

COLD-CRUCIBLE DESIGN PARAMETERS FOR NEXT GENERATION HLW MELTERS

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ABSTRACT

The cold-crucible induction melter (CCIM) design eliminates many materials and operating constraints inherent in joule-heated melter (JHM) technology, which is the standard for vitrification of high-activity wastes worldwide. The cold-crucible design is smaller, less expensive, and generates much less waste for ultimate disposal. It should also allow a much more flexible operating envelope, which will be crucial if the heterogeneous wastes at the DOE reprocessing sites are to be vitrified.

A joule-heated melter operates by passing current between water-cooled electrodes through a molten pool in a refractory-lined chamber. This design is inherently limited by susceptibility of materials to corrosion and melting. In addition, redox conditions and free metal content have exacerbated materials problems or lead to electrical short-circuiting causing failures in DOE melters. In contrast, the CCIM design is based on inductive coupling of a water-cooled high-frequency electrical coil with the glass, causing eddy-currents that produce heat and mixing. A critical difference is that inductance coupling transfers energy through a nonconductive solid layer of slag coating the metal container inside the coil, whereas the joule-heated design relies on passing current through conductive molten glass in direct contact with the metal electrodes and ceramic refractories. The frozen slag in the CCIM design protects the containment and eliminates the need for refractory, while the corrosive molten glass can be the limiting factor in the JH melter design. The CCIM design also eliminates the need for electrodes that typically limit operating temperature to below 1200°C.

While significant marketing claims have been made by French and Russian technology suppliers and developers, little data is available for engineering and economic evaluation of the technology, and no facilities are available in the US to support testing. A currently funded project at the Idaho National Engineering and Environmental Laboratory (INEEL), is providing preliminary data on the CCIM technology using a small laboratory unit at the Khlopin Radium Institute in St. Petersburg Russia with INEEL Sodium Bearing Waste surrogate. The task includes both the baseline borosilicate glass and a new iron-phosphate glass developed at the University of Missouri-Rolla, which may offer significant advantages in compatibility with greater concentrations of highly refractory oxides. This project is integrating two disparate advances to develop a system with strong potential for benefit to the Department of Energy. Collaborative development of basic physical parameter data on the CCIM using promising glass formulations is being conducted by University of Missouri - Rolla, Russian and American researchers.

INTRODUCTION

The current technology in use in the USA and Europe for converting highly radioactive wastes into glass for final disposal is based on joule-heated melting. A JHM operates by passing electricity between water or air-cooled electrodes submerged in a molten pool of glass in a brick-lined chamber. This design is inherently limited by the susceptibility to corrosion and melting of the refractory bricks and metal electrodes, respectively. In addition, the glass chemistry must be carefully controlled or it can increase the materials problems and lead to the electrical short-circuiting and glass leaks that have caused failures in test melters.¹ The need for a more rugged glass melter with a broader operating range is recognized across the DOE complex and echoed by Russian researchers, who point out that a new melter concept will be needed to process some of the unique high activity wastes still in storage and unsuitable for processing with their JHMs at Mayak.² Joule-heated melters are simply too limited by materials considerations to allow adequate operating flexibility for cost-effective processing of the wide variety of high-activity wastes in the global nuclear complex. A radically different design that has been in use for several years in Russia for a different kind of wastes is called a cold-crucible induction melter or CCIM. The CCIM design eliminates many of the materials and operating limitations inherent in the joule-heated melter. The cold-crucible design is smaller, less expensive, and generates much less waste for ultimate disposal.^{3,4} It should also allow a much more flexible glass chemistry, which will be crucial if the heterogeneous wastes in the DOE Complex are to be processed.

In contrast to the joule-heated melter, the CCIM design melts glass by induction inside of a water-cooled high-frequency electrical coil. Similar to the heating of food in a microwave oven, the glass absorbs energy from an electrical field caused by the coil without ever actually touching it. Not only does the coil not touch the molten glass, the melt is contained in a water-cooled crucible, which creates a coating of solidified glass that protects the crucible from corrosion. The frozen glass in the CCIM design also contains the molten pool in a shell, or so called "cold-skull" of the same chemistry, eliminating the need for any refractory. Change out of the corroded refractories and electrode materials from a failed melter results in significant radiation exposure and lost operating time during maintenance, as well as generating voluminous highly radioactive cumbersome wastes. Much of this is eliminated with the CCIM design. These benefits present a compelling argument for more thorough evaluation of a potentially next-generation design for high-level waste vitrification.

