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J. E. Surma D. L. Smatlak

P. P. Woskov J. K. Wittle D. R. Cohn P. Thomas C. H. Titus R. A. Hamilton

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Pacific Northwest Laboratory Richland, Washington 99352

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J. E. Surma
Pacific Northwest Laboratory
Richland, Washington

D. R. Cohn
D. L. Smatlak
P. Thomas
P. P. Woskov
Plasma Fusion Center
Massachusetts Institute of Technology
Cambridge, Massachusetts

C. H. Titus
J. K. Wittle
R. A. Hamilton
Electro-Pyrolysis, Inc.
Wayne, Pennsylvania

ABSTRACT

A "National Laboratory-University-Industrial" three-way partnership has been established between the Pacific Northwest Laboratory (PNL), Massachusetts Institute of Technology (MIT), and Electro-Pyrolysis, Inc. (EPI) to develop graphite electrode DC arc technology for the treatment of buried wastes. This paper outlines the PNL-MIT-EPI program describing a series of engineering-scale DC arc furnace tests conducted in an EPI furnace at the Plasma Fusion Center at MIT, and a description of the second phase of this program involving the design, fabrication, and testing of a pilot-scale DC arc furnace. Included in this work is the development and implementation of diagnostics to evaluate and optimize high temperature thermal processes such as the DC arc technology.

INTRODUCTION

Across the U.S. Department of Energy (DOE) complex, the past practices of DOE and its predecessor agencies in burying radioactive and hazardous wastes have left DOE with the responsibility of remediating large volumes of buried wastes and contaminated soils. The Buried Waste Integrated Demonstration (BWID), a program within DOE's Office of Technology Development (OTD), has chosen to evaluate treatment of buried wastes at the Subsurface Disposal Area (SDA) of the Radioactive Waste Management Complex (RWMC) at the Idaho National Engineering Laboratory (INEL). Many scenarios are being evaluated for the treatment of buried wastes. Because of the characteristics of the buried wastes within INEL's SDA, the potential for using high-temperature thermal treatment technologies is being evaluated.

The Pacific Northwest Laboratory (PNL), Massachusetts Institute of Technology (MIT), and Electro-Pyrolysis, Inc. (EPI) have entered into a collaborative "National Laboratory-University-Industrial"

three-party program to develop and demonstrate the technology using a graphite electrode DC arc furnace for the treatment of buried wastes. The program, which is funded through BWID, has involved testing an engineering-scale DC arc furnace to gain preliminary operational and waste processability information. It has also included the design, fabrication, and evaluation of a second-generation, pilot-scale graphite electrode DC arc furnace.

The program was initiated in 1992, and 13 test melts have been conducted at the MIT Plasma Fusion Center in the latter part of FY92 using EPI's engineering scale (Mark I) furnace. The design and initial fabrication of the second generation (Mark II) pilot scale furnace have paralleled the Mark I testing during FY92. Work will continue in FY93 with the installation of the Mark II furnace at MIT. A large-scale test is planned in this furnace to fully evaluate the DC Arc Technology with respect to processing buried wastes. A set of plasma process diagnostics is being developed, drawing upon the experience of the Plasma Fusion Center to characterize the operation of the plasma arc furnace and to optimize the operation of the overall system.

Widely ranging simulants of INEL buried waste were prepared and processed in the Mark I furnace. The tests included melting of soils with metals, sludges, combustibles, and simulated drums. Very promising results in terms of waste product quality, volume reduction, heating efficiency, and operational reliability and versatility were obtained. The results indicate that the graphite electrode DC arc technology would be very well suited for treating high melting point wastes such as those found at the INEL SDA. The graphite electrode DC arc furnace has been demonstrated to be very simple, yet effective, with excellent prospects for remote or semi-remote operation.

The Mark II furnace design incorporated many of the features that will be required for a semiremote or remotely operable furnace, including prototype provisions for alpha-containment and the capability of continuous processing at power levels of over 1 MW with a waste processing rate of 500-700 pounds per hour. Much of the buried waste across the DOE complex contains a high weight fraction of metals. For this reason, the Mark II furnace was designed with the capability to separate the IEB slag (glass) from the metal phase through an auxiliary furnace tapping arrangement. The complete testing and optimization of the Mark II furnace will provide DOE with the required information to fully evaluate the graphite electrode DC arc technology for the treatment of buried and mixed wastes.

