Melting, Solidification, Remelting, and Separation of Glass and Metal

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ACRONYMS

CS	Centrifugal Separation
DCS	Differential Centrifugal Separation
DDFA	Decontamination and Decommissioning Focus Area
DOE	U.S. Department of Energy
DTA	Differential thermal analysis
FIU	Florida International University
GTA	Gradient thermal analysis
HCET	Hemispheric Center for Environmental Technology
SAIC	Science Applications Information Corporation
SEM/EDS	Scanning electronic microscopy/Energy disperse type X-ray spectrometry
TRU	Transuranic

1. INTRODUCTION

Several kinds of radioactive waste exist in mixed forms at DOE sites throughout the United States. These wastes consist of radionuclides and some useful base materials. One purpose of waste treatment technologies is to vitrify the radionuclides into durable, stable glass-like materials to reduce the size of the waste form requiring final disposal. The other purpose is to recycle and reuse most of the useful base materials. Thus, improved techniques for the separation of molten metal and glass are essential.

Several high temperature vitrification technologies have been developed for the treatment of a wide range of mixed waste types in both the low-level waste and transuranic (TRU) mixed waste categories currently in storage at DOE sites throughout the nation. These processes include the plasma hearth process, which is being developed by Science Applications International Corporation (SAIC), and the arc melter vitrification process, which is being developed at Idaho National Engineering Laboratory. The products of these processes are an oxide slag phase and a reduced metal phase. The metal phase has the potential to be recycled within the DOE Complex. Enhanced slag/metal separation methods are needed to support these processes. A separation method is also needed for the radioactively contaminated scrap metal recycling processes in order to obtain highly refined recycled metals.

1.1 PROJECT OBJECTIVES

The ultimate goal of this project is to find an efficient way to separate the slag phase from the metal phase in the molten state. A basic knowledge of the thermal, transport, and metallurgical phenomena that occur in the melting and solidification of glass, metal, and their mixture is the foundation of the development of the separation methods.

1.2 REVIEW OF THE LITERATURE

The melting behavior and properties of selected glasses have been studied by Skrovankova et al. (1993). The melting behavior of glasses based on the system SiO_2 - B_2O_3 - Al_2O_3 - Na_2O prepared from natural aluminosilicate raw materials was explained using data obtained from differential thermal analysis (DTA), gradient thermal analysis (GTA), and heating microscope examinations. The chemical durability of the glasses was also studied, and metal oxide additions were investigated for their effects on the properties of SiO_2 - PbO -20% BrO_3 -5% SrO -5% MgO -10% Al glass systems. Bulk glass samples were measured for physico-chemical properties such as linear thermal expansion coefficients, water resistance, density, and softening point. Some measurement physicochemical properties were provided, and the effect of each metal oxide addition on the properties was noted.

Viskanta (1994) and Nemec (1994) have reviewed and summarized the modeling of glass melting. Nemec (1994) addressed several aspects of the process, including heat transfer, reaction, sand dissolution, bubble generation, and homogenization. Viskanta's review emphasizes the simulation of flow patterns and heat transfer in the molten stage.

The complex melting and remelting processes are characterized by a number of features which make in-situ measurement difficult. These include 1) high temperatures, 2) the reactivity of the alloys, and 3) the important contribution of heterogeneous reactions. Thus, few references are available regarding the basic melting phenomenon of metals. Wu (1993) presents a one-dimensional, time-dependent heat transfer model for the melting process in a circular scrap melter. The phase-change problem is tackled using an enthalpy-based method, and the complex scrap geometry is described with the porosity concept (i.e., the blocking effect of the scrap is represented by a blocking factor that is a function of the scrap's porosity).

Due to measurement difficulties, the solidification process of metals has not been studied extensively. Several experiments on low melting temperature metals and alloys have been conducted (Garcia and Prates 1978; Gau and Viskanta 1985; Winegard and Cole 1964 and; Wolff and Viskanta 1988).

Few studies are available on the solidification of glasses. Nishioka et al. (1990) investigated the solidification of glass powder with simulated high-level radioactive waste during hydrothermal hot pressing.

