Cold Crucible Induction Melter Testing at the Idaho National Laboratory for the Advanced Remediation Technologies Program - 9337

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ABSTRACT

AREVA Federal Services (AFS) is performing a multi-year, multi-phase Advanced Remediation Technologies (ART) project, sponsored by the U.S. Department of Energy (DOE), to evaluate the feasibility and benefits of replacing the existing joule-heated melter (JHM) used to treat high level waste (HLW) in the Defense Waste Processing Facility (DWPF) at the Savannah River Site with a cold crucible induction melter (CCIM). The AFS ART CCIM project includes several collaborators from AREVA subsidiaries, French companies, and DOE national laboratories. The Savannah River National Laboratory and the Commissariat a l'Energie Atomique (CEA) have performed laboratory-scale studies and testing to determine a suitable, high-waste-loading glass matrix. The Idaho National Laboratory (INL) and CEA are performing CCIM demonstrations at two different pilot scales to assess CCIM design and operation for treating SRS sludge wastes that are currently being treated in the DWPF. SGN is performing engineering studies to validate the feasibility of retrofitting CCIM technology into the DWPF Melter Cell. The long-term project plan includes more lab-testing, pilot- and large-scale demonstrations, and engineering activities to be performed during subsequent project phases.

This paper provides preliminary results of tests using the engineering-scale CCIM test system located at the INL. The CCIM test system was operated continuously over a time period of about 58 hours. As the DWPF simulant feed was continuously fed to the melter, the glass level gradually increased until a portion of the molten glass was drained from the melter. The glass drain was operated semi-continuously because the glass drain rate was higher than the glass feedrate. A cold cap of unmelted feed was controlled by adjusting the feedrate and melter power levels to obtain the target molten glass temperatures with varying cold cap levels. Three test conditions were performed per the test plan, during which the melter was operated with a target melt temperature of either 1,250°C or 1,300°C, and with either a partial or complete cold cap of unmelted feed on top of the molten glass. Samples of all input and output streams were collected for analysis. Laboratory analyses and mass balances will be used to determine the fate of feed constituents, especially Cs.

The melter off-gas composition was measured at the melter outlet duct. Sample analyses are still in progress; but preliminary conclusions are possible using the continuous emissions monitoring system (CEMS) data. The concentrations of CO_2, CO, CH_4, total hydrocarbons (THC), and NO_x increased with increasing feedrate of the feed containing water, nitrates, and formate. Over 90% of the formate (a reductant used in the simulant feed) was converted to CO_2 and water vapor. Under 6-9% of the H in the formate converted to H_2, and under 1% of the formate decomposed to gaseous hydrocarbons. This small degree of formate conversion to potentially flammable off-gas species reduces off-gas flammability concerns. About 36-61% of the NO_x in the off-gas (evolved from nitrites and nitrates in the feed) was destroyed.

INTRODUCTION

AREVA Federal Services is performing a multi-year, multi-phase ART project, sponsored by the U.S. Department of Energy, to evaluate the feasibility and benefits of replacing the existing joule-heated melter (JHM) used to treat HLW in the DWPF at the Savannah River Site with a CCIM. Compared to JHMs, CCIMs have features that can lead to (a) higher specific waste throughputs, (b) higher waste loadings in vitrified glass, (c) smaller corresponding volumes of glass that require permanent disposal, and (d) potentially lower overall HLW treatment costs.

The AFS ART CCIM project includes several collaborators from AREVA subsidiaries, French companies, and DOE national laboratories. The key project components and performers include:

- Laboratory-scale studies and testing performed by the Savannah River National Laboratory and the CEA to determine a suitable, high-waste-loading glass matrix

- Pilot-scale demonstrations using existing CCIM test systems operated by CEA in Marcoule, France, and by the INL, to assess CCIM design and operation for treating SRS sludge wastes that are currently being treated in the DWPF
- Engineering studies by SGN to validate the feasibility of retrofitting CCIM technology into the DWPF Melter Cell
- Development of a comprehensive plan (including cost and schedule) for lab-testing, pilot- and large-scale demonstrations, and engineering activities to be performed during subsequent project phases.

TEST OBJECTIVE

The INL Off-gas System Evaluation (OGSE) test was performed to measure the off-gas composition and other properties to provide data to determine how the CCIM retrofit design can interface with, operate within the constraints of, the existing DWPF off-gas control system (1). The fate of radioactive cesium (Cs) is of particular interest to determine its impact on the existing DWPF off-gas system. This is important because the CCIM is expected to operate at a higher temperature range of around 1,250^{o}C, thus potentially increasing the volatility of the Cs, compared to JHMs which typically operate at or below 1,150^{o}C. The simulant recipe includes non-radioactive Cs to measure this key parameter.

Test features that will accomplish this objective include:

- Operate the melter system while feeding a non-radioactive simulant of the SRS HLW sludge waste, during operating conditions selected to represent potential conditions expected for larger CCIM systems. Operating conditions that could influence off-gas emissions and the fate of feed constituents include (a) glass and freeboard temperatures, (b) melt agitation, (c) cold cap coverage, (d) melt residence time, and (g) freeboard gas turbulence and velocity. A single bubbler will be used to provide melt agitation in addition to the convection currents in the glass caused by temperature differentials.
- Determine the melter off-gas source term to the off-gas system. This includes measuring the off-gas composition and also measuring amounts and composition of entrained and volatilized particulate matter at the outlet of the melter, for selected melter operating conditions listed above.
- Determine compositions and masses of the product glass and scrub solution, and the glass REDOX levels as indicated by the REDOX ratio (Fe^{+2}/Fe^{total}).
- Determine the fate of key feed constituents (glass formers, radionuclide surrogates, toxic metal surrogates, nitrates/nitrites, organics, and acid gases).

