumina does not normally sinter at temperatures lower than 1750° to 1800°C.

Analysis of the data indicated that the cake did not form at a temperature rise of 600°C while moving 18 inches in the ceramic muffle but did form in a 1200°C rise. It appears that the temperature differential between the preheat section and the high temperature zone may be the controlling factor. Since the amount of pressure caused by height of the column is only a few pounds per square inch, column height should not be a factor in the caking of the carrier grain.

The packed carrier column must expand as its temperature is raised and in an essentially straight-walled muffle, the area for expansion is limited. Considering these factors, forces created by a large temperature increase may be sufficient to cause partial sintering. If this is the cause, caking could probably be avoided by using a muffle that has been tapered sufficiently to compensate for the thermal expansion of the carrier and thus eliminate this pressure buildup.

The muffle was removed and its diameter was found to increase from 20 to 22 mils, but it had been installed with the small end down. It was placed back in the furnace with the small end up, then further high-temperature tests were performed to determine whether this reversal had a positive effect on the caking. However, fragile or partial cakes were still formed at the higher temperatures.

NORMAL FURNACE OPERATION

In operation, a mixture of pellets and carrier or just pellets are fed into the open hopper at the top of the furnace. The rate of downward movement of the mixture through the preheat, sintering, and cooling muffles is controlled by a belt feeder situated in a gas-tight compartment in the furnace base. Argon-5% hydrogen gas is introduced into the base compartment. Sufficient gas pressure is applied to overcome the pressure drop across the moving bed of pellets and carrier. The flow of gas through the bed produces the intimate contact between pellets and gas that is necessary for proper sintering and control of the O/M ratio when using the low hydrogen content furnace atmosphere.

The upward flow of furnace gas through the matrix of pellets and carrier also prevents diffusion of oxygen from the open feed hopper, so an air lock is unnecessary at the feed end. An air lock mounted below the belt feeder in the base compartment allows the pellet-carrier material to pass from the furnace environment to the outside atmosphere. The automatic cycle of the air lock includes a nitrogen purge of the chamber preventing introduction of air into the furnace gas.

A control console that can be mounted away from the furnace contains operating controls, temperature indicators and recorders, and power controllers with an over-temperature cutoff safety feature. The panel adjacent to the console contains gas flow meters, pressure indicators, and valves to control water cooling and gas flows.

Startup Procedures

The following is a list of procedures for starting the VSF:

1. Cycle the butterfly valve air lock to assure that the normal 60 to 65 psi air pressure is available to the cylinders and that the valves are operable.

2. Stop the air lock cycle in the purge phase. (The indicator light is on when both valves are closed and the air lock is purging.) Turn the air-lock switch off.

3. Turn on the furnace cover gas (argon-5% hydrogen) and purge all lines, including the water bubbler.

4. Set flow rate at 20 cubic feet per hour (cfh) for furnace gas, 2-1/2 cfh for insulation purge gas, and 2-1/2 to 3 cfh for air lock purge gas. Purge for three to four hours.

5. Attach oxygen monitor to the base probe and monitor oxygen content of furnace during purging at adequate intervals.

6. When the O_2 content approaches 100 ppm, start the air lock cycling to check the atmospheric seal. (O_2 content should not change.) Stop the air lock cycling.

7. Turn power on the furnace winding and gradually raise the temperature at 50°C/hr to between 500 and 600°C. Leave power on at least 12 hours to bake out moisture from furnace insulation.

8. Continue raising the furnace temperature at 300°C/hr to the desired operating temperature or a maximum temperature of 1800°C.

9. As the furnace temperature approaches 1000°C,
   a. turn power on to the preheat windings
   b. start water flow through cooling coil
   c. start belt feeder, then
   d. start airlock cycling to remove pellets/carrier.

10. As the furnace temperature rises,
    a. check the O_2 content often to assure low O_2 content in furnace cover gas, then
    b. check the skin, flange, and preheat muffle temperature, and record them.

11. After approximately one hour at the desired operating temperature or if the furnace reaches the maximum temperature of 1300°C, reduce the control temperature to 1700°C and allow it to stabilize with carrier grain moving through at a setting of 2.5 on the motor speed control.

12. When equilibrium conditions are assured, probe the entire length of the preheat and sintering zones with a tungsten-rhenium thermocouple to verify the furnace profile.

13. Reduce the furnace gas flow that has been bubbled through water to moisturize to a level of 15 cfh.

14. Place several molybdenum pellets in the top hopper and operate the furnace through a complete cycle to confirm the presence of the reducing atmosphere before starting any sintering tests that require such an atmosphere.