BACKGROUND

The DOE, through its Office of Technology Development (OTD), has initiated a comprehensive program to develop and demonstrate advanced remediation technologies to support DOE's environmental restoration objectives. The BWID has been established within the OTD to develop and demonstrate technologies applicable to the remediation of DOE sites containing buried wastes. Across the DOE complex, the past practices of DOE and its predecessor agencies in burying radioactive and hazardous wastes have left DOE with the responsibility for remediating large volumes of buried wastes and contaminated soils. The BWID program has chosen to evaluate treatment of buried wastes at the Subsurface Disposal Area (SDA) of the Radioactive Waste Management Complex (RWMC) at the Idaho National Engineering Laboratory (INEL). Many scenarios are being evaluated for the treatment of buried wastes. Because of the characteristics of the buried wastes within INEL's SDA, the potential for using high-temperature thermal treatment technologies is being evaluated.

The soil-waste mixture at INEL, when melted or vitrified, produces a glass/ceramic referred to as iron-enriched basalt (IEB). The only waste form presently approved for the disposal of long-lived radionuclides is borosilicate glass (BSG). Since studies have determined that large additions of

additives would be required to adjust the INEL waste composition to produce a BSG form, which would lead to a substantial waste volume increase, it would be advantageous if the INEL waste could be processed into an IEB waste form. One potential problem with producing the IEB material is the high melting temperature of the waste and soil (1400-1600°C). One technology that has demonstrated capabilities to process high melting point materials is the graphite electrode or plasma arc heated furnace. Technology based on the graphite electrode DC arc furnace is particularly attractive because it could provide the advantages of simplicity, high availability, safety, a high processing rate, and versatility.

Since the early 1900s, AC-powered graphite electrode arc furnaces have been used for reprocessing scrap metal for the steel industry. Drawing upon steel-making experience and advances in solid-state power conversion equipment, EPI has developed and refined a DC arc furnace for treating waste materials.

The graphite electrode DC electric arc is used to deliver thermal energy into the material to be processed. The ionized gas (plasma) of the arc generally will be maintained between an electrode and the material to be processed (called the "transfer" arc mode). EPI has designed a graphite electrode assembly that allows the arc to be maintained between two concentric electrodes (called the "nontransferred" arc mode). When the electrode is operated in this manner, the energy supplied to the material being melted is the result of thermal energy transfer only and is not as efficient as operation in the transferred arc mode. Although future programmatic activities plan to use the concentric electrode design, this technology was not used for the tests described in this report. Use of a currentcontrolled graphite electrode DC arc provides the high-temperature environment required for melting a range of heterogeneous waste forms (temperatures in excess of 1700°C can be produced in the material to be processed). In addition, the high-temperature region around the DC arc will destroy organic species and vapors that evolve from the material being processed. The volume of off-gas produced in the DC arc treatment of solid waste is greatly reduced relative to combustion treatment--typically by more than a factor of 10. Since the volume of gas is substantially reduced, there are fewer entrained particulates in the off-gas, and the size of the off-gas scrubbing equipment can be reduced substantially.

Graphite electrodes offer important advantages for buried waste treatment applications. Graphite electrodes are robust. These electrodes, as used in the simplified design of EPI devices, are well suited to the requirements of remote operation. Graphite electrodes do not have to be water cooled, in contrast to metal electrode plasma torches, thus eliminating any possibility of an accident that could cause pressurized water to contact molten slag or metal. Moreover, short DC arcs, which are highly effective in transferring thermal energy into the material to be processed, can be used. Graphite arc electrodes also permit the use of higher levels of DC arc current, providing the capability to transfer large amounts of the heat into the material to be processed. These factors make possible the high processing rates required for full-scale implementation of this technology at the INEL site.

