INDUCTION MELTING FOR VOLUME REDUCTION OF METALLIC TRU WASTES

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ABSTRACT

Volume reduction of metallic transuranic wastes offers economic and safety incentives for treatment of wastes generated at a hypothetical commercial fuel reprocessing facility. Induction melting has been identified as the preferred process for volume reduction of spent fuel hulls, fuel assembly hardware, and failed equipment from a reprocessing plant. Bench-scale melting of Zircaloy and stainless steel mixtures has been successfully conducted in a graphite crucible inside a large vacuum chamber. A low-melting-temperature alloy forms that has demonstrated excellent leach resistance. The alloy can be used to encapsulate other metallic wastes that cannot be melted using the existing equipment design.

INTRODUCTION

Metallic radioactive wastes may be generated at facilities for the handling and treatment of spent fuel including hypothetical commercial fuel reprocessing plants and facilities for the consolidation of spent fuel rods. Spent fuel hulls, fuel assembly hardware, and failed equipment are large-volume, transuranic (TRU) wastes that will be generated at these facilities. Significant incentives exist for reducing the volume of these wastes prior to their shipping and disposal. The Nuclear Waste Treatment Program at the Pacific Northwest Laboratory (PNL) is chartered to develop and evaluate methods for treating these wastes safely and economically.

A recent evaluation of alternative treatments has identified the incentives for treatment of the TRU wastes from reprocessing.[1,2] This evaluation indicated that extensive treatment (compared to no treatment) could save about $1.7 billion in the treatment, transportation, and disposal of wastes from reprocessing 70,000 MTU of spent fuel. The potential savings result in large part from the reduction in the volume of the metallic wastes by melting. The study also identified that potential risks during handling and disposal are reduced if the wastes are melted.

Melting of metallic wastes has been evaluated in the United States and abroad as a method to achieve volume reduction of low-level and TRU wastes. The studies have considered ferrous and nonferrous wastes and have investigated volume reduction alone and volume reduction with decontamination.[3-8] Induction melting of low-level beta- and gamma-contaminated steel wastes has been demonstrated at the Waste Experimental Reduction Facility (WERF) at the Idaho National Engineering Laboratory (INEL).[9]

Initial volume reduction studies at PNL emphasized the melting of Zircaloy cladding wastes using the Inductoslag process.[10-13] A recent study evaluated seven techniques, including the Inductoslag process, for volume reduction of metallic wastes from fuel reprocessing.[14] Vacuum coreless induction melting was rated highest. This evaluation was based upon the following criteria: complexity of process, state and type of development required, safety, process requirements, and facility requirements. Therefore, current development activities at PNL are directed toward completing the development of the vacuum coreless induction process for applications requiring the volume reduction of contaminated Zircaloy and other metallic TRU wastes.[15,16] This report presents a summary of that work.

DEFINITION OF METALLIC WASTES

Metallic TRU wastes from the reprocessing or consolidation of spent fuel will include hulls and assembly hardware, failed equipment, and miscellaneous metals remaining from the treatment of high-efficiency particulate air (HEPA) filters and combustible TRU trash. The hulls and hardware represent the largest volume of waste with an estimated volume of 640 m^3 generated annually at a 1500-MTU/yr reprocessing plant. Failed equipment would add an additional 26 m^3/yr to the metallic waste volume. Melting these wastes would reduce their annual volumes to 93 m^3 and 3.4 m^3, respectively.[1]

The nonfuel components of spent fuel are comprised of Zircaloy 4 (80 to 87 wt%), stainless steel (11 to 14 wt%), Inconel (1 to 5 wt%), and other alloys (1 to 3 wt%). The combined metal content for the fuel hulls and assembly hardware is shown in Table I. Failed equipment is assumed to be 100% ferrous alloys.[3] The radionuclide content of the spent fuel hulls is dependent upon the burnup of the fuel, the

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TABLE I

Combined Metal Content of Fuel Hulls and Assembly Hardware

Element	Metal Content, wt%
Zr	80.7
Fe	9.9
Ni	3.0
Cr	3.0
Sn	1.3
Cu	0.9
Other	1.3

storage time since discharge from the reactor, and the efficiency of the fuel dissolution during reprocessing. To determine the radionuclide content on typical spent fuel hulls, six 5-cm pieces of fuel cladding from a pressurized-water reactor (PWR) fuel bundle processed during the Nuclear Waste Vitrification Project at PNL were dissolved and analyzed radiochemically.[17] The average radionuclide content and standard deviation are shown in Table II.

