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The Cladding Hull Decontamination and Densification Process

Part 1

The Prototype Cladding Hull Decontamination System

**T. M. Lambright
D. R. Montgomery**

April 1980

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

**Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute**

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Richland, Washington 99352

SUMMARY

A prototype system for decontaminating Zircaloy-4 cladding hulls has been assembled and tested at Pacific Northwest Laboratory. The decontamination process consists of treatment with a gaseous mixture of hydrogen fluoride (HF) and argon (Ar) followed by a dilute aqueous etch of ammonium oxalate, ammonium citrate, ammonium fluoride, and hydrogen peroxide. The continuous cleaning process described in this report successfully descaled small portions of most charges, but was unable to handle the original design capacity of 4 kg/hr because of problems in the following areas:

- Control of HF reactor temperatures
- Regulation of HF and argon mixtures and flows
- Isolation of the HF reactor atmosphere from the aqueous washer/rinser atmosphere
- Regulation of undesirable side reactions
- Control over hull transport through the system

Due to the limited time available to solve these problems, the system did not attain fully operational status.

The work was performed with unirradiated hulls that simulated irradiated hulls. The system was not built to be remotely operable.

The process chemistry and system equipment are described in this report with particular emphasis on critical operating areas. Recommendations for improved system operation are included.

CONTENTS

SUMMARY	iii
INTRODUCTION	1
THE HYDROGEN FLUORIDE DECONTAMINATION PROCESS.	3
FEED MATERIAL	3
FEED MATERIAL FLOW	3
Basic Process Chemistry	8
OPTIMUM PROCESSING PARAMETERS	8
HF Reactor Residence Time.	8
Aqueous Washer/Rinser Residence Time	10
DEVELOPMENT OF THE CLADDING HULL DECONTAMINATION SYSTEM.	11
SYSTEM DESCRIPTION	11
Critical Operating Areas	13
SYSTEM MODIFICATIONS.	25
ANALYSIS OF OPERATING DATA	29
TEST #1	29
TEST #2	29
TEST #3	31
TEST #4	32
TEST #5	32
TEST #6	32
TEST #7	32
TEST #8	33
CONCLUSIONS	35
REFERENCES.	39
ACKNOWLEDGMENTS.	41

TABLES

1	Operation Sequence for Airlock Valve System	7
2	Operating Data From Prototype Decontamination System . .	30

FIGURES

1	Flow Diagram For Cladding Hull Decontamination Process . .	4
2	Pictorial Flow Diagram of Cladding Hull Decontamination Process	5
3	Cladding Hull Decontamination Assembly	6
4	Typical Hulls After Treatment in Prototype Decontamination System	9
5	Prototype Decontamination System	12
6	HF Gas Supply Subsystem	14
7	Key to Control Instrumentation of Prototype Decontamination System	15
8	Control Instrumentation of Prototype Decontamination System	17
9	Blueprint of Process Flow Diagram	19
10	Safety Control Logic For Cladding Hull Decontamination System	21
11	Feed Chute to Reactor Clogged With Hulls and ZrF_4 Crystals	24
12	Modification to Reactor Gas Removal System	28

INTRODUCTION

Current United States policy prohibits the reprocessing of nuclear fuels. The work described in this report defines a possible method for handling metallic wastes from such fuels in the event that the technology is needed.

Fuel element cladding residues from the chop-leach, head end process constitute a low-density, high-surface-area metallic waste with sufficient transuranic nuclides to classify them as TRU contaminated wastes (as defined by the Department of Energy limit of >10 nCi/g). The residues represent a waste volume of approximately $0.3 \text{ m}^3/\text{MT}$ of uranium, weighing approximately 324 kg/MT of uranium. Separating the long-lived actinide element contamination from large amounts of radioactive cladding material reduces the volume of TRU waste that requires special handling. Once this is done, a considerable potential exists for reduced disposal costs.

Analyses⁽¹⁾ of spent fuel cladding showed that hull contamination arises from two sources: 1) surface contamination on the inside and outside surfaces of cladding hulls, and 2) irradiation of uranium (U) and thorium (Th) impurities in the zirconium metal.

Laboratory-scale work conducted between 1975 and 1977 proved the feasibility of removing the transuranic surface contamination from Zircaloy-4 cladding pieces in 600°C hydrogen fluoride (HF) argon (Ar) gas followed by etching with ammonium oxalate, ammonium citrate, ammonium fluoride, and hydrogen peroxide. However, surface treatment (descaling) alone could not reduce typical cladding contamination to less than 10 nCi/g because the irradiated cladding material contains 100 to 200 nCi/g of transuranic nuclides. These transuranic nuclides arise from U and Th impurities that are distributed uniformly throughout the zirconium metal.^(a) The removal of these contaminants during melting by zone refinement or by extraction into a slag (CaF_2) was not evaluated because it required moving the melting operation into a hot cell.

