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HOT ISOSTATIC PRESSING OF CERAMIC WASTE FROM SPENT NUCLEAR FUEL

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ABSTRACT

Argonne National Laboratory has developed a process to immobilize waste salt containing fission products, uranium, and transuranic elements as chlorides in a glass-bonded ceramic waste form. This salt was generated in the electrorefining operation used in electrometallurgical treatment of spent Experimental Breeder Reactor-II fuel. The ceramic waste process culminated with a hot isostatic pressing operation. This paper reviews the installation and operation of a hot isostatic press in a radioactive environment. Processing conditions for the hot isostatic press are presented for non-irradiated material and irradiated material. Sufficient testing was performed to demonstrate that a hot isostatic press could be used as the final step of the processing of ceramic waste for the electrometallurgical spent fuel treatment process.

INTRODUCTION

Within the Department of Energy, there is a quantity of spent nuclear fuel containing elemental sodium that was used within the fuel elements to provide a thermal bond between the fuel matrix and cladding. This fuel was generated during operation of the Experimental Breeder Reactor II (EBR-II) at Argonne National Laboratory West (ANL-W) in Idaho and Fermi I in Michigan. Both were fast reactors using metallic fuel and sodium coolant. The driver fuel is highly enriched uranium, and the blanket fuel is depleted uranium [1].

The sodium metal within the fuel matrix is highly reactive. Because of its presence, the fuel is generally believed to not be suitable for direct disposal in a geological repository and to require treatment[2]. Argonne National Laboratory has demonstrated the electrometallurgical treatment technology to prepare these fuel types for eventual disposal. The waste salt stream consisted of spent electrolyte (LiCl – KCl eutectic) containing chlorides of the alkali, alkali earth, lanthanide fission products and some actinides[3]. The waste salt was immobilized in a glass-bonded ceramic waste form. In making the ceramic waste form, these salts were first occluded into the pores of the zeolite-4A using a high temperature blending process. Salt loaded zeolite was then mixed with glass frit and loaded into cylindrical stainless steel canisters. These canisters were evacuated and heated to remove residual moisture. The canisters were then sealed. Finally, they were subjected to elevated temperature and pressure in a hot isostatic press (HIP). The resulting ceramic waste form (CWF) was a glass-bonded sodalite suitable for disposal in a repository.

The HIP process was scaled-up to produce CWF samples that measured 11.4 cm in diameter and 15 cm in height. This effort included HIP processing of non-irradiated material and continued through the processing of irradiated material in the Hot Fuel Examination Facility (HFEF). The lessons learned while installing and operating a HIP in a radioactive environment is the primary focus of this report.

HOT ISOSTATIC PRESS MANUFACTURING

A HIP is a high-pressure vessel having closures to seal both ends. The HIP, model MIH-9, used for demonstration-scale testing, was purchased in 1995 from Asea Brown Boveri (ABB) Autoclave Systems Inc. Flow Autoclave Systems Inc. later bought ABB in 1999. The MIH-9 used a monolithic vessel. Though much larger and heavier than a wire-wound vessel, Section VIII-3 of the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code covering wound vessels was not approved at that time. The maximum operating temperature of the unit was dependent on the type of heating element that was installed. The molybdenum heating element provided a maximum temperature of 1450 °C. Maximum pressure of the unit was 200 MPa. The unit had a working diameter of 15 cm and a working depth of 30 cm. The industry would classify a unit with such small working dimensions as a small or "mini" HIP.

These smaller units are typically used in research facilities. The units are assembled with the pressure vessel, the pumps, compressors, and valves all in the same cabinet. The only separate piece of equipment might be an operator terminal. Maintenance on any component that is located in a radioactive hot-cell would be more difficult than on that of a component located out of the hot-cell.

The HIP would be installed in the decon cell in HFEF. While this portion of the hot-cell is designed for occasional entry, the equipment must still be designed for fully remote maintenance and recovery from faulted conditions because failure while processing a radioactive sample would preclude entry due to excessive radiation levels. The in-cell portions of the system must be modularized to simplify maintenance and allow fully remote replacement of components.

To minimize the amount of components that would be located in the hot-cell, the major systems of the HIP unit were evaluated, and the pressure vessel and a few other components (described below) were separated from the system before construction.

The overpressure relief valve and the pneumatic valves were located on a gas panel and attached to the side of the vessel. These valves controlled the pressurization and de-pressurization of the unit. The valves were left with the vessel because once gas entered the vessel, it would be potentially contaminated gas; therefore, if a line or valve leaked, this gas would have to be contained. Additionally, if the valves were located in a "clean" area, the exhaust gas during normal depressurization would have to be monitored and filtered. With the valves in the hot cell, exhaust gas would pass through existing cell monitoring and filtering systems.