TABLE II

Weight Fraction of Radionuclides Attached to Spent Fuel Hull

Radionuclide	Weight Fraction g radionuclide/g hull
Cesium-137	4.1E-6 ± 5.7E-7
Cesium-134	5.3E-9 ± 1.1E-9
Antimony-125	9.8E-8 ± 8.3E-9
Cobalt-60	7.9E-9 ± 2.2E-9
Europium-154	3.8E-8 ± 7.1E-9
Strontium/Yttrium-90	2.5E-6 ± 3.1E-7
Plutonium-239+240	5.8E-6 ± 2.3E-6
Americium-241/Pu-238	3.6E-7 ± 1.6E-7
Curium-244	3.3E-9 ± 2.0E-9
Uranium-total	1.6E-4 ± 4.7E-5
Ruthenium-106	5.9E-11 ± 7.2E-12

The PWR fuel was discharged on May 10, 1974 with a burnup of 29500 MWd/MTU.

PROCESS DESCRIPTION

PNL is developing the vacuum coreless induction melting process for treatment of TRU metallic wastes. Induction melting requires the generation of an electric current in the metal using electromagnetic induction. Coreless induction means that the induction coil surrounds the metal charge, and vacuum means that the melting is conducted in a partial vacuum. The coreless induction melting process requires a crucible to contain the molten metal. For Zircaloy, the crucible is usually made of graphite; and for steel, the crucible is made of an oxide refractory material. The crucible can be drained by either a tilt-and-pour technique or through a pour nozzle in the bottom of the crucible. PNL is developing the latter method.

Figure 1 shows the induction furnace used at PNL. The system includes a single induction coil (23-cm high, 20-cm diameter) around a graphite crucible. A water-cooled cold finger is used to close the bottom pour nozzle. The molten metal is drained into a stainless steel canister beneath the crucible. The crucible, coil, and canister are contained within a large vacuum chamber. A hopper and feed mechanism adjacent to the vacuum chamber are used to add scrap metal to the crucible while maintaining the vacuum. The 200-kW, 3-kHz power supply and the vacuum system are located outside the vacuum chamber.

In a typical experiment, the melt chamber is evacuated to 0.1 Pa and is then backfilled with argon to 33 kPa (one-third atmosphere). The crucible is then heated and the initial metal waste charge is fed into the crucible. As the initial charge melts, additional feed is added until the crucible is full of molten metal. The water-cooled cold finger is then removed; and, after about 15 seconds, the frozen metal plug in the sprue melts and the crucible drains. The cold finger is then replaced and another melt cycle is started.

The majority of tests have been performed using a mixture of 18 to 20 wt% 316 stainless steel and 80 to 82 wt% Zircaloy 4. Although zirconium metal has a melting point of 1852°C and iron has a melting point of 1536°C, a mixture of 85 wt% Zr and 15 wt% Fe forms a eutectic with a melting point of 948°C. Nickel and chromium form similar eutectics with zirconium. Fortuitously, this composition is approximately the same as the combined fuel hull and assembly hardware mixture expected from a fuel reprocessing plant. The Zircaloy/stainless steel mixture melted rapidly at ~1260°C. The best pouring temperature was 1300 to 1325°C. When melting oxidized Zircaloy hulls in our bench-scale system, the highest melting rate was 41 kg/h at 40 kW with a power consumption of 1.03 kWh/kg. Unoxidized hulls melted at 53 kg/h with a power consumption of 0.75 kWh/kg. The 1.1-cm-thick graphite crucible easily withstood eight melt cycles with only a 0.08-cm-deep by 0.64-cm-wide erosion zone at the melt line.

One test was conducted with a simulated PWR fuel bundle skeleton with a composition of 52 wt% stainless steel, 30 wt% Zircaloy 4, and 18 wt% Inconel. This mixture was completely melted at 1593°C but would not melt through the freeze plug with the bulk melt temperature as high as 1700°C. A second induction coil around the pour nozzle is needed to aid in melting the plug when the crucible is to be drained.

Another melting test was conducted on a mixture similar to that expected from a TRU waste incinerator. The residue from the incineration of simulated process wastes included 44 wt% carbon steel, 19 wt% stainless steel, 19 wt% glass, and 19 wt% electric motor parts. A molten pool with a viscous slag on top was established at a temperature of 1537°C. Within 15 minutes the molten metal had dissolved through the 1.7-cm-thick graphite crucible, demonstrating the need for an oxide refractory crucible for melting ferrous metal wastes.