INL ENGINEERING SCALE CCIM TEST SYSTEM

The INL has developed and constructed a fully integrated CCIM test system including solid and liquid/slurry feed systems, and a complete off-gas treatment system designed to comply with the most rigorous air emission regulatory requirements. This test system includes sampling access and monitoring capabilities to measure and characterize off-gas emissions. It includes an integrated off-gas system that can destroy NO_x and flammable gas emissions and scrub acid gases and particulate matter. It provides the capability to generate data to determine how the feed constituents partition throughout the system. The INL CCIM test system (Figure 1) includes these subsystems:

- Induction power system
- Feed system
- Melter system
- Glass product tapping system
- Cooling water system
- Off-gas control system
- Process monitoring and control system

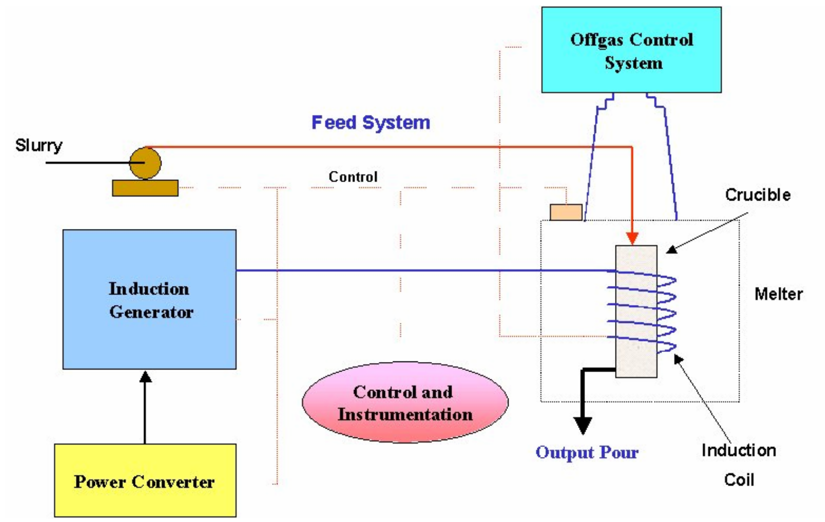


Figure 1. Simplified process schematic of the CCIM test system.

Induction Power System

Major components of the induction power system include a Taylor-Winfield Thermionic C-6000 radio frequency (RF) generator, a coil transfer relay, and the induction coil. The frequency generator itself consists of three subsystems – an enclosed plate transformer unit, a high frequency generator chassis, and a control workstation. The enclosed transformer receives 480 V input power from the INL Engineering Demonstration Facility (IEDF) and supplies nominal 15-kV, 3-phase power to the generator chassis. The 15 kV transformer output, which is rectified with a 3-phase, full-wave bridge in the chassis, is subsequently filtered to provide low-ripple direct current to the triode oscillator. Varying the angle of the firing system allows programming the plate voltage to the triodes, thus controlling output power.

The induction power system provides induction power of up to 60 kW. The power level is adjustable via the control panel or 4–20 mA computer interface continuously over the range of 10–60 kW. The generator can also be configured to provide full power at a single frequency over a frequency range of 200–400 kHz and 1.7–4 MHz. The maximum design power output is 75 kW, but overpower interlocks limit the maximum power to 60 kWe without readjustment. The target frequency for this test, to best emulate the energy deposition profile in the Commissariat a l'Energie Atomique (CEA) CCIM test system in Marcoule, France, is 1.6 MHz (2). The frequency for the OGSE tests has been adjusted to about 1.7 MHz to closely match the target frequency.

Feed System

The test system includes feed systems for delivering liquid, slurry, or solid granular or powdered materials. The liquid feed system includes a feed tank for mixing and feeding liquids or slurries. The feed tank is equipped with a stirrer, sparger, and recirculation loop to maintain homogeneity in slurry feed mixtures. A recirculation pump draws feed solution from the feed tank and recycles it back to the feed tank, which improves mixing in the feed tank. A separate metering pump provides melter feed flow metering and control, drawing feed from the recirculation loop.

Melter System

The melter crucible contains the molten melt material. It is constructed of 304L stainless steel, and consists of three primary components: the lower manifold, the upper manifold, and the cooling tubes. The crucible wall consists of vertical water-cooled tubes oriented to form a crucible cylinder with a nominal 26.7 cm (10.5 inches) internal diameter. The overall crucible height is 40.6 cm (16 inches).

During operation, the water-cooled tubes cause glass adjacent to the tubes to solidify into a solid skull wall that forms the crucible enclosure, while maintaining appropriate temperatures in the crucible walls. The crucible water cooling system can provide adequate cooling at the nominal full generator power (60 kW) and with a glass temperature up to 2,000°C.

Liquid and solid feeds are fed to the top of the molten melt inside the melter crucible, using tubes that penetrate through the melter lid. In this test, only a liquid slurry feed was used. Solid glass frit used at initial startup was added manually prior to test startup. As the water in the slurry feed is evaporated, REDOX reactions occur between reductants and oxidants in the feed, and the dried residues heat to melt temperatures and become incorporated into the melt. New fresh feed is continuously fed. The feed is heated and melted via heat transfer from the molten bath, which in turn is inductively heated by the electrical induction field.

The crucible lid consists of an internal inconel shell that encloses the crucible freeboard space, which is covered with an outer stainless steel shell. The internal space between the inner and outer shells of the lid can be air or steam-cooled, or uncooled.

The melt height inside the crucible varies as the feed is continuously fed and the glass is semi-continuously tapped. The melt height can range between 10.2 – 30.5 cm (4 – 12 inches), but the nominal design height is 26.7 cm (10.5 inches), equal to the melt diameter.

Glass Product Tapping System

As the molten bath volume increases from added melted feed, the glass is drained semicontinuously. The crucible includes a bottom drain assembly, which allows draining the crucible down to about a 4-inch depth, so that a sufficient height of molten glass remains in crucible to remain coupled with the induction field.

The tapped glass exits the tapper and gravity drains into a catch basin. The catch basin is sized to contain the full volume of glass in the melter if necessary, to avoid spilling molten glass onto the laboratory floor.

Cooling Water System

Several of the power supply and melter components are water cooled:

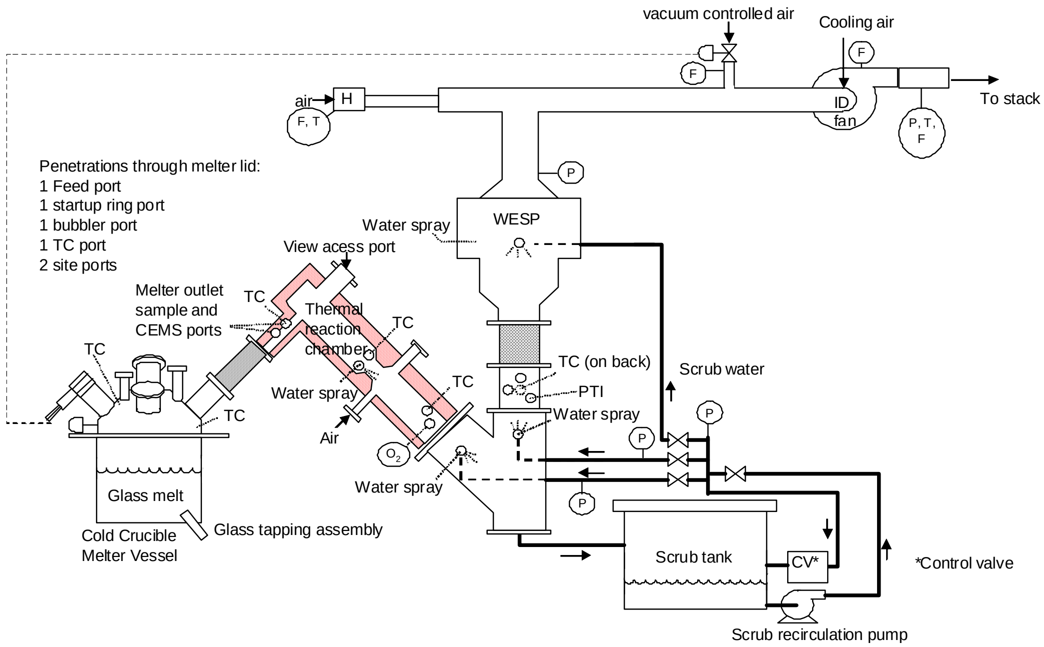
- The RF generator
- The induction coil
- The melter crucible
- Depending on design, the glass tapper