From the review, it can be concluded that little research has been conducted on borosilicate glass; little work has been conducted on the melting and solidification of high-temperature metals and alloys; and no experimental investigations have been carried out on the melting and solidification of glass/metal mixtures.

1.3 REVIEW OF EXISTING GLASS/METAL SEPARATION TECHNOLOGIES

A number of technologies are available to separate substances from molten metal. These technologies include the gravity mode, the vibratory mode, and the differential cooling method of separation. In addition, directional solidification is used to separate the useful scrap metal from the glass matrix. These methods are not appropriate for the waste vitrification process, however.

Purohit et al. (1995) conducted a research project to define the design bases and validate the final design of the slag and metal removal and handling system to be built for the field-scale radioactive plasma hearth process system developed by Science Applications International Corporation (SAIC) and Retech, Inc. Proof-of-principle testing of the slag and metal separation and slag pouring concepts was completed by SAIC at facilities in Ukiah, CA and Idaho Falls, ID. The current practice involves the installation of a glass/slag tap and molten metal tap on the upper and bottom portions of the furnace, respectively, to separate the two phases by means of gravity. The draining of the metals in vitrified nuclear waste is accomplished using a melter on a sloping floor with a 45° angle.

The use of ceramic filters to remove inclusions in melted alloys has been investigated in casts weighing up to four tons at Special Melted Products in Sheffield. No problem of filter blockage was experienced, and oxide inclusions were successfully captured. The success was only partial for TiN inclusions since these can also precipitate after solidification. The filtration bed contained flux-covered alumina balls, and tabular alumina granules were used to remove the indigenous and exogenous inclusions (Bates 1995).

A vacuum technique has been developed by Kubota Ltd. for removing slag from molten metals. This slag removal process employs suction to remove the molten slag from the top of the metal matrix. The slag is immediately cooled and turned to granules using water. It is carried out via the suction pipē using air as the transport medium. This processing unit consists of a main slag suction unit, the transport unit, and the main slag removal unit. The slag separation discharge and withdrawal device separates the granular slag from the air and discharges the slag into a water tank. The slag is then transported by a conveyor system to the disposal tank area. This process involves three independent moving systems as well as air and steam transport systems. As a result, this technology fails to fulfill the present DOE requirement specifying that the technology have as few moving parts as possible.

Lakomskii et al. (1985) proposed another method for the separation of slag/metal emulsions. It is suggested for the removal of liquid, nonmetallic inclusions from the metal phase in the field of centrifugal forces. As this investigation was limited to theoretical analyses, experimental work must be performed to verify the feasibility of this method.

In order to improve or develop new separation technologies to meet the recycled metal free release standard, the transport phenomena in the melting and solidification processes must be studied to provide basic knowledge with which to develop or improve the separation technologies.

In high-temperature mixed waste treatment processes, a melt consisting of an oxide phase and a reduced metal phase is produced in crucibles. Generally, the metal phase is two to three times more dense than the slag phase. The gravity separation method is currently being considered and tested as a treatment process. SAIC has developed and tested a crucible construction design to separate these two phases. This design is described by SAIC (1994) as follows:

An under weir is used to separate the metal from the slag. The weir dams off a portion of the crucible and has a slot at the base. On the feed side of the weir, molten metal and slag produced during the drip melt process separate into two distinct layers due to the difference in densities. Molten metal - the bottom layer - flows under the weir and rises up on the other side. After flowing under the weir, the molten metal rises to a higher level that is determined by the hydrostatic force exerted by the molten slag and metal on the feed side of the weir. From the metal only side of the weir, the metal can be removed from the hearth by continuous overflow or by intermittent tilting and pouring. The molten slag can be removed from the opposite side of the crucible either by continuous overflow or by intermittent tilting and pouring.

