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Modified IRC Bench-Scale Arc Melter for Waste Processing

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ABSTRACT

This report describes the INEL Research Center (IRC) arc melter facility and its recent modifications. The arc melter can now be used to study volatilization of toxic and high vapor pressure metals and the effects of reducing and oxidizing (redox) states in the melt. The modifications include adding an auger feeder, a gas flow control and monitoring system, an offgas sampling and exhaust system, and a baghouse filter system, as well as improving the electrode drive, slag sampling system, temperature measurement and video monitoring and recording methods, and oxidation lance. In addition to the volatilization and redox studies, the arc melter facility has been used to produce a variety of glass/ceramic waste forms for property evaluation. Waste forms can be produced on a daily basis. Some of the melts performed are described to illustrate the melter's operating characteristics.

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ACRONYMS

A-40	"average" TRU-contaminated waste (60 wt. %) plus soil (40 wt. %)
ASG	aluminosilicate glass
BWID	Buried Waste Integration Demonstration Program
DC	direct current
DCAM	direct current arc melter
EPA	Environmental Protection Agency
FWF	final waste form
HVPM	high vapor pressure metal
IEB	iron-enriched basalt
IEB4	iron-enriched basalt with ZrO ₂ and TiO ₂
INEL	Idaho National Engineering Laboratory
IRC	INEL Research Center
IWPF	Idaho Waste Processing Facility
PSPI	Private Sector Participation Initiative
RFP	Rocky Flats Plant
RWMC	Radioactive Waste Management Complex
SCFM	standard cubic feet per minute
TRU	transuranic
WSL	Waste Stream Limit project

MODIFIED IRC BENCH-SCALE ARC MELTER FOR WASTE PROCESSING

1.0 INTRODUCTION

This report describes the modified Idaho National Engineering Laboratory (INEL) Research Center (IRC) bench-scale arc melter facility. The facility is used to test various melter processes and to make glass/ceramic waste forms for composition and property tests. Some of these experiments are briefly discussed to illustrate the melter's performance characteristics.

The original IRC arc melter facility¹ was constructed to identify some of the problems that may be encountered in melting materials simulating wastes from the Radioactive Waste Management Complex (RWMC) at the INEL. The design selected allows a wide range in the amount of contact between the electrodes and the melt (non submerged to submerged), while providing the options of predominantly Joule-heating, predominantly arc heating, or a combined heating mode. Due to fiscal limitations, the measurement systems were minimal and much of the instrumentation was borrowed from other projects. However, the increased interest in melting simulated waste materials and in processing conditions has provided the additional support necessary to construct a self-sufficient arc melter facility. Studies of volatilization of high vapor pressure metals (HVPM) and different slag redox states, and production of waste forms with HVPM, transuranic (TRU) surrogates, and cesium, required better offgas control and access to the melt. The next section describes the arc melter, as modified to perform this work.

2.0 MODIFIED ARC MELTER FACILITY

The arc melter facility consists of a stainless steel melt chamber with a removable crucible, three 80 kW DC power supplies, a control and monitoring console, an auger feeder, a gas flow control and monitor, a dual stepper motor electrode drive, a slag sampling system, a thermocouple for measuring the slag temperature, cooling water flow and temperature measurement devices, a data logger for recording measurements with time, video monitoring and recording equipment, an insertable lance for oxidation, an offgas sampling train and control system, and a baghouse filter system. The photograph in Figure 1 shows the system as it was configured for HVPM testing; a schematic of the system is shown in Figure 2. The facility is very flexible and can be used to study many different arc melting processes and to produce a variety of waste forms for further testing.

The arc melter has two graphite, DC-supplied, electrodes that maintain an arc above the melt as shown in Figure 2. The capacity of the melter is approximately 20 kg, with a maximum power input of about 40 kW (limited by the heat transfer capability of the cooling water). In normal

Figure 1. Photograph of the modified bench-scale arc melter.

Figure 2. Schematic of the modified arc melter system.

operation, the electrodes are close to the melt, with short arcs occurring between the electrodes and the melt. The current passing through the relatively high resistance melt results in a predominantly Joule-heating mode with small arc radiation losses to the chamber. The arc gap above the melt can be constantly adjusted by a stepper motor to maintain stable operation.

The water-cooled crucible assembly has a removable stainless steel insert. This design allows fast turnaround of batch operations. The bottom of the chamber consists of a water-cooled stainless steel crucible (10 in. diameter and 14 in. high) immersed in a stainless steel pot through which high velocity water flows. A refractory pot with 1/2 in. thick walls can also be inserted into the crucible if desired. Seven ports are available in the top of the melt chamber, which is also water cooled.

A small entry port through the crucible containment chamber provides thermocouple access to the melt for temperature measurements. A hand-held optical pyrometer is used for melt surface temperature measurements. Thermocouples are installed to measure the heat loss to the chamber, crucible cooling water, electrodes, and offgas. The thermocouple readings, arc voltage, current, and water flows are recorded with time on a data logger. Gas flow rates are recorded by hand. Electrode feed-throughs into the melt chamber are insulated and protected by boron nitride. An ice or liquid

nitrogen cold trap is used to condense volatile particles that enter the chamber exhaust. A high-efficiency baghouse filter collects the remaining condensed and solid particulate before the gas exhausts from the building.