SYSTEM DESIGN AND DESCRIPTION OF MAJOR COMPONENTS

Electromagnetic induction is a method of heating conductive materials that relies on electrical currents that are induced into the material being heated. These currents, called eddy currents, heat the material. A generic electromagnetic induction furnace, or melter, consists of a few major process components: a melter body to physically contain the material being melted, an induction coil, and a high-frequency alternating current power source called an induction generator. Another required component is a power supply or power conversion system that conditions commercial 50 or 60 Hertz line power to meet the input requirements of the high frequency power source. A "cold-crucible" melter physically contains the molten material within a structure of cooling coils, or cooled panels forming a crucible internally coated by a frozen layer of the feed material. An offgas treatment system is required for applications where hazardous or radioactive volatiles may be released. In addition, continuous process CCIM systems require a means to pour the resulting melt into storage or transport containers. An integrated control and engineered safety system will insure safety and melt quality.

Figure 1 is a generic block diagram of a cold crucible induction heated melter system. Note that there are eight major system components. Each of these components performs a key function, and together they form a system. The system configuration and each of the components perform distinct functions and must respond to certain design requirements to ensure overall system performance.

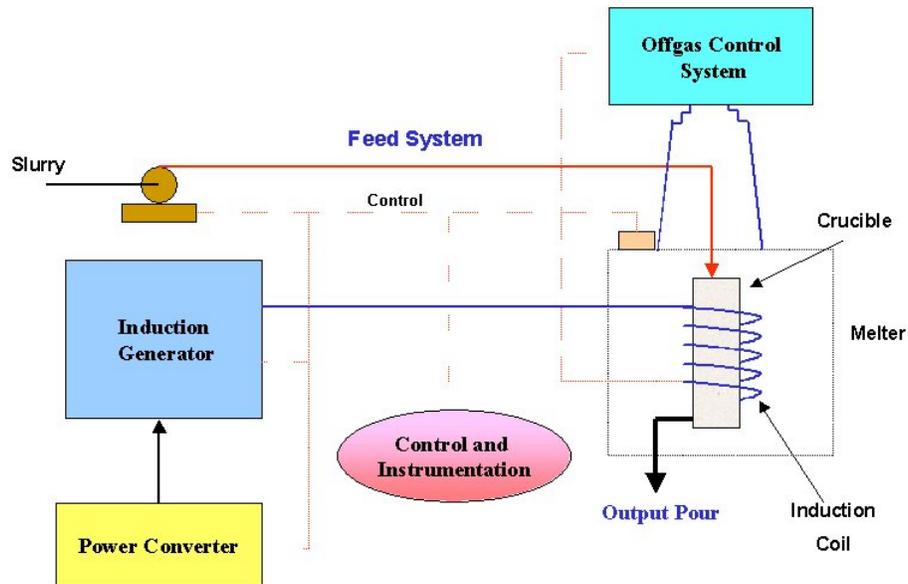


Fig. 1. Cold Crucible Induction Melter System

CCIM Crucible

The crucible contains the melt material. In a CCIM system the crucible consists of cooled sections, closely spaced, but not touching, configured such that an internal volume is created. This can be a simple right-cylinder, oval, or a more complex shape such a cruciform. An example is shown in Figure 2, a photograph taken in Dr. Albert Aloy's laboratory at the Khlopin Radium Institute, St. Petersburg, Russia.