The cooling water system consists of two primary and two secondary loops. One of the primary loops circulates cooling water through the cooled components of the RF generator and the induction coil. This primary cooling loop exchanges heat through a liquid-liquid heat exchanger to a secondary cooling system that includes an outdoor chiller and uses a mixture of water and antifreeze. The other primary cooling loop cools the melter crucible. It transfers heat through a liquid-liquid heat exchanger to a secondary loop that includes an outdoor radiator.

Off-Gas Control System

The CCIM off-gas system is shown in Figure 2. The off-gas system includes these components:

- Heated duct to the thermal reaction chamber (TRC)
- Thermal reaction chamber
- Off-gas quench section
- Wet scrubber system
- Induced draft fan



[ccim offgas system design calculations aug 25 08 08.xls]process diagram 4

Figure 2. INL CCIM test system off-gas control system.

A prototype heated inconel duct connects the melter to the TRC. This 79 cm (31 in) long, 7 cm (2.8 inch) inside diameter duct is angled at 45 degrees from horizontal, and heated using a high-temperature electric resistance heating element to heat the duct wall up to 800°C if desired. This duct is designed to minimize gas condensation and particulate deposition, and, if heated hot enough, will encourage particulate deposits to melt and drain back into the melter. An observation/cleanout port is located in the wall of the TRC opposite the end of this duct, to enable monitoring and cleanout of this duct, if necessary, during operation.

The sample port for collecting process off-gas samples downstream of the melter is located near the outlet of this heated duct, 10 duct diameters downstream of the melter outlet and 1 diameter upstream of the inlet to the TRC, which is a suitable location for isokinetic particulate sampling in small ducts according to U.S. EPA Method 1A ("Sample and Velocity Traverses for Stationary Sources with Small Stacks or Ducts," 40 CFR 60, Appendix A). Two sample points are available for continuous emissions monitoring system (CEMS) measurements. One location is at the inlet of the heated duct, and the other is at the inlet of the TRC.

The TRC is designed to perform nonselective, noncatalytic, thermal NO_x reduction and also fully oxidize any reduced gas species such as H_2, CO, or CH_4. The off-gas is heated in the first stage to NO_x reduction reaction temperatures (of around 800-1,000°C) using an electric immersion heater. The immersion heater has a maximum temperature rating of 1,200°C. The reaction chamber is sized and configured to provide adequate heat transfer surface area, mixing, and residence time to heat and maintain the gas at the design temperature for at least 2-seconds residence time.

The second stage of the TRC uses evaporative cooling of a water spray into the off-gas to cool the NO_x-reduced off-gas to about 800°C, if the TRC is operated in the NO_x-reduction mode. In the oxidizer section, air is normally added to provide oxygen for complete oxidation of CO and other products of incomplete combustion (PICs) that were remain in the off-gas. The oxidizer section is designed consistent with typical efficient thermal oxidizer designs, with a residence time of at least 2 seconds. During this test series, the TRC was not operated for NO_x destruction because off-gas NO_x control was not a test objective.

Following the TRC, the off-gas is quenched by water spray evaporation to the adiabatic dewpoint of the off-gas with the added evaporated water. This section is constructed of Hastelloy alloy to tolerate high off-gas temperatures and to provide good resistance to corrosion at both high and low temperatures.

The off-gas then passes through a wet electrostatic precipitator (WESP) to remove acid gases and particulate matter. The WESP uses electrical energy to charge entrained particulate matter and condensed water droplets, causing them to migrate to the collector walls. The WESP is self-cleaning. The scrubber system includes a scrub tank that collects and holds scrub solution, and the scrub solution recirculation system that pumps scrub solution to the spray quench nozzles in the off-gas quench section, and to the scrubber.

The induced draft (ID) fan provides the motive force to draw the off-gas from the melter through the off-gas system. Since off-gas flowrates may vary widely under different test conditions, the ID fan is equipped with flow/pressure control dampers, and upstream, damper-controlled addition of ambient air to augment and control the total gas flowrate and vacuum.

Process Monitoring and Control System

The CCIM test system is continuously monitored and controlled using a computer-based data acquisition and control system (DACS). The system includes a control computer, LabVIEW software, P&ID displays, the instrument interface, plant instruments, the control interface, plant controls, and a video monitoring system. Key parameters for all subsystems are continuously monitored and controlled. Specific parameters that are continuously measured and recorded include temperatures, pressures, voltages, power levels, flowrates, and off-gas composition.

FEED SIMULANT

Approximately 137 million L (36 million gallons) of high-level radioactive waste (HLW) is stored in underground tanks at the SRS. These wastes are from spent nuclear fuel and target treatment, and contain dissolved salt (dissolved solids) and 8% undissolved sludge in an alkaline solution which consists mainly of metal hydroxides (mostly Fe, Al, Mn, and Ni). This sludge is the waste that is being processed in the DWPF. Sludge Batch 4 (SB4), which is high-aluminum sludge from the HM process, was processed at DWPF until October 2008. The DWPF is now processing Sludge Batch 5.

The simulant feed for the OGSE tests was designed to represent the SB4 DWPF feed. The SB4 simulant used in the OGSE tests is shown in Table I. Glass frit 503-R6 was mixed with the simulant to achieve a waste loading of 46% weight percent of oxides from the waste simulant in the product glass (3). The remaining 54 wt% of the glass is from the added frit. This waste loading is higher than typically achievable in joule heated melters. The calcined oxides composition of the simulant is shown in Table II. The frit composition was tailored to provide the desired concentrations of B, Li, Na, and Si in the glass. The frit composition was 14 wt% B_2O_3, 9 wt% Li_2O, 3 wt% Na_2O, and 74 wt% SiO_2.

Table III shows the calculated composition of the feed slurry including the Frit 503 R6 for a waste loading of 46% oxides in the product glass. This composition is nearly identical to the measured composition of glass from crucible tests with a waste loading of 45% (3). Non-radioactive Cs was spiked into the simulant as surrogate for radioactive Cs. The Cs was spiked at a level such that, assuming no frit addition, the Cs_2O concentration in the glass is 0.5 wt%, high enough to ensure accurate analysis of Cs concentrations.