The major modification to the original arc melter was removing the external chamber and enclosing the melt chamber itself. The new facility also includes an auger feeder, a gas flow control and monitoring system, a modified electrode drive, a modified slag sampling system, improved temperature measurement and video monitoring and recording methods, an improved oxidation lance, an offgas sampling and exhaust system, and a baghouse filter system. These modifications allow the arc melter facility to process HVPM, Cs, and TRU surrogates. The modifications made to each part of the arc melter facility are discussed next.

2.1 Melt Chamber

The reactor chamber's external enclosure, employed in the previous test series, was removed to provide more direct access for process control, sampling, and monitoring functions required during HVPM testing. A cover for the top of the crucible now provides containment; offgases from the melt are conveyed to the filter system through closed ducting, reducing personnel hazards associated with high vapor pressure metals. The melt chamber, which opens by lowering, is held to the cover by eight clamps. The chamber is mounted on a hydraulic lift table operated via a foot pedal control. After the melt has cooled sufficiently, the clamps are released and the water jacket and crucible are lowered from the fixed melter cover. This greatly decreases the labor to change crucibles and prepare for another run.

As shown in Figure 3, the melt chamber cover has ample channeling for water cooling, two ports for inserting electrodes, two for viewing the melt, one each for feeding test materials and process air, and one for exhaust. Two auxiliary ports provide for *in situ* melt sampling and temperature monitoring. The view ports are equipped with slide-seal mechanisms that allow the inside surface of the viewing port to be cleaned while maintaining containment. Additional passages were drilled in each slide assembly so that the region between the closed slide and the port could be purged of gases before cleaning the view port window.

Figure 3. Schematic of the melt chamber lid penetrations.

The cooling water control baffle in the stainless steel water cooling jacket was modified to accept a 14 in. high stainless steel crucible. A coat of high-alumina mortar (WO-2813) or a ceramic refractory pot with 0.5 in. thick walls lines the crucible to 1) provide better heat and electrical insulation, minimizing heat transfer through the wall (and thus total heat requirements and melt time), 2) obtain molten metal in the bottom of the melt, and 3) produce slow-cooled waste forms. The ceramic refractory pot can be easily removed, intact, from the stainless steel crucible after melting. The composition of the two refractories used as crucible insulation are shown in Table 1.

Table 1. Composition of the two refractories used to line the crucible of the arc melter.

	Al ₂ O ₃	SiO ₂	TiO ₂	Fe ₂ O ₃	CaO	MgO	Na ₂ O, K ₂ O
Mortar	8.0-62.0	34.0-38.0	0.5-1.0	1.0-2.0	trace	trace	1.0-3.0
Ruby Refractory	93.3				1.4	5.3	

All values in weight percent

2.2 Electrode Assembly

A schematic drawing of the electrode drive system is shown in Figure 4. Tapered-end graphite electrodes, 1 in. in diameter by 6 in. long, connect to a vertical traverse mechanism via copper threaded rods. The traverse mechanism provides a way to manually adjust the position of the electrode assembly during the experiments. An additional stepper motor with a controller was added to the vertical traverse mechanism to allow adjustment of the anode position with respect to the cathode. The electrical connectors are water-cooled brass blocks with brass extension tubes.

The length of the electrodes, crucible height, and volume melted determine how long the test can operate. Depending on the amount of material loaded in the crucible at the start of the run, the length of electrodes is limited to between 6 and 8 in. The melting time is limited by anode electrode erosion, since it is consumed at a higher rate than the cathode. It has been found that coating the electrodes with 0.060 in. of zirconia will inhibit erosion,¹ therefore all of the electrodes used in the modified arc melter have been thus coated. Typical operating times are 2 to 3 h before an electrode change is required.

Figure 4. Schematic of the electrode drive system.

2.3 Auger Feeder

The feed system transports the waste from the material reservoir into the arc melter. This system consists of a volumetric powder feed auger, an adapter tube, and a distribution rotor and assembly. The powder feed auger unit, K-Tron Model S200, is equipped with a 25 mm diameter auger and custom-shortened hopper. The adapter tube is the interface between the auger and the arc melter. The feeder calibration curve is given in Figure 5.

Figure 5. Calibration for the arc-melter feeder.

2.4 Offgas Sampling and Exhaust System

The offgas system consists of a sample train, a parallel cold trap, and a baghouse filter unit (Figure 6). If samples are being collected, the hot offgas exits the system through the offgas sampling train and proceeds to the baghouse filter system. Alternatively, the offgas may exit through the cold trap and then go to the baghouse filter system, bypassing the sampling train. Ball valves located in the lines prior to both the sampling train and the cold trap provide the switching option. The seals in these valves are fabricated from Teflon, chosen for its ability to sustain performance at temperatures between 200 and 300°C. The metal-to-glass connection at the sampling train junction uses a compression fitting combined with Teflon ferrules to assure an adequate seal.

Figure 6. Diagram of offgas sampling and exhaust system.

The cold trap condenses water vapor and captures fine particulate entrained in the exhaust, as well as cooling the exhaust gas to a reasonable temperature. The cold trap consists of a stainless steel trap partially submerged in ice water. The melter exhaust is fed into the trap, exposing the hot offgases to the cold water temperatures. This cools the gases before they enter the baghouse filter system and provides the first opportunity to collect condensable matter. Liquid nitrogen has also been used as the coolant.