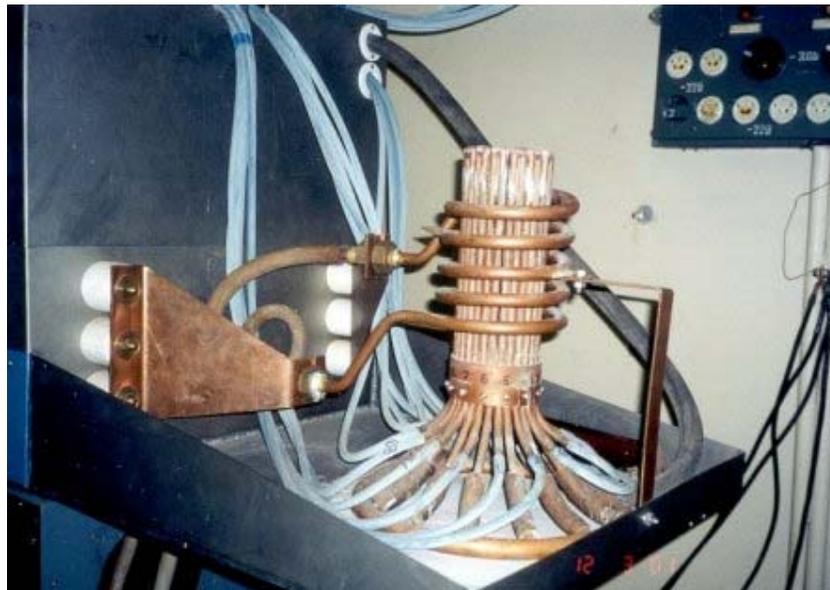


Fig. 2. Cold-crucible at the Khlopin Radium Institute

The vertical copper tubes forming the walls of the crucible in the photograph are parallel cooling loops fed by the copper ring manifold that acts as a supporting base for the crucible. The hairpin cooling loops act as the staves of a wooden barrel. The staves do not touch, but are sufficiently close that by freezing a layer of glass in direct contact with the cooled surfaces they effectively seal the crucible. Because the cooling loops do not touch, they cannot conduct current around the crucible, and are essentially transparent to the induced field. Thus, the inductive field generated by the surrounding coil passes through the crucible wall and interacts with the crucible contents. The material to be melted is placed inside the crucible assembly. When the high-frequency induction coil is energized, the field generated induces eddy currents in any conductive material in the crucible. For non-conductive glasses and glass forming materials, startup will require the addition of sacrificial materials conductive at ambient temperatures such as metal, carbide, or graphite. As heating begins, the coolant removes enough heat such that the material directly adjacent to the wall remains unmelted or freezes, forming a protective shell known as a "cold-skull," that encapsulates the melt around its periphery and on the bottom.

There are a number of crucible design issues. The crucible must contain the waste feed prior to initiation of melting as well as the molten pool during processing. It must allow the varying magnetic field associated with eddy current heating to pass through in a known and predictable manner from the external induction coil to the waste matrix inside the crucible. It is desirable that electrical energy losses due to the heat exchanger be as small as possible. The crucible must remove heat rapidly enough to maintain a solid glass layer (or unmelted feed particulate) at the external surface of the melt. The coiling coils must also preserve their own integrity by maintaining their surface temperature below the material melting point. Lastly, the coil materials must be corrosion resistant to the feed material on top of the glass (liquid, slurry, paste or solids) as well as gases and any condensate that may contact the crucible in the plenum over the waste. Corrosion is reduced due to the relatively cool crucible surface, but the materials of construction must withstand gases that may be highly acidic or basic, as well as containing free halogens like fluoride and chloride. Note that the crucible need not be operated at temperatures that cause significant condensation of the offgases and reflux of acidic solutions. The crucible wall can be maintained at any temperature within the acceptable range of the materials of construction.

Induction Coil

As noted above, the induction coil is placed outside the crucible. Referring once again to Figure 2, it can be seen that the coil surrounds the crucible. The purpose of the induction coil is to transform the output of the high frequency generator into a varying magnetic field. The magnetic flux passes through the crucible and produces eddy currents in the conductive contents inside the crucible. The eddy currents are dissipated by the electrical resistivity of the material causing joule-heating and melting by the following relationship:

$$\delta = C(\rho/\pi\mu f)^{1/2} \quad (\text{Eq. 1})$$

Where:

δ - is the effective skin depth

C – is a proportionality or conversion constant

ρ - is the electrical resistivity of the melt

μ - is the magnetic permeability of the melt

f - is the operating frequency of the melter

Approximately 86% ($1-1/e^2$) of the energy transferred by the induction coil is deposited in the skin-depth estimated by this equation. Thus, design of an induction coil is not trivial. It should be configured (diameter, number and spacing of turns) to maximize efficiency of energy transfer from the generator to the melt. The design is constrained by the physical dimensions of the crucible, the electrically conductive

properties and volume of the melt, the desired thermal profile of the melt, and the output electrical properties of the generator itself. Because of the large amount of energy conducted through the coil and the electrical resistivity of common conductive materials used to make the coil such as copper, a coil as part of a CCIM system will likely require active thermal cooling. Induction coils may also require a mechanical support structure due to both the size and weight.

Induction Generator

The induction generator is typically rated in terms of maximum power output and frequency(s) of operation. Its purpose is to generate high frequency alternating current to drive the induction coil. Typical frequencies for CCIM application range from 100 kHz to 2 MHz, with power levels from 20 kilowatts for laboratory melters to a 10 megawatt design by the ST. Petersburg Elektrotechnical Institute.⁵ For this frequency and power range, designs are based on either metal oxide semiconductor field effect transistor (MOS-FET) power devices or vacuum transmitting tubes. Both transmitting tubes and MOS-FET's dissipate significant power for all circuit configurations, and active cooling will be required. Air is commonly used in the broadcast industry but water has a much higher specific heat and is likely to prove superior. However, a recirculating high-purity water system may be required because common utility water is somewhat conductive and will lower the quality factor (Q) of the induction coil.