TEST METHODOLOGY

Tests were performed to accomplish the test objectives. Process conditions were automatically and manually controlled to maintain stable conditions at each test condition. The test system was operated at selected operating conditions, while test data were continuously, electronically logged, and also manually recorded onto data sheets. Samples from process input and output streams were collected for analysis. Two different test series were performed:

- An initial parametric evaluation of operating conditions was performed to enable the operators to determine how to operate each subsystem within target ranges, and in some cases determine what those ranges are.

Table I. SB4 simulant composition (no frit).

Parameter	Units	Value	Comments
Total Solids	gm/L	486	
Insoluble Solids	gm/L	270	
Soluble Solids	gm/L	217	
Calcined Solids	gm/L	298	solids because the hydroxides and nitrates are converted to oxides.
Density (g/ml)	gm/ml	1.30	
pH	---	6.70	
Water content	gm/L	814	Calculated from the density and total solids content.
Cations			
Al	gm/L	43.61	
B	gm/L	< 0.30	
Ba	gm/L	0.22	
Ca	gm/L	7.40	
Ce	gm/L	0.00	
Cr	gm/L	0.35	
Cs	gm/L	0.00	
Cu	gm/L		Not measured
Fe	gm/L	69.21	
K	gm/L	< 0.30	
Li	gm/L	< 0.30	
Mg	gm/L	7.69	
Mn	gm/L	15.02	
Na	gm/L	38.85	
Ni	gm/L	3.30	
P	gm/L	0.05	
S	gm/L	0.81	
Si	gm/L	3.36	
Ti	gm/L	< 0.03	
Zn	gm/L	0.14	
Zr	gm/L	0.20	
Total	gm/L	191.14	
Anions			
Fluoride	gm/L	< 0.13	
Chloride	gm/L	7.93	
Nitrite	gm/L	7.97	
Nitrate	gm/L	49.66	
P2O5	gm/L	< 0.13	
Formate (COOH)	gm/L	90.74	
Sulfate	gm/L	1.35	
Oxalate (C2O4)	gm/L	< 0.13	

Notes:
1. Data source, unless otherwise noted: Stone, Mike, 2008, Savannah River National Laboratory, "Batch 1 SRAT Sample Results.xls" email communication on December 8, 2008.

[CCIM feed comp and emissions estimates, frit 503R6 23dec08.xls]Batch 1 SRAT

- Following the initial parametric evaluation, a selected number of operating conditions was defined for longer-term operation, process measurements, and process sampling to meet test objectives during the Off-gas System Evaluation (OGSE) test series.

The OGSE test series included sample collection for analysis of all input and output streams, so that laboratory analyses and mass balances could be used to determine the fate of feed constituents, especially Cs:

- Simulant feed
- Product glass
- Material recovered from the melter freeboard, lid, and off-gas pipe after the test
- Off-gas and particulate matter entrained in the off-gas
- Scrub solution.

Table II. Calcined solids composition for the simulant for Sludge Batch 4 (no frit).

Parameter		Value	Comments
SRAT product data			
Total Solids (wt. %)		37.4	
Insoluble Solids (wt. %)		20.7	
Soluble Solids (wt. %)		16.7	
Calcined Solids (wt. %)		22.9	This value is not adjusted for the added Cs spike.
Density (g/ml)		1.30	
Water (wt%)		62.6	Calculated from total solids value.
Total Solids wt% calcined			
Al		14.65	
B	<	0.10	
Ba		0.07	
Ca		2.49	
Ce		0	No Ce in SRAT Batch 1
Cr		0.12	
Cs		0.00	
Cu		---	Not measured
Fe		23.25	
K	<	0.10	
Li	<	0.10	
Mg		2.59	
Mn		5.05	
Na		13.05	
Ni		1.11	
P		0.02	
S		0.27	
Si		1.13	
Ti	<	0.01	
Zn		0.05	
Zr		0.07	
Total		64.21	
Oxides (wt%) - calcined solids basis			
Al_2O_3		27.69	
B_2O_3		0.00	"Less than detection limit" values are converted to zero.
BaO		0.08	
CaO		3.48	
Ce_2O_3		0.24	
Cr_2O_3		0.17	
Cs_2O		0.50	Spike added per SRNL recommendation that the glass from the SB4 feed contain 0.5 wt% Cs_2O.
CuO		---	Not measured.
Fe_2O_3		33.25	
K_2O		0.00	
Li_2O		0.00	
MgO		4.29	
MnO		7.97	
Na_2O		17.62	
NiO		1.41	
P_2O_5		0.04	
SO_4		0.82	
SiO_2		2.42	
TiO_2		0.00	
ZnO		0.06	
ZrO_2		0.09	
Total		100.12	

Notes:

1. The anions chloride, nitrite, nitrate, and formate are not included in the calculation of calcined solids, as they are not expected to remain in the calcined solids. Phosphorous pentoxide and sulfate are included because they may stay in the glass.
2. Data source, unless otherwise noted: Stone, Mike, 2008, Savannah River National Laboratory, "Batch 1 SRAT Sample Results.xls" email communication on December 8, 2008.

[CCIM feed comp and emissions estimates, frit 503R6 23dec08.xls]test plan calcine table

Table III. Calculated product glass composition including glass frit 503 R6 for 46% waste loading.

Elemental basis		Oxide basis		Measured glass composition, 45% waste loading from Marra 2008
Element	wt%	Oxide	wt%	wt%
Al	6.74	Al_2O_3	12.73	12.68
B	2.39	B_2O_3	7.71	7.70
Ba	0.03	BaO	0.04	0.04
Ca	1.14	CaO	1.60	1.38
Ce	0.00	Ce_2O_3	0	0.11
Cr	0.05	Cr_2O_3	0.08	0.10
Cs	0.47	Cs_2O	0.50	---
Cu	0.00	CuO	0	0.03
Fe	10.70	Fe_2O_3	15.30	14.42
K	0.05	K_2O	0.06	0.03
Li	2.30	Li_2O	4.96	5.50
Mg	1.19	MgO	1.97	1.38
Mn	2.32	MnO	3.00	2.88
Na	7.20	Na_2O	9.71	10.40
Ni	0.51	NiO	0.65	0.83
P	0.01	P_2O_5	0.02	---
S	0.13	SO_4	0.38	0.43
Si	19.20	SiO_2	41.07	42.05
Ti	0.00	TiO_2	0.01	0.03
Zn	0.02	ZnO	0.03	0.03
Zr	0.03	ZrO_2	0.04	0.05
Total	54.50	Total	99.84	100.03

Notes:
1. No volatilization of any elements is assumed.
2. No anions are included in this calculation.