The separation of the glass from the molten metal slag and scrap metal requires that a motion be imparted on the fluid or semi-fluid constituents. The evaluation of the type of motion took considerable analysis and thought. The methods that show promise are the Centrifugal Separation and a Differential Centrifugal Separation techniques. These methods meet the DOE's requirement of having the least amount of equipment possible with the consideration of recontamination. This evaluation resulted in the development of the two methods proposed in this project.

2. PROJECT DESCRIPTION

The aim of this project is to investigate the melting and solidification characteristics of selected glasses, metals, and their mixtures and to develop new or improved separation technologies for glass/metal separation. The primary goals of this project are:

- Development and implementation of measurement methods for the metal and glass melting and solidification tests;
- Investigation of the melting and solidification characteristics of glass, metals, and their mixture;
- Simulation testing; and
- Molten-state, high-temperature testing and demonstration of the new or improved technologies.

2.1 FISCAL YEAR 1996 TASKS

The following objectives were established for FY96:

- 1) Survey of existing glass/metal separation methods and measurement methods for the melting and solidification experiments involving glass and metals;
- 2) Completion of the experimental setup, consisting of melting and solidifying equipment, temperature measurement devices, and characterization devices;
- 3) Preliminary experiments to verify the performance of the system; and
- 4) Development of a mathematical and numerical model to be used to simulate the solidification process.

Most of these objectives were achieved. Due to a delay in the delivery of the induction furnace, the melting and solidification experiments were rescheduled for FY97. In their place, the conceptual design of the separation technologies was finalized, and the numerical simulation of solidification was conducted.

The following objectives were achieved:

1. The survey of existing glass/metal separation methods and measurement techniques for melting and solidification experiments of metal, glass, and their mixtures (March 1996);
2. The conceptual design of the separation technologies (April 1996);
3. Establishment of the numerical simulation model of solidification using the enthalpy-porosity method (April 1996).
4. The acquisition of the melting and solidification experimental measurement and characterization devices (September 1996);
5. The simulation of molten aluminum and iron steel (September 1996); and
6. The proposal of the simulation demonstration plan, and the design and construction of the setup (September 1996).

2.2 PROPOSED FISCAL YEAR 1997 TASKS

The following tasks are scheduled for completion in FY97:

1. Experimental investigation of the melting, remelting, and solidification of glass and the mixture of the metal and glass to obtain the phase distributions during the process, thereby providing the information for the final design of the DCS and CS technologies;
2. Verification of the concept used in the DCS and the CS technologies by means of tests in low temperature environments using simulants;
3. Improvements in the design of the DCS and CS and techniques; and
4. Testing of the DCS and CS technologies at high temperatures using glass and metal.

The DCS and CS technologies are still in the development stage. Since both gravity and centrifugal force are used in these technologies, their separation potential is much higher than that currently achieved with the baseline technology. In the technology development process and laboratory-scale demonstration, the operational performance of the DCS and CS technologies is being evaluated in the following areas:

- Effects of the viscosity of the glass phase;
- Required density difference between the glass/metal phases to achieve satisfactory separation efficiency;
- Separation efficiency for different glass phase formulations;
- Optimal rotation speed for specific glass/metal combinations;
- Optimal operational temperature for specific glass/metal separation processes;
- Comparison of the compositions and microstructures of the metal ingots resulting from the application of baseline technology and the innovative separation technologies;
- Effectiveness of the flow control system; and
- Duration of the separator crucible subjected to high temperature molten metal conditions.

In the FY97 continuation of this project, the DCS and CS technologies will be tested and demonstrated in three stages. These follow:

- I. Simulation demonstration tests using simulated liquids will be conducted to verify the principle upon which the designs are based. These tests will provide constructive information regarding system design improvements;
- II. Metal and glass separation tests will be performed to obtain the above-mentioned performance data. The parameters will be adjusted to determine the best operation conditions for the different ratios of glass and metal;
- III. Optimal design for glass/steel separation will be based on the demonstration tests utilizing both the simulated liquids and metal/glass in order to increase the separation efficiency.

All the tests and demonstrations will be conducted at the Hemispheric Center for Environmental Technology (HCET) at Florida International University (FIU).

