The Cladding Hull Decontamination and Densification Process

Part 2 Densification by Inductoslag Melting

**R. G. Nelson
D. R. Montgomery**

April 1980

**Prepared for the U.S. Department of Energy
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SUMMARY

The Inductoslag melting process was developed to densify Zircaloy-4 cladding hulls. It is a cold crucible process that uses induction heating, a segmented water-cooled copper crucible, and a calcium fluoride flux. Metal and flux are fed into the furnace through the crucible, located at the top of the furnace, and the finished ingot is withdrawn from the bottom of the furnace. Melting rates of 40 to 50 kg/hr are achieved, using 100 to 110 kW at an average energy use of 2.5 kWh/kg.

The quality of ingots produced from factory supplied cladding tubing is sufficient to satisfy nuclear grade standards. An ingot of Zircaloy-4, made from melted cladding tubing that had been autoclaved to near reactor exposure and then descaled by the hydrogen fluoride decontamination process prior to Inductoslag melting, did not meet nuclear grade standards because the hydrogen, nitrogen, and hardness levels were too high.

Melting development work is described that could possibly be used to test the capability of the Inductoslag process to satisfactorily melt a variety and mix of materials from LWR reprocessing, decontamination, and storage options. Results of experiments are also presented that could be used to improve remote operation of the melting process.

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THE CLADDING HULL DECONTAMINATION AND DENSIFICATION PROCESS DENSIFICATION BY INDUCTOSLAG MELTING

INTRODUCTION

The technology presented in this document was developed to handle metallic wastes from a plant designed to reprocess current light water reactor (LWR) fuels. Present United States policy does not permit the reprocessing of LWR fuels because of the danger of nuclear weapons proliferation. Should a proliferation-resistant fuel cycle be developed, the technology presented here will be applicable to these wastes.

During the reprocessing of LWR fuels, fuel bundles are sheared in short lengths for acid leaching of the fuel pellets. The residues from the acid leach include fuel cladding, as well as massive end fittings, fuel spacer grids, assorted springs, guide thimbles, and flow channels. These materials constitute a high-volume waste representing about 22% of the original fuel bundle weight and have a bulk density of about 1.1 kg/L. Without densification, they occupy a substantially larger volume than vitrified high-level wastes. Densification by melting will reduce this volume by a factor of 6. As recovered from the dissolver, these hardware residues represent about 325 kg/MT of uranium, and consist of 70 to 80 wt% Zircaloy, 12 to 22 wt% stainless steel (SS) and 8 wt% Inconel. They contain sufficient actinide contamination to be classified as transuranic wastes.

Pacific Northwest Laboratory (PNL) has evaluated the decontamination and melt densification process for cladding hulls as a means to minimize waste storage requirements. A method for decontaminating the hulls prior to melting to remove TRU element contamination has been developed and is described in a companion document.⁽¹⁾ This report describes the design, installation and the semi-remote operating results of an Inductoslag melting furnace. The selection of the Inductoslag melting process and the preliminary melting results have been previously reported.^(2,3)

The work described was performed at PNL. Supporting melting development work was performed through subcontract to the United States Bureau of Mines, Albany Metallurgical Research Center, Albany, Oregon.

DESCRIPTION OF THE INDUCTOSLAG MELTING PROCESS

Because of the chemical activity of Zircaloy at its melting point (1850°C), melting must be done in a vacuum or inert atmosphere. Conventional crucible materials react vigorously with molten Zircaloy. For example, Zircaloy melted in graphite crucibles picks up excessive carbon. Consequently, cold crucible processes were developed to melt Zircaloy.⁽⁴⁾ These processes have also been used for melting and remelting iron, nickel, and cobalt alloys. Alternate cold crucible melting techniques have been evaluated for possible use in cladding hull densification.⁽²⁾ Conclusions drawn from this evaluation led to the selection of the Inductoslag process as the most promising process for cladding hull densification.

The Inductoslag melting process (Figure 1) was developed originally by Clites and Beall^(5,6) of the U.S. Bureau of Mines, Albany, Oregon. The process uses induction heating, where the melt is insulated from a segmented, water-cooled crucible by a layer of frozen slag. A pool of Zircaloy-4 and CaF_2 is melted on Zircaloy-4 "starting stub" and subsequent fuel material is fed into the top of the crucible as the ingot is withdrawn through the bottom of the crucible.

Previous work⁽³⁾ with the furnace developed by the Bureau of Mines successfully demonstrated that simulated cladding hulls of segregated, clean or oxidized Zircaloy, stainless steel, and Inconel could be melted at rates up to 28 kg/hr. The remelted, clean Zircaloy-4 ingots were still within nuclear grade chemical and hardness specifications. Corrosion tests of tubing fabricated from the 10-cm-dia ingots were also within nuclear specifications. These successful demonstrations were the basis for proceeding with the design and construction of an Inductoslag melting furnace specifically for melting cladding hulls.

FIGURE 1. The Inductoslag Melting Process Developed Originally by Clites and Beall at the Albany Metallurgical Research Center, U.S. Bureau of Mines, Albany, Oregon. (5)

DESCRIPTION OF THE PNL INDUCTOSLAG MELTING FURNACE

The PNL Inductoslag melting furnace was designed to be installed in a radioactive facility so that radioactive demonstration melting could be conducted after suitable testing and operation in a nonradioactive environment. Figure 2 shows the layout of the melting and decontamination system, and Figure 3 shows the major components of the melting system in place. The furnace rests on a 0.6 M high stand. The height of the stand represents the difference in elevation between the floor of the hot cell facility and the floor outside the facility.

FIGURE 2. Layout of the Chemical Decontamination and Densification System

FIGURE 3. The Inductoslag Melting Furnace

FURNACE DESIGN

The furnace was constructed from stainless steel with smooth surfaces and rounded corners to aid remote cleaning. Bolt heads are uniform in size and conically shaped to facilitate remote maintenance and operation, and the large vertical door is designed to expose the interior of the furnace chamber for remote maintenance and operation.