OBJECTIVE OF THIS WORK

The primary objective of the work was to determine the suitability of the graphite electrode DC arc process to the treatment of INEL buried wastes through first an evaluation of an engineering-scale DC arc furnace followed by pilot-scale evaluation and demonstration of the technology for the treatment of buries waste simulants. A secondary objective of this work is to develop and implement a full range of diagnostics for high temperature thermal processes.

MARK I FURNACE EXPERIMENTAL PROGRAM

An engineering-scale (Mark I) graphite electrode DC arc furnace was used in the first phase of the PNL-MIT-EPI program. This furnace system consists of the furnace, off-gas handling components, a recirculating water system, a furnace shroud air exhaust system, DC power supply, nitrogen purge system, and instrumentation.

Furnace Equipment Description

The Mark I furnace shown in Figure 1, consists of a graphite crucible with an inside dimension of 13-1/2-in.-diameter tapering to 12-1/2-in.-diameter at the bottom. The inner height of the crucible is 21-in. A movable 2-in.-diameter graphite electrode is located in the center of the crucible The entire configuration is contained in a square steel box with hot face brick and insulating brick surrounding the graphite crucible. The Mark I furnace was capable of operating at DC power levels of 300 Kw. The furnace was operating under an inert atmosphere by purging 3-4 CFM of N₂ through the furnace chamber.

The Mark I furnace could either be operated in batch processing or a semi-batch processing mode. For this work the furnace was in all cases operated in the batch mode. A typical batch involved charging the furnace crucible with approximately 100 pounds of buried waste simulant.

Mark I Test Program

The DC arc technology is thought to be broadly applicable to the treatment of generic buried waste at many DOE complexes. However, because of the initial charter of this BWID program, this work focused on evaluating the technology's applicability to a specific buried waste stream, that of the INEL SDA.

The SDA is 88 acres of land within a larger waste burial site known as the RWMC at the INEL site. Waste of various categories was disposed of in the SDA beginning in 1952 when the burial ground, or RWMC, was opened. Wastes disposed of at the SDA include organics, inorganics, toxic metals, and radionuclides. Much of the SDA waste was generated at many off-site locations and shipped to the INEL RWMC. The largest percentage of offsite waste buried at the SDA was derived from the shipment and subsequent burial of Rocky Flats Plant (RFP) waste. The waste simulant development effort for the DC arc testing program used all known characteristics to establish a full complement of waste simulants, thereby permitting a complete evaluation of the DC arc technology relative to its capabilities to treat this type of waste.

Thirteen separate arc furnace runs were carried out to evaluate the processability of a wide range of waste simulant materials under a variety of operating conditions. The waste simulants developed and used in the Mark I testing program are grouped into five primary categories as shown below:

- · INEL soil only
- · INEL soil plus metals
- · INEL soil plus combustibles
- · INEL soil plus RFP 74 series sludges
- · INEL soil plus high vapor pressure metals.

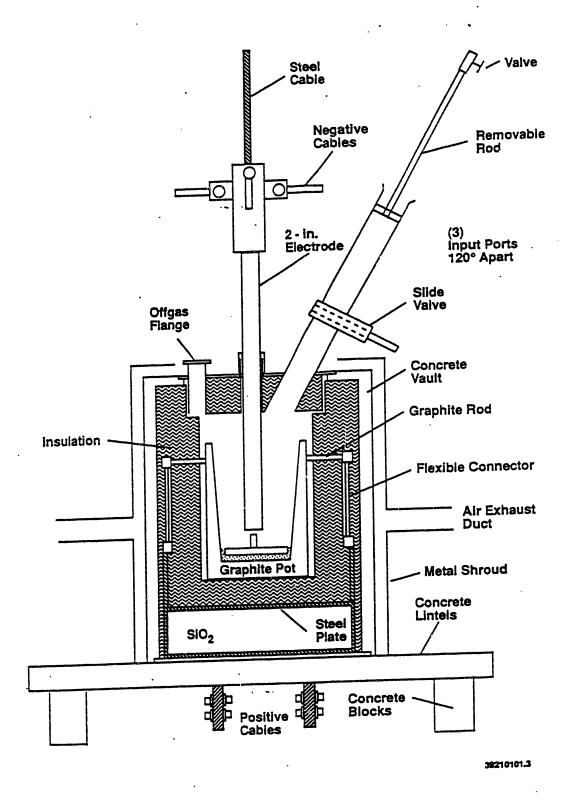


Figure 1. Mark I Engineering-Scale Furnace

Although other waste categories exist, these five categories cover the major fraction of waste at the INEL SDA, and would be sufficient to complete a nearly comprehensive evaluation of the treatment of SDA waste using the DC arc process.