Feed System

Cold crucible melters used in Russia and France receive calcined (dry) or paste (moist) feed materials to maximize the use of the inductive energy for melting glass. Current high-level waste operations in the United States at the Savannah River Site, SC and West Valley, NY feed liquid slurries to joule-heated melters (JHM). Offgas and volatility concerns, and complexity of design drive this departure in operational philosophy. Some waste constituents, notably cesium, ruthenium, and lead are quite volatile at common melter operating temperatures, causing a significant fraction of these materials to partition to the offgas system. Recycling the materials caught in the offgas treatment system to the melter reduces these losses but some fraction is revaporized and ends up back in the scrub solution. Disposal of the liquid scrub waste requires that it must be treated to reduce the leachability of hazardous components (lead) to disposal standards. This is further complicated by the radioactive constituents (cesium and ruthenium).

By feeding an aqueous slurry onto the melt, a boiling layer can be maintained over a transitional layer of drying feed solids on top of the molten glass itself. Materials volatilized from the molten glass must pass upward through this "cold cap" which causes condensation and reflux back into the melt, thereby maximizing retention of these species in the glass. Cold cap operation can reduce cesium losses from several tens of percent to just 2-3%, but the added cost is the melter heat required to vaporize all of the waste liquids. This additional heat load is reported to cause a reduction in melter throughput (derating) by 50% or more versus feeding calcined materials. Offsetting this reduction in melter capacity is the fact that the additional equipment needed to dry or calcine the liquid waste is avoided. The waste feed can also be accurately metered and mixed with a reducing agent (probably sugar) to promote reduction of nitrate to nitrogen and glass formers (either in chemical form or as frit) added to yield the final vitrified waste form. This mixing of constituents is not a trivial process; the quality assurance program to assure consistent melter feed at Savannah River is a large and complex operation.⁶

Offgas Control System

Offgas control for thermal treatment has been the subject of extensive study and development. Key issues are acid gases, mercury, condensation of semi-volatile alkali metals, organic byproducts such as products of incomplete combustion (PICs) notably dioxins, and a variety of lower concentration radioactive and

hazardous elements including cesium, ruthenium, iodine, lead, and cadmium. Offgas from a CCIM is not expected to be significantly different from a JHM operated at the same temperature with the same feed. However, one of the potential operating advantages for the CCIM is the capability to melt glasses at significantly higher temperatures. Though the practical benefits in increased feed rate or greater solubility of refractory oxides such as alumina and zirconia have not been quantified, glass melts have been completed in CCIMs at up to 2000°C, and other materials have been processed at up to 3000°C. Offsetting these potential advantages is the higher temperature exacerbating the volatility of problematic constituents.

Operation of a glass melter with a cold cap can drastically reduce offgas temperature, even though the melt temperature is quite high. Personnel from the Radon facilities outside Moscow report an offgas temperature of just over 100°C when processing a sodium nitrate waste dried to a paste containing 20-25 wt% moisture. At these temperatures, the primary issue would be keeping the gases hot enough to keep acid gases from condensing and refluxing on the upper part of the cold-crucible and plenum surfaces. Radon personnel report a cesium loss of about 1-3%. Offgas is filtered through a <100 micron metal mesh, followed by a HEPA grade filter. The essentially particle free gases are then scrubbed with nitric acid to remove NO_x, with the residual NO_x reduced over a vanadium impregnated catalyst. If supplemental plenum heaters were employed to accelerate evaporation of the liquid feed as is typical in JHM applications, offgases could be several hundred degrees higher and any threat of aqueous condensation problems would be eliminated. This increased temperature would be expected to mirror the results seen with JHM experience.

Output Pour

Joule-heated melters, currently in use for HLW processing, pour glass in a fashion similar to a teapot, with molten glass exiting from a level near the bottom of the pool, up through a heated pour spout, and dropping into a receiving canister. This type of design is also possible with a CCIM, but this is an area where innovation could make significant improvements. The teapot design allows good control over melt level, but the concept has had to overcome several challenges. Without sufficient heating of the spout area, strings of glass can form due to rapid cooling of the high surface tension falling stream. With wear of the point where the glass disengages from the melter, the glass can wick back on the surface of the pour spout instead of separating cleanly to pour into the canister. These phenomena can plug the pour spout and halt operations until the required maintenance is complete.