The 10-cm-dia crucible (Figure 4) has 24 vertical water-cooled, coaxial copper tubes attached to water headers. The tubes are slightly flattened on the crucible inside diameter (ID). Not shown in Figure 4 is a layer of glass tape wrapped around and embedded in a thin layer of ceramic on the outside of the vertical section of the crucible. The tape strengthens the crucible and provides electrical insulation between the segments.

Zircaloy-4 cladding hulls (melt stock) are loaded into the feed hopper through a hinged door on top of the furnace. The hulls are fed into the furnace through a trough and the ingot is withdrawn from the furnace using twin ball lead screws. Maximum withdrawal is 80 cm. Air motors are used to power the ingot withdrawal, melt stock feeder, and flux feeder. An electric motor powers an agitator in the feed hopper to prevent the melt stock from collecting together (bridging).

There are two viewing ports on top of the furnace door. A television camera is mounted on each viewing port, which allows an operator to view the molten pool and feed trough through television receivers mounted on a control console. The console is located outside the simulated cell. The system can be monitored and operated remotely by one operator from the console. Loading the furnace with hulls before melting and ingot removal after melting require manual operation at the present time.

A closed loop recirculating water system is used to cool the crucible, withdrawal table, the stool, heat shields, chamber, and the chamber door. Once-through water is used to cool the power source and induction coil. Water filters are used on the water lines that supply water to the crucible and power source.

FIGURE 4. The Inductoslag Melting Crucible

Power for melting is supplied by 200 kW 3 kHz solid state system by way of a 16-cm long, 16-turn induction coil fabricated from 1.3 cm-dia flattened tubing.

The vacuum pumping system uses a 100 L/sec blower backed by a 10 L/sec mechanical pump.

SAFETY SYSTEMS

Cold crucible melting processes require cooling water. A water leak in a critical component could result in a violent molten metal water reaction. The safety systems described below were designed primarily to prevent such an occurrence. Component protection in the event of a service or component failure is of lesser importance.

- An air pressure gage on the supply line is visible from the control panel. Lack of air to any one of the three systems mentioned above, or to the material feeder, the flux feeder, or the ingot withdrawal will light the respective panel light and sound an alarm.
- Interlock switches within the power source will render the furnace inoperative if the inlet water temperature is too high or the pressure too low. The induction heating coil, which receives its cooling water from the power source, has an open sight drain and water temperature indicator on the outlet line.
- The melting furnace cooling water is recirculated through the heat exchanger. The recirculator has a reservoir with a gage that indicates the level of reserve water. When the water in the reservoir is low, a panel warning light comes on and an alarm sounds.
- The crucible assembly, withdrawal table, heat shield, furnace chamber, and the furnace door are cooled by five separate parallel water circuits. Each circuit has a flow switch connected to a light and audible alarm on the control panel. Each circuit also has a vane type flow meter that measures the rate of flow of water. The crucible assembly is also interlocked into the melting power source. The inlet and outlet water temperature of the crucible are indicated by gages on the control panel.

DEVELOPMENT OF THE INDUCTOSLAG MELTING SYSTEM

Melting development experiments were designed and conducted to test the capability of the Inductoslag process to satisfactorily melt a possible variety of material mixes. Material mixes were derived from variations in LWR reprocessing, decontamination and storage options. Work was also conducted to improve the remote operation of the melting process.

ALLOY MELTING RATE STUDIES

Alloy melting rate studies were conducted at the U.S. Bureau of Mines laboratory using their 100 kW - 10 kHz Inductoslag melter while the PNL melting furnace was being assembled.

Feed material was comprised of an approximate average mixture of 85 wt% Zircaloy - 10 wt% 304 stainless steel and 5 wt% Inconel 718. Previous work⁽²⁾ has shown that this mixture has a slow melting rate of 5 to 8 kg/hr when using 85 kW of power and a flux mixture of 48 wt% CaF_2 to 52 wt% MgF_2 . Feed stock comprised of this average metal mixture resulted in some unmelted Zircaloy potted in a matrix of eutectic. The resulting ingots were free of voids and suitable for direct storage.

A series of melts was made in an attempt to increase the melting rate of this average metal mixture. Two crucible sizes were used, one 10 cm (4 in.) in dia and another 13 cm (5 in.) in dia. Three flux compositions were used having liquidus temperatures of 950° , 1100° , and 1150°C , respectively. Table 1 shows that the experiments did not yield improved melting rates.

1. The 13 cm ingots at near constant power did not yield higher melting rates. In fact, melting rates for the 13 cm ingots were lower than the melting rates achieved for the 10 cm ingots.
2. Variations in slag composition yielded no significant changes in melting rates. The ingot was never completely coated with slag in any of the melts. It was also evident that contact had occurred between the crucible wall and the ingot. This metal-to-metal contact causes high rates of

TABLE 1. Zircaloy-Stainless Steel Alloy Melt Rate Experiments for the Inductoslag Process

Melt Number	Ingot (cm) Dia	Flux Composition Weight	Slag C Liquidus	Metal Composition Weight % ^(b)	Weight Metal Melted, kg	Power Input kW	Time Min ^(a)	Melt Rate kg/hr	Comments
538-2	10	73 CaF ₂ /27 MgF ₂	1150	Zr-15Fe+Cr++Ni ^(b)	5.0	85	39	7.7	Fuel hulls + 304 SS to make 15% Fe alloy
538-3	10	67 CaF ₂ /33 MgF ₂	1100	Zr-15 SS	5.0	80	60	5	Fuel hulls + 15% 304 SS
538-4	10	67 CaF ₂ /33 MgF ₂	1100	Zr-15 SS		85			Run terminated, pool dumped into bottom of furnace
548-1	13	52 CaF ₂ /48 MgF ₂	950	Zr-15 SS		95			Bridged
548-2	13	52 CaF ₂ /48 MgF ₂	950	Zr-15 SS	5.2	90	71	4.4	Chopped Zr sheet + 15% 304 SS
548-3	13	67 CaF ₂ /33 MgF ₂	1100	Zr-15 SS	5.8	95	100	3.5	Chopped Zr sheet + 15% 304 SS

(a) Time measured from start of feeding scrap to an established molten pool until last scrap added was completely melted.