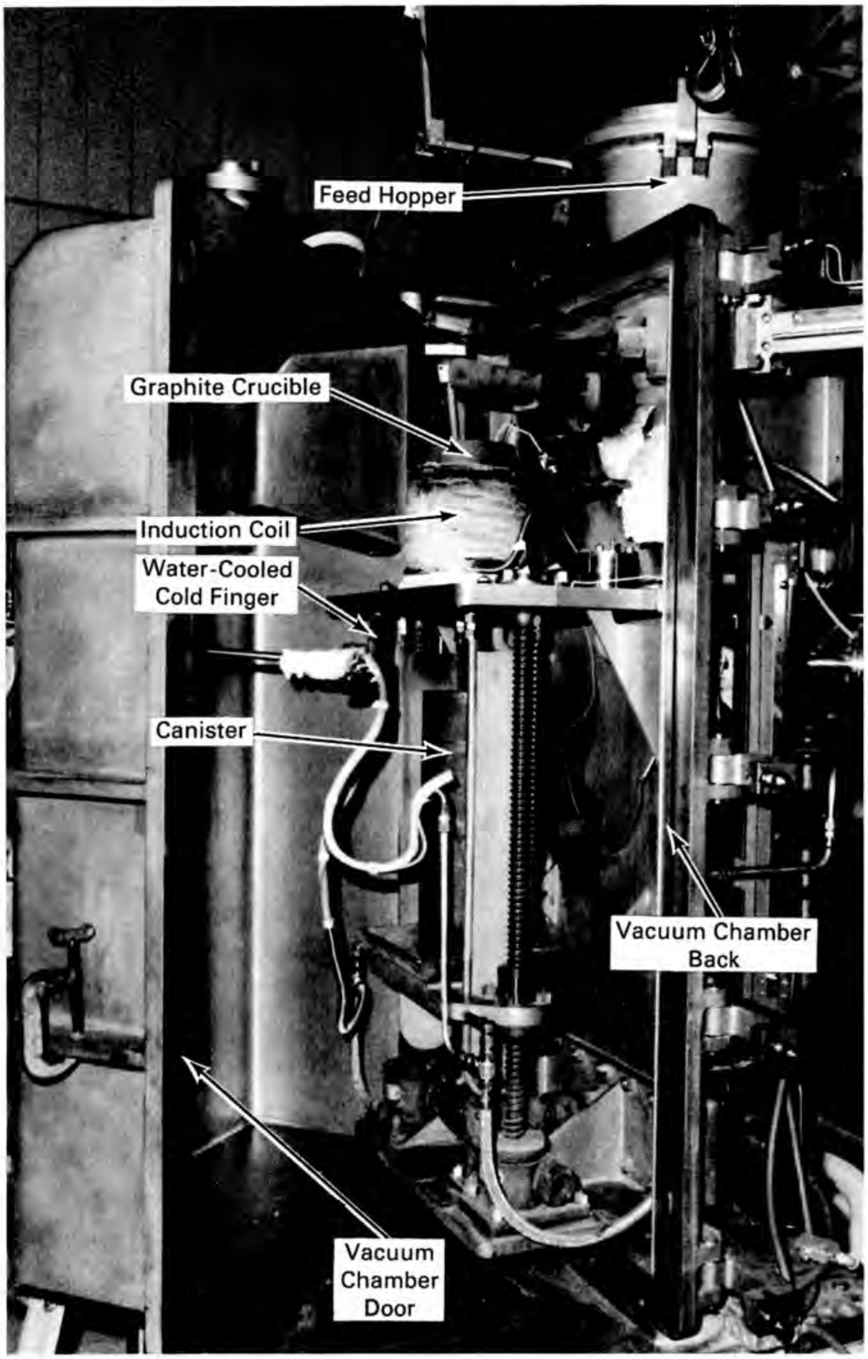
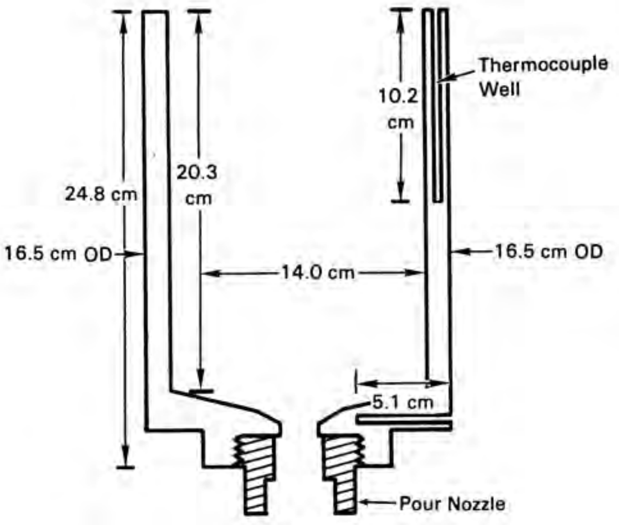