A vacuum pump assisted in purging the vessel of impure gas by pulling gas from it. For reasons similar to the pneumatic valves, the vacuum pump was selected to be with the vessel to avoid containing suspect gas. This had the additional benefit of minimizing the length of pipe that was purged before each run, thus reducing purge time.

To allow the HIP to be moved without any dangling cords that might get caught and damaged, the power connectors, and other electrical lines were modified with quick disconnects. Similarly, all cooling hoses and gas lines were modified to have quick disconnects.

Both the thermal barrier and the furnace, shown in Fig. 1, are inserted together into the pressure vessel for a HIP run. The thermal barrier slips over the furnace element and provides an insulated barrier between the furnace and the cooling jacket of the HIP. The thermal barrier was locked to the furnace by using a "keyhole." This design had three studs protruding from the bottom of the thermal barrier. These were inserted into three keyholes on the furnace. When the furnace and the thermal barrier were to be removed together, the thermal barrier was rotated clock-wise. This trapped the studs in the narrow portion of the keyhole. With the studs trapped, the thermal barrier would be removed with the furnace secured inside. To load a sample into the furnace, the thermal barrier would be removed, leaving the furnace in the HIP. The sample would be loaded into the HIP and would rest in the furnace on a raised pedestal. If only the thermal barrier was to be removed, it was rotated counter-clockwise. This enabled the studs to clear the large opening of the keyhole. A new interface was developed to remove the thermal barrier and the furnace. A double J slotted interface was welded to the top of the thermal barrier. A T-Bar handle could be inserted into the slot. The handle could be manually rotated to the desired location.

Fig. 1 Thermal barrier and furnace modified to include lifting fixtures.

A change was made to the cooling system. Normally, the open loop circulates plant water through a heat exchanger and then drains the water to a plant drain. On the other side of the heat exchanger, rust inhibited water is cooled. This cooled water is pumped through the vessel in a closed loop. If the pump failed, an alarm would sound and two valves could be manually switched to allow plant water to directly cool the vessel. This water was still drained into a plant water drain. Since the unit would be in a hot-cell, the water would became suspect once it entered the vessel and could not be drained into regular plant drains. To avoid this pumping failure, a second pump was installed. The second pump would activate if the flow through the vessel was low. The two by-pass valves were disabled.

Typical hydraulic fluid is flammable. Hydraulic actuators are used to move the vessel yoke and to lift the top plug. To prevent the possibility of fluid igniting, the hydraulic fluid was changed to non-flammable hydraulic fluid.

Out of cell the HIP vessel was positioned with a forklift, but no forklift was available in the hot cell. A removable lifting frame was designed so that an overhead crane could lift and position it.

INITIAL HIP OPERATION

In early December 1995, the HIP arrived at ANL-W and was unloaded into research building 789. In January 1996, ABB began set-up and checkout of the HIP in the building.

After checkout, assembling and operating instructions were developed. Step-by-step instructions were developed for the operators of the unit. It became increasing apparent that the existing control system was not compatible with existing data archival techniques. Additionally, the components of the controller were becoming obsolete, making repairs difficult. The HIP control system was upgraded to include a new user interface and an interface to the standard data archival system. As part of this, drawings of the unit were created, including electrical wiring and junction boxes diagrams. Several operators were trained and readied for operation.

During the month of April 1998, the HIP began the Phase I Qualification. The purpose of the Phase I Qualification was to prove that the instructions for operation and assembling the equipment were accurate. Additionally, disassembling and assembling the unit ensured that parts and assemblies fit properly. During the checkout, six additional equipment changes were logged. These included the addition of personal protection barriers to the power leads and modifications of handling equipment for the thermal barrier. The Phase I Qualification was completed and approved in April 1998.

IN-CELL PREPERATIONS

Now that the major HIP components had been modified and the HIP was again fully operational in a research building, other components of the HIP were evaluated to see if they would be functional in a hot-cell. In addition to typical difficulties that are encountered with equipment in-cell, the HIP would be located in the hot-cell between two tables. This restricted access to any component that was lower than a meter from the floor. In this location, access to the left-rear corner would be difficult for the electrical mechanical manipulators (EMM). Components that might fail, wear out, or be damaged were examined. Several additional components were identified that needed modifications.