(b) Sufficient 304SS added to make a 15 Fe alloy.

heat loss from the metal pool and cladding residues. Therefore, alloy compositions for melting rate studies were chosen at 0.5, 3.0, and 4.0 wt% Inconel balance Zircaloy-4. Reject Inconel 750 plenum springs were procured from a spring manufacturer for use as the Inconel in the alloys. The Zircaloy-4 consisted of 3.04 kg of chopped cladding tubing and 1.5 kg of simulated end cap material. Pure calcium fluoride was used as flux.

The results of these melt rate studies are shown in Table 2. All three compositions melted at rates between 21 and 27 kg/hr and thus, compared favorably with the results obtained from Heat Numbers 660 and 661, in which pure Zircaloy-4 was melted at 23 and 27 kg/hr, respectively. Figure 5 shows an etched cross section of Heat Number 662, in which Zircaloy-4, 4 wt% Inconel, melted at 25 kg/hr. Previous work⁽²⁾ showed that the Zircaloy-4, 4 wt% Inconel alloy had a melt rate of only 4.7 kg/hr when a 52 wt% CaF_2 - 48 wt% MgF_2 eutectic flux was used. Observations at that time were that the ingot, after melting, had no visible slag on the ingot side wall. The melting of these alloys with the pure CaF_2 flux showed a thin uniform layer of flux on the ingot side wall. This thin uniform layer of flux provided a layer of insulation between the ingot and the water-cooled crucible. This layer of insulation was similar to, but thinner than, the flux layer found on the pure Zircaloy ingots. The tapering at the top of the Zircaloy Inconel alloy ingot indicated that the upper part of the ingot melted with a thick slag layer. The metal at the top of the ingot, the last to solidify after feeding had stopped, was free of unmelted Zircaloy.

Heat Number 662 was melted without the simulated end caps. Melting without the simulated end caps demonstrated that the end cap material had no effect on melting rate when present in the feed material. Inconel springs were encased in cladding tubing sections in this heat to simulate the way they might be received from the dissolver. Heat Number 639B was made with loose Inconel springs in the feed material. No appreciable effect on melting rate was observed.

Prior to the above melting experiments, feed tests were conducted to determine if separation of feed material occurred in the feed hopper during

TABLE 2. Experiments on Inductoslag Melting(a)

Heat Number	Composition	Melt Rate kg/hr	Melt Charge Starting Pad Configuration	Remarks
637	Zircaloy-4, 3 wt% Inconel		Dovetail with "Z" strips mixture of end caps and tubing	Aborted - insufficient inductive coupling to start melt; "Z" strips fused to melt stock for a good grip.
638	Zircaloy-4, 3 wt% Inconel	21	Dovetail with "Z" strips + solid block of Zircaloy-4, 2.8 cm thick	Withdrawal pad separated from ingot and then gripped, allowing continued withdrawal.
639A	Zircaloy-4, 4 wt% Inconel	27	Dovetail with "Z" strips + solid block of Zircaloy-4	Melt progressed without incident; dovetail grip OK for withdrawal.
639B	Zircaloy-4, 4 wt% Inconel	19	Melt started on top of Heat 639A	Inconel springs loose in the charge rather than encased. Withdrawal difficulty encountered because of lack of bonding between Heats 639A and 639B. This accounts for the lower melt rate. Withdrawal movement due to gravity, some difficulty in removing ingot from the dovetail.
640	Zircaloy-4, 0.5 wt% Inconel	27	Dovetail with "Z" Strips + 40 of tubing with two 1-in. thick x 3 in. dia compacts of tubing on top	Gripping and withdrawal with dovetail was excellent. Fused but unmelted pieces of cladding were on the ingot bottom. Some difficulty in removing ingot from the dovetail.
641	Zircaloy-4, 0.5 wt% Inconel		16 pieces of cladding placed in and around the dovetails with two solid pieces (1903 g)	Aborted after losing contact with the withdrawal pad. Flux ran down and around the dovetail and did not have the strength for withdrawal.
642	Zircaloy-4, 0.5 wt% Inconel	23	Preshaped Zircaloy-4 starting block slipped into dovetail	Simulated end caps removed from melt stock to check their effect on melt rate--no real effect.
659	Zircaloy-4, 4 wt% Inconel	16	3/8-in. steel stud; Heat 639B starting block (4.457 kg) + 875 g Zircaloy-4 + 75 g Inconel springs	Aborted after 15 min because ingot pulled apart below junction of starting stub and metal added during melting.
660	Pure Zircaloy-4	23	3/8-in. Zircaloy-4 stud + Zircaloy-4 block	For pure Zircaloy-4 stock for refabrication development and BNW personnel training. Melted by PNL personnel on USBM equipment.
661	Pure Zircaloy-4	27	3/8-in. Zircaloy-4 stud + Zircaloy-4 block	For pure Zircaloy-4 stock for refabrication development and BNW personnel training. Melted by PNL personnel on USBM equipment.
662	Zircaloy-4, 4 wt% Inconel	25	3/8 in. steel stud + Zircaloy-4 4 wt% Inconel starting block (3.3 kg) + 1.037 kg of Zircaloy-4, 4 wt% Inconel alloy	Melt progressed without incident. Simulated end caps removed from melt stock.