[CCIM feed comp and emissions estimates, frit 503R6 23dec08.xls]glass with frit

The off-gas was continuously monitored using a continuous emissions monitoring system (CEMS) for O_2 CO_2, H_2, CO, CH_4, total hydrocarbons (THC), and NO_x. The off-gas samples included EPA Method 29 for metals (including Cs) and total particulate matter emissions, and EPA Method TO-14 for redundant H_2 and CH_4 measurement, and measuring speciated volatile organic compound (VOC) emissions. EPA Method 29 was performed isokinetically, and also measured off-gas velocity, flowrate, and H_2O concentration.

OFF-GAS SYSTEM EVALUATION (OGSE) TEST RESULTS

Figure 3 shows a summary of the OGSE test temperatures and power levels. The system was operated continuously over a time period of about 58 hours. Starting glass, manually added to the crucible prior to the test, became essentially completely molten and reached the target operating temperature of about 1,250^{o}C in about 3 hours after induction power startup.

As shown in Figure 4, simulant feed was started at 3.75 hours, and continued for a total of about 52 hours. The simulant feedrate was varied from 1.0 - 4.5 kg/hr.

Three test conditions were performed per the test plan:

1. Essentially complete cold cap, nominal 1,250^{o}C melt temperature, simulant feedrate nominally 3.8 kg/hr.

2. Less complete cold cap, nominal 1,250^{o}C melt temperature, simulant feedrate at nominally 2.3 kg/hr (lower feedrate to enable less complete cold cap coverage).

3. Essentially complete cold cap, nominal 1,300^{o}C melt temperature, simulant feedrate at 4.1 kg/hr (slightly higher feedrate to maintain cold cap).

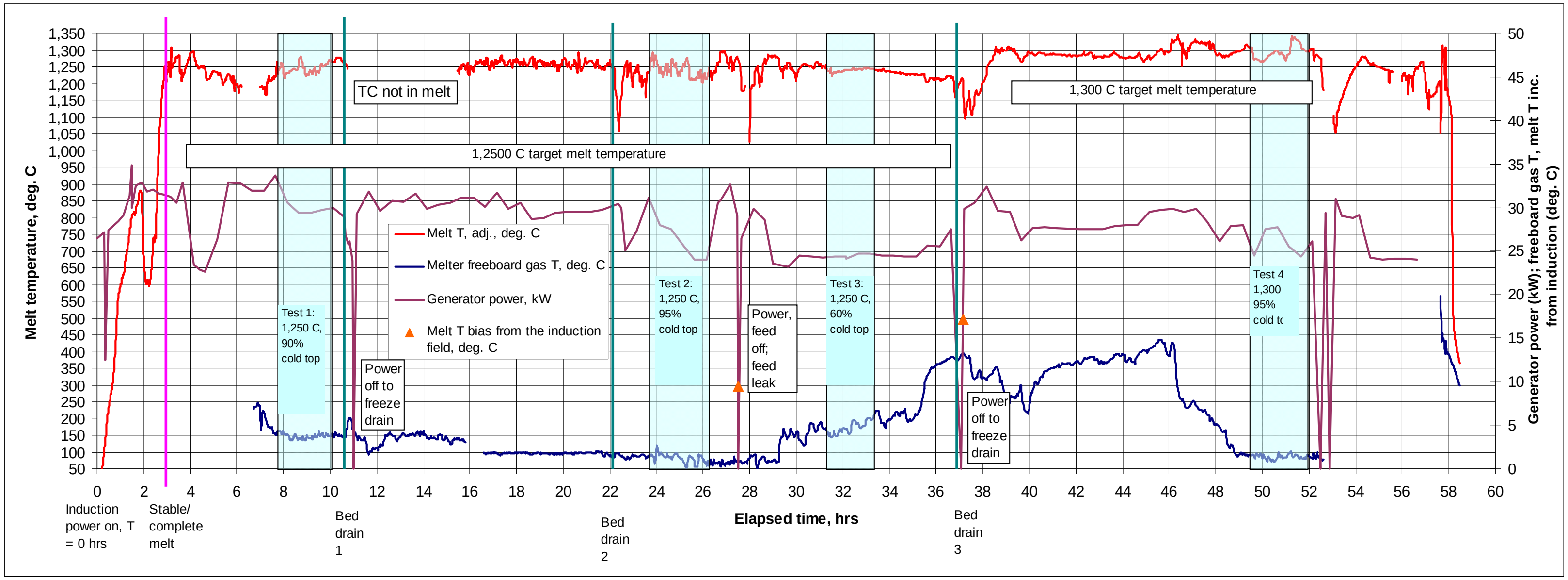


Figure 3. Off-gas System Evaluation test temperature and power levels.

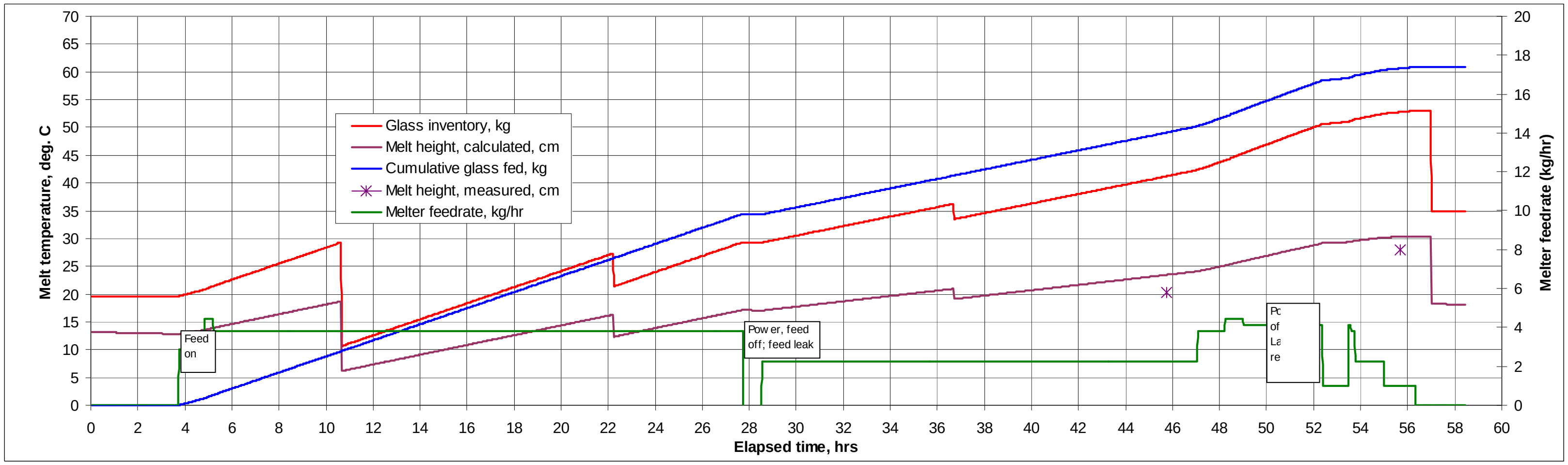


Figure 4. Off-gas System Evaluation test feed and melter parameters.