Compared to similar equipment, the prototype VSF requires much greater operating time because of the time required to purge the atmosphere (3 to 4 hr) before applying power to the molybdenum windings, the rate at which the furnace can be brought to operating temperature (8 to 10 hr), and the time for cooling the furnace down so that it may be opened up to air atmosphere (3 to 4 dy).

CONCLUSIONS FROM FURNACE OPERATION

Specific conclusions can be made from the operations of the prototype vertical sintering furnace to date. They include the following:

1. The pressure drop through the granular carrier grain and/or a mixture of carrier and pellets is adequate to prevent air from entering the furnace. Thus, the VSF can be operated without an airlock on the top entrance, and an oxygen-free environment can be maintained around the windings. Also, a controlled environment can be maintained in the muffle.

2. A butterfly-type air lock at the furnace exit permits pellets and carrier to pass from the furnace to the room without degrading the furnace environment with an inward flow of air.

3. The carrier grain's protection of pellets through the furnace cycle exceeds all expectations. Normal density pellets pressed with sharp edges show no chipping or other visible flaws. Several kilograms of extremely fragile alumina pellets, which could be crushed between the fingers, exhibited only minor damage after a cycle through the furnace.

4. The exit belt conveyor is adequate to handle carrier grain, a combination of carrier and fuel pellets, and simulated waste pellets.[a] Its variable speed provided good control of the material moving through the furnace.

5. Carrier grain can pick up volatile contaminants such as silica and fluorine during the initial startup and bake-out of a green muffle. Therefore, the carrier used for this purpose should be replaced with fresh carrier grain for subsequent furnace operations.

6. For high-temperature operation, the inside diameter of a muffle in a vertical furnace should be tapered sufficiently to allow for the thermal expansion of the packed column.

7. No long-term, high-temperature sintering tests were possible because the carrier caking. However, results of the brief tests indicated the feasibility of fast sintering.

8. Because there is too great a temperature differential between the spot outside the muffle where the control thermocouple was attached and the inside of the muffle, the control thermocouple should penetrate the muffle wall.

9. The exterior furnace skin temperature is $<200^{\circ}F$, indicating that a lesser amount of insulation should be adequate to protect the elastimer seals and components that are required for the design of a remotely operated furnace.

10. The 25-kVa power supply used should be adequate for a much higher capacity furnace. In typical usage, 15 kVa will raise the temperature at a rate of 300 to $400^{\circ}C/hr$, and $\sim$10 kVa is sufficient to maintain the temperature at $1750^{\circ}C$.

(a) See section, "Other Applications for the Vertical Sintering Furnace."

PRELIMINARY AND MOCKUP TESTS

Since the furnace is designed to use a granular carrier instead of boats, the first tests were designed to determine the effects of carrier grain size and the presence of pellets on gas pressure drop through the furnace. Tests were also performed to determine the bulk density of the furnace matrix using steel pellets, and to make similar calculations for UO_2 pellets and alumina carrier. Finally, tests were run on alumina and urania pellets to determine the adequacy of the VSF to sinter without damaging pellets. The carrier and both alumina and urania pellets were observed for signs of reaction. Tests were run to determine the effects of grain size and the presence of pellets on gas pressure drop through the furnace. No crushed fused alumina was available so bubble, insulating-grade alumina was used. The alumina was screened to three sizes for preliminary tests in an apparatus consisting of a 1-3/4-in. diameter glass tube, a N_2 gas supply, a flowmeter, and a pressure gage. These are the results:

Screen Size	Pressure Drop
12/28	0.15" H_2O/ft.
28/60	1.09" H_2O/ft.
60/80	2.36" H_2O/ft.

The gas flow rate used was 2-1/2 standard cubic feet per hour, which was extrapolated from the amount of gas required to properly reduce the expected maximum amount of pellets in a 4-inch diameter furnace (2 kg/hr). An arbitrary mixture of 60/80 mesh alumina and steel pellets sized for pressurized water reactors (PWRs) yielded a pressure drop of 4.25 inches of water per ft. This confirmed the expected trend.

On the basis of the preliminary results, solid fused alumina grain was ordered: 20F - (20/40 mesh) and 46F - (40/70 mesh). Repeated pressure drop tests indicated 0.17 and 0.99 inches of water per ft., respectively in the two meshes. A 50/50 mixture of the two sizes yielded 0.47 inches of water per ft. and 0.83 inches of water per ft. when approximately 33% volume of PWR-sized pellets were added.