3. INNOVATIVE GLASS/METAL SEPARATION TECHNOLOGIES

In association with this project, two methods have been designed to separate different slag/metal mixtures -- the Centrifugal Separation (CS) technology and Differential Centrifugal Separation (DS) technology. The Differential Centrifugal Separation (DCS) technology and Centrifugal Separation (CS) technologies involve the following principles and processes:

- The mixture of the molten metal and glass is subjected to centrifugal action;
- Physical laws or cohesive properties cause differential movement of the substances due to their inherent properties;
- Materials with like properties coagulate and the aggregates increases;
- Separation of the substances with distinct identities;
- Physical removal of the separated phases; and
- Solidification of the separate phases.

In both processes, the glass phase and metal phases are solidified in separate containers.

3.1 CENTRIFUGAL SEPARATION TECHNOLOGY

A schematic representation of the Centrifugal Separation (CS) technology is shown in Figure 1. This technology is designed to be the second stage of the separation process after the dam/gravity tapping. The first stage is described above. After the metal melt accumulates in the melting hearth, the metal tap is opened and the metal melt flows into the separation pot continuously. The metal container and the separation pot installed in a rotating platform rotate at a high speed. As a result of gravity and centrifugal force, the metal melt is pushed into the metal container, and it solidifies. After the metal flows out from the pot, the molten slag can be poured into the slag container. The remaining impurities in the metal container are then pushed toward the inlet, where they can be removed easily. The solidified metal inside the container is very clean.

Figure 1. Schematic of the Centrifugal Separation (CS) method.

In the experimental system, an induction furnace is used to obtain the glass and metal melt mixture. The melt is poured into a separation pot. The separation pot rotates with the solidification container. Both the pot and the container are driven by a motor and rotate at a high speed. The separation process depicted in Figure 1 then occurs. After separation, elemental and structures analyses are conducted to evaluate the effectiveness of the separation techniques.

3.2 DIFFERENTIAL CENTRIFUGAL SEPARATION (DCS) TECHNOLOGY

The Differential Centrifugal Separation (DCS) technology achieves separation by means of the combination of a plunger insertion mechanism and a specially conceived and designed rotating container which holds the molten constituents.

The schematic of the differential centrifugal separation method and the details of the furnace assembly in DCS are shown in Figures 2 and 3, respectively.

Figure 2. Schematic of the Differential Centrifugal Separation (DCS) method.

1. Motion control plunger link	8. Lug assembly
2. Plunger shaft	9. Insulation washer
3. Plunger bracket	10. Steel casing
4. Graphite plunger	11. Air/convective port
5. Crucible outer insulation	12. Refractory insulation
6. Crucible inner profile	13. Casing support shaft
7. Assembly bolts	14. Collection bins

Figure 3. Details of the furnace assembly used in the DCS technology.

This system employs a simple rotating crucible that acts as a secondary furnace holding the contents to be melted. The innovative aspects of this design are its inventive internal shape and speed combination, which can effect the separation of steel. Upon rotation of the furnace, the separated effluent is ejected out by differential plunger assembly. The separated effluent can be collected in stationary bins with continuous feed operation so as to enable 24-hour operation for an ideal production scenario.

3.3 TECHNOLOGY APPLICABILITY

The Differential Centrifugal Separation (DCS) and Centrifugal Separation (CS) technologies separate the glass and metal phases to improve the separation efficiency of the baseline technology used in the mixed waste vitrification process. The DCS and CS technologies can be applied in related applications such as the recycling of radioactively contaminated metals. These technologies can be implemented in all kinds of slag/metal separation processes in the metallurgical industry.

These technologies will therefore be tested for the different kinds of glass and metals that are contained in the mixed waste in DOE sites. Important parameters such as the rotation speed and separation rate can then be adjusted according to the different operating conditions. The DCS

technology is suitable for the separation of molten glass with lower viscosity. The CS method is applicable to molten glass with higher viscosity.