The baghouse filter system, Torrit Model VS-550, is used to capture essentially all the particles entrained the exhaust prior to its exit into the building exhaust system. The filter medium can be shaken to collect the particulate matter. The system, which has a capacity of 550 cubic feet per minute, has been modified to include a HEPA filter to ensure total particulate removal.

2.5 Slag Sampling

Slag samples were collected using a quartz tube aspiration device, shown schematically in Figure 7. The aspiration device consists of four components: a quartz tube, a venturi, a vacuum control valve, and a vacuum cut-off valve. The quartz tube is used to collect the sample. Since it is transparent, the operator can see the amount of slag collected. A vacuum is created in the quartz tube as 90 psi air is passed through the venturi. Pressure in the quartz tube is controlled by adjusting the vacuum control valve; the vacuum cut-off valve provides a means to put a vacuum lock on the sample. To prevent slag samples in large tubes from running back into the melt, the arc melter was shut off to allow the sample to become more viscous prior to extraction.

2.6 Data Logger

Various measurements are recorded using a Fluke Model 2285B data logger, with capacity to handle 120 separate data channels. The data logger is remotely controlled by an IBM personal computer, with software provided by Fluke. The active channels on the data logger are programmed to transmit data through the RS-232 communications port to the computer, where it is displayed for monitoring the process as well as stored. Data recorded includes voltage, current, cooling water temperatures, melt thermocouple and optical pyrometer temperatures (optional), and gas flow rates. Records are usually made at 1 min intervals.

2.7 Air Flow Control

Air and other gas flows into the arc melter are monitored and controlled by a MKS M062-LS06E mass flow valve and a MKS 440 series mass flow controller. The mass flow controller displays the flow into the arc melter on a screen located in the instrument rack and on an auxiliary monitor located near the metals sampling train. This allows flow into the arc melter to be

Figure 7. Sketch of slag sampling system.

conveniently monitored by the operators of both the arc melter and the metals sampling train. The flow rate is recorded manually.

2.8 Temperature Measurement

Melt temperature is determined with a Type C, tungsten-rhenium (W-5%Re/W-26%Re) sheathed thermocouple, equipped with an insulating alumina tube, that is periodically immersed in the melt by the operator. The temperature is displayed on a digital monitor. The thermocouple measurements are supplemented with continuous digital readouts of melt surface temperature from an IRCON two-color optical pyrometer. The optical pyrometer is flexibly mounted and can be directed at the melt through one of the viewing ports. The temperature is displayed on its controller, located in the instrument rack. Both temperatures are usually recorded manually by the operator but may also be recorded by the data logger.

2.9 Video Monitor

The arc melter is instrumented with a Pulnix high-resolution color CCD camera [Model TMC-50 RGB (NTSC)] and a Fujinon manual, fixed focal length lens (Model CF 25). Light from the arc melter is attenuated using a 2.5 to 3.0 neutral density filter. Video signals from the camera are viewed on a Sony PVM-1344Q high-resolution, rack-mountable monitor and recorded on a Panasonic AG1960 super VHS tape recorder.

2.10 Air/Oxygen Lance

An air/oxygen lance was purchased to insert into the slag to create oxidizing conditions. However, it was marginally too large to be used with this experimental configuration and its water requirements could not presently be met in the Plasma Processing Laboratory. Therefore, a smaller lance was fabricated to provide submerged air/oxygen injection into the slag. The air lance was constructed from three concentric stainless steel tubes. As shown in Figure 8, the inner and outer tubes were welded to a copper end piece, while the intermediate tube provides a flow path for the cooling water. Air is supplied to the inner tube, which delivers the gas to the copper nozzle. The copper nozzle contains five 0.025 in. diameter holes, four being directed at 30 degrees from center and one on center as shown in the diagram. Air flow into the lance is monitored by a MKS M062-LS06E

mass flow valve and a MKS 440 series mass flow controller. During operation the lance is grounded to the arc melter frame for safety.

Figure 8. Drawing of the air/oxygen lance.

3.0 ARC MELTER EXPERIMENTS

Several arc melter experiments are briefly discussed in this section to illustrate the melter's operating characteristics. The melter has been used to study volatilization of HVPMS, both with and without a cold cap of feed materials, as well as the effects of the reducing or oxidizing state of the melt on the retention of toxic and TRU waste components in the melt for IWPF.² The volatilization experiments investigated the actual effects of high-temperature melting of IEB4/A-40 on the volatile emissions of TRU surrogates (CeO_2 and Sm_2O_3), cesium, and selected toxic and volatile elements/oxides designated as HVPM (Cd, Cr, Pb and Zn). The melter has also been used to prepare samples for waste form devitrification studies for the IWPF, and to produce waste forms for a waste stream limits (heterogeneity extremes) study for BWID and a waste form comparison study for PSPI.