(a) ^{241}Pu , a transuranic nuclide contributes most of this induced radioactivity. However, it may not be included in a regulatory definition of transuranics because it is a beta emitter and has a relatively short (13-1/2 yr) half life. If so, surface decontamination may be sufficient to reduce the alpha activity of cladding to near or below 10 nCi/g.

A semi-continuous processing system was designed and built to demonstrate the technique of surface decontamination. The system was installed at PNL during the latter part of 1978, and eight nonradioactive descaling^(a) tests were performed in 1979. Data gathered from these tests are the subject of this report.

A companion report⁽²⁾ has been prepared on the densification of cladding hulls by melting in the Inductoslag Furnace. Please refer to the second volume of this report for further information on the densification process.

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

THE HYDROGEN FLUORIDE DECONTAMINATION PROCESS

The hydrogen fluoride (HF) decontamination process consists of exposing autoclaved Zircaloy-4 tubing pieces to a flowing mixture of 10 to 80% HF and 20 to 90% Ar at $600^{\circ}\text{C} \pm 50^{\circ}\text{C}$ for $1 \text{ hr} \pm 1/2 \text{ hr}$. This treatment is followed by etching the hulls in an aqueous solution of 0.4M ammonium oxalate-0.16M ammonium citrate-0.1M ammonium fluoride-0.3M hydrogen peroxide at $100^{\circ}\text{C} \pm 5^{\circ}\text{C}$ for $1 \text{ hr} \pm 1/2 \text{ hr}$. The tubing pieces are then rinsed in hot water and dried before melting in the Inductoslag furnace.⁽²⁾

FEED MATERIAL

For nonradioactive operations, 1.2-m (4-ft) lengths of scrap Zircaloy-4 cladding material were autoclaved to produce oxide layers ranging from 21 mg/dm^2 to 74 mg/dm^2 . The autoclaved cladding was then chopped into 2.5- and 5.0-cm (1- and 2-in.) lengths for use in the prototype decontamination system. The resultant product is referred to as "hulls" in this document.

For oxide removal purposes, earlier research work⁽¹⁾ determined that zirconium oxide layers on unirradiated, autoclaved Zircaloy adequately simulated oxide layers on irradiated fuel cladding. The unirradiated oxides proved to be slightly more difficult to remove, possibly because of the absence of radiation-induced defects.

FEED MATERIAL FLOW

Figure 1 is a schematic of the descaling process. The figure shows various reactant and product flows that are quantified over the acceptable ranges. A more simplified flow diagram is shown in Figure 2.

Prior to treatment in HF gas, the hulls (feed material) are manually placed into a stainless steel canister where they are heated and then purged with argon to eliminate moisture and oxygen in the feed material. The canister is then manually attached to the hull inlet port which initially faces downward. When the operator is ready to admit hulls to the reactor chamber, the canister is manually rotated into the vertical position shown in Figure 3.

FIGURE 1. Flow Diagram For Cladding Hull Decontamination Process

FIGURE 2. Pictorial Flow Diagram of Cladding Hull Decontamination Process

FIGURE 3. Cladding Hull Decontamination Assembly

Next, the hulls are fed into the reactor chamber as the chamber rotates in a counterclockwise direction. The reactor chamber is maintained at a slightly higher pressure than the washer/rinser to prevent water vapor from entering the HF reactor. The screw feeder, an integral part of the chamber, then moves the hulls through the reactor. As the hulls pass through the reactor (heated to 600°C), the HF-Ar gas mixture is admitted. The HF gas penetrates heat caused imperfections in the oxide layer to form a zirconium fluoride layer at the metal-oxide interface. Unreacted HF gas is scrubbed from the off-gas stream by a potassium hydroxide aspirator. The treated hulls then leave the chamber through an opening in the hollow rotating shaft and are then transferred to the aqueous washer/rinser through two ball valves that serve as an airlock. The ball valves, the associated vibrators, and the argon purging system are controlled by a programmable sequencing system. The operation sequence for the airlock valve system (Table 1) is initiated by the closure of a microswitch which is tripped once for every revolution of the reactor chamber.

The hulls (delivered by the reactor) are then transported through the aqueous washer/rinser where the zirconium fluoride layer is dissolved, allowing the oxide layer to slough off the hulls. The clean hulls fall into a second canister and are dried before melting takes place in the Inductoslag

TABLE 1. Operation Sequence for Airlock Valve System

<u>Time (min:sec)</u>	<u>Top Valve</u>	<u>Argon Purge</u>	<u>Spool Vibrator</u>	<u>Bottom Valve</u>	<u>Washer/Rinser Vibrator</u>
0:00	On				
5:00	Off				
5:10		On			
5:30		On		On	
6:00		On	On	On	
6:30		On	Off	Off	
7:50		On			On
8:00		On			Off
10:00		Off			

furnace. Cleaned hulls are shown in Figure 4. The patches of black oxide film on some of the hulls is evidence that optimum conditions were not attained for all hulls.