All of the electrical components were examined and if needed, modified to allow handling by an EMM. As a general rule, all wiring was moved so that it could be removed and installed without snagging on other equipment. Quick-disconnects were added to the end of wires and cords, including the power cord of the vacuum pump. The mounting brackets of limit switches, junction boxes and other electrical components were modified with a handling fixture and/or re-located so they could be removed and installed. Electrical drawings were updated to reflect all changes.

The vacuum pump was located in the left rear of the HIP. Because a vacuum pump is a component that is likely to fail, it was raised to allow easier access. The bolts that secured the pump in place were removed, and tapered pins were used. These kept the pump from moving side-to-side while the weight of the pump held it down. A lifting fixture was added and the pump could now be positioned onto the pins with an overhead crane as shown in Fig. 2.

Fig. 2 Vacuum pump after modifications.

The pneumatic valves on the gas panel were found to be difficult to replace individually because the threaded fasteners needed to be torqued. The panel, shown in Fig. 3, was modified to allow the entire unit to be removed. If a valve failed, the panel would be removed from the vessel and taken to a repair area. There the valve could be repaired or replaced. If the damage was extensive, a spare panel could be installed while repairs on the primary panel continued.

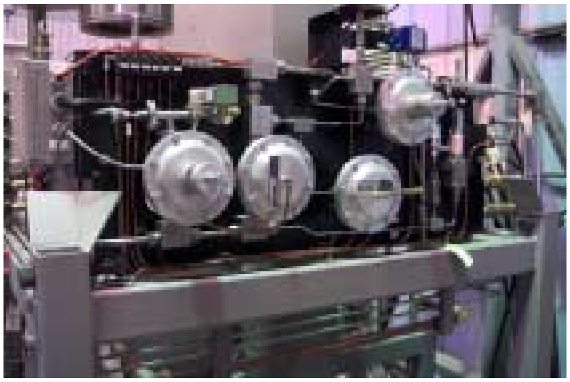

Fig. 3 Gas panel containing pneumatic.

A large oval yoke, weighing almost 2 tons, is nearly one half of the total weight of the in-cell HIP components. The yoke, when centered over the pressure vessel, holds the top and bottom end plugs in place to maintain vessel pressure. The yoke is positioned by a hydraulic ram, located at the bottom of the yoke, near the floor. If the ram failed with the yoke centered over the end plugs, the contents of the vessel could not be removed until after the ram was repaired. To repair the ram, either the HIP would be transferred to a repair area or a manned entry would be required. If the HIP was loaded with a radioactive sample, it could not be transferred to the repair area and a man entry would be prohibited. Additionally, to move the HIP the yoke must be centered over the vessel to maintain the proper center of gravity. If the ram failed with a yoke not centered, the center of gravity would not be correct, preventing the HIP from being transferred. The ram was modified to allow remote removal and assembly. Changing the bolts to pins, adding quick-disconnect, and adding a lifting fixture (see Fig. 4) did this.

Fig. 4 Modified hydraulic ram used to move the yoke.

The top end plug is a precision-machined plug equipped with pressure-sealing O-rings. The plug is located in the top of the pressure vessel and moves up for removal and down for insertion into the pressure vessel by means of a hydraulic ram. The frequent use wears out the O-ring, preventing the plug from sealing pressure, and requiring replacement. Remote changing of O-rings is possible, but due to location would be very awkward in this case. The plug was not altered, but the mounting fixture was modified, as shown in Fig. 5, so that the plug could be removed and taken to the repair area. A spare plug was purchased to allow for processing during maintenance of this item.

Fig. 5 Top end plug with modified mounting bracket.

The keyhole design worked well to lift and position the thermal barrier and furnace by hand, but some additional modifications were required for handling with the in-cell equipment. The T-Bar lifting handle was modified to include a longer handle, an alignment pin and a channel guide as shown in Fig. 6. The studs that secured the thermal barrier to the furnace were easily bent when the EMM was used for turning. Mechanical stops were welded to the furnace to prevent over-rotating the thermal barrier.

When the furnace is lowered into place, furnace contactors engage with protruding power blades located in the bottom of the HIP vessel. Visual markings on HIP vessel were used to align the two. This was difficult to do at a remote distance, and damage to the contactors resulted. To assure that the furnace was properly aligned before the contactors were engaged, pins were added on the top of the pressure vessel and ten centimeter long guide pins were installed in the bottom of the HIP vessel.

Fig. 6 Lifting handle used to remove the thermal barrier and the furnace.

Various designs were evaluated for the canister that would be used to contain powders during HIPing. If a container or a weld on the container failed during processing, the container might expand. If the expansion was sufficient, the canister could damage the furnace elements requiring the installation of a new furnace. As an additional measure of protection, an expansion cage was designed and fabricated (see Fig. 7). This cage was made of mild steel. The sides utilized perforated steel, allowing gas flow. An expansion cage was used in all HIP cycles.