In all these experiments, the feed simulated the residue expected from complete oxidation of TRU-contaminated RWMC waste in an incinerator. This oxidized "average" waste stream has been designated A-0.³ If 40 wt% RWMC soil has been added to A-0, the designation is A-40. Other waste streams of importance, based on the amount stored at the RWMC, are the Rocky Flats Plant sludges described in Table 2.²

The slag that results from melting the waste with soil has a composition similar to basalt but with increased iron, and hence has been called iron-enriched basalt (IEB).³ Adding oxides of Zr and Ti results in a similar composition but a different mineral structure.⁴ This is called IEB4⁵ (because Zr and Ti are from Group IV of the Periodic Table). The Zr/Ti minerals have an affinity for uranium and the TRU radionuclides. The designations IEB/A-40, IEB4/S-40, etc. are used below to identify the waste streams being melted and the addition of Zr and Ti oxides (the amounts of these oxides may vary in different experiments).

Table 2. Principal Rocky Flats Plant sludges stored at the RWMC.

Rocky Flats Plant Sludge	Waste Stream Designation	Content
RFP 741 and 742	H1-0 and H2-0	Hydrated metal oxides
RFP 743	S-0	Calcium silicated organics
RFP 744	P-0	Portland cemented sludges
RFP 745	N-0	Alkali nitrates

3.1 Volatilization without a Cold Cap

For the volatilization experiments without a cold cap, IEB4/A-40 was added to an already melted IEB4/A-80 mixture so that a high-temperature melt in the 1 to 10 Pa.s viscosity range could be obtained initially, with lower temperatures for the subsequent additions. Also, over the course of several experiments, the melted material would approach the composition of the IEB4/A-40 being fed. Feed material was added slowly so that it melted essentially at contact. The effects of melting over a range of temperatures from 1200 to 2300°C were determined. During these experiments, the power input and alkali additions were varied to change the temperature and viscosity of the melt. These tests were conducted with two different types of crucible refractory, a coating of mortar and a ceramic refractory liner as described in the melt chamber section.

During these tests, slag and offgas were sampled to provide information on partitioning of the TRU surrogates, Cs, and HVPM at the various operating temperatures without a cold cap. The offgas was collected in an Environmental Protection Agency (EPA) "Modified Method 5" metals sampling train. Slag samples were extracted with a quartz tube as described above.

A considerable amount of entrained or condensed fine dust was carried along with the offgas. The quantity of dust greatly exceeded the amount of potentially volatile materials and precluded use of the video monitor for most of these tests. This problem may be related to the size of the simulated waste materials, which were pulverized to <1.0 mm particles. Prior experiments without auger feeding had also produced occasional dust storms with a similar size distribution of simulated waste particles (but without HVPM or surrogates). The input air flow rate, about 1 scfm, should not have caused the dust storm. The soil used in the waste feed had not been incinerated, which would have removed the CO₂ and other gases. Small particles in the auger-fed material, which is released at the top of the melt chamber, may have been entrained by the out gas from soil and volatile matter already in the melter.

3.2 Volatilization with a Cold Cap

The cold-cap experiments were conducted with the same IEB4/A-40 mixture to determine the effect of this technique on the emission of the TRU surrogates, Cs, and HVPM, as well as of the more basic species in the mixture. The mixture was fed at a high rate to keep it all from melting. Offgas sampling occurred only over the period of feeding; the offgas was collected in an EPA "Modified

Method 5" metals sampling train. Slag samples were taken before and after feeding to provide information on the partitioning effects at the various operating temperatures with a cold cap.

The amount of entrained or condensed fine dust carried along with the offgas was greatly reduced by the cold cap of waste materials. It was thus easier to see and control the electrode/melt gap and the sample train filters did not plug up and shorten the test period.

With simple experiments such as these, it is difficult to determine the relative importance of the contributing factors to emission from the melter. Future work will investigate additional methods to retain both condensate and particles in the melter.

3.3 Redox Experiments

These tests were performed with IEB4/A-40 with TRU surrogates, HVPM, and Ce to determine the effect of the redox state on retention of the TRU surrogates, toxic, and volatile metals and on the waste form phase structure. Strong oxidation or reducing conditions can drastically affect the minerals formed and the properties of the waste form. Two levels of reducing states were generated by adding different amounts of carbon particles. Two levels of oxidizing states were provided by running the air lance at different flow rates. The results of the volatilization and redox tests are being evaluated.^a

3.4 Waste Form Production

The arc melter facility has been used to produce a variety of different glass/ceramic waste forms. The IWPF studies on devitrification processes and on the optimum amount of Ti/Zr in IEB4 have used the arc melter to generate uncontaminated base material from an IEB/A-40 mixture. Various amounts of TiO₂ and ZrO₂ can be added to this base material, the mixture remelted and processed over a prescribed heat treating schedule, and the resulting phase structure characterized.^{6,7} The Ti/Zr minerals formed scavenge the TRU and U radionuclides into an essentially insoluble species. The amounts of Ti, Zr, and Ca are critical in producing sufficient amounts of desirable minerals and reducing undesired species concentrations.

a. J. W. Sears, J. M. Hillary, P. C. Kong, and A. D. Watkins, "Volatilization and Redox Testing in a DC Arc Melter," unpublished results, August 1993.

The PSPI is doing comparative testing of waste forms, including Portland cement concrete, sulfur polymer concrete, aluminosilicate glass (ASG), IEB and IEB4.⁸ The arc melter is being used to produce ASG/S-40, IEB/A-40, IEB4/A-40, and IEB4/H1-40. The ASG is prepared in the melter without the refractory lining. The predominantly crystalline (ceramic) IEB and IEB4 waste forms are being made with the refractory lining. Immediately after a IEB/IEB4 run, the melter cooling water is turned off and drained, giving a slow cooling condition.