BULK DENSITY AND COMPATIBILITY

A 2/3 volume of alumina carrier and 1/3 volume of pellets was assumed to represent a reasonable production load. Tests were run to determine the bulk density of the furnace matrix using steel pellets as substitutes for fuel pellets. Then, calculations were made to adjust this bulk density to that of the same matrix containing UO_2 pellets. This figure was used to determine the static pressure that might be exerted on pellets in the hot zone of the furnace. A test apparatus that simulated a bed of alumina grain containing a UO_2 pellet was fired at 1700°C for 4 hr. The pellet density was only 89% of theoretical but there was no evidence of the grain adhering to or reacting with the UO_2 not of it sticking to itself. This result confirmed prior experience in the firing of both uranium and mixed oxides of uranium-plutonium contained in alumina sand.

SINTERING TESTS

Even though there were still problems with carrier grain caking, sintering tests were conducted using alumina and urania pellets. The alumina pellets were obtained from a batch that was made during a press set-up, so they varied in green density. They were fired at 1700°C at a rate that exposed them to this temperature for two hours. The theoretical density of the sintered pellets varied from 86 to 96%. This firing was assumed to be successful since the final variance in density could have been caused by the difference in green densitities.

Alumina pellets that were so poorly pressed and so fragile as to be crushed by manual handling were mixed with carrier grain and fired for two hours at 1700°C. Most of the pellets came out of this cycle and through the furnace without physical damage. This demonstrated that the vertical furnace and carrier grain concept should be capable of handling pellets of normal makeup without damage, even to sharp edges. It also verified results of preliminary tests that indicated there was no reaction between the carrier and either alumina or urania pellets.

A few UO_2 pellets were placed in the furnace and were sintered at the same temperature and rate as the alumina pellets. Their sintered density was only 91.3% average, compared with the 94.5% average occurring in pellets from a control firing conducted in a laboratory furnace.

This result indicated inadequate sintering or control, which resulted from UO_2 pellet exposure to either improper timing or temperature. In any event, the partial sintering or caking of the carrier grain caused erratic feed and shut down of the furnace to clean out the blockage. This problem prohibited conducting effective high temperature sintering tests.

TEST CONCLUSIONS

Tests were performed in sufficient number and variety to establish that if the carrier caking problem were eliminated, the VSF could adequately sinter nuclear fuel pellets in an efficient, remote-control mode. The proof-of-principle tests have been completed satisfactorily with this exception: confirmation of an effective muffle configuration and/or carrier composition that will not cake at the higher temperatures.

Only a few preliminary tests were performed on the sintering of nuclear fuels. All of these tests indicated positive results; but additional tests are necessary. Tests that are especially important are those that would verify an adequate environment in the furnace to assure the proper O/M ratio in fuel pellets of differing compositions, large-scale tests to establish throughput rates, and those that would determine whether there is possible refluxing of volatiles from binders, etc.

OTHER APPLICATIONS FOR THE VERTICAL SINTERING FURNACE

The goals for development of the VSF under the FRAD Program included verification of its capability to sinter nuclear fuel pellets. When the FRAD Program ended, research on the VSF was extended to include testing of the furnace concept's adaptability to processes for pellets other than nuclear fuels.

ADAPTABILITY OF DESIGN FOR NUCLEAR WASTE PELLETS

From the start it was felt that the design of a furnace to sinter waste pellets would not be as involved as a furnace to sinter fuel pellets. Waste pellets can be sintered at lower temperatures, in less stringent environments, and under fewer atmosphere control requirements. Waste pellets might, however, require additional furnace provisions for removal and handling of effluents.

SINTERING OF NUCLEAR WASTE PELLETS

A batch of simulated zirconia waste pellets with the theoretical composition as indicated in Table 1 was obtained from Idaho National Engineering Laboratory. These pellets are chemically similar to those made from nuclear wastes, but they contain no radioactivity. The pellets were made on a disc pelletizer using rather large amounts of the liquid phosphoric and nitric acid binders and solid binders such as boric oxide. Since these pellets had only been air dried, most of the components were in the oxide form and the binder compounds had not reacted with the calcine.

Two-Hour Tests

Simulated waste pellets, alone, and a mixture of pellets and alumina grain were put through trial firings in a laboratory box furnace. Both batches were contained in stainless steel cans. The tests were conducted in air at $\sim 850^{\circ}C$ for a period of two hours. There was no indication of either agglomeration of the pellets or of any reaction with the alumina. Another test was conducted at $850^{\circ}C$ for two hours on waste pellets in a die loaded with a weight to simulate a five-foot column of waste pellets and alumina carrier. There was also no indication of agglomeration or distortion of the pellets under this load.