The CCIMs seen in Russia are typically operated in a semi-batch mode, and glass is removed by literally tipping the melter and decanting, or by using a water-cooled metal plug valve. Experimental melters range from the current KRI unit that melts a starting charge and is then cooled to remove glass fragments, to slightly larger units at the scientific institutes that are poured from the top either by tipping or overflow. Note that pouring from the top precludes continuous feed, unless an underflow weir is used to separate the feed chamber from the pouring or fining chamber. Russian developers have also built a water-cooled underflow weir design in a CCIM. The commercially offered French designs use water-cooled gate valves to effectively control pouring from a bottom outlet.

The Russian design philosophy seems to emphasize smaller high-frequency (MHz) units, versus the French approach using larger diameter, relatively lower-frequency (KHz) melters. With the lower inventory in process in the Russian units, it may not be practical to operate in a continuously pouring mode, and semi-batch operation with a cycling melt depth may be most advisable. Without refractory to consider, the consequences of thermal shock are drastically reduced. A trade study is probably warranted to evaluate the relative benefits of semi-batch operation with a small inventory versus continuous pouring with a much larger molten inventory. Beyond the operational aspects are the QA issues around small versus large batches, and the safety aspects due to the consequences of catastrophic failure.

Power Converter

The generator requires a direct current voltage supply. The key components of the power conversion system are the voltage converter and rectifier, filters, regulator, and control and protection circuitry. Some of these components may be combined in the same physical hardware. The capacity of the supply must exceed the output of the induction generator because of inefficiencies in the generation of RF power. This excess capacity requirement may be as high as 100%. For a lamp tube configuration, the voltage requirement will typically be between 1000 and 3000 Volts. For a MOS-FET based amplifier the voltage requirements will be an order of magnitude lower, on the order of hundreds of volts. Note that for the latter case the current will be proportionately higher. For most designs the voltage supply must be maintained within a certain accuracy and likely must be programmable over a range of values. Design issues include 1) power and voltage output requirements (including the requirement for excess capacity), 2) specification of accuracy of programmed value, regulation, and purity of output, 3) heat dissipation, 4) engineered safety system including interlocks and crowbar circuit protection, 5) reliability, and 6) the necessity of providing backup power during interruptions of commercial power.

Controls and Instrumentation

The controls and instrumentation element of a CCIM system serves three functions: 1) maintain temperature within the melt to a specified tolerance during steady state operation; 2) supply information to operators and to a system that records operating parameters during operation, 3) provide the means for control of the process during non steady-state operation such as startup, shutdown, and melt pour, and 4) acquire and transmit data necessary for operation of engineered system-level safety systems. Control of a CCIM system may be done manually or automatically. The design of an automated control system requires the steady state values and dynamic responses of the power supply, RF generator, and melter itself along with the associated sensors.

MASS AND ENERGY BALANCE

The primary design variable for both mass and energy balances is operating temperature. If the potential benefits of higher temperature operation including higher throughput and higher waste loading outweigh the potential disadvantage of higher volatility, the offgas treatment system for the CCIM could be made somewhat more complex. Recycle of offgas scrub solution back to the melter to reduce overall losses due to volatility and entrainment could also be less efficient. Current operation at the Defense Waste Processing Facility (DWPF) returns a scrub waste stream comparable in size to the feed going to the melter. The scrub solution is concentrated in an evaporator for eventual feed back to the melter. A higher melter operating temperature could reduce the potential for effective recycle, and the scrub waste stream may have to be treated by some other method of immobilization such as grout. The ramifications of higher temperature operation on the overall material balance should certainly be evaluated during experimental development.

The energy balance with a CCIM is drastically different from a JHM. First the efficiency of the energy deposition in the melt is a function of the both the power source, which has to convert line voltage to that required for the high frequency generator, and the coupling efficiency of the high frequency coil with a relatively low-conductivity glass melt. In a JHM, essentially all of the applied power is dissipated directly in the glass melt. Second, in a JHM, refractory brick and several layers of low-density silica insulating board surround the melt. The outer metal containment may be water-cooled, but the multiple insulating layers drastically reduce the total heat transfer. In the CCIM, there may be a 1000+°C temperature gradient across a 2-5 mm layer of solidified glass "cold-skull" between the melt and the cooled crucible. This lower energy efficiency is not necessarily a significant cost issue, but the energy balance, thermal profile, and operational energy deposition model must be understood to do detailed design. This knowledge should be a primary focus in laboratory and pilot-scale development of the CCIM technology.