The DCS and CS technologies meet safety requirements to ensure that the workers are adequately protected during remediation activities involving with hazardous wastes. Both technologies are characterized by smooth operation owing to the use of a minimum number of moving parts. Both employ remote operation and involve just one moving assembly for separation purposes. The removal and disposal of the vitrified matter can be handled with minimum risk as the containers in which the material solidifies can be transported directly to waste burial sites via trailer.

4. MELTING AND SOLIDIFICATION EXPERIMENTS

The objective of the melting and solidification experiments is to provide fundamental information for the development of the slag/metal separation technologies. The thermal, transport, and metallurgical phenomena in the melting, solidification processes of glass, metal and their mixture will be studied in FY97.

The basic experimental system has been designed. Its capabilities include melting and solidification of ferrous and nonferrous metals; the use of different solidification cooling patterns; temperature measurement during the melting and solidification processes; and characterization of the material structure and phase distribution of the test materials.

During the melting, the melting speed is measured by inserting thermocouples into the molten liquid pool to monitor the temperature histories of the inside and the surface of the crucible. In addition, samples are taken from different positions inside of the crucible to analyze the phase or composition distribution in the molten state. When the cooling process begins, the fluid and thermal characteristics of the solidification are measured and observed. Once solidified, the ingot is characterized using SEM/EDS to obtain the composition or phase distribution and material structure.

4.1 APPARATUS AND INSTRUMENTATION

The details of each portion of the system are described in the following subsections.

4.1.1 Furnace

The furnace employed is a model is LFC 30 with a capacity of 30 pounds of steel alloys and a melt rate of 180 pounds per hour. The furnace equipment consists of the following:

- 50 kW, 3000 Hz, LP Pachydyne integral power supply, with power factor correction capacitors and an output transformer;
- Remote operator panel complete with on/off push buttons, power adjust dial, digital meters, and fault annunciation light;
- One set of water-cooled power leads, approximately 12 feet long;
- Lift coil style coreless furnace complete with coil cast in refractory, a structural steel support table with counterbalance weights, coil lifting guide framework, a fume ring, and a differential water pressure switch;
- Two sets of refractory materials consisting of alumina refractory crucibles and support blocks; and
- Indoor water pump recirculating water system with a pump/motor, a water/water heat exchanger, and complete system controls.

4.1.2 Cooling System

Solidification is conducted in the melting crucibles. Because of the high temperature, the air jet has been chosen as the preferred cooling method. The process is controlled by using the air jets and adjusting the heating power of the coil. The support table of the furnace was modified to install four spray nozzles around the crucible. This method offers the advantage of immediate cooling with heating as a means of control.

4.1.3 Crucibles

Alumina crucibles and their liners will be used for the melting and solidification of steel. These will also serve as the solidification holder. These durable crucibles can be heated quickly, and they withstand temperatures of up to 1750°C.

4.1.4 Temperature Measurement

Two methods for conducting high temperature measurements are used:

- One-time use B-type thermocouples, and
- A Hughes thermal video monitoring system for surface temperature measurement and visualization at high temperatures.

4.1.5 Characterization Method

An SEM/EDS system will be used to characterize the solidified glass and metals to obtain information about the material structure and phase distribution. The devices are described below.

The SEM/EDS, JSM-35CF high-performance scanning electronic microscope with an Energy Disperse Type X-ray Spectrometer (EDS) and Image Analyzer. The SEM has the spatial resolution for a secondary electron image of 60 Å (working distance 15 mm) and 150 Å (working distance 39 mm). It has a detector system consisting of scintillator-photomultiplier system complete with a preamplifier and a collector. The EDS attachment allows microscope expansion to a multichannel microanalyzer for X-ray elemental analysis of micro-regions with a attainable sensitivity of 0.5 percent.

The Image Analyzer (Micro-Particle Analyzer) can calculate the area percentage of metallic precipitates and the number, diameter, and area of particles from scanning images. The direct analysis of video signals greatly reduces measurement time and minimizes error. Prompt data processing is ensured by special logic circuits and a built-in microprocessor.

SEM/EDS is primarily used to characterize metallurgical phenomena such as segregation and oxide particulate distribution.