The BWID Waste Stream Limits project is experimentally obtaining the significant properties of waste forms from extremely heterogeneous wastes, such as the H1-, H2-, S-, P-, N-series sludges discussed above. Present arc melter runs include IEB4/H1-0, H1-20, H1-40, H1-60, H1-80, and soil (H1-100) with the same HVPM, Cs, and TRU-surrogates used above.⁹

4.0 MELTER OPERATING CHARACTERISTICS

The report describing the original melter¹ discusses several of its operating characteristics, most of which are unchanged. General observations and additional information regarding electrode erosion and energy balances obtained in the tests described in the previous section are discussed here. These tests are listed in Tables 3 and 4.

Table 3. Operating conditions and electrode erosion data for the volatilization and redox experiments.

Test	Time (min)	Ave. power (kW)	Total A·h	Anode loss (g)	Cathode loss (g)	Anode loss rate (g/A·h)	Cathode loss rate (g/A·h)	Max. Temp. (°C)
Volatility Test without Cold Cap (Mix of IEB4/A-80 and IEB4/A-40)								
HVPM01	169	15.7	230	73.2	43.3	0.32	0.19	1460
HVPM02	125	18.8	223.7	73.2	43.3	0.33	0.19	1650
HVPM03	98	-	-	-	-	-	-	1900
HVPM04	60	16.1	113.4	22.6	9.7	0.20	0.09	-
HVPM05	136	16.3	251	61.1	37.9	0.24	0.15	1600
HVPM06	144	12.7	213	48.9	31.6	0.23	0.15	1460
Volatility Test with Cold Cap (Mix of IEB4/A-80 and IEB4/A-40)								
CC01	62	14.3	96.2	37.3	31.2	0.39	0.32	>2300
CC02	112	9.5	82.2	31	10.7	0.38	0.13	1400
CC03	86	14.4	144.2	69.1	24.3	0.48	0.17	2000
CC04	62	14.6	66.5	35.8	25.1	0.54	0.38	-
CC05	80	13.6	83	48.7	21.3	0.59	0.26	2100
Reduction Tests (IEB4/A-40)								
RED01	110	10.9	103.5	51.5	35.6	0.50	0.34	2000
RED02	95	11.5	106.1	22.8	11.3	0.21	0.11	1800
Oxidizing Tests (IEB4/A-40)								
OX01	115	6.6	84.4	36	3.3	0.42	0.04	1665
OX02	125	12.8	161.3	77.2	51	0.48	0.32	1850
OX03A	95	13.0	36.5	57.3	43.2	1.57	1.18	1700
OX03B	120	13	-	133.1	37.7	-	-	1900

Table 4. Operating conditions and electrode erosion data for PSPI and BWID:WSL waste form preparation.

Test	Time (min)	Ave. power (kW)	Total A·h	Anode loss (g)	Cathode loss (g)	Anode loss rate (g/A·h)	Cathode loss rate (g/A·h)	Max. Temp. (°C)
PSPI Waste Forms								
IEB/A-40-A	72	13.7	88.8	38.4	37.9	0.43	0.43	1622
IEB/A-40-B	102	13.3	138.8	37.3	37.1	0.27	0.27	1750
ASG/S-40-A	43	12.7	34.5	28.5	24.3	0.82	0.70	1600
ASG/S-40-B	105	10.8	114.5	38.1	28.9	0.33	0.25	-
ASG/S-40-C	68	15.5	113.2	39.1	17.8	0.35	0.16	1600
IEB4/A-40-A	28	7.0	26.8	-	-	-	-	-
IEB4/A-40-B	90	5.2	65.7	52.5	40	0.57	0.43	1490
IEB4/A-40-C	135	11.7	161	57.0	25.1	0.35	0.16	1620
IEB4/H1-40	152	13.8	242	49	31.4	0.20	0.13	1650
BWID:WSL Waste Forms								
IEB4/H1-0-A	93	8.1	86.5	37.5	27	0.43	0.32	1020
IEB4/H1-0-B	75	8.2	92.1	16.7	15.4	0.18	0.17	1210
IEB4/H1-20-A	100	10.7	104.8	52.5	42.3	0.50	0.40	1103
IEB4/H1-20-B	118	8.2	106.9	20.9	19.5	0.20	0.18	1414
IEB4/H160-A	113	10.0	106.8	-	-	-	-	1650
IEB4/H1-60-B	75	12.7	127.6	30.7	18.6	0.24	0.15	-
IEB4/H1-80	195	10.8	147.4	68.3	36.7	0.46	0.25	1820
IEB4/H1-100-A	140	9.2	97.9	52.6	11.1	0.54	0.11	-
IEB4/H1-100-B	45	11.8	140	49	31.4	0.29	0.15	-

The temperature range was fairly easily changed by altering the power level or adding alkali. Reliably measuring the temperature with the Type C thermocouple probe posed some problems. Measurements were taken by inserting the thermocouple probe into the slag until the reading stabilized and then removing it. Initial measurements were taken with an uncoated thermocouple, which provided erratic readings and did not survive many measurements. A dual alumina well was subsequently used to protect the thermocouple, resulting in longer life and more stable readings. The maximum melt temperatures for the volatilization tests without the cold cap (Tests HVPM01 through

HVPM06) ranged from 1460 to 1900°C. With the cold cap, there was a marked increase in temperature, even with a decrease in power, and less emission of condensate and particles.