Mark I Test Results

The results of the Mark I furnace testing program indicate the graphite electrode DC technology is well suited to the treatment of buried wastes. The technology has been demonstrated to be effective for treating a diverse mixture of materials, including metals, combustibles, sludges similar to those found at the INEL site, and mixtures containing high vapor pressure metals.

The Mark I tests that were completed all produced products that were in a highly reduced state, primarily because the Mark I furnace operating conditions were not optimized. It is not anticipated that the pilot-scale (Mark II) furnace will produce a highly reduced waste form. At this time it is unclear whether or not the reduced state of the glass would diminish the quality of the waste form. The only concern with a reduced redox state is the capability to contain adequate amounts of metal in the glass.

The Mark I tests using large quantities of combustible material resulted in the nearly complete pyrolysis of the carbonaceous material and the formation of a densified glass product from the resultant inorganic ash and soil mixture.

The capability to treat wastes containing large quantities of metals was demonstrated with extremely favorable results. The metal phase separates cleanly from the glass phase. Both the glass and metal products are highly densified and would serve as a suitable waste form. If it is later determined that the majority of the radioactive contaminants remain in the glass phase, it may be advantageous to operate an arc furnace in a mode (reducing environment) that will promote the separation of metals from the glass. This determination will be made under future program activities and in conjunction with ongoing programs within the BWID program.

The unique capability of the graphite electrode to operate in the submerged mode will substantially improve the partitioning of materials processed or treated using this technology. This was effectively demonstrated with operation of the arc in both the submerged and unsubmerged modes. The results of these two runs showed conclusively that the emissions from an arc furnace could be reduced by over a factor of two. The volatility of HVPMs from a DC arc furnace is comparable to that for the high-level waste treated in liquid fed ceramics melters (LFCMs) and by ISV. For operation of the furnace in the nonsubmerged mode, the retention of Cs within the glass product was measured to be 98.1 wt%. For submerged arc operation, 99.12 wt% Cs retention was observed. These values compare favorably to 90% to 99.25% for the LFCM technology and 99.63% for ISV (Carter 1988, Peterson 1992, Goles 1983). The total mass fraction lost to the off-gas was measured to be 0.3 wt% and 0.54 wt% for submerged and nonsubmerged arc operation, respectively, based on all inorganic constituents of the feed material.

MARK II PILOT-SCALE DEMONSTRATION

To completely evaluate the graphite electrode DC arc technology for the treatment of buried wastes the Mark II pilot-scale furnace was installed at the Plasma Fusion Center at MIT. This furnace will provide required data from a furnace system designed specifically for the treatment of waste materials. The furnace system incorporates features for waste feeding, containment, glass and metal pouring in a sealed chamber, and a complete prototypic off-gas system.

The Mark II furnace design incorporated many of the features that will be required for a semiremote or remotely operable furnace, including prototype provisions for alpha-containment and the capability of continuous processing at power levels of over 1 MW with a waste processing rate of 500-700 pounds per hour. Much of the buried waste across the DOE complex contains a high weight fraction of metals. For this reason, the Mark II furnace was designed with the capability to separate the IEB slag (glass) from the metal phase through an auxiliary furnace tapping arrangement. The complete testing and optimization of the Mark II furnace will provide DOE with the required information to fully evaluate the graphite electrode DC arc technology for the treatment of buried and mixed wastes.