4.2 TEST MATERIALS

Two kinds of glass-like materials, alumophosphate and borosilicate glass, are commonly used in waste treatment. Alumophosphate glass is used in Russia, whereas borosilicate glass is being tested and used by the DOE.

Borosilicate glass systems have been studied the most for the immobilization of radioactive waste. Many different compositions have been evaluated. A summary of the composition ranges of the high-level radioactive waste that has been investigated is given in Table 1 (Tomozawa et al. 1985). The effects of waste-glass components on processing and product performance have been reviewed by Tomozawa et al. (1985) (see Table 2). Borosilicate glass will be used in the experiments. The approximate compositions and thermal properties of commercial borosilicate glasses and borosilicate glass for high-level radioactive waste treatment and the metal slagging/refining process are listed in Table 3.

Table 1.
Composition of Glass Frits

Component	Compositional range, wt. %
SiO ₂	27-59
B ₂ O ₃	10-20
Na ₂ O	1-20
Li ₂ O	0-6
K ₂ O	0-3
CaO	1-6
MgO	2-3
ZnO	7-30
CuO	0-4
TiO ₂	0-5
ZrO ₂	0-1
La ₂ O ₃	0-1
Al ₂ O ₃	0-3

Source: Tomozawa et al. 1985.

Table 2.
Effects of Waste-Glass Components on Processing and Product Performance

Frit Components	Processing	Product Performance
SiO ₂	Increases viscosity greatly; reduces waste solubility	Increases durability
B ₂ O ₃	Reduces viscosity; increases waste solubility	Increases durability in low amounts; reduces in large amounts
Na ₂ O	Reduces viscosity and resistivity; Increases waste solubility;	Reduces durability
Li ₂ O	Same as Na ₂ O, but greater effect; increases tendency to devitrify	Reduces durability, but less than Na ₂ O does
CaO	Increases then reduces viscosity and solubility	Increases and then reduces durability
MgO	Is same as CaO; reduces tendency to devitrify	Same as CaO
TiO ₂	Reduces viscosity slightly; increases and then reduces waste solubility; increases tendency to devitrify	Increases durability
ZrO ₂ , La ₂ O ₃	Reduces waste solubility	Increases durability greatly
Al ₂ O ₃	Increases viscosity and has a tendency to devitrify	Increases durability
Fe ₂ O ₃	Reduces viscosity; hard to dissolve	Increases durability
U ₃ O ₈	Reduces tendency to devitrify	Reduces durability
NiO	Hard to dissolve	Reduces durability
MnO ₂	Hard to dissolve	Increases durability
Zeolite	Slow to dissolve; produces foam	Increases durability
Sulfate	Antifoam, melting aid	Too much causes foam or formation of soluble second phase

Source: Tomozawa et al. 1985.

Table 3.
Compositions and Thermal Properties of Borosilicate Glasses

Borosilicate Glasses							
Composition, Nominal %							
Type	SiO ₂	Al ₂ O ₃	Na ₂ O	K ₂ O	B ₂ O ₃	TiO ₂	Others
Metal refining-1	80	2	4	0.5	13		
Metal refining -2	80	2	4		13		
Metal refining -3	80	2	4	1	13		
Pyrex	81	2	4		13		
Sodium	68	2	6		24		
Defense waste Frit 131	57.9		17.7		14.7	1.0	5.7-Li ₂ O, 2.0-MgO, 0.5-ZrO ₂ , 0.5-La ₂ O ₃
Commercial waste Frit 76-101	59.7		11.2		14.2	4.45	3.0-CaO, 7.45-ZnO
Thermal Properties							
Type	Density (g/cm ³)	Working Temp. (°C)	Thermal exp. coe.(x10 ⁷ /°C) 0-300°C		Thermal con. (W/cm °C) 0°C 100°C		Heat capacity (J/g °C) 25-175°
Pyrex	2.23	1250	33		11.2	12.9	0.85
Sodium	2.24	1020	46		10.4	12.1	

Sources: Scholze 1991; Tomozawa et al. 1985; and Bansal and Doremus, 1986.