Fig. 7 Mild steel expansion cage used in the HIP to help protect the furnace.

During May 1998, the HIP began Phase II Qualification. The purpose of the Phase II Qualification was to demonstrate that the HIP was ready for installation in HFEF. The qualification demonstrated that fixtures could be remotely handled. Panels, pumps and hoses were connected and disconnected. Items to be HIPed were loaded and unloaded from the HIP vessel. Maintenance items were simulated. The Phase II Qualification Plan was approved in November 1998. Over fifty-five additional drawings were prepared and approved to document the final configuration of the HIP.

Eighty-three canisters were processed through the HIP out-of-cell before it was installed in the hot-cell. The reference material and reference parameters for processing the CWF in the HIP were established. The reference material contained simulated salt occluded into granular (75-250 µm) zeolite 4A at 500°C in a V-mixer. This zeolite was then mixed with 25 wt% of granular glass frit at ambient temperatures. The reference processing parameters began with purging the HIP vessel several times, then ramping at 5°C/min to a temperature of 750°C and then soaking for an hour. This preheated the material allowing the glass frit to soften. The vessel was also pressurized to 2 MPa. This compressed the canister in the axial direction and minimized compression in the radial direction. At the end of the soak, pressure and temperature were again ramped up. The maximum pressure was 100 MPa, because previous runs and modeling proved that more pressure was not required. This cycle had a maximum temperature of 850°C. Both pressure and temperature reached their maximum at approximately the same time. After an hour hold, both pressure and temperature were ramped down. This cycle proved to minimize material cracking.

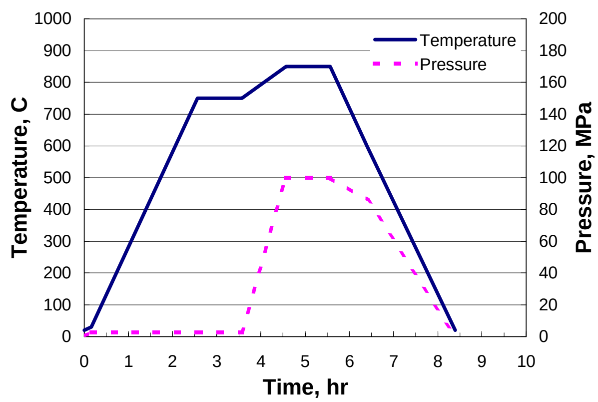

Fig. 8 Reference temperature and pressure parameters for out of cell HIP processing.

Using the reference material and processing parameters, density values of the processed CWF ranged from 2.32 to 2.47 g/cc. XRD indicated sodalite as the major phase with minor phases of nepheline and halite. The microstructure of these HIP canisters was satisfactory. Leach release results were generally low. All of these results are detailed in Ref. 4.

HOT CELL PROCESSING

The HIP was installed in the HFEF decon cell in front of window 5D (see Fig. 9). This is the portion of the cell that has an air atmosphere. A series of rapid venting tests were performed with the HIP in the decon cell. Additionally, these experiments supplied data used in a HIP depressurization analysis. In January 1999, the HIP was pressurized to 34 MPa (5000 psi) and then allowed to vent as quickly as possible. The decon cell pressure changed from approximately -0.62 in. w.g. to approximately -0.55 in. w.g. The next day, the HIP was pressurized to 103 MPa (15000 psi) and then allowed to vent as quickly as possible. The pressure in the decon cell changed from approximately -0.63 in. w.g. to approximately -0.52 in. w.g. With the data from these venting tests, the analysis concluded that the HIP would safely vent pressure and the cell would not become pressurized, even during a worst case scenario.

Fig. 9 ANL-West HIP installed in the HFEF "Hot Cell."

The Phase III Qualification for the HIP was completed in March 1999. This qualified the HIP for in-cell operations. No modifications were made to the HIP during the qualification.

The first seven canisters processed in HFEF were filled with surrogate reference material. The canisters were loaded with non-radioactive material and sealed out-of-cell. All of these canisters were processed using the same reference HIP cycle used previously. These canisters were processed to verify that the HIP had been installed properly and was operating normally. Additional runs were performed examining material issues. A total of fourteen HIP canisters were processed through the HIP in HFEF using surrogate loaded canisters.

The next series of experiments used one-inch containers filled with irradiated powder. The material was prepared in a small furnace in the Hot Fuel Dissolution Apparatus (HFDA) located in HFEF. These were the first irradiated samples prepared for the HIP. The first of these containers was prepared in April 1999.