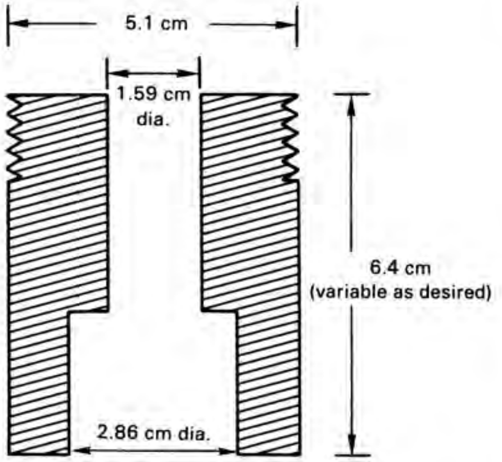
Fig. 1. Interior of vacuum coreless induction melting furnace chamber.

Operational problems observed while processing these simulated metallic wastes have centered around the pour nozzle. One problem has been the formation of a solidified drop of metal at the tip of the nozzle, which prevented replacement of the water-cooled plug prior to the next melt cycle. Figure 2 shows the current nozzle design that has mitigated the problem. The plug must also be replaced before the metal has cooled. A second problem results when the metal plug in the pour nozzle fails to melt when the water-cooled plug has been removed. It is expected that a second induction coil surrounding the nozzle will remedy this problem.

The largest volume of wastes from fuel reprocessing will be the hulls and assembly hardware. However, a small volume of failed equipment will also be generated. Because these wastes are primarily stainless steel, they cannot be melted in the graphite crucible used to melt the Zircaloy/stainless steel eutectic. One solution would be to change the crucible material to suit the material being melted. A second solution would be to place the steel waste in the receiving canister and simply pour the molten Zircaloy/stainless steel eutectic over the wastes, thereby encapsulating them. We have successfully demonstrated this second technique, encapsulating



A. Graphite Crucible and Pour Nozzle



B. Pour Nozzle Detail

Fig. 2. Schematic of current design for the bench-scale graphite crucible and pour nozzle.

simulated failed equipment in the Zircaloy/stainless steel eutectic as shown in Fig. 3. Minimum densities observed are at least 93% of the theoretical density for the eutectic, indicating complete encapsulation with little or no void formation.

The product of the melting is a monolith of the eutectic or mixture inside a stainless steel canister. No reactions between the canister and the eutectic have been observed. Several melt batches are required to fill a canister. Each casting forms a small contraction pipe at the top, as shown in Fig. 4. The pipe is easily filled by the next casting and observed densities are typically greater than 99% of theoretical.

As part of the initial characterization of the metal melting product, specimens cut from an ingot of the Zircaloy/stainless steel eutectic were exposed to deionized water, silicate water, and brine at 90°C in accordance with the MCC-1P leach test procedure.[18]

Fig. 3. Example of encapsulation showing ferrous alloys encapsulated in the Zircaloy/stainless steel eutectic mixture.

The test was run for 27 days rather than the standard 28 days. The ingot was prepared by melting a mixture of 18 wt% 316 stainless steel and 82 wt% Zircaloy 4 at a maximum temperature of 1371°C for 22 minutes. Nonradioactive tracers including 1.8 g $Sr(NO_3)_2$, 1.1 g $CsNO_3$, and 5.7 g $Ce(NO_3)_3$-$6H_2O$ were added to the 1001 g of metal mixture prior to melting. Table III shows the results of the test. The specimens showed only small weight changes and very little material was released to the leachates. None of the tracers were detected in the leachates at the conclusion of the test. Longer test intervals of 6 and 12 months are planned for FY 1986 to determine the long-term behavior of this waste form.