(a) CaF₂ Flux used on all heats.

FIGURE 5. Etched Cross Section of a Zircaloy-4 (4 wt% Inconel) Inductoslag Ingot Melted at 25 kg/hr

melting. These tests revealed that the encased springs did not separate. However, the simulated end cap material did separate considerably and sank toward the bottom of the feed hopper.

MELTING FURNACE STARTUP AND OPERATION

The major problems encountered in startup and operation of the PNL Inductoslag melt furnace were associated with initiating and maintaining the flux layer and with finding the optimum height of the molten pool.

Proper operation of an Inductoslag melting furnace requires that on initial heating, the flux should melt first. The flux should then flow to the water-cooled crucible wall, and should then freeze in the annulus between the starting stub and crucible wall. When the flux freezes in the annulus between the starting stub and crucible wall it forms a seal, which prevents additional flux and metal from flowing down the side of the starting stub to the bottom of the furnace (runout). To achieve this seal, the melting point of the flux

must be lower than the melting point of the metal and the annulus must not be too wide or too short. Otherwise, the flux will not freeze at the proper location to form the seal. After the seal and molten pool have been established and feeding and ingot extraction are occurring alternately, the position of the molten pool must then be maintained high enough to prevent runout. If the position of the molten pool is allowed to go too high, liquid metal will splash toward the top of the crucible, where it will freeze and form a bridge with the incoming feed material. This bridge will inhibit further feeding.

Table 3 provides a summary of the heats made on the PNL Inductoslag melting furnace. All Heats, except Numbers 1 and 2, were melted using Zircaloy-4 cladding tubing 2.5 and 5.0 cm in length.

Major problems associated with startup and development of successful Inductoslag melting practices were induction coil design, induction coil position, clogged crucible water passages, melt stock feed control, crucible taper, and proper flux composition for stainless steel melting.

Experiments conducted in an attempt to match the induction coil design to the power source resulted in a 16-turn coil 16 cm (6.3 in.) long made from flattened 1.3 cm (0.5 in.) dia tubing.

In Heat Number 1, an attempt was made to establish a full molten pool on a starting stub of stainless steel (CG 27). Calcium fluoride was used as a flux. In the event of a water leak or component failure on initial startup, the consequences would be less severe when melting stainless steel than when melting Zircaloy-4. The stainless steel melts at a lower temperature and is less-reactive. In Heat Number 1, the liquid metal ran out the bottom, leaving a metal bridge across the top of the crucible. The runout was caused by the proximity of the melting point of the flux (1360°C) and the melting point of the stainless steel (1370°C). The flux did not melt and freeze to form the seal between the stub and the crucible wall before the stainless steel melted. Another small problem was a water leak in a crucible segment which was detected after the furnace was opened. During disassembly of the crucible metal chips, Teflon tape, and other debris were found to have clogged 10 of the 24 coaxially water-cooled crucible segments. Insufficient water flow in a few segments had

TABLE 3. PNL Inductoslag Melting of Zircaloy-4(a)

Heat Number	Melted Weight, kg	Melt Rate, kg/hr	Approximate Power, kW	Energy Use, kWh/kg	Hardness BHN	Remarks
1	3.0		100			Stainless steel charged for equipment checkout; metal runout due to flux not melting; crucible water leak.
2			85			Mild steel charge for equipment checkout; magnetic field from the coil pulled excess metal from the feed chute, causing a metal bridge.
3	2.3	10.7	83	7.8		USBM expert present; no major problems; heat terminated for examination.
4	10.9	28.4	90	3.2	189	USMB expert present; no major problems; smooth ingot side wall typical of USBM Inductoslag ingots.
5			82			Frozen metal ring formed above the melt zone; efforts to melt this ring failed; crucible recoated; segment gaps reduced.
6	14.1	20.7	100	4.9	185-194	Power limited because of hot induction coil cooling water; rough ingot side wall.
7						Melt stopped because of bridge formation; restarted and ingot pulled away from the bridge.
8	14.1	28.4	100	3.5	187-189	Crucible segment gaps reduced to 0.13 - 0.25 mm; recoated with ZrO ₂ ; starting stub position at 7 cm below crucible top; power increased to 94 kW when flux began to melt; rough ingot sidewall.
9	15.9	45.3	110	2.4		Lost power due to momentary low water pressure in heat exchanger cooling water; regained molten pool within 30 sec; rough ingot sidewall.
10	15.5	51.8	110	2.1	185-186	Good melt; rough ingot sidewall.
11	17.8	56.6	110-120	1.9-2.1	183-186	120 kW increases the instability of the cone-shaped pool; vapor-degreased hulls used; rough ingot sidewall.
12	19.3	38.5	110	2.9		Melting OK; rough ingot sidewall.
13	20.5	47.2	108-110	2.3	187	Melting OK; rough ingot sidewall.
14	16.21	18.7	76-110	4-6		Varied kW and material feed rate for sidewall finish experiment; material feeder problems.

(a) All feed stock is chopped Zircaloy-4 cladding tubing except Nos. 1 and 2.