Mark II Furnace Description

The Mark II furnace is approximately 23 ft in height and 7 ft in diameter. The furnace is a refractory lined carbon steel vessel shown in Figure 2. Because this furnace is a research tool four soft patch panels are located around the circumference of the furnace 90 degrees apart to provide access to the furnace chamber. A waste feed system is attached to one of the ports, and a glass discharge chamber is located directly (180°) opposite the feed system. The other two soft patch locations will be utilized for the furnace diagnostic equipment described in a following section. The off-gas system incorporates components that are anticipated to be in a final arc or plasma furnace system used to process actual radioactively contaminated buried waste. The electrode assembly in the Mark II furnace incorporates a unique coaxial arrangement with a outer graphite electrode that is 14 inches O.D. and 10 inches I.D.. The inner electrode is a solid 6 inch diameter piece of graphite. This electrode assembly posses the capability to operate in both the transferred arc mode (arcing from the electrode to the material being processed) or the nontransferred mode (arcing between the inner and outer electrodes. This capability will allow the furnace to easily be started by operating the torch in the nontransferred mode to initially melt the material in the hearth. When the molten material become sufficiently electrically conductive the operation of the electrode can then be changed to the transferred mode of operation which is a more efficient method of providing heat-energy to a material being melted.

Mark II Testing Program

The testing of the Mark II furnace will be similar in many respects to the Mark I program described previously. The waste simulants to be processed in the Mark II furnace will closely resemble those processed in Mark I. The difference in the Mark II tests are that the material will be processed in larger quantities and in a continuous processing mode. In addition the operation of the furnace will be evaluated from the standpoint of materials feeding, glass and metals pouring, and the overall furnace operability including off-gas. A complete material balance will be established for the various waste simulants that will be process in the Mark II furnace including electrode material consumption. The Mark II furnace like the Mark I can operate with the electrode submerged in the overburden. Further evaluations with respect to submerged and unsubmerged arc operations will be conducted.

Mark II Furnace Diagnostics

A significant effort for the Mark II furnace testing program involves the development and application of furnace diagnostics. The diagnostics effort includes the development of analytical equipment for measurements of furnace and glass temperatures, online off-gas emissions both in the furnace chamber and off-gas line, spatially resolved gas temperatures and arc zone temperatures, etc.. The diagnostics program is drawing largely from the Plasma Fusions Centers experience in high temperature diagnostics from previous work in plasma fusion programs.

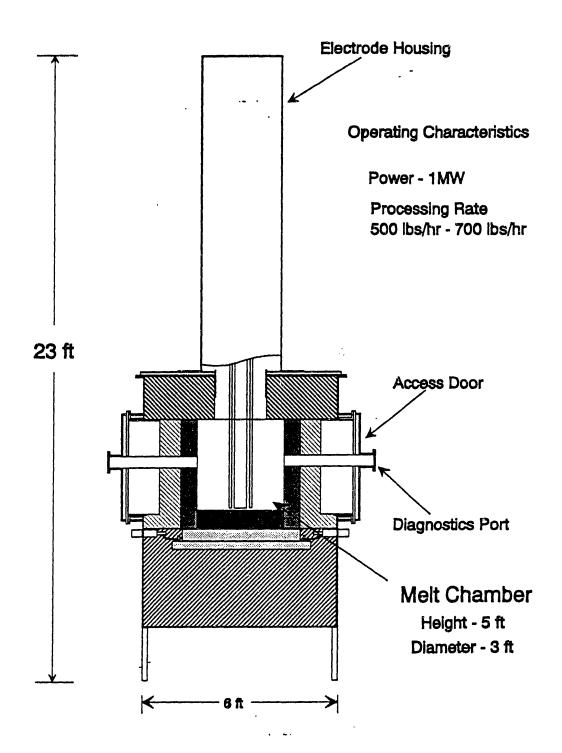


Figure 2. Mark II Pilot-Scale Furnace.

SUMMARY

The overall operability of the Mark I furnace proved to be a success. Information gained from the Mark I program was used to enhance the design of the Mark II furnace. The Mark II furnace is an optimized furnace that has been specifically designed with the goal of treating buried wastes such as those found at the INEL. The future activities planned for the Mark II furnace will build on the information obtained during the Mark I experiments.

The Mark I furnace system, Mark II furnace system, and the diagnostics equipment are now in place at the Plasma Fusion Center at MIT and available as test bed for evaluation of the graphite electrode DC arc furnace technology for the treatment of various DOE wastes.

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