Equipping a Glovebox Form Testing and Characterization of Plutonium Bearing Materials

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EQUIPPING A GLOVEBOX FOR WASTE FORM TESTING AND CHARACTERIZATION OF PLUTONIUM BEARING MATERIALS

Abstract: The recent decision by the Department of Energy to pursue a hybrid option for the disposition of weapons plutonium has created the need for additional facilities that can examine and characterize waste forms that contain Pu. This hybrid option consists of the placement of plutonium into stable waste forms and also into mixed oxide fuel for commercial reactors. Glass and glass-ceramic waste forms have a long history of being effective hosts for containing radionuclides, including plutonium. The types of tests necessary to characterize the performance of candidate waste forms include: static leaching experiments on both monolithic and crushed waste forms, microscopic examination, and density determination. Frequently, the respective candidate waste forms must first be produced using elevated temperatures and/or high pressures. The desired operations in the glovebox include, but are not limited to the following: 1) production of vitrified/sintered samples, 2) sampling of glass from crucibles or other vessels, 3) preparing samples for microscopic inspection and monolithic and crushed static leach tests, and 4) performing and analyzing leach tests *in situ*.. This paper will describe the essential equipment and modifications that are necessary to successfully accomplish the goal of outfitting a glovebox for these functions.

INTRODUCTION

The need for facilities to produce, prepare samples of, and test waste forms that contain plutonium and small levels of other radionuclides is increasing. Facilities to perform this type of work are not plentiful and are decreasing on an annual basis. We have recently brought on-line a glovebox equipped for this type of work. Our goals were to have the following abilities: 1) production of small scale glass and glass/ceramic Pu-containing waste forms, 2) the ability to sample these waste forms once produced, 3) the ability to prepare samples for scanning electron microscopy (SEM) examination, and 4) the ability to conduct in their entirety the ASTM methods C1220-92 [1],

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materials characterization center test #1 (MCC-1), and C1285-94, the product consistency test (PCT) [2].

The above mentioned ASTM methods are tests for the durability and the consistency of waste forms. The MCC-1 is a static leach test to be conducted on a monolithic sample using water or another selected leachant at a specified surface area of sample to volume of leachant ratio, 10 m^{-1}. The PCT is also a static leach test which is performed on a powdered sample using leachants similar to the MCC-1 test with the primary difference being the physical state of the sample when starting the test. The PCT sample is a powdered sample obtained from sieving the material and collecting the size fraction that passes through a 100 mesh screen but does not pass through a 200 mesh, i.e., 150 to 75 µm. The tests are conducted at a temperature of 90 ^{o}C for a time period of 28 days (MCC-1) or 7 days (PCT), although other time periods are allowed.

The tasks delineated above involve several challenges when applying them to plutonium containing samples. It was with a thorough understanding of performing these procedures on the bench top with cold samples that we outfitted the glovebox described in this paper for work on Pu waste forms. This paper will detail the equipment we installed in our glovebox to perform the required tasks involved in the production, testing, and characterization of Pu-containing waste forms.

EXPERIMENTAL ASPECTS

The different operations that can be performed in the glovebox fall in to three categories: 1) waste form production, 2) sample preparation, and 3) leach testing including specialized sample preparation and some characterization methods. These technical areas will be discussed completely below with emphasis on the descriptions of the equipment involved and the operational aspects. An additional section describing the glovebox itself will precede these sections so that the reader is familiar with the glovebox in question.

I) Physical Description of the Waste Form Testing Glovebox

The glovebox that was outfitted and recommissioned as the Waste Form Testing Glovebox (WFT GB) is of stainless steel manufacture by the Stainless Equipment Co. (Englewood, CO). The glovebox was manufactured in the late 1970's and used for other purposes until the late 1980's when it was decommissioned and stored. The recommissioning activities began in 1995 and took 11 months.

The box is 12 ft in length and 4 1/2 ft wide. The total height is 8 ft with approximately 9 ft of the box exhibiting a raised floor at a height of 5 ft. The portion of the box that does not sport a raised floor is located at the far end away from the door interfacing to the bag-out port. The bag-out port contains a window, which is fitted on hinges for use as an entrance to the bag-out port, and a pair of gloves as well as a 15 inch bag-out ring (Central Research Laboratories, WI). Please see Fig. 1 for a side view of the WFT GB. The "deep well" portion of the box was outfitted with a "French can" style transfer port/container,(Central Research Laboratories, WI), for transferal of contaminated materials to other facilities that are similarly outfitted with a minimal threat of contamination release.