TABLE 3. (contd)

Heat Number	Melted Weight, kg	Melt Rate, kg/hr	Approximate Power, kW	Energy Use, kWh/kg	Hardness BHN	Remarks
15	10.21	15.7	88	5.6		Feed chute altered for better control of feed rate; reduced power level to check effect on sidewall finish.
16	9.18		92			Low power with molten zone as low as possible in the crucible to check the effect on sidewall finish; worse surface finish to date.
17	10.0	13.0	90	6.9	181-189	Demonstration heat; 2.5 cm and 6 cm long cladding melted as low as possible in the crucible with low power level; rough ingot surface.
18			96			Reshaped crucible and raised coil 1.5 cm; objective to melt as high as possible in the crucible to form a flux pool; good flux pool formed; no metal feed.
19	10.0	40.0	100	2.5		Objective of melting ingot without losing flux pool was successfully met; 2.5 cm and 5 cm long cladding with end caps; smooth sidewall. Thick flux layer on ingot.
20	20.0	43.0	108	2.5	181-187	20 kg of cladding and end caps melted at varying heights within the crucible; flux pool lost at lowest position but regained after rebuilding pool height; smooth sidewall. Thick flux layer on ingot.
21	20.0	41.0	114	2.8		Melted in helium atmosphere; flux ring not visible in T.V. monitor; need different optical filtering; all other melting and ingot appearances the same as argon.
38						Operator training; melting was too high in crucible splashed metal causing shorting of induction coil, resulting in water leak.
39	30.0					Operator training; three separate starts were successfully accomplished.
40	2.5	5.3	100	18.9	207-217	HF descaled hulls slow melt rate due to gas evolution.
41	10.5					Operating; four separate starts were successfully accomplished.

caused overheating of some segments resulting in slight localized surface melting and water had penetrated one of these segments. The damaged segments were replaced and water filters installed on the inlet lines of the crucible and power source.

In Heat Number 2, an attempt was made to increase the spread between the calcium fluoride flux and metal melting points by using mild-steel feed stock with a melting point of 1500°C . This would allow time for the flux to melt and form the seal at the crucible wall before the mild steel melted. Inductive forces from the coil deflected the ferro-magnetic feed stock so that proper feeding was not possible. Efforts to melt mild steel were discontinued, as ferro-magnetic materials are not constituents of the fuel bundle residues. Melting was then concentrated on Zircaloy-4.

Heat Number 3 was conducted to establish the technique of pool initiation with a proper flux seal. The heat was initiated on a Zircaloy-4 starting stub. No feeding was attempted. Observations during melting indicated that a good pool and flux seal had probably been established. The heat was terminated to examine the flux and molten metal freezing configuration. Relationships were then drawn between the freezing configuration and the conditions observed on the television monitor during melting. This examination revealed that a flux seal had been established.

Heat Number 4 was a continuation of Heat Number 3 with material feeding and ingot extraction. This successful heat resulted in a typical Inductoslag Zircaloy ingot.

Subsequent heats resulted in a series of interrelated problems associated with operation, feed control, and crucible taper. The inductive forces on the molten pool produce a column of molten metal several inches high. High power levels cause the column to swing and if the column touches the crucible wall near the top, a frozen metal ring is formed that cannot be remelted. This occurred in Heat Numbers 5 and 7, making it necessary to re-apply the ceramic coating on the outside of the crucible. After each of these heats, a workable

melting procedure was established for Heat Numbers 8 through 13. However, the ingot side walls produced in each of these heats were rough and were not typical of Inductoslag melted Zircaloy.

Heat Numbers 14 through 17 were attempts to improve the side wall quality of the ingots by varying melting power, feed rate, and pool position. On Heat Number 15, the feed chute was altered by the addition of an adjustable baffle which gave the operator an improved range of feeds with more precise control. The experience gained from heats 14 through 17 resulted in establishing a melting procedure that would produce ingots at rates of approximately 30 to 55 kg/hr using power levels of 85 to 110 kW. Energy usage varied from 2 to 5 kWh/kg. Ingot soundness and hardness were excellent. The average Brinell hardness values of 187 were within the American Society for Testing and Materials (ASTM) specification of 200 maximum for nuclear grade Zircaloy-4. This indicated that the material was also within acceptable chemical and corrosion resistance standards. However, the ingot side wall roughness was still apparent. This roughness made flux removal difficult and was not desirable for leaching resistance during long-term storage. The rough ingot surface was caused by the metal freezing against the crucible wall without the benefit of a CaF_2 flux layer between the freezing metal and the crucible wall.

A light taper in the crucible (large diameter down) is desirable to ensure a free and unobstructed ingot withdrawal. A re-examination of the techniques used to determine crucible taper during crucible recoating after Heat Number 7 revealed that the taper was much larger than was intended. This could result in the flux freezing too low in the crucible.

The crucible segments were then reshaped to produce a diametrical taper of 1.2 mm over a 20.4 cm span (0.046 in. over 8 in.), and the induction coil was repositioned upward with one coil above the top of the crucible.

Heat Numbers 18 through 21 yielded ingots with excellent side wall quality. The flux pool was visible throughout each heat. During Heat Number 20, the pool was purposely lowered until the flux pool was lost. By stopping ingot extraction and continuing to feed, the pool level was raised and the flux pool re-established.

The melting procedure established for making 20 kg Zircaloy-4 ingots is as follows:

1. Position the starting stub 7 cm from the top of the crucible.
2. Add 200 g of CaF_2 to the top of the stub.
3. Add 800 g of CaF_2 to the flux feeder.
4. Mix 800 g of CaF_2 with the metal feed.
5. Evacuate the furnace to less than 0.005 torr and close main vacuum valve.
6. Backfill the furnace with Argon to 250 torr.
7. Turn power on at 40 to 60 kW, hold for five min or until flux starts to melt. Increase power to 100 kW to establish molten pool. If flux ring has not formed, add flux from flux feeder until a flux ring does form. Add metal feed and operate with molten pool as high in the crucible as operator is comfortable with. Normal operating power will be between 100 to 120 kW. After all metal has been added, turn power down.

Heat Number 21 was made using the established melting procedure described above, except that helium was used as the protective atmosphere. No visible difference in melting characteristics or ingot quality was detected. When melting in a helium atmosphere the television monitor does not show the slag ring. It is believed that a change in the monitor's optical filtering will show the contrast between the slag ring and molten metal.

After the successful melting practice described above had been developed, Heat Numbers 38, 39, and 41 were melted to provide operator training.