The experiments performed in the arc melter have produced much more auxiliary information than can be evaluated based on the original scope and resources available. Nevertheless, the understanding of the mechanisms and operating conditions increases with each experiment, particularly with those that have new objectives.

4.1 Electrode Erosion

Electrode erosion is one of the factors that limit the operating time of the arc melter. Data from the volatilization-redox experiments and waste form production presented in Tables 3 and 4, respectively, show that the anode consumption rate is usually 25 to 50% higher than that of the cathode. Although it is clear that the anode is consumed faster, without information about arc length or submerged arc, it is difficult to formulate a precise relationship between power input, temperature, chamber atmosphere (redox conditions), and electrode erosion rates.

Table 3 also shows that the electrode erosion rate (in mass per A·h) when using the cold cap is approximately double that without the cold cap. It therefore appears that electrodes submerged in a cold cap will probably have a larger reducing effect on the melt than electrodes held above a liquid melt without cold cap. The consumption rates under the reducing state (Tests RED01 and RED02, no cold cap) appear to lie between those of the volatilization tests with and without the cold cap, but the data are not definitive. Higher consumption occurs with the oxidizing state (Tests OX01 to OX03, no cold cap) than with similar conditions and a cold cap.

In the volatilization runs without the cold cap, it is difficult to see the position of the electrodes above the melt because of the dust. The cold cap material also makes it difficult to observe the position of the electrodes and, therefore, to keep them just above the melt. Some initial oscilloscope measurements were made in an attempt to identify a signature related to electrode position, but they were inconclusive. The problem of determining electrode position should be addressed in future work.

4.2 Energy Balance

An understanding of the energy or heat balance of the system is critical to operating the arc melter. During the volatilization experiments, changing conditions in the melter were evident from energy balance measurements. Crucible heat transfer calculations were made for the original melter.¹ The voltage and current traces with time are also informative and included in the following discussion.

4.2.1 Base Case

During the first volatility tests, a coat of mortar was used to line the stainless steel crucible to provide thermal insulation for the slag. Figure 9 shows typical time variations of voltage and current for the base case with the thin mortar liner and no cold cap covering the melt (Test HVPM02). Figure 10 gives the corresponding heat balance. Each curve in Figure 10 represents a cumulated sum of energy. The bottom curve is the heat loss of the electrodes. The next higher curve is the electrodes plus the chamber lid. The next higher curve includes the above plus the crucible jacket, and the solid curve is the input electrical power. The difference between curves represents the energy loss by the added component. The energy to the melt is the essentially the difference between the input electrical power and the sum of the losses to the crucible, lid and electrodes because the energy carried away in the offgas is negligible due to its low flow rate. Note that the heat losses to the electrodes and the chamber lid are relatively constant.

Figure 9 shows the initial open-circuit voltage and zero current at time zero. Ignition occurs by conduction through a graphite strip immersed in the crushed waste/soil mixture between the two electrodes. During the initial starting period of about 10 minutes, the current is relatively high (with a lower voltage) through the graphite strip to heat the soil or slag sufficiently to form a good conducting path in the slag. As the graphite strip is consumed and the slag conducting path is formed, the voltage increases and the current is decreased to maintain conduction. Often when initiating a melt with new material, foaming would be observed and power would be reduced to limit foam formation. The input power curve in Figure 9 shows such an event at about 10 min into the melt. After another 10 min, sufficient conductivity is attained and the power can be raised to the desired heating level to melt the simulated waste and soil mixture. The power then slowly increases as the melt resistance decreases, the current increases and the voltage drops.

Figure 9. Voltage and current curves showing the power input for HVPM02.

Figure 10. Heat balance for the HVPM02 test (mortar coated crucible, no cold cap).

Similar effects are observed on the energy balance plots in Figure 10. After ignition, the heat losses from the electrodes and the lid increase slowly to a plateau, but are small compared to the energy going to the melt (until steady state is reached) and the crucible. During this period, the power increases rapidly with the energy loss from the crucible close behind as the current drops and the voltage increases. Conduction and radiation from the graphite strip are heating the unmelted mixture. The main portion of the mixture acts as an insulator, so a large portion of the electrical energy goes into heating the melt. As time proceeds, steady state thermodynamics takes over and most of the electrical energy is transferred to the cooling water in the crucible. After 18 min., the power was increased to approximately 21 kW. The heat losses through the crucible lag behind the input power, finally converging after 40 min. At this point the mixture is essentially melted and input power equals losses.

At 70 min., the feed of IEB4/A-40 plus HVPM, Cs, and TRU surrogates commences and the power is raised to 31 kW. Note the fluctuations in the crucible heat transfer that are probably due to the feeding and resultant melting. Note also that the electrode and chamber lid loss values remain nearly constant. At 113 min. the current signal to the data acquisition system was lost, shown by the power curve going to zero. After 125 min, the slag sampling measurements were completed so the power was turned off.