Due to the glovebox having a "deep well" portion and there being some equipment stationed on the floor of this portion an overhead crane was positioned in the box. This crane was positioned on tracks and had a lifting capacity of 250 lb.. The control for the crane was located outside the box. The lateral travel range of the crane was the entire length of the box and it was capable of transversing the entire width of the box.

The glovebox is an air flow-through box, with a single HEPA filter at the inlet and a double HEPA filtered exhaust. The static pressure of the glovebox is maintained at 0.5 to 1.0 inches of water negative to the room by means of a suspect exhaust ventilation system.

The glovebox was fitted with some additional features to facilitate operation of the equipment. Electronic feedthroughs were installed allowing various pieces of equipment to be controlled remotely. Because some equipment requires compressed gases for operation, the glovebox was also plumbed with three compressed gas lines--one for argon, helium, and compressed air or nitrogen. These lines are equipped with check valves to minimize the migration of contamination in the gas lines.

II) Waste Form Production Equipment

The production of various glass and glass/ceramic waste forms containing plutonium required a high temperature furnace in the glovebox. A Lindberg/Blue M box furnace, model 51524 (Lindberg/Blue M, Watertown, WI) was installed. The programmable furnace, maximum temperature of 1700°C, has a separate instrument control console enabling it to be remotely controlled. The programmable nature of the furnace maximizes its versatility since the appropriate ramps and hold times could be entered without undue operator interaction. The furnace controller is equipped with an automatic cut-off for high temperatures as a safety feature. This furnace was stationed in the deep portion of the glovebox. This allowed for use of the furnace without compromising the more readily accessible upper working surface. The amount of interaction with the furnace once it was loaded with an experiment was minimal so this arrangement was a wise use of space. Opening the furnace door is administratively controlled so that it does not take place at temperatures exceeding 200°C.

Some of the samples required the application of a hydraulic press before they were heated, i.e., press and sinter. A small hydraulic press, Carver Laboratory Press, Model 3393 (Fred S. Carver, Inc., Menomonee Falls, WI) was used for this operation. The press was operated on the raised portion of the floor and stored in the lower portion. The installation of an "eye" hook facilitated the movement from level to level using the overhead crane.

III) Sample Preparation Equipment

There were several distinct types of samples that were required from the typical waste form in the glovebox. It is pertinent first to understand that the majority of the waste forms were contained in a crucible of some sort, usually of alumina composition. Thus our sampling technology had to be sufficient to deal with the container as well as the sample material. The specific types of samples required for our characterization needs were: 1) small monolithic samples to be mounted and polished for microscopic examination using a SEM, 2) larger monolithic samples, i.e., right circular cylinders 1 cm in diameter and height, and 3) crushed samples in the 150 to 75 µm size range.

An ideal way in which to obtain raw samples for all three needs was to use a modified drill press to core drill samples. Using a diamond core drill allowed for quick sampling of glass and glass/ceramic materials inside the glovebox. A self-contained water circulation system for cooling of the bit using a peristaltic pump for water circulation proved effective and reliable. A commercial drill press was modified at nominal expense, less than five hundred dollars total cost. It was fitted with a core drill bit. A cross vise was installed on the swing table to clamp the samples securely in place, allowing multi-directional adjustments. A splash guard was built around the swing table. Several two-part holders were built using silicone caulk and Plexiglas to hold crucibles for core drilling.

The MCC-1 procedure requires that the monolithic samples have a uniform surface finish. The core drilled right circular cylinder monoliths are prepared in final form in the following manner. The ends of the monoliths are cut square using a Buehler Isomet low speed saw (Buehler Inc., Lake Bluff, IL). The ends of the sample are then polished using a LECO VP-160 polisher/grinder with AP-60 attachment (LECO, St. Joseph, MI). This is an automated polisher which allows for polishing without holding the sample, thus minimizing the threat of a glove tear during the polishing operation. This is driven off the compressed gas line mentioned above, either air or nitrogen works well. The outside of the cylindrical shaped sample is polished using a home built polisher. This polisher consists of a variable speed motor that drives a cylinder containing an open drum with sandpaper installed inside. The drum has a rotating pedestal inside it where the sample sets and is held against the inside of the drum. The sandpaper can be changed by installing a new liner for the drum. Different grit liners were pre-assembled so that a variety of surface finishes could be obtained quickly and easily. Please see Fig. 2 for an end-on view of this polishing device. The monolithic sample is ready for the MCC-1 test when it is completely polished.