PROTOTYPE MELTER DESIGN

A conceptual design for a full-scale, remotely operated, vacuum coreless induction melter system has been prepared. The system includes the induction furnace plus a canister-handling subsystem and mechanisms to feed 55-gallon drums directly into a crucible or unpackaged wastes into the crucible through a side-fed airlock vibratory system. The crucible assembly consists of an inner replaceable graphite liner, an outer permanent graphite susceptor, insulation, crucible frame, and induction coil. The crucible has a maximum capacity of 1400 kg. A second induction coil surrounds the pour nozzle. Power for the primary coil is provided by a 400-kW, 1-kHz power supply and a 100-kW,

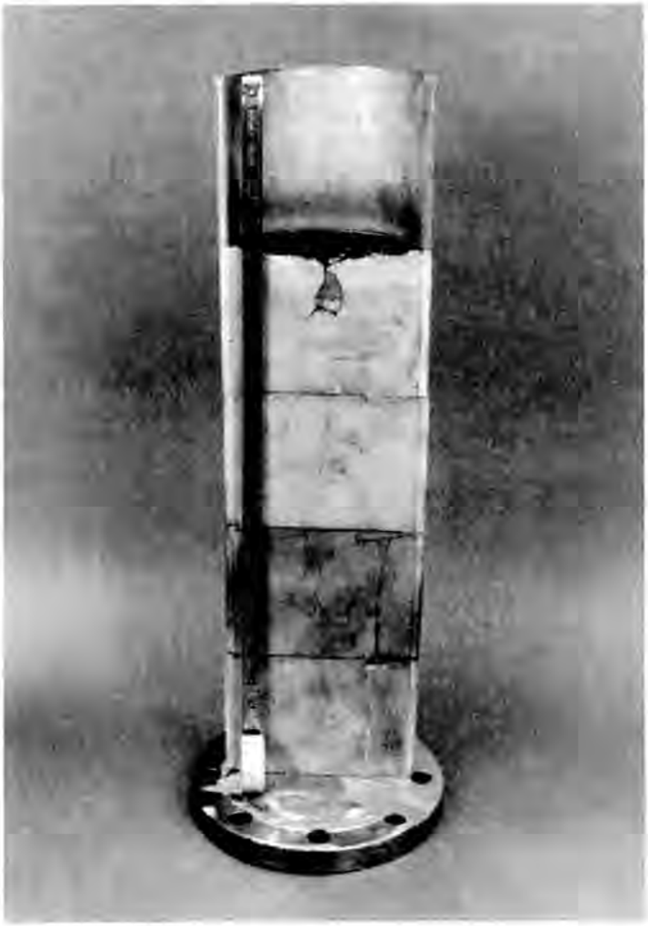


Fig. 4. Cross section of a Zircaloy/stainless steel ingot inside a stainless steel canister. Ingot was cut into four sections corresponding to four consecutive pours. Contraction pipe from the last pour remains at the top.

TABLE III

Results of MCC-1P Leach Test on Zircaloy/Stainless Steel Waste Form*

	Deionized Water	Silicate Water	Brine
Percent Weight Change	-4E-4	+4E-3	+8E-3
pH Change	+0.3	+1.9	+1.7
Normalized Elemental Mass Loss (g/m^2)			
Fe	0.02		0.18
Mn	0.33		
Ni			0.23
Zr			0.03

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*27 days rather than standard 28 days.

3-kHz power supply is planned for the pour nozzle coil. The crucible and coils will be located on the chamber door of the vacuum chamber such that they are accessible when the door is opened. The detailed design for this prototype melter will be completed in 1986.

SUMMARY

Volume reduction of metallic TRU wastes offers economic and safety incentives for treatment of wastes generated at a commercial fuel reprocessing and fuel fabrication facility. Of the numerous processes that might be applicable for the consolidation of metallic wastes, vacuum coreless induction melting was identified as the preferred technique considering the complexity and safety of the process, the state of development, and process and facility requirements. Bench- and laboratory-scale tests are under way to evaluate the performance of the melting system with simulated wastes typical of those that would be generated during fuel reprocessing.

Fuel hulls and assembly hardware represent the largest volume of TRU wastes, and the waste volume can be reduced by a factor of seven by melting. The major components of these wastes, zirconium and iron, form a low-melting eutectic with a composition of 85 wt% Zr and 15 wt% Fe. This ratio is approximately the same as the ratio of Zircaloy to stainless steel in the fuel hulls and associated assembly hardware.

Vacuum coreless induction melting in a graphite crucible with a bottom pour nozzle is a simple and reliable method for reducing the volume of Zircaloy hulls and stainless steel hardware. The product is a leach-resistant monolith contained within a stainless steel canister. Other metallic wastes that are not compatible with the graphite crucible can be encapsulated in the eutectic by placing the wastes in the canister prior to filling. This melting technique may also be applicable to other metallic wastes including those that would be generated during spent fuel consolidation activities.

Studies are needed to demonstrate the technique on a production scale. Off-gas treatment systems must be designed and tested. Testing using actual radioactive wastes is needed to understand the volatility and tendency of the radionuclides to form slags.

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