Basic Process Chemistry

As determined by Griggs,⁽¹⁾ the following reactions define the metal and oxide film modifications that occur during hull processing:

Reaction 1 is considered to be the primary reaction. Reactions 2 and 3 are small side reactions, and Reactions 4, 5, and 6 are undesirable corrosion reactions. Experiments with the prototype equipment showed that Reactions 5 and 6 occurred to a great extent in nearly every run. A change described in the System Modifications section of this report was made for Test #8 to help minimize the reaction with hydrogen. However, zirconium hydrides were still present as evidenced by the agitated molten pool observed during hull melting in the Inductoslag furnace.

OPTIMUM PROCESSING PARAMETERS

The following operating conditions were found to be best for this system.

HF Reactor Residence Time

A residence time for the hulls of approximately 30 min in the HF reactor seems to be sufficient for oxide film removal. The hulls were adequately cleaned during Test #8, where a 15% HF/85% Ar mix was used at $550^\circ\text{C} \pm 50^\circ\text{C}$. Earlier tests yielded similar results: the ratio of HF to Ar can be less than that indicated by the laboratory-scale experiments. However, reactor temperature was excessively high during hull processing, indicating that undesirable

FIGURE 4. Typical Hulls After Treatment in Prototype Decontamination System

reactions were taking place. Total gas flow should be increased to purge the reactor of any hydrogen that is produced, therefore preventing an exothermic reaction which is indirectly caused by hydrogen.

Although it is not a critical parameter, reactor pressure was maintained at 5 in. of H_2O above the washer rinser atmosphere.

Aqueous Washer/Rinser Residence Time

Residence time in the aqueous washer/rinser should be about 90 min total with nearly 60 min in the wash solution. The wash solution flow rate should be near 16 liters/hr. The rinse water flow rate should be near 12 liters/hr.

DEVELOPMENT OF THE CLADDING HULL DECONTAMINATION SYSTEM

It was decided early in the project to construct a nonradioactive mock-up of the processing system to facilitate hull handling without the complicating influence of radiological hazards. The original goal of constructing a completely remote nonradioactive facility was abandoned in mid-1978 because of funding and time constraints. The prototype system was finally built in late 1978, but necessitated substantial manual operation.

SYSTEM DESCRIPTION

The prototype cladding hull decontamination system consists of the following subsystems:

- HF reactor and aqueous washer/rinser
- chemical storage, mixing and transfer equipment
- gas supply subsystem
- control console and associated safety circuits.

The HF reactor and aqueous washer/rinser subsystem is detailed in Figure 3. This subsystem consists of a rotating Inconel treatment vessel where the gaseous HF reaction takes place and a rotating stainless steel etching-rinsing vessel where removal of the treated oxide film occurs. The two vessels are separated by an airlock valve assembly which is described later.

The chemical storage, mixing and transfer equipment is shown in Figure 5. It is an assembly of stainless steel tanks, mixers and pumps used to prepare solutions for the aqueous washer/rinser.

The HF reactor and washer/rinser and the chemical storage, mixing and transfer equipment were arranged on the 0.6-m-high platform shown in Figure 5 because the equipment was originally intended to be used in a hot cell facility. The platform height represents the difference in elevation between the floor of the hot-cell facility and the floor outside the facility.

The gas supply subsystem consists of separate equipment for HF supply and Ar supply. The HF gas supply equipment was located in an enclosed ventilated

FIGURE 5. Prototype Decontamination System

cabinet (Figure 6). The ventilation system assured that gas leakage would be properly handled. Since hydrogen fluoride is a liquid at room temperature, it was necessary to vaporize the gas by heating the cylinders. Since a large volume of argon was necessary for process operation, a liquid argon storage tank was used. Ambient temperature supplied sufficient heat to vaporize the argon. The gas was piped to various subsystems using appropriate fittings, valves and meters.

The control console and associated safety circuits are shown in Figures 7, 8 and 9. The control console records and controls process temperatures, material flows, system pressures and system tank levels. The console also contains alarms for selected parameters. The alarms terminate power to critical subsystems when process conditions exceed control limits. Figure 10 explains the safety control logic.

Critical Operating Areas

During system testing, the HF gas supply subsystem, the air lock valve assembly that isolates the HF reactor from the aqueous washer/rinser and the hull feeding and transport functions were found to be essential for controlled operation. Problems in one or more of these areas forced termination of each test before a complete charge of hulls could be processed. These operating areas are described here and reasons given for their critical nature. Solutions are presented in the System Modifications section of this report.