The SEM sample is prepared by simply using the saw mentioned above and removing a small piece from a core sample or sawing the core axially if desired. The sample is then mounted using epoxy into a phenolic ring for polishing using the above LECO polisher. Once this is finished the sample is cleaned and removed from the glovebox for coating and examination in the SEM.

The PCT sample is by definition a powdered sample of size fraction -100 mesh to +200 mesh. This is accomplished by crushing a selection of the core drilled pieces using a mortar and pestle. The mortar was modified to minimize the "debris scatter" by mounting it onto a platform and placing a Plexiglas cover over it with an over-sized hole for the pestle in the top. This crushed material is then sorted using an ultrasonic sifter, ATM Sonic Sifter, Model L3P (ATM Corporation, Milwaukee, WI), with the appropriate selection of sieve pans. The method of capturing the fines was modified so that they are

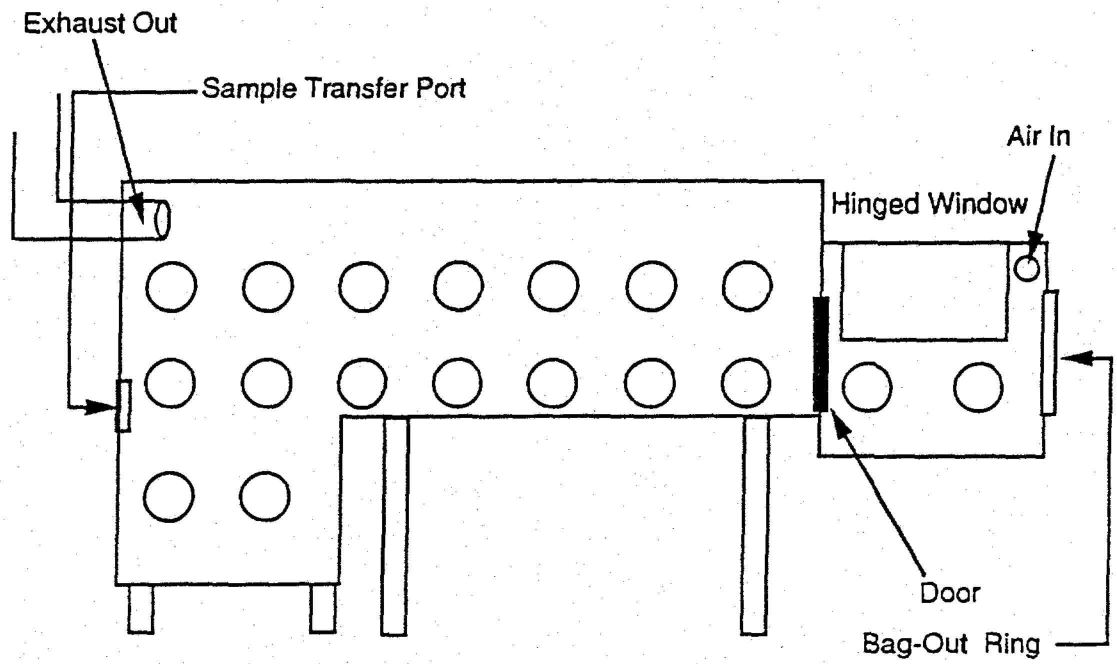

Figure 1. Side View of Waste Form Testing Glovebox.