Off-gas sampling and continuous off-gas concentration monitoring were performed for each of these test conditions. Two sets of off-gas samples were collected for the first test condition.

The average test conditions measured for each off-gas sampling period are shown in Table IV. The melt temperature averaged within 11^oC of the target temperature. The reported melt temperatures have been corrected for a relatively small bias in the temperature measurement caused by the induction field, and average within 11^oC of the target melt temperatures.

Table IV. Melter system conditions during the melter off-gas sampling periods.

					Average operating conditions						Average generator power			Specific feedrate	
Test	Test Con-dition	Start COT	End COT	Duration, hrs	Melt T, ^oC	Cold top coverage, %	Free-board T, ^oC	Calc'd melt height, cm	Feed rate, kg/hr	Melter vacuum, cm H_2O	Anode current, A	Plate voltage, kV	Power, kW	kg/hr/m2 surface area	kg/hr/kW generator power
1	1	7.91	9.87	1.95	1,243	90	146	17	3.8	0.1	5.1	6.0	30.4	68	0.13
2	1	23.66	26.41	2.75	1,239	95	81	15	3.8	0.5	4.7	5.8	27.3	68	0.14
3	2	31.39	33.06	1.67	1,239	70	168	19	2.3	0.5	4.3	5.6	24.4	40	0.09
4	3	48.65	51.91	3.26	1,294	95	89	27	4.2	0.7	4.5	5.9	26.4	75	0.16

Notes:
1. COT = Continuous operating time (elapsed time since test start).
2. The sampling period includes the time during which the Method 29 metals and particulate sampling train and the TO-14 sample are collected, and the time between these two sample collection times.
3. The melt temperature has been corrected for the average amount of bias to the TC reading caused by induction field.
4. The melter cold top coverage is a subjective estimate made by looking through the site ports.
5. The melter freeboard temperature TC is shielded from melt top radiation effects.

[jan09 ccim test data 18feb09.xls]test conditions

A total of 161 kg of feed was processed during the test, producing about 61 kg of product glass. As the feed was fed to the melter, the glass level increased until a portion of the molten glass was drained from the melter. The glass drain could not be operated continuously because the glass drain rate was higher than the glass feedrate. Four glass drains were performed during the test series. Figure 5 shows molten glass being drained from the melter. Residual glass that did not drain out at the end of the test was manually removed from the melter after cooling.

The cold cap coverage was achieved by adjusting the feedrate and melter power levels to obtain the target molten glass temperatures with varying cold cap levels. Figure 6 shows that the specific feedrate can be increased with higher cold cap levels. The melter freeboard temperature was sensitive to the extent of cold cap coverage, because the cold cap reduced thermal radiation from the molten glass.

The melt temperature was measured using a thermocouple (TC) that could be raised or lowered so that it provided a representative melt temperature. Figure 7 shows a vertical temperature traverse in the melter from the bottom of the crucible to the top of the melt, when the melt depth was 28 cm. The melt temperature was relatively constant in the melt between the melt bottom up to 12 cm high (16 cm below the melt surface). The melt temperature TC was typically 5-8 cm above the bottom of the melter, at a depth where the melt temperature was fairly constant.

Simulant and Product Properties

The composition and other properties of the feed simulant, glass product, calcined/sintered material recovered from the crucible walls and lid above the melt and in the off-gas pipe, and off-gas particulate matter are being analyzed at this time. Analytical results presently available include:

- Feed simulant: Average density = 1.36; pH = 5.5; water content = 54.0 wt%; total solids content = 46 wt%; and total calcine = 37.8 wt%. These values are close to the intended target compositions.

- Glass product: Average density = 2.66.

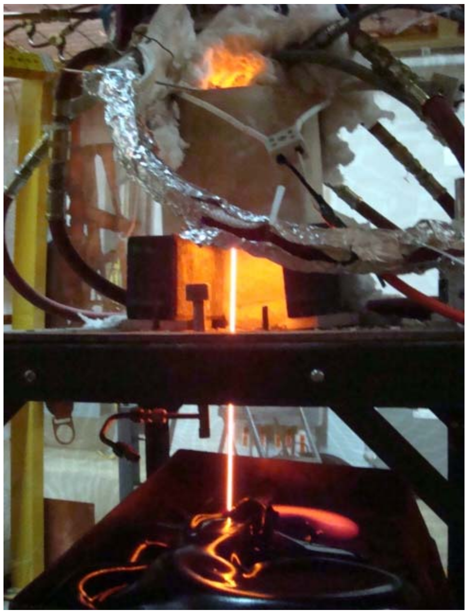


Figure 5. Molten glass draining from the melter through the bottom tap hole.

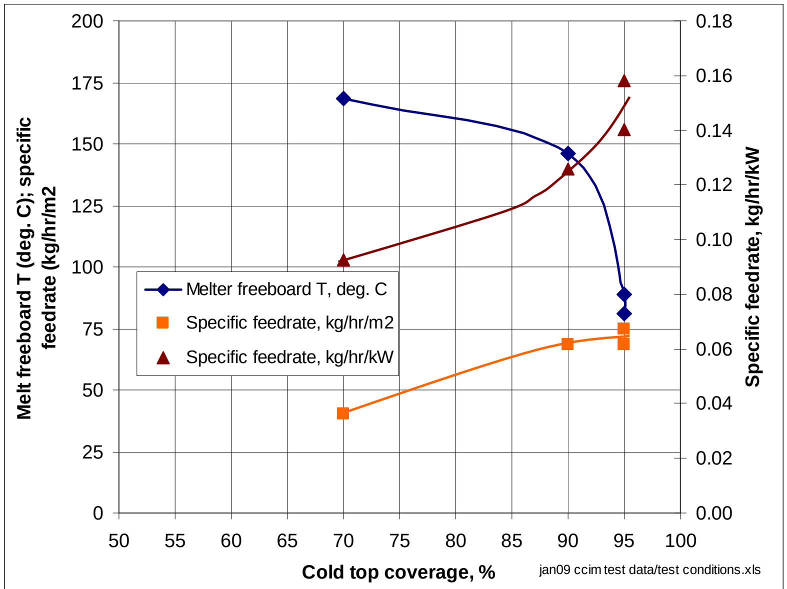


Figure 6. Specific feedrate and freeboard temperature for different cold cap coverage levels.