Problems with the HF gas supply subsystem encompassed three major areas:

1. Insufficient Gas Supply Capability. Heavy wall 1/4-in.-diameter Inconel tubing was used as the main HF gas supply line. With two exceptions, flow rates above 2.8 SLPM could not be maintained in a continuous basis. Higher HF gas flow rates are necessary to maintain sufficient total gas flow through the reactor to remove reaction products and heat. One cause for the low flow rates was the flow restrictions imposed by elbows, bends, valves and filters. Another factor contributing to low flow rates was that heating the HF cylinders and supply lines and maintaining them at the desired temperature proved to be more difficult than expected.

FIGURE 6. HF Gas Supply Subsystem

NO	DESCRIPTION	FUNCTION	COMPONENT	NO	DESCRIPTION	FUNCTION	COMPONENT
1	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	REACTOR ZONE 1	47	VARIAC	POWER LIMITER	HULL DRYER HOT PLATE
2	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	REACTOR ZONE 1	48	HASTINGS TR-100P TOTALIZER	RECORD TOTAL HT FLOW	HT FLOW INTO REACTOR
3	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	RINSE HEAD TANK	49	L & N ELECTROMAX 131 CONTROLLER CAT NO 6432-A-4019-200-0-20	CONTROL HF REACTOR TEMPERATURE	REACTOR ZONES 1 THROUGH 5
4	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	REACTOR ZONE 2	50	HASTINGS ALL-10RPGA FLOWMETER	DISPLAY HF GAS FLOW	FLOW TO HF REACTOR
5	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	REACTOR INLET SEAL	51	HASTINGS FC-1P FLOW CONTROLLER	CONTROL HF GAS FLOW	MOTORIZED FLOW CONTROL VALVE
6	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	WASH HEAD TANK	52	HASTINGS ALL-10RPGA FLOWMETER	DISPLAY ARGON GAS FLOW	FLOW TO HF REACTOR
7	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	REACTOR ZONE 3	53	FUSE BOX	PREVENT DAMAGE TO REACTOR FURNACE POWER PACKS	0 VOLTAGE POWER PACKS
8	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	REACTOR OUTLET SEAL	54	L & N ZERO VOLTAGE POWER PACK	INTERFACE CONTROLLER (NO 49) TO HEATER	REACTOR ZONE 3
9	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	WASH SOLUTION INLET TO WASHER/RINSE	55	L & N ZERO VOLTAGE POWER PACK	INTERFACE CONTROLLER (NO 49) TO HEATER	REACTOR ZONE 2
10	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	REACTOR ZONE 4	56	L & N ZERO VOLTAGE POWER PACK	INTERFACE CONTROLLER (NO 49) TO HEATER	REACTOR ZONE 1
11	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	HULL DRYER	57	L & N ZERO VOLTAGE POWER PACK	INTERFACE CONTROLLER (NO 49) TO HEATER	REACTOR ZONE 5
12	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	WATER JACKET	58	L & N ZERO VOLTAGE POWER PACK	INTERFACE CONTROLLER (NO 49) TO HEATER	REACTOR ZONE 4
13	DIGITEC 2770 HT SERIES, DC PANEL METER	PERCENT TOTAL FLOW	INDICATES WASH SOLUTION FLOW RATE	59	FOXBORO 196R-1A-AF MAGNETIC FLOW TRANSMITTER	MONITOR RINSE WATER FLOW	FLOW TO WASHER/RINSE
14	DIGITEC 2770 HT SERIES, DC PANEL METER	PERCENT TOTAL FLOW	INDICATES WASH SOLUTION FLOW RATE	60	FOXBORO 196R-1A-AF MAGNETIC FLOW TRANSMITTER	MONITOR WASH WATER FLOW	FLOW TO WASHER/RINSE
15	DIGITEC 2770 HT SERIES, DC PANEL METER	LIQUID LEVEL (PERCENT TOTAL CAPACITY)	DRAIN TANK	61	INCANDESCENT INDICATOR LIGHT	INDICATES WHEN TO PLACE SWITCHES 35-40 IN AUTOMATIC POSITION	SIGNAL RECEIVED FROM SEQUENCER OUTPUT Y1
16	DIGITEC 2770 HT SERIES, DC PANEL METER	LIQUID LEVEL	WASH SOLUTION	62	RATIOTROL 1/6 HP MOTOR SPEED CONTROL	CONTROL MOTOR SPEED	HF REACTOR
17	DIGITEC 2770 HT SERIES, DC PANEL METER	LIQUID LEVEL	RINSE WATER	63	RATIOTROL 1/6 HP MOTOR SPEED CONTROL	CONTROL MOTOR SPEED	AQUEOUS WASHER/RINSE