RESULTS TO DATE

While significant marketing claims have been made by French and Russian technology suppliers and developers, little data is available for detailed engineering and economic evaluation of the technology, and no facilities are available in the US to support testing. Further, as a direct result of their responsibilities to safely manage all facets of the fuel cycle, the DOE is unlikely to invest in technology without significant understanding of its operation and control. The lack of CCIM expertise in the complex probably limits the potential for realizing the benefits of this technology. Thus, a small project is currently underway at the INEEL under the Laboratory Directed Research and Development (LDRD) Program to develop a basic theoretical understanding of the CCIM theory of operation. The current test program at the Khlopin Radium Institute is limited to a preliminary evaluation of the Russian CCIM technology. The intent of the LDRD is to provide enough factual information to the HLW Program, to have this technology considered as an option to the baseline joule-heated melter. The LDRD is also exploring the potential for an iron phosphate glass alternative to the baseline borosilicate composition for immobilizing SBW. An iron phosphate glass matrix has been developed by Dr. Delbert Day at the University of Missouri, Rolla, and samples have been provided to the INEEL HLW Program for verification analyses, which are to be completed in FY-02. If cost-effective, qualification of a non-borosilicate host glass could be pursued.

Results to date for the LDRD include:

- Three experimental melts were performed with Run 78 calcine chemistry and the borosilicate baseline composition at waste loading of 33, 38 and 43 mass % of simulated INEEL waste oxides in the final product.
- All three glass compositions display a considerable increase in electrical conductivity as temperature is increased above the melting point, requiring the operator to adjust power to avoid overheating the melt.
- The product glasses were 99+% amorphous.
- PCT results improved with higher waste loading. Preliminary PCT results are:

Table I. Preliminary PCT Results.

Nominal PCT Results Averaged over Melt Depth (g/m ²)				
Run 78 Calcine Waste Loading	Γ_B	Γ_{Si}	Γ_{Li}	Γ_{Na}
33 wt%	0.662	0.187	0.835	1.020
38 wt%	0.623	0.162	0.729	0.943
43 wt%	0.252	0.094	0.491	0.620

- Three melts were also performed with SBW chemistry and the borosilicate baseline composition at waste loadings of 25, 30, and 35 wt% of simulated INEEL waste oxides in the final product.
- Melting time was reduced at higher waste loading due to the fluxing properties (higher conductivity) of the SBW surrogate. Preliminary PCT results are:

Table II. Preliminary PCT Results.

Nominal PCT Results Averaged over Melt Depth (g/m ²)				
SBW Chemistry Waste Loading	Γ_B	Γ_{Si}	Γ_{Li}	Γ_{Na}
25 wt%	0.376	0.276	0.959	0.592
30 wt%	0.766	0.296	0.892	1.083
35 wt%	0.907	0.395	0.994	1.521

- Iron phosphate glasses were also evaluated at the University of Missouri, Rolla for immobilizing SBW. The recommended iron phosphate glass (IPG) matrix melts at 1000°C and immobilizes 40 wt% SBW based on acceptable glass durability.
- Glass liquidus temperature for the IPG is 740°C, and the lower viscosity (0.55 Poise at 1000°C), melting time is reduced to 2 hours to produce a homogeneous melt under laboratory conditions.
- The PCT protocol was designed specifically for borosilicate glass. Thus, results with the iron phosphate matrix are not directly comparable. The results for aluminum and phosphorous are offered in the table below in the position of boron and silicon. Iron and silicon would be the comparable primary cation and anion in the host matrix, but the iron release rates are essentially at detection levels. Results for potassium and sodium are shown in place of lithium and sodium, the primary fluxing cations. Preliminary PCT results are:

Table III. Preliminary PCT Results.

Nominal PCT Results Averaged over Melt Depth (g/m ²)				
SBW Chemistry Waste Loading	r _{Al}	R _P	r _K	r _{Na}
40 wt%	0.06	0.13	0.16	0.32
48 wt%	0.26	3.72	5.55	6.03

- The IPG glass will now be analyzed for sulfate retention to compare to the baseline borosilicate composition, which has been limited to about 20 wt% due to sulfate solubility.⁷ Thus, additional evaluation will be necessary in FY-02.
- One CCIM experiment has been performed with the IPG glass and the electrical properties are quite favorable to this type of processing. Again, this work will continue in FY-02.

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