A plutonium HIP experiment used fifteen small canisters that were filled, evacuated and welded at ANL-East and then shipped to ANL-West to be processed. These canisters were 2.54 cm (1 inch) diameter by 7.62 cm (3 inch) long. Processing of the canisters occurred in May 1999, using the reference CWF HIP cycle and reached a maximum pressure of 100 MPa (14,500 psi) with a maximum temperature of 850 °C. Upon completion of the HIP cycle, the 15 canisters were removed from the HIP. One canister did not compact. A hole was found in the evacuation tube seal weld. The remaining 14 tubes compacted and were sent to be analyzed. The samples support waste qualification testing.

After material preparation equipment was installed into HFEF, a batch of irradiated salt, from the processing of 100 EBR-II drivers, was blended with zeolite and later mixed with 25% glass frit. Canisters were loaded, crimped and welded in HFEF. The first canister loaded with irradiated ceramic waste material was processed in the HIP in June 1999 and is shown in Fig. 10. To demonstrate the repeatability, ten canisters were processed using the reference HIP cycle (see Fig. 8). The tenth canister was processed in August 1999.

After processing the canisters in the HIP, the average density of the irradiated CWF's was 2.43 g/cc with values ranging from 2.33 to 2.51 g/cc. The microstructure of the radioactive CWF is very similar to the surrogate and U/Pu-doped CWF's studied previously. It consists of large sodalite and glass regions with actinide/rare earth/yttrium-containing phases and halite decorating the boundaries between these regions. The uranium, plutonium, yttrium and rare earth fission products are observed in a mixed oxide phase. On a finer scale, this phase tends to be displaced into the glassy regions, distributed uniformly in the waste form.

Based on the 7-day Product Consistency Test (PCT) results, the durability of the radioactive CWF is comparable to good high-level waste glass and is much better than the reference EA glass [5].

Fig. 10 The first demonstration scale canisters, loaded with irradiated material, processed at ANL-W.

A canister was partially processed in July 1999. During processing, the cycle reached 750 °C. From here, temperature ramps up and pressure begins to ramps up. Unfortunately, an incorrect cycle was loaded into the computer and the pressure never ramped up. The cycle was aborted at this point. The canister was re-HIPed in December 1999. The canister had a normal collapse pattern, verifying that a HIP canister could be processed multiple times.

CONCLUSION

In late 1994, ANL began considering using a hot isostatic press as the last step in the final processing of ceramic waste from the electrometallurgical spent fuel treatment process. In support of this, temperature, pressure, and other cycle conditions for a hot isostatic press were developed for demonstration scale operations. The demonstration scale operations were performed first using non-irradiated material. Approximately one-hundred demonstration scale canisters were processed using non-irradiated material. Intermediate and production scale canisters were processed to demonstrate that the CWF could be HIPed on a much larger scale.

A HIP was modified to operate in a radioactive environment. The pressure vessel and other components were evaluated and configured to remain in the hot-cell, while the control cabinet remained out-of-cell. In addition to the standard equipment modifications made to in-cell equipment, modifications specific to a HIP were also implemented. A hot isostatic press was installed in a radioactive environment. Demonstration scale canisters were loaded with irradiated material from the electrometallurgical spent fuel treatment process. Eleven irradiated canisters were processed using identical temperature, pressure, and other HIP cycle conditions.

The compositions and microstructures of the ceramic waste form made with the various salts, both non-irradiated and irradiated, were determined by several techniques: X-ray diffraction, scanning electron microscopy, transmission electron microscopy, and other microscopy examinations. Additionally, the corrosion behavior of both non-irradiated and irradiated samples was monitor with a variety of tests: Material Characterization Center No. 1 (MCC-1) tests, Product Consistency Tests (PCT), density measurements, and cracking measurements. Sufficient data were obtained to demonstrate that a HIP could be used as the final step of the processing of ceramic waste for the electrometallurgical spent fuel treatment process.

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NOMENCLATURE

ABB	=	Asea Brown Boveri
ANL	=	Argonne National Laboratory
ASME	=	American Society of Mechanical Engineers
CWF	=	Ceramic Waste Form
EBR	=	Experimental Breeder Reactor
EMM	=	Electrical Mechanical Manipulator
HFDA	=	Hot Fuel Dissolution Apparatus
HFEF	=	Hot Fuel Examination Facility
HIP	=	Hot Isostatic Press
MCC-1	=	Material Characterization Center No. 1
PCT	=	Product Consistency Test

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