18	DIGITEC 2770 HT SERIES, DC PANEL METER	LIQUID LEVEL	STORAGE TANK	64	1/4" SWAGelok BULKHEAD FITTING	CONNECTOR	REGULATED AIR PRESSURE TO RINSE SOLUTION BALL VALVE OPERATOR
19	DIGITEC 2770 A DC PANEL METER	SPECIFIC GRAVITY	WASH SOLUTION	65	1/2" SWAGelok BULKHEAD FITTING	AIR SUPPLY	
20	DIGITEC 2770 A DC PANEL METER	SPECIFIC GRAVITY	STORAGE SOLUTION	66	1/4" SWAGelok BULKHEAD FITTING	CONNECTOR	REGULATED AIR PRESSURE TO WASH SOLUTION BALL VALVE OPERATOR
21	DIGITEC 2770 A DC PANEL METER	PRESSURE, INCHES OF WATER	HF REACTOR				
22	AIR PRESSURE REGULATOR	ADJUST WASH SOLUTION FLOW RATE	FLOW TO WASHER/RINSE				
23	NEWPORT 268-KC1 DIGITAL THERMOMETER	TEMPERATURE °C	HF ABSORBER				
24	AIR PRESSURE REGULATOR	ADJUST RINSE WATER FLOW RATE	FLOW TO WASHER/RINSE				
25	TOGGLE SWITCH	POWER SUPPLY	HF ABSORBER PUMP				
26	NEON LAMP	WARNING LIGHT	PUMP FAILURE OR HF ABSORBER				
27	PUSHBUTTON SWITCH WITH INDICATOR LIGHTS	RESETS SAFETY CIRCUIT	ENABLES OPERATION OF AIRLOCK VALVES AND REACTOR MOTOR AFTER SAFETY CIRCUIT HAS DROPPED OUT AND TIMER HAS TIMED OUT				
28	TOGGLE SWITCH	SAFETY CIRCUIT BYPASS	EMERGENCIES REACTOR MOTOR & AIRLOCK VALVES WHEN REACTOR PRESSURE IS BELOW LIMITS				
29	TOGGLE SWITCH	SAFETY CIRCUIT BYPASS	EMERGENCIES SAFETY SYSTEM CIRCUIT BOX WHEN REACTOR PRESSURE IS ABOVE NORMAL LIMITS				
30	TOGGLE SWITCH	SAFETY CIRCUIT BYPASS	EMERGENCIES SAFETY SYSTEM CIRCUIT BOX WHEN ARGON FLOW IS BELOW NORMAL LIMITS				
31	TOGGLE SWITCH	POWER SUPPLY	EMERGENCIES AUDIO ALARM OUTPUT WHEN SAFETY SYSTEM CIRCUIT BOX IS DEENERGIZED				
32	SECTION SWITCH	EMERGENCY POWER KILL #1	DEACTIVATES SAFETY SYSTEM CIRCUIT BOX				
33	TOGGLE SWITCH WITH LIGHT	EMERGENCY KILL #2	DEACTIVATES POWER TO AIRLOCK SYSTEM				
34	L & N CEMERY CAT NO 440-1-23-31-51-A104	PRESSURE CONTROLLER	HF REACTOR				
35	TOGGLE SWITCH WITH LIGHT	CONTROL MODE OF OPERATION (AUTOMATIC, OFF, OR MANUAL)	ARGON PURGE ON SPOOL PIECE				
36	TOGGLE SWITCH WITH LIGHT	CONTROL MODE OF OPERATION	REACTOR HULL FEEDER				
37	TOGGLE SWITCH WITH LIGHT	CONTROL MODE OF OPERATION	SPOOL PIECE PNEUMATIC VIBRATOR				
38	TOGGLE SWITCH WITH LIGHT	CONTROL MODE OF OPERATION	WASHER/RINSE HULL FEEDER				
39	TOGGLE SWITCH WITH LIGHT	CONTROL MODE OF OPERATION	TOP AIRLOCK VALVE				
40	TOGGLE SWITCH WITH LIGHT	CONTROL MODE OF OPERATION	BOTTOM AIRLOCK VALVE				
41	L & N SPEEDOMAX 3-PEN RECORDER, 0-100 MV INPUTS	TEMPERATURE RECORDER	1. HF ABSORBER TEMP 2. REACTOR INLET SEAL 3. REACTOR ZONE 1				
42	L & N SPEEDOMAX 3-PEN RECORDER, 0-5V INPUTS	PRESSURE & FLOW RECORDER	1. REACTOR PRESSURE 2. HF GAS FLOW 3. ARGON FLOW				
43	ELUKE 2240A DATALOGGER	DATA RECORDING	TEMPERATURES, FLOWS, ETC.				
44	TOGGLE SWITCH WITH LIGHT & FUSE	PROCESS ALARM RELAYS (SOLID STATE)	DEACTIVATES POWER TO SAFETY SYSTEM CIRCUIT				
45	VARIAC	POWER LIMITER	WASH SOLUTION HEAT TAPE				
46	VARIAC	POWER LIMITER	RINSE WATER HEAT TAPE				

FIGURE 7. Key to Control Instrumentation of Prototype Decontamination System (Drawing No. H-3-41891)

FIGURE 9. Blueprint of Process Flow Diagram

FIGURE 10. Safety Control Logic For Cladding Hull Decontamination System

Because of the high air flow maintained in the storage cabinet (Figure 6) by the ventilation system, excessive heat was removed from the cabinet. Insulation of the cylinders and supply manifold, and application of a heat tape to the manifold helped somewhat, but erratic indications from the flow transducer persisted. The source of this erratic behavior can most likely be related to condensation of HF gas in the transducer bore.