The melting behavior of glass is usually described by the viscosity curve which is well represented by the Vogel-Fulcher-Tammann equation as follows:

$$\log \eta = A + B/(T - T_0) \quad (1)$$

where, η represents the viscosity, A and B are correlating constants, T denotes temperature, and T_0 is the reference temperature. The melting properties of the two main components of borosilicate glasses, SiO₂ and B₂O₃, are given by Horst Scholze (1991). For SiO₂, A equals -2.49,

B equals 15004, and T_0 equals 253, that is, the transformation temperature is 1203°C ($\log \eta = 13.3$), the Littleton temperature is 1740°C ($\log \eta = 7.6$), and the working point is 2565°C ($\log \eta = 4.0$). For B_2O_3 , the transformation temperature is 254°C, the Littleton temperature is 1740°C, and the working point is 2565°C.

The common metal phases resulting from the vitrification process are steel, copper, and aluminum. Aluminum and steel will be used in these experiments. The experiments with aluminum will be conducted first since this metal has a lower melting point, and it is easier to run the experiments. The experience gained from the aluminum experiments will help improve the experimental system and serve as preparation for the high temperature steel melting and solidification tests. In addition, aluminum and steel represent low density and high density metals, respectively. The properties of aluminum and steel are listed in Table 4.

Table 4.
Thermal Properties of Aluminum and Iron Steel in Solid and Molten Phase

Property	Aluminum	Iron
Melting point (K)	933.1	1548
Latent heat (KJ/kg)	394	220
Specific heat (KJ/kg K)	1.08	0.829
Density in solid (kg/m ³)	2700	7200
Thermal conductivity in solid, (W/m K)	238	80.2
Density in liquid (kg/m ³)	2385	7000
Thermal conductivity in liquid, (W/m K)	94.03	44
Viscosity in molten liquid, (kg/m s)	1.3×10^{-3}	7.18×10^{-3}
Thermal expansion coefficient in liquid, (1/K)	1.174×10^{-4}	6.0×10^{-5}
Surface tension, (N/m)	0.914	0.845
Surface tension gradient, (N/m K)	-3.5×10^{-4}	-3.5×10^{-4}

Source: American Society for Metals, 1978-c1989

5. MAJOR FISCAL YEAR 1996 ACCOMPLISHMENTS

In FY96, the following work was completed:

1. The survey and review of the research on the melting and solidification of metal, glass, and their mixture.
2. Installation of the test system. All the melting and cooling equipment, temperature sensors, and other measurement instruments are ready for use;
3. Preliminary tests on aluminum and aluminium/glass mixture melting/solidification; and
4. Numerical simulation of molten aluminum and iron steel solidification.

5.1 PRELIMINARY EXPERIMENTAL RESULTS

Prior to formal melting and solidification tests using the induction furnace, aluminum and an aluminum/glass mixture were melted and solidified in a heating and cooling controlled graphite resistance furnace. These tests were conducted to observe the aluminum-glass-crucible interaction. Figure 5 presents an example of the final solid products of the aluminum/glass mixture tests. Two separate phases still existed even though diffusion and interaction occurred. The metal phase can easily be separated from the crucible, but the glass phase adhered to the crucible and could hardly be removed. From the appearance of the product, it can be inferred that the aluminum vapor greatly increased the viscosity of the molten glass. Hence, it is theorized that some ingredients are needed in the glass formula to reduce the viscosity of the molten glass for separation purposes.

Figure 5. Melting/solidification ingot sample of the glass/metal mixture.

5.2 PRELIMINARY NUMERICAL SIMULATION RESULTS

To study the flow, transport, and solidification patterns, the solidification of molten aluminum and iron steel in a crucible 0.1m in diameter and 0.1m in height have been simulated under the convective cooling boundary condition. The initial temperature was 1000 K for molten aluminum and 1600 K for molten iron steel. The heat transfer coefficient was selected as 250 (W/m² K) for the air jet cooling; and the cooling temperature was 300 K.