The pellets fired in an air atmosphere for two hours at 850°C changed in color from a tan to a light grey. There were no other apparent changes. A furnace atmosphere of one-third argon-hydrogen and two-thirds nitrogen was used for the waste pellet runs - mainly to assure protection of the furnace windings. The furnace preheat muffle temperature was set at 750°C to drive off most of the volatiles. The "hot zone" of the furnace muffle was maintained at ~850°C. an internal probe indicated a temperature of over 800°C for approximately 10 inches in the muffle so five inches/hr feed rate was used.

TABLE 1. Theoretical Composition of Air-Dried Pellets

Compound	Composition, wt%
ZrO_2	15.0
Al_2O_3	11.3
CaF_2	28.8
CaO	13.3
B_2O_5	3.7
Cs_2O	0.6
SrO	0.5
No_3	6.6
Bentonite	4.0
Metakaolin	1.6
P_2O_5	11.2
H_2O	3.4
	100.0%

The first trial was made with 2 to 3 kgs of waste pellets, which formed a train that was 12 inches long in the muffle. The test pellets were preceded and followed by alumina carrier grain. However, when the pellets came out of the furnace, it was found that the carrier had filtered through the train of waste pellets, thoroughly mixing with the pellets.

After being exposed to a ~800°C temperature for two hours, the zirconia waste pellets changed from a tan to a grey color that varied from light to dark. A few of the pellets were largely black, but when they were broken, this black color was found to be only on the surface. There was no apparent change in size or density of the pellets. The fired pellets were more resistant to breakage, and when broken there were fewer fines.

Extended Tests

The next run was made over a period of several days. During the day shift, waste pellets only were fed into the furnace. The feed was stopped at the end of the shift and the furnace was held at temperature overnight. In this way it was possible to completely fill the muffle with pellets (6 to 7 ft). The run progressed very smoothly the first day with the pellets being fed from the hopper without bridging at a rate of approximately 1.5 kg/hr. The next day the exit gas coil plugged with excessive amounts of condensate and this forced volatiles up through the bed of pellets in the hopper. A plate was installed in an attempt to seal the top of the hopper, and the coil was flushed to clear it. However, the coil continued to plug and the plate did not stop the flow of gas, so condensate was formed and dripped back into the pellets. This caused some of the pellets to stick together and restrict the flow into the furnace muffle. These problems continued for two days before the run was stopped, the temperature was lowered, the exit feeder was taken out, and the remaining pellets were removed from the furnace.

The pellets from this test run had the same general appearance and physical characteristics as those of the previous run. The last portion of this sintering run produced pellets that were similar; however, this part included chunks up to 2 inches in cross section that were formed from an agglomeration of individual pellets. A black coating was apparent on the surface of most of the pellets that formed the agglomerate and throughout the mass. As with the individual black pellets produced earlier, the black color or coating was limited to the surface.

Approximately 20 kg of waste pellets were processed through the furnace and the positive results obtained from this run were that 1) the furnace atmosphere was adequately maintained with a full column of pellets alone (no carrier); 2) the first 10 to 12 kg of pellets processed through the furnace showed no agglomeration and; 3) the waste pellets exhibited no apparent physical damage from passing through the furnace without carrier.

Large Batch Tests

After an insulated cover was installed to seal off the hopper, the outside of the hopper was insulated and a larger coil was installed to handle the liquid condensate, another test run was set up to sinter the zirconia waste pellets at >800^{o}C for two hours at an approximate rate of 1.5 kg/hr. As with the previous tests, waste pellets were fed into the furnace in sufficient amounts (approximately 15 kg) to fill the muffle. Again, carrier grain preceded and followed the waste pellets. Only once during the run was there any indication that the exit coil was plugging. There was also no indication of condensate forming in the feed hopper.

As with pellets from the previous runs, these zirconia pellets had changed in color from a tan to a grey. However, the grey was a lighter shade than in previous runs and there were only a few pellets with black spots or coatings on their perimeters. Other physical attributes were the same as pellets from the previous runs. No agglomerations were present.

ADVANTAGES OF NOT USING A CARRIER

Using the vertical furnace to sinter a product without using a carrier, offers the following advantages:

- more product capacity for given furnace size
- less production waste or scrap generated (replacement carrier)
- simplified operations (no mixing and separation of carrier and product or handling of carrier).

However, the elimination of the carrier in these tests either resulted in or added to the problem of handling the condensate that was driven from the simulated zirconia pellets.

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