2. Nonuniform Mixing. As determined in the laboratory-scale work,⁽¹⁾ the use of undiluted HF gas resulted in exothermic reactions between the HF gas and zirconium hydrides. The zirconium hydrides are formed when hydrogen is generated by the reaction $4\text{HF} + \text{Zr} = \text{ZrF}_4 + 2\text{H}_2$. The hydrogen reacts with zirconium to form hydrides as described by the reaction $\text{Zr} + x\text{H}_2 = \text{ZrH}_{2x}$. If argon is insufficiently mixed with the HF, and insufficient quantities of argon are present to remove the hydrogen from the reactor, then zirconium hydrides will form, indirectly causing the reactor temperature to rise uncontrollably.
3. Insufficient Gas Removal and Poor Pressure Control. Variations in the gas removal rate caused fluctuations in reactor pressure. The gas removal rate varied as the off-gas filter became clogged with ZrF_4 powder. The filter was installed to prevent the powder from clogging the reactor pressure control valve. Excessive backpressure developed in the filter as it became loaded with powder and reactor pressure could not be maintained at a constant level. In addition, the aspirator pump used to remove gas from the reactor was not large enough and had a tendency to surge when used at full capacity. This also made pressure control very difficult.

The airlock valve assembly that isolates the HF reactor from the aqueous washer/rinser is necessary to prevent the introduction of water vapor into the HF reactor. Reaction 4 in the Basic Process Chemistry Section of this report shows that zirconium is severely corroded by water at system operating temperatures. The airlock valve assembly also serves to prevent excessive HF gas from leaking into the washer/rinser. The airlock valve assembly must, therefore, be gas tight when the valves are closed but must also allow the free flow of sharp, abrasive cladding pieces and ZrF_4 powder when the valves are open.

Problems with the hull feeding and transport functions encompassed five major areas:

1. Hull Loading. Hulls are loaded into the reactor with a canister that is rotated upward by the operator at startup. The problem with this method is that the hulls tend to bridge when they are fed as a large group. Vibrators were needed to dislodge the bridges so the hulls could flow. Another disadvantage is that once the equipment has been heated to operating temperature, more hulls cannot be added because the loading canister also seals the reactor feed chute. This makes the process an essentially batch-type operation with existing equipment.
2. Hull Entry to the Reactor Chamber. Hull transfer from the reactor feed chute into the reactor chamber is accomplished through an opening in the hollow rotating shaft of the reactor chamber (Figure 3). As the opening rotates to the downward position, hulls are fed into the chamber. However, the hulls tend to bridge at this opening and ZrF_4 powder produced during hull process enables the hulls to agglomerate. This opening became plugged in three of the eight tests and a typical result is shown in Figure 11.
3. Hull Bridging in the Airlock Valve Assembly. The spool piece between the two airlock valves (Figure 3) stores hulls while the top valve goes through its operating cycle. When the bottom valve opens, the hulls should fall out of the spool piece into the washer/rinser. However, the hulls bridge in the spool aided by ZrF_4 powder and unless vibrated will not come out.
4. Hull Bridging at the Washer/Rinser Entrance. The entrance of the washer/rinser is another area where a restricted diameter caused the hulls to bridge. The hulls were again vibrated free with the pneumatic vibrators.
5. Hull Detection. Devices that detect the presence and/or movement of hulls are difficult to design and implement because of the extremely corrosive nature of HF gas at 600°C . The absence of such a device made it impossible to determine whether or not the hulls were jammed at the

FIGURE 11. Feed Chute to Reactor Clogged With Hulls and ZrF₄ Crystals

reactor entrance until sufficient time had elapsed (2.5 hr) to allow hull transport to the washer/rinser discharge. Even after hull discharge was observed, jamming was imminent throughout the test. Furthermore, since it was desirable to terminate the flow of HF gas as soon as possible after the last hull had been processed, a hull detection device would have been very useful.

SYSTEM MODIFICATIONS

Several modifications were made to the decontamination system as operating experience was gained. The changes are described here to provide a record so that design deficiencies will not be repeated.