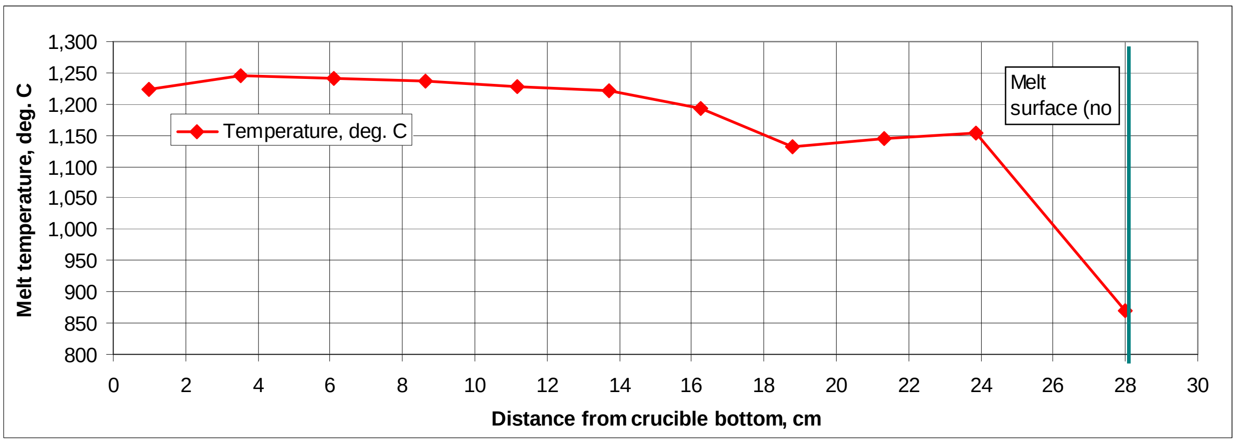


Figure 7. Melt temperature vertical traverse.

Melter Off-gas Composition

The off-gas composition and flowrate were measured at the melter outlet duct, so that the fate of feed constituents including Cs, other metals, total particulate, nitrite, nitrate, and formate could be determined. The off-gas sample analyses are still in progress; but preliminary conclusions are possible using the CEMS data.

Figures 8 and 9 show the melter off-gas composition measurements. The CO_2, CO, CH_4, THC, and NO_x levels tended to increase with increasing feedrate, consistent with the increase in the feedrate. The NO_x level was also higher during Test Condition 3 when the target melt temperature (1,300^{o}C) and the feedrate were higher. The H_2 levels were very close to detection limits based on the range of the analyzer, and so experienced more scatter than the other measurements.

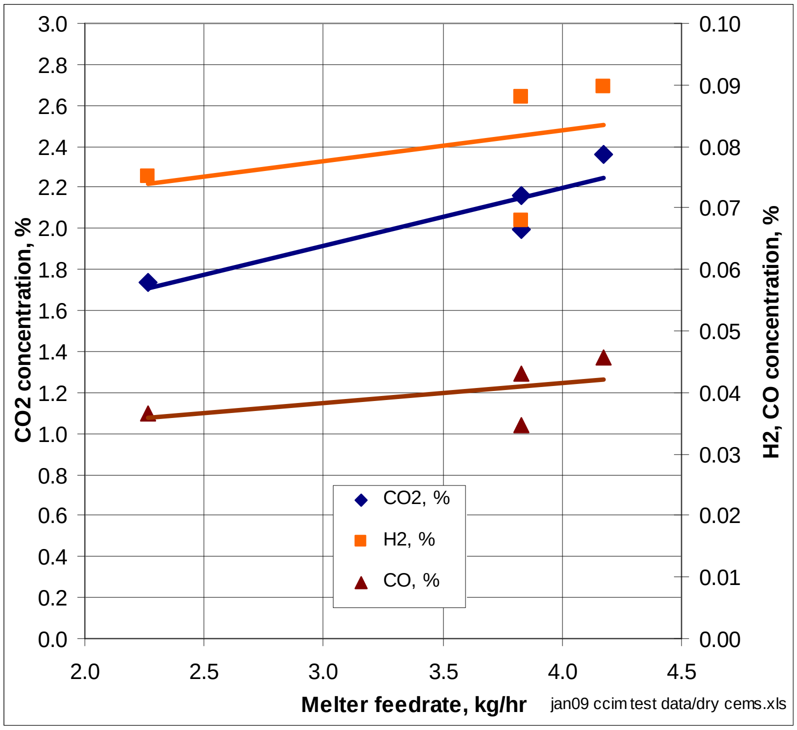


Figure 8. CO_2, H_2, and CO concentrations in the melter off-gas on a dry basis.

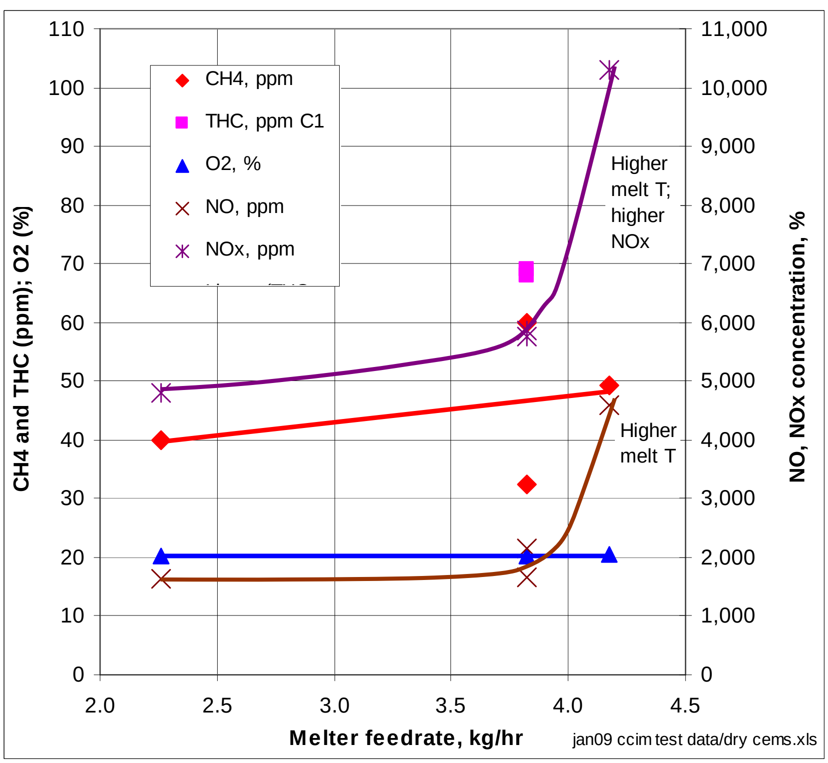


Figure 9. CH_4, THC (reported as CH_4), O_2, NO, and NO_x concentrations in the melter off-gas on a dry basis.