INGOT EVALUATION

One objective of the program was to evaluate ingots made from melted hulls that had been descaled by the hydrogen fluoride decontamination process described in Vol. 1 of this report. The starting material for Heat Number 40 was composed of Zircaloy-4 cladding tubing that had been exposed to 1000 psi steam at 450°C for 400 hr to an average weight of 62 milligrams per square decimeter.^(a) Afterward, the tubing was chopped into 2.5 and 5.0 cm lengths

(a) Eighty milligrams per square decimeter is an estimated weight gain for normal reactor exposure.

and processed through the hydrogen fluoride decontamination system to remove the oxide coating. During the melting of the 2.5 kg of descaled hulls, an abnormal amount of spitting and gas evolution was observed. After each addition of hulls to the melt, excessive metal spitting and gas evolution occurred which slowed the melting rate of 5.3 kg/hr compared to 40 to 50 kg/hr on as-received hulls. Observation of the melt became difficult because of condensation on the viewing ports. The interior of the furnace was coated with an unusual amount of condensed products, and the results of spitting in the form of frozen metal BB-like shot were on the top of the crucible and around the base of the crucible. Flux conditions during melting appeared normal, the ingot was easily removed and the side wall was smooth and bright.

The effect of improper and proper flux conditions during metal freezing on Zircaloy ingot surface quality are shown in Figures 6 and 7, respectively. Typical sections of Inductoslag Zircaloy-4 ingots melted at a fast rate of 62.2 kg/hr and at a slow rate of 15.7 kg/hr are shown in Figures 8 and 9, respectively. The finer grain size and shallower pool of the fast melting rate are probably a result of a lower molten metal temperature.

Figure 10 is a section of ingot made from melted oxidized hulls that had been descaled using the hydrogen fluoride decontamination process. While the melting characteristics of this ingot were distinctly different from previously melted hulls, the macro structure is nearly identical to ingots made from melted as-received hulls (Figure 9).

The chemical analyses and ingot hardnesses of Inductoslag ingots produced during this program are shown in Table 4. These are compared to the ASTM Specification B350 for reactor grade Zircaloy-4 and the vendor analyses for the as-received hulls used as starting material.

Heat Numbers 6 through 17 were melted using as-received hulls degreased prior to melting. The surface of this material was bright with no visible signs of oxidation. The analyses and hardness of ingots melted from this material are still within ASTM B350 Zircaloy-4 ingot specification. The only significant increase in impurity levels are those of fluorine, hydrogen and nitrogen, the source of which is probably the calcium fluoride flux used in melting.

FIGURE 6. Typical Rough Surface of Zircaloy-4 Inductoslag Ingot Caused By a Lack of a CaF₂ Flux Layer During Metal Freezing

FIGURE 7. Typical Smooth Surface of Zircaloy-4 Inductoslag Ingot
Resulting From Proper CaF_2 Flux Pool During Metal Freezing

FIGURE 8. Heat Number 13. Zircaloy-4 Melting Rate of 62.2 kg/hr Achieved. Note the finer grain and shallower pool as compared to Figure 9.

FIGURE 9. Heat Number 15. Melting Rate of 15.7 kg/hr achieved. Note the coarser grain and deeper pool as compared to Figure 8.

FIGURE 10. Heat Number 40. Zircaloy-4 melted from oxidized hulls that had been descaled by the hydrogen fluoride process.

Heat Number 40 was melted from hulls that had been oxidized in 1000 psi steam at 400°C for 400 hr to an average weight gain of 62 milligrams per square decimeter, then descaled by the hydrogen fluoride decontamination process. The surface of this material appeared nearly descaled with a mat-like texture having patches of black zirconium oxide covering an estimated 5% of the surface area. Substantial increases in hydrogen, oxygen, nitrogen, and in hardness were noted for Heat Number 40 compared to Heat Numbers 1 through 17. Some increase in fluorine is also evident. The hydrogen, nitrogen, and hardness levels are above the maximum allowable limits in ASTM Specification B350.

The problem of gas evolution during the melting of Heat Number 40 can be attributed to hydrogen evolution and possibly to some fluorine. The high oxygen content is probably caused by incomplete oxide removal during the hydrogen

TABLE 4. Chemical Analyses of Inductoslag Melted Zircaloy-4

Alloy Element	ASTM B350	Weight Percent								As-Received Cladding Hulls
		Inductoslag Melted Heats								
		6	8	9	10	11	15	17	40	
Tin	1.20 to 1.70	1.55	1.47	1.47	1.47	1.50	1.42	1.47	1.60	1.55
Iron	0.18 to 0.24	0.217	0.205	0.207	0.203	0.204	0.188	0.202	0.232	0.20
Chromium	0.07 to 0.24	0.117	0.106	0.110	0.105	0.106	0.100	0.109	0.108	0.11
Fe + Cr + Ni	0.28 to 0.37	0.335	0.312	0.318	0.309	0.311	0.289	0.312	0.341	0.31
Impurity	Maximum	Parts Per Million								
Aluminum	75	<75	<75	<75	<75	<75	<75	<75	<25	<35
Boron	0.5	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.25	<0.25
Cadmium	0.5	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	<0.25
Calcium	<100	<100	<100	<100	<100	<100	<100	<100	<10	<10
Carbon	270	160	170	150	200	120	170	160	230	170
Cobalt	20.0	<5	<5	<5	<5	<5	<5	<5	<5	<10
Copper	50.0	<20	<20	<20	<20	<20	<20	<20	<20	<10
Fluorine		14	56	36	36	25	38	37	55	<5
Hafnium	100.0	<100	<100	<100	<100	<100	<100	<100	90	<50
Hydrogen	25.0	18	23	20	23	17	20	16	55	11
Oxygen		1160	1300	1220	1210	1220	1260	1150	1900	1210
Lead		<5	<5	<5	<5	<5	<5	<5	<5	<25
Magnesium		<10	<10	<10	<10	<10	<10	<10	<10	<10
Manganese	50.0	<10	<10	<10	<10	<10	<10	<10	<10	<25
Nickel	70.0	<10	<10	<10	<10	<10	<10	<10	<10	<35
Nitrogen	65.0	50	50	50	40	40	50	50	90	29
Silicon	120.0	70	80	70	70	70	70	80	40	60
Thorium		<4	<4	<4	<4	<4	<4	<4	<3.5	0
Titanium	50.0		<20	<20	<20	<20	<20		<20	<25
Tungsten	100.0	<40	<40	<40	<40	<40	<40	<40	<40	<25
Uranium	3.5	<1.0	1	2	1	2	2	1	1.4	<1.0
Vanadium		<20.0	<20	<20	<20	<20	<20	<20	0	0
Brinell Hardness	200	185	189		185	186		184	208	187