The power input and total loss curves follow each other fairly closely. In this test (Test HVPM02), the mortar provides little thermal resistance to the system; therefore, the heat is quickly transferred from the slag to the cooling water by conduction through the crucible and radiation to the lid.

4.2.2 Refractory Liner Case

Later volatilization tests were performed with a ruby refractory liner (1/2 in. thick). A photograph of the refractory liner containing an IEB/A-40 waste form is shown in Figure 11. The slag waste form usually makes a good bond, without the thin soil skull found in the mortar-lined melts. The refractory is easily removed from the stainless steel crucible, but the waste form is difficult to separate from the refractory.

A typical voltage and current trace with the refractory pot is shown in Figure 12 (Test HVMP06); the typical energy balance is shown in Figure 13. Note that the power input to the

Figure 11. IEB/A-40 waste form produced during the PSPI proof tests.

crucible is lower (about 15 kW instead of 21 kW) for the refractory liner than the mortar, but the time to melt (to get the sum of losses curve to the power input curve) is about the same (40-45 min). Test HVPM06 was a continuation, remelting the material used in the previous test. In remelts, there is little or no foaming and there is less of a problem in getting the solid matter liquified and conducting. As a result, higher currents are possible throughout the initial starting period.

At 50 min into Test HVPM06, the offgas sample train was opened and feeding of the IEB4/A-40 plus additives commenced at an average rate of 7.5 kg/h. Feeding continued for 8 min. The immediate changes in voltage and current are seen in Figure 12, as well as the corrections. The temperature in the melt, measured with a Type C thermocouple, dropped from 1740°C before the addition to 1720°C afterward. In the previous example (Test HVPM02), the higher power level and the less insulating mortar coating did not result in the type of voltage/current response seen in Figure 12 over the 50 to 60 min region.

Figure 12. Voltage and current curves showing the power input for HVPM06.

Figure 13. Heat balance for the HVPM06 test (ruby refractory lined crucible, no cold cap).

At 80 min, one of the two power supplies was turned off and the set point on the remaining unit doubled, resulting in a drop in the melt temperature to 1420°C. Sampling and feeding commenced at 81 min for a period of 11 min. The temperature after feeding was 1460°C. Figure 13 shows the reduced power and energy losses over this period. At the end of the test, the power was increased and the remaining hopper material fed at a high rate to simulate conditions for the cold cap experiments discussed below.

4.2.3 Cold Cap and Refractory Liner Case

The cold cap provides a thermal barrier on the surface of the melt and also shields the arc from the lid. The typical voltages and current traces are shown in Figure 14 (Test CC03). The fluctuations vary between tests and can be large under the conditions employed here (manual control, variable visibility, and feed from the top of the chamber). The corresponding heat loss curves are shown in Figure 15.

For Test CC03, the initial melting was considered complete after 50 min. The power was then adjusted to maintain temperatures around 1800°C. Temperatures of 1870 to 2000°C were measured before the offgas sampling and 1750°C thereafter. The cold cap was initiated by feeding at 168 kg/h for 40-80 s and then reducing the feed rate to 1/4 to 1/2 of that to maintain the cold cap. For Test CC03, the operating parameters are quite stable. Some other tests were less stable and were affected by the manual control limitations. It was more difficult to operate at the low melt temperatures (<1500°C) because of the large changes in conductivity with temperature and the low currents required under these melter conditions.

4.3 Temperature Gradient in Slag

During the first PSPI run on IEB/A-40, measurements were made to quantify the temperature gradient in the molten slag. After operating at 13 kW for about 40 min, the first temperature measurements were taken (labeled t=11:55 in Figure 16). This curve shows that the temperature of the surface of the slag is slightly lower than that just beneath the surface. The data taken 20 min later (t=12:15) show that the system is now closer to equilibrium--the peak temperature is closer to the average temperature. In both cases the temperature is the highest just below the slag surface. This test incorporated a ceramic refractory liner, so a smaller thermal gradient occurs than would be expected without a refractory liner. With time, the thermal gradient will reach a steady state value

Figure 14. Voltage and current curves showing the power input for CC03.

Figure 15. Heat balance for the CC03 test (ruby refractory lined crucible, with cold cap).

Figure 16. Plot of temperature gradient in IEB/A-40 with refractory liner in place at two different times during the PSPI waste form production.

based on the thermal-electrical input, convection within the melt, insulating effect of the refractory, and heat extracted by the cooling water. The temperature profile at 12:15 should be near the steady state value, based on the time passed and the energy balance plots above.

4.4 Slag Cooling and Devitrification of Waste Forms

For the PSPI, BWID-WSL, and IWPF-Redox programs, it was determined that the waste forms should be tested after appropriate heat treatment. The glass waste forms (ASG) should be cooled rapidly to limit the formation of crystals. Glass-ceramic or ceramic waste forms need to be cooled more slowly to give the crystals time to grow. Previous reports have indicated that substantial crystal structures will not form unless they have time to nucleate.^{4,b} A desirable devitrification cycle, developed previously,¹ that provides different temperatures for nucleation and growth is shown in Figure 17. Three general methods for heat treatment are considered for these experiments at this time. The first is a fast-cooling in the mortar-lined, water-cooled crucible, which usually generates a predominantly glassy waste form. The second is slow-cooling with the refractory lining, accomplished by draining the cooling water from the crucible jacket when the power to the electrodes is turned off. This has been designated as refractory slow-cooled and is used to obtain an intermediate crystalline

b. J. Flinn, et al., "IEB Peer Review," TPT bibliography No. T-1840, August, 1981.