The average dry basis off-gas concentrations for the time periods of the off-gas sample trains trended around:

- O2: 20.2-20.4% (due to dilution of the melter feed from purge air and air inleakage)
- CO_2: 1.7-2.4%
- H_2: 0.07% to 0.09%
- CO: 0.03-0.05%
- CH_4 and THC reported as CH_4, 32-69 ppm
- NO: 1,600-4,600 ppm
- NO_x: 5,800-10,000 ppm

Melter off-gas flammability is sometimes a concern when an organic reductant such as formic acid (HCOOH) is used, as in this test. Figure 10 shows that over 90% of the H in the formate converted to water vapor. Under 6-9% of the H in the formate converted to H_2, and under 1% of the formate decomposed to gaseous hydrocarbons in the melter off-gas. This small degree of formate conversion to potentially flammable off-gas species reduces off-gas flammability concerns.

The formate reductant is added to the feed to achieving the desired glass reducing – oxidizing (REDOX) conditions. The formate, and decomposition products of the formate, also react with nitrite, nitrate, and NO_x decomposition products. Figure 11 shows that a portion (about 57-61%) of the NO_x was destroyed when the target melt temperature was 1,250^{o}C. NO_x destruction was lower, about 36%, for the higher nominal 1,300^{o}C melt temperature.

CONCLUSIONS

Additional results from this test will be included in future reports after receipt of feed, glass, and off-gas analyses. At that time, the preliminary results and conclusions presented here may be revised. Meanwhile, preliminary conclusions are possible from the presently available data.

A simulant of the DWPF SB4 feed was successfully fed and melted in a small pilot-scale CCIM system.

The melter off-gas was characterized at the melter outlet for selected melter operating conditions where the melt temperature was controlled at about 1,250^{o}C and 1,300^{o}C, with controlled levels of cold cap of unmelted feed on the melt surface.

The melter off-gas composition measurements included O_2, CO_2, H_2, CO, CH_4, THC, NO_x. The concentrations of CO_2, CO, CH_4, total hydrocarbons (THC), and NO_x increased with increasing feedrate of the feed containing water, nitrates, and formate.

Over 90% of the formate (a reductant used in the simulant feed) was converted to CO_2 and water vapor. Under 6-9% of the H in the formate converted to H_2, and under 1% of the formate decomposed to gaseous hydrocarbons. This small degree of formate conversion to potentially flammable off-gas species reduces off-gas flammability concerns.

About 36-61% of the NO_x in the off-gas (evolved from nitrites and nitrates in the feed) was destroyed.

REFERENCES

1. SOELBERG, Nick and Jay Roach, “Art CCIM Off-Gas System Evaluation Test Plan,” INL-EXT-08-14449, January 2009.

2. GIROLD, C. et al, CEA memo to the ART CCIM Phase II-A team, “Proposed Configuration and Operating Conditions for the INL Test,” 10 September 2008.

3. MARRA, J. C., et al, 2008, "Glass Formulation Development and Testing for Cold Crucible Induction Melter Technology Retrofit and Deployment Project, Phase II-A – Demonstrations, Lab-Scale Evaluation and Assessment Final Report," Draft, SRNS-STI-2008-00036, October 2008.

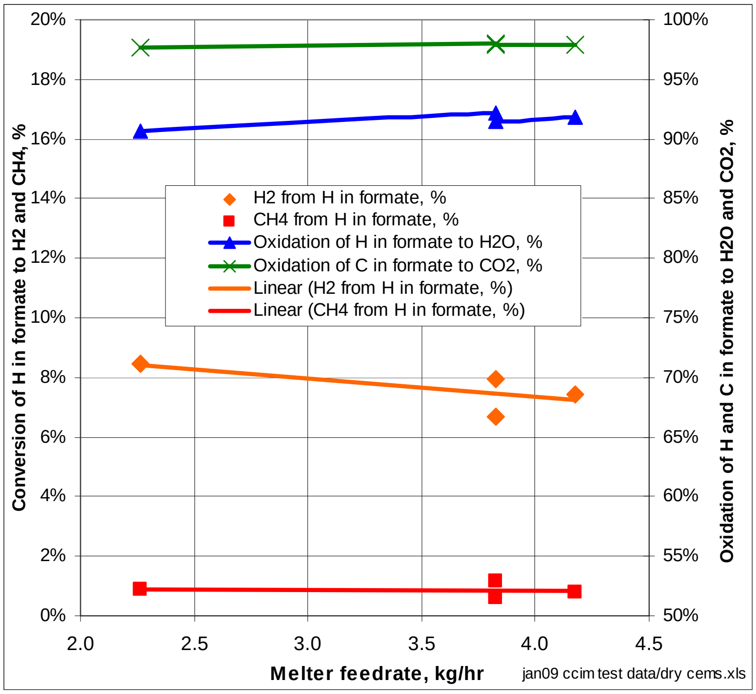


Figure 10. Conversion of C and H in the formate (COOH) in the feed to CO_2, H_2O, H_2, and CH_4.

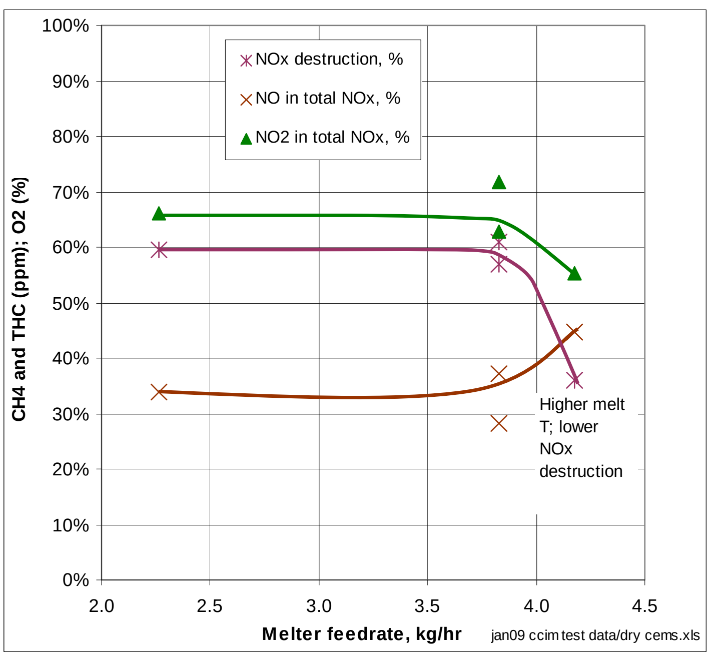


Figure 11. NO_x destruction and NO_x speciation in the melter off-gas.