fluoride decontamination process. The high hydrogen content may be the result of an excessive amount of hydrogen present during the high temperature phase of the descale process. The high fluorine could be the result of incomplete low temperature stripping of the treated surfaces, and the high nitrogen the result of incomplete removal of the stripping solution by the final rinse.

The evaluation of the quality of the Zircaloy-4 ingot melted from descaled hulls is based on a single, small 2.5 kg ingot made from the earliest product of the descale process. Improvements and refinements in process control should yield improved product quality. While the resulting ingot does not meet nuclear grade Zircaloy-4 standards, its chemical composition and hardness indicate that it possesses adequate engineering properties for reuse in a non-critical application within the nuclear environment.

WITHDRAWAL GRIPPING EXPERIMENTS

Inductoslag melting is started on a "starting stub" threaded to the water-cooled base. The starting stub fuses to the ingot during the melt and must be machined off for recycle or left on the ingot, where the stub becomes an addition to the waste stream. Experiments were performed that were aimed at 1) simplifying the remote removal of the ingot from the withdrawal grip, and 2) reducing or eliminating the amount of added material in the starting stub.

Two methods were investigated to achieve the aims: the dovetail water-cooled base and the threaded stud or bolt in the water-cooled base (Figures 11 and 12). Either method will work satisfactorily provided sufficient mass is present for an inductive coupling to initiate the molten pool. Chopped simulated cladding hulls alone do not provide the mass required for pool initiation. Compacted or flattened hulls or solid end cap material or other scrap is required.

Heat Numbers 637, 638, 639A, and 640 (see Table 2) were made using "Z" strips of Zircaloy sheet on the dovetail as shown in Figure 11. The results of those heats showed that 1) bonding and successful withdrawal can be achieved if there is enough solid or compacted material placed on the top of the "Z"

FIGURE 11. Z-Shaped Inserts for Gripping the Inductoslag Ingot for Withdrawal

strips to obtain an inductive coupling and to start a molten pool, and 2) without the "Z" strips flux will flow around the dovetail and form a connection too weak for withdrawal (Heat Number 641).

The alternative mechanism for ingot withdrawal consists of a stud screwed into the water-cooled base as shown in Figure 12. This method requires a starting technique that consists of 1) melting small increments of solids, 2) allowing the mass to cool, 3) lowering the starting stub after cooling, and 4) repeating the procedure until about 10 to 13 cm of ingot length has been melted. Melting can then proceed normally.

It appears that either gripping method can be made to work, providing that a charge with sufficiently high density is present to start a molten pool. The

FIGURE 12. Threaded Stud Method of Gripping the Inductoslag Ingot for Withdrawal

choice of the ingot gripping method will depend on the ease of remote operation and the overall melting cycle time. The dovetail design allows the finished ingot to be knocked off the dovetail on the water-cooled base, while the stud design will require the ingot to be unscrewed from the water-cooled base. The best method of operation will need to be determined during cold remote demonstration.

Heat Numbers 639B and 659 were the Zircaloy-4 - 4 wt% Inconel (see Table 2) alloy started on solid blocks of the same alloy. These ingots pulled apart at the junction between the starting blocks and the added metal. The reasons for this are not clear at this time. It is possible that a low melting eutectic mush was formed, causing a weak zone in the ingot. This phenomenon should be investigated further.

Heat Number 660 and 661 were pure Zircaloy-4 cladding tube melts made for refabrication development stock. The stud method was used for gripping and a solid Zircaloy-4 block was used for pool initiation. These were melted by PNL personnel during training on the U.S. Bureau of Mines Inductoslag melting equipment.

INDUCTOSLAG SCALEUP TEST

A test to demonstrate that the Inductoslag melting process can be scaled up was performed using a different Inductoslag furnace in contract work supervised by the U.S. Bureau of Mines. A 36 kg ingot of mild steel was melted in an Inductoslag crucible 20.3 cm in dia by 21.6 cm deep using CaF_2 slag. Total melt time was 23 min using 240 kW of power at a frequency of 2 kHz. Based on the result of this test and Zircaloy Inductoslag melting experience, it is estimated that 260 kW would melt a 20.3 cm dia Zircaloy ingot.

GRAPHITE CRUCIBLE MELTING

One experiment was conducted using a different melting process. Previous work⁽²⁾ had shown that vacuum induction melting of surface cleaned 85 wt% Zircaloy, 10 wt% 304 stainless steel, and 5 wt% Inconel in graphite crucibles could be performed at temperatures of 1300°C . This low melting process was possible because of the formation of low melting eutectics between zirconium and iron, nickel, and chromium in the stainless steel and Inconel. To determine the effect of oxidized surfaces on the temperature required for melting, two 5 kg heats of 85 wt% Zircaloy-4, 10 wt% 304 stainless steel and 5 wt% Inconel 718 were made using the graphite crucible process. Oxidized sheet material was used as melt stock. Temperatures in the range of 1500°C to 1600°C were required to melt the material. This is above the melting point of the stainless steel and Inconel.