Figure 17. IEB devitrification cycle used to promote zirconolite crystal growth.

waste form. The third method, similar to that in Figure 17, is to remove the solidified waste form, remelt it in an oven, hold at a specified temperature(s) for a specified period(s) for crystal nucleation and growth, and air quench.

Typical cooling curves for the first two methods are shown in Figure 18, where a refractory slow-cool performed on IEB/A-40 is compared with a (mortar) fast cool of an ASG/S-40. If the temperature profiles are normalized using the overall temperature difference, the curves will be even further apart. A significantly larger fraction of the material became crystallized in the slow-cooled case, but the fraction was also found to be dependent on the waste loading.^b

The time-temperature devitrification schedule for work now being performed is shown in Figure 19. The schedule is remelting at 1400°C for 1 h, followed by a 16 h soak at 1200°C, a furnace cool slowly to 700°C and soak for 2 h, and furnace cooling to ambient temperature. This devitrification schedule is intended to promote the formation of stable crystals in IEB and IEB4 glass-ceramics to decrease the radionuclide leach rate from these waste forms.

Figure 18. Temperature response of waste forms under slow and fast cooling conditions.

Figure 19. Devitrification schedule used for the Redox, PSPI, and BWID:WSL waste form preparation.

Selected photomicrographs for the fast-cooled ASG/S-40, slow-cooled IEB/A-40, and devitrified IEB/A-40 are shown in Figure 20 at 200x. The ASG/S-40 (Figure 20a) is not as glassy as other slags that have been poured onto cold sand, but the crystalline structure is small. Figure 20b shows the structure of slow-cooled IEB/A-40. The crystalline structure is more evident, with a larger concentration of spinels (white crystals). Figure 20c shows the devitrified IEB/A-40, which looks much like the slow-cooled specimen, only the white crystals are slightly larger. This suggests that the cooling time may not be as critical as originally thought.

(a)

Figure 20. Photomicrographs of (a) fast-cooled ASG/S-40, (b) slow-cooled IEB/A-40, and (c) devitrified IEB/A-40 at a magnification of 200x.

(b)

(c)

Figure 20. Continued

5.0 CONCLUSIONS AND RECOMMENDATIONS

The modified arc melter facility is capable of operating with toxic metals and a wide composition of materials. Processes involving volatilization and dust can be handled safely. Waste forms can be produced with a wide variety of reduced or oxidized states, and in relatively well-insulated refractory pots that can be easily removed from the stainless steel crucible. Various rates of feeding are possible. Slag samples can be taken with minimum effort as standard procedure. Melts can be processed rapidly, with a turnaround period of less than one day. The energy balance can provide useful information about melting, feeding, and maintenance problems.

Additional research is needed in the following areas to improve the control and consistency of arc melting of simulated wastes.

1. The voltage-current characteristics associated with desired electrode/melt configurations, the appropriate power supply characteristic, and the techniques required to control the arc/Joule heating process in the arc melter.
2. The emission of particulate and volatile matter from the melt chamber. This understanding is needed to identify ways to minimize offgas particulate and enhance the retention of HVPM in the melt. This will influence selection of the optimum method of feeding the waste materials into the melter.
3. The role of the skull (cold slag between the refractory and the melt) in minimizing the corrosion and dissolution of the refractory into the molten slag and the buildup of contaminant radionuclides in the skull or refractory.

The present system has been designed to perform the functions necessary for funded research. Additional modifications that would enhance the capabilities of the melter facility include the following.

1. Provisions for tapping the slag for continuous operation and study of the skull/refractory interaction, transient buildup of contaminants, skull removal, and other longer-term processes.
2. An additional tap for molten metal for steady-state metal decontamination studies.
3. Provisions for heating the melt chamber to retard and/or control waste form cooling, thereby eliminating the separate reheating and devitrification steps with their potential

for contamination by the ceramic crucibles for used reheating. An alternative is to rapidly transfer the crucible containing the melt to an oven.

Some of the lessons learned in operating the present arc melter system are the following.

1. For the initial heating period, two 80 kW power supplies set for 400 VOC are required to provide stability due to potentially large shifts in current and voltage. After operation has stabilized, one power supply may be turned off.
2. Operation at low temperatures ($<1400^{\circ}\text{C}$) and low power with the compositions studied is not conducive to stable operation and should be avoided.
3. Temperatures in excess of 2000°C can be easily obtained. The melt temperature may require monitoring with feedback control to maintain the desired set point.
4. When adding feed at low power and with manual control, it may be necessary to increase the power so that the cold, nonconducting feed does not significantly alter the operating conditions or produce a low current mode. For this system 100 A, or more, is recommended.
5. Solidified slag melts more easily and can be operated at higher currents than the initial simulated-waste/soil mixture.
6. Exhaust lines must be cleaned out after each test to ensure that plugging will not occur.
7. The cold trap must be kept at the appropriate temperature range so that ice will not form, but the exhaust gas is sufficiently cooled. A water and ice mix is adequate.

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