1. Band heaters attached to a length of pipe comprised the original heating system for the HF reactor. However, a gap of approximately 2.5 cm (1 in.) existed between the length of heated pipe and the rotating reactor vessel. This method of heating proved to be ineffective because the heaters were operating at their upper limit to maintain the reactor vessel at 600°C and the heater ends were not insulated, causing excessive heat loss. Electrical shorting also occurred during the first test and a replacement furnace was sought. The equipment loan pool provided a resistance-type heater with Kanthal wound elements. It consisted of five individually controlled zones, each rated at 5 kW and 220 V. The furnace body was fully insulated and end covers were fabricated to minimize heat losses. This furnace operated in a satisfactory manner throughout the testing phase.
2. The airlock valve assembly that isolates the HF reactor from the washer/rinser underwent two major modifications. The original assembly consisted of two vacuum gate valves. During the first test with HF gas, the gate valves broke and would not provide a gas-tight seal. The hulls became trapped in the inner valve mechanisms, causing the valves to jam. In addition, ZrF_4 powder on the O-rings prevented proper sealing. The airlock assembly was then changed to incorporate a rotary airlock valve. Hull feeding proved to be a problem with this design since the valve

could only handle a few hulls at a time without jamming. After much design work, a feeding device was installed, but it had a tendency to become plugged unless vibrated. One test with the rotary airlock valve indicated excessive HF and water vapor leakage through the valve. The valve and feeding device were subsequently replaced. The final airlock valve assembly design employed two short-pattern ball valves with an argon-purged spool piece separating the valves. The ball valves do not have any crevices where hulls can become trapped and cause damage. The major concern with the ball valves is that the spool piece between them must not be filled with hulls to the point where the hulls interfere with proper closure. With both valves closed, argon is used to purge the spool piece of both HF gas and water vapor, preventing undesirable reactions from occurring when the valves are reopened. The ball valves performed well for several tests, but long-term performance is unknown. The conclusions section provides further information on this type valve.

3. The HF gas supply system was modified by providing more turns on the cylinder heating coils. Before this change, it took too long to heat the cylinders to operating temperature, and the cylinders could not be maintained at temperature as the gas was removed from them. A heat tape was added to the HF manifold to prevent gas from condensing and producing erratic signals in the flow transducer. Insulation was also installed around the cylinders and on the HF manifold to reduce the cooling effects created by the flow of air through the cabinet. For Tests 7 and 8, the HF and argon gases were mixed in a tubing tee before being admitted to the reactor. This solved the problem of excessive hydrogen production (see section entitled Basic Process Chemistry). However, a mixing valve would be a better solution.
4. The reactor gas removal system required the installation of a filter to trap ZrF_4 powder before it clogged the bore of the reactor pressure control valve. The filter is a baffle-type filter, augmented with stainless steel wool. The filter trapped the powder efficiently but created back-pressure that prevented adequate gas removal rates.

Another modification was made for the last test to help remove the excessive amounts of hydrogen. An extension was inserted into the feed chute to allow gas removal near the mouth of the reactor (Figure 12). The extension prevented hydrogen and HF gases from passing through the bed of hulls in the feed chute, minimizing the production of zirconium hydrides which react exothermically with HF gas. In addition, the argon flow entering the canister was greatly increased to flush all gases down toward the reactor mouth.

5. Vibrators had to be added to the hull supply canister, the spool piece between the airlock valves and the entrance pipe to the washer/rinser. Without vibration, these operating areas allowed hulls to bridge, preventing hull flow. However, a problem arose from the vibration. The reactor pressure control system employs an electro-pneumatic converter to translate electrical signals into air pressure outputs. When vibrated, the converter received and generated false signals. The converter was mounted on vibration isolating pads to eliminate the problems.
6. As the overall decontamination system became more complex, various timers, counters and switches were needed to properly control its operation. A programmable sequencer was purchased to handle the elaborate sequencing steps of the airlock valve system. Once the sequencer was installed, other uses became apparent and it was eventually used for major portions of equipment control logic.

FIGURE 12. Modification to Reactor Gas Removal System

ANALYSIS OF OPERATING DATA

Table 2 shows all the pertinent data that was gathered for each test. Below are brief comments on each test.

TEST #1

This test indicated that the production of ZrF_4 powder would cause gas removal problems so a filter was fabricated to remove the powder from the exit gas. Insufficient quantities of HF gas were vaporized from the cylinders, preventing the desired ratio of HF:Argon and frustrating efforts to maintain a total gas flow high enough to purge the reactor of hydrogen. Vibration of control equipment was found to cause poor process control, and vibration isolation pads were used to eliminate the problem.

The total percent of hulls cleaned in Test #1 (Table 2) gives a false impression because so few hulls were used in the original charge. Since the process was usually controllable for approximately 10 min, the hulls which were reacted during that time period were adequately cleaned. The rest of the hulls in the charge (no matter how many) were not cleaned. Therefore, if the charge is small, the amount that are cleaned comprise a larger percentage of the total.

The original oxide weight gain on the hulls varied widely because of poor autoclaving conditions. It was subsequently decided not to use hulls from that batch for later tests.