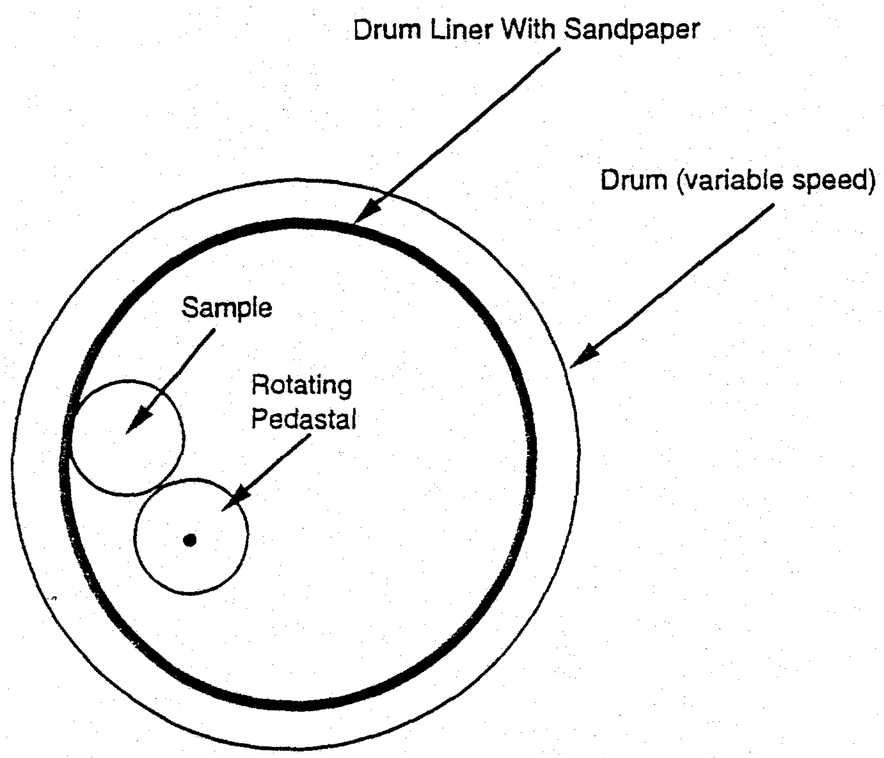

Figure 2. Drum Style Polisher (End View).

collected into a Plexiglas dish and not the flexible bag assembly that is commercially available. This material is then cleaned using the ultrasonic bath, see below, before use in the PCT procedure.

The ultrasonic bath, Fisher Scientific Model FS14H (Fisher Scientific, Pittsburgh, PA) was used anytime cleaning of the sample was necessary. A variety of cleaning fluids was used, but primarily water, methanol and ethanol.

IV) Leach Testing Equipment

It is not the intention of this paper to describe, in full, the MCC-1 or PCT procedures. The reader is referred to references 1 and 2 for that detail. In brief, it is pertinent for the reader to know that both procedures require that the sample be loaded in to a Teflon or steel vessel, the lid tightened and the vessel placed into a constant temperature oven for a set period of time, one or several weeks. It is necessary that the temperature of the oven be monitored for quality control purposes. When the test is completed the sample must be separated from the leachate, the liquid remaining, and the liquid analyzed. The leachate is filtered, manually, in the case of the PCT test using a syringe equipped with a 0.45 μm filter. The leachate solutions can be analyzed for pH and conductivity in the box. The more extensive analysis, such as elemental, require that the leachate be sent to an analytical laboratory facility.

To provide the necessary environment for the leach tests, 90 °C, two Thelco ovens, model number 70DM, (Precision Scientific, Chicago, IL) were selected. The ovens are capable of maintaining +/-2.0 °C uniformity throughout the interior of the oven, including the samples, at the test temperature. Using the manufacturer supplied interface board temperature monitoring is accomplished using a RS232 cable and a PC. The computer is located adjacent to the glovebox and a program written in Quick BASIC is used to gather the temperature data.

An analytical balance was installed into the glovebox for support of the leach tests being run there and also to support the use of special nuclear materials which require an accurate mass measurement. An Ohaus Analytical Plus Electronic balance, model AP 200 (Ohaus Corporation, Florham Park, NJ) This balance had the capability of measuring to +/- 0.1 mg for masses up to 200 g. A set of calibrated weights was also put in the glovebox to enable daily calibration checks.

CONCLUSIONS

A pass-through air atmosphere glovebox outfitted for waste characterization efforts on plutonium containing waste forms is described. This glovebox was equipped to perform the following tasks:

1) Manufacture small quantities of plutonium containing waste forms using a press and a high temperature furnace.
2) Prepare samples for microscopic examination using a SEM.
3) Conduct MCC-1 and PCT tests.

The facility is equipped in such a manner as to perform these functions in a manner consistent with the ASTM standard for plutonium gloveboxes, C-852-93 [3].

REFERENCES

[1] ASTM C1220-92, Standard Test Method for Static Leaching of Monolithic Waste Forms for Disposal of Radioactive Waste, Annual Book of ASTM Standards, Philadelphia, PA, 1995.

[2] ASTM C1285-94, Standard Test Methods for Determining Chemical Durability of Nuclear Waste Glasses: The Product Consistency Test (PCT), Annual Book of ASTM Standards, Philadelphia, PA, 1995.

[3] ASTM C852-93, Standard Guide for Design Criteria for Plutonium Gloveboxes, Annual Book of ASTM Standards, Philadelphia, PA, 1995.

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