The results indicate that the solidification of iron occurs much more slowly than that of aluminum under the same cooling condition since iron steel has much lower thermal conductivity and diffusivity. Natural convection in the liquid metal pool is significant in the early stage of the solidification process. The temperature of the liquid pool becomes more uniform in the later stages, so the convection inside is very weak. This can be seen from the maximum velocities inside of the molten metal pool, which are listed in Table 5.

Table 5.
Maximum Velocities in the Molten Metal Pool (m/s)

Solidification Time (s)	Aluminum	Iron Steel
40	0.00867	0.0135
80	0.00292	0.00666
160	0.000643	0.00398

Graphic results of temperature distribution, solidification patterns, and velocity fields are shown in Figure 6 (a)-(b) for aluminum and in Figure 7(a)-(b) for iron steel.

(a) Temperature distribution and solidification pattern.

Velocity Vectors (M/S) Max = 6.427E-04

(b) Stream function and velocity vectors.

Figure 6. Temperature distribution, solidification patterns, and flow field for molten aluminum solidification ($t = 160$ s).

Liquid phase
 Solid phase
 V max = 0.00147 m/s

(a) Temperature distribution and solidification pattern.

(b) Stream function and velocity vectors.

Figure 7. Temperature distribution, solidification patterns, and flow field for molten iron solidification ($t = 160$ s).

In these figures, it can be seen that the solidification front surface is parabolic like, and the solidification is faster for aluminium than for iron steel.

6. CONCLUDING REMARKS

Mixed waste vitrification and radiologically contaminated scrap metal recycling are two significant, integrated demonstrations related to the United States Department of Energy's Decontamination and Decommissioning Focus Area (DDFA). These two processes produce a melt consisting of a slag (glass) phase and a reduced metal phase. The separation of metal/glass represents a significant problem in these two processes. Although various slag/metal-phase separation technologies exist in the steel industry, more efficient separation technologies are still needed to produce reusable metal materials with minimum radioactive contamination levels. Since melting, remelting, and solidification happen either sequentially or simultaneously during the melting and solidification process, the fundamental study of the melting, remelting, and solidification of metal and glass and their mixture is essential for the development of a new separation technology. Thus, this two-year experimental investigation involves the fundamental study of the melting and solidification of metal, glass, and their mixture and the development of a slag/metal separation technology to enable the clear separation of molten metal and glass.

Two innovative technologies are being developed concurrently at the Hemispheric Center for Environmental Technology (HCET) at Florida International University (FIU) in an effort to produce a successful method to separate the vitrified matters from the useful metals and thereby facilitate the recycling of the metals. These technologies are the Differential Centrifugal Separation (DCS) technology and the Centrifugal Separation (CS) technology.

Both the DCS and the CS technologies use centrifugal force to separate the glass from the metals as a result of the density difference between the two phases. In the DCS process, the glass phase is forced to flow out of the crucible container by means of a plunger movement. The metal phase then remains in the bottom of the container and can be poured out intermittently. In the CS process, as the separator rotates, centrifugal force impels the denser metal phase to flow out of the separator from a tap on the bottom. The glass phase, which exhibits less density, remains in the separator and can be poured out intermittently. In both processes, the glass and metal phases are solidified in separate containers. The solid glass phase is disposed of or buried as a final waste form. The clean metal phase can be reused to fabricate waste containers for internal DOE use or provided to commercial industry, depending on the contamination level of the metal phase.

This two-year project commenced in October 1995 (FY96). In FY96, an assessment of the baseline glass and metal separation technologies was performed. The results indicate that the baseline technology yields a high percentage of glass in the metal phase, necessitating further separation treatment. A number of melting and solidification tests were conducted. In addition, temperature distribution, solidification patterns, and flow field in the molten metal pool were simulated numerically for the solidification processes of molten aluminum and iron steel. Initial designs of the laboratory-scale DCS and CS technologies were also completed. The principle demonstration separation units using simulants were constructed. Application for the patenting of these technologies is pending.

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