The first ingot was allowed to cool in the crucible, resulting in extensive crucible reaction. The second ingot was poured into a graphite mold. While crucible reaction is apparent, both the crucible and mold are reusable. This ingot cracked. Ingot of this composition melted with clean material and under similar vacuum condition did not crack.⁽²⁾

CONCLUSIONS

The Inductoslag melting process developed at Pacific Northwest Laboratory (PNL) will remelt clean Zircaloy-4 cladding tubing into reactor-grade ingots at acceptable melting rates. Mixtures of Zircaloy-4 and Inconel can also be Inductoslag melted at acceptable rates. The Inductoslag melting process is capable of remote operation, but additional development work is needed to improve the remote operation of the ingot and feed material handling, cleaning, and furnace preparation phases of the process.

Descaled^(a) Zircaloy-4 product quality attained in the hydrogen fluoride decontamination process must be improved before melted reactor-grade ingots can be made at an acceptable melting rate using the Inductoslag method.

The chemical analyses and hardness of the metal produced from the melted, oxidized and descaled hulls do not meet reactor-grade ingot standards. However, this does not preclude the possibility of using this material in a non-critical nuclear environment.

(a) "Descaling" is used here because the work reported in a companion document was pilot-scale work conducted with unirradiated hulls, making "decontamination" a misnomer.

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APPENDIX A

MELTING OF SIMULATED UNIRRADIATED LIQUID METAL
FAST BREEDER REACTOR (LMFBR) STAINLESS STEEL CLADDING

APPENDIX A

MELTING OF SIMULATED UNIRRADIATED LIQUID METAL FAST BREEDER REACTOR (LMFBR) STAINLESS STEEL CLADDING^(a)

The work reported here was sponsored by the Department of Energy and performed for the Union Carbide Corporation, Oak Ridge, Tennessee, Nuclear Division, under the direction of Mr. John Van Cleve. The summary of this work is presented because of its significance to the melting of stainless steel, a major constituent of the LWR fuel bundle residues.

The Pacific Northwest Laboratory (PNL) has performed experiments to demonstrate that Liquid Metal Fast Breeder Reactor (LMFBR) stainless steel cladding can be melted in an Inductoslag melting furnace that produces ingots acceptable as densified waste. Simulated, unirradiated LMFBR stainless steel cladding was melted to determine which procedures would produce the best quality ingots. Four types of flux were evaluated, based on their effectiveness in starting and maintaining the melt without runouts, on ingot sidewall quality; on ease of slag removal after melting; and on ingot quality as shown by macro-etch disc samples of the ingots.

Fifteen 10 cm dia heats were made in two Inductoslag furnaces; eleven heats at PNL and four heats at the Bureau of Mines in Albany, Oregon. The heats performed at the Bureau of Mines compared the melting characteristics of a 10 kHz power supply with the melting characteristics of PNL's 3 kHz power supply.

The experiments established that:

- An ingot of acceptable quality can be produced from LMFBR-type stainless steel cladding in a 10 kHz Inductoslag melting furnace with a melting

(a) D. R. Montgomery, "Melting of Simulated Unirradiated LMFBR Stainless Steel Cladding." Letter report prepared by Pacific Northwest Laboratory for Union Carbide Corporation, 37830-Nuclear Division, Oak Ridge, TN.

rate of 18 kg/hr and a power consumption of 3.9 kWh/kg. A flux of 33 wt% MgF_2 + 67 wt% CaF_2 can be successfully used to produce these stainless steel ingots.

- A furnace with a 10 kHz power supply is significantly more efficient than a furnace with a 3 kHz power supply, although satisfactory ingots can be produced with either power supply.
- The melting characteristics of slightly oxidized and unoxidized stainless steel cladding feedstock are not discernibly different.

APPENDIX B

VOLUME REDUCTION BY PRESS COMPACTION

APPENDIX B

VOLUME REDUCTION BY PRESS COMPACTION

Press compaction received a limited evaluation as an alternate method of densifying chop-leach cladding residues. Although press compactions of zirconium and zirconium alloys are routinely made in industry on relatively clean, oxide-free material, very little information is available on the compaction of zirconium materials that have an oxide coating and brittleness typical of goal-exposed PWR cladding hulls emerging from the chop-leach process. Using press compaction equipment at the U.S. Bureau of Mines in Albany, Oregon, several compacts were made from unirradiated Zircaloy-4 PWR cladding (2.5 cm lengths) with oxygen levels ranging from a clean, non-oxidized condition to a very brittle, highly oxidized condition (Table B.1).

The tubing has steam autoclaved in 91 cm lengths and sheared into 2.5 cm lengths for compacting. The compaction tests were performed in a cylinder 10 cm in dia, using 750 g of hulls (starting height of 10 cm) for each run. A slow rate of compaction was used to allow the recording of the press ram pressure and displacement.

A plot of percent volume reduction versus unit compaction pressure is shown in Figure B.1 from test data. The plot shows that more than 90% of the volume reduction is accomplished with a unit pressure of 15,000 psi, although the compacts were pressed to 45,000 psi before being ejected from the die. Some cladding lengths flaked from the compact surfaces while being ejected from the die and during a subsequent 60 cm drop test, but the compacts remained reasonably intact. This was true for all groups of material. A small residue of fines was generated in compactions of the low and medium oxidized material with a considerable quantity of fines being produced with the highly oxidized material.

Table B.1 Oxygen Levels for Cladding Hulls
Used in Press Compaction Tests

Designation	O ₂ Level
Clean (as-manufactured)	1200 ppm
Low O ₂	34 to 80 mg/dm ² gain
Medium O ₂	50 to 112 mg/dm ² gain
High O ₂	3 to 870 mg/dm ² gain

FIGURE B.1 Volume Reduction Achieved by Press Compaction
of Clean and Oxidized Unirradiated PWR Cladding
Hulls

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