During the test, a higher-than-expected reaction temperature was noted in the region where the hulls first entered the reactor. At this point, it was assumed that water vapor leaking through the airlock valves was causing the temperature problems.

TEST #2

First incidence of hull bridging. Hulls became jammed at the entrance port to the reactor (Figure 3) because hull feeding by vibration was unknowingly attempted when the port was facing upward. The hulls are normally

TABLE 2. Operating Data From Prototype Decontamination System

Test Number	1	2	3	4	5	6	7	8
Charge Weight (kg)	4.52	6.67	18.30	15.90	8.99	8.99	9.5	13.6
Weight of Clean Hulls (kg)	0.5 ^(a)	0.36	--	0.2 ^(a)	--	0.2 ^(a)	1.27	1.33
Total Percent Cleaned	11	5	--	1	--	2	13	10
Percent of Hull Surface Cleaned:								
Outside Diameter	50	90	--	50	--	80	95	95
Inside Diameter	10	90	--	10	--	60	95	95
Average Oxide Weight (mg/dm ²)	21 to 317	49	49	49	74	74	74	50
Percent HF:Argon Mix (average)	13:87	49:51	25:75	18:82	--	17:83	21:79	15:85
Minutes Maintained	~60	~65	~10	~32	--	~148	~80	~18
Maximum Maintained HF Flow Rate (SLPM)	2.6	8.4	7.6	2.8	0.6	1.4	2.0	1.6
Minutes Maintained	~4	~9	~10	~3	~15	~7	~6	~4
Average Reactor Temperature	584°C	577°C	567°C	568°C	--	554°C	519°C	553°C
Standard Deviation	36°C	62°C	96°C	73°C	--	58°C	57°C	64°C
Peak Reactor Temperature	637°C	700°C	703°C	688°C	--	690°C	654°C	729°C
Zone Number (No. 2 at Reactor Entrance Five Zones Total)	1	2	2	1	--	1	1	2
Residence Time of Hulls in Reactor:								
Total Minutes at Temperature	60	60	37	60	--	60	60	27
Minutes With HF Flow	60	60	10	32	--	60	60	18
Residence Time of Hulls in Wash Section of Washer/Rinser (minutes)	~40	~20	--	~20	--	~20	~68	~20
Average Wash Solution Temperature	63°C	79°C	--	89°C	--	88°C	75°C	86°C
Weight Gain (Loss) of Identified Hulls (mg/dm ²):								
1	(b)	(b)	(b)	(b)	(b)	(b)	(c)	(78)
2	(b)	(b)	(b)	(b)	(b)	(b)	(c)	4
3	(b)	(b)	(b)	(b)	(b)	(b)	(770)	566
4	(b)	(b)	(b)	(b)	(b)	(b)	(c)	(110)
5	(b)	(b)	(b)	(b)	(b)	(b)	(c)	1625

(a) Estimated data.

(b) No hulls marked.

(c) Marked hulls not retrieved.

admitted to the reactor by gravity when the port is facing downward. Vibration aids the flow of hulls when the port is facing downward but tightly packs the hulls in the feed chute when the port is facing upward. Later, when the port moves into the downward position, the hulls are so tightly packed that they cannot fall out. A microswitch was added to the reactor assembly to activate the vibrator only when the opening faced downward.

For the second time, the reactor temperature in one of the five zones was much higher than the control limits. Later comparison of airlock valve operation to temperature showed a recurring rise in temperature with each valve operation. This clearly indicated that water vapor was causing undesirable corrosion reactions. However, it was not known at this time that hydrogen was also indirectly contributing to the temperature control problems.

Improved heating coils on the HF cylinders enabled more HF to be volatilized and the off-gas filter prevented the clogging of the reactor pressure control valve. No pressure control problems were caused by the vibration of equipment.

TEST #3

This was the first test using the rotary airlock valve. Reactor pressure and temperature were very difficult to maintain within process limits. Either the lack of a consistently good airlock seal between the reactor and washer/rinser or a misalignment of the front rotating bellows seal, or both, could conceivably explain why the reactor pressure decreased to zero several times during the test. A poor airlock seal would admit water vapor to the reactor chamber, causing undesirable corrosion reactions and an excessive rise in temperature. A misalignment of the front bellows seal would admit oxygen to the reactor chamber, causing additional undesirable reactions.

The test was terminated 37 min after it began because of erratic pressure and temperature conditions. Too many variables could not be controlled during the test to make any valid assumptions about the process. The rotary airlock valve was replaced with two ball valves and an improved argon purge system.

TEST #4

This was the first test with ball valves serving in the airlock valve assembly. Several problems were encountered, but none was attributed to the ball valves. A workable solution was finally developed for the airlock valve assembly.

A pump on the HF gas scrubber began surging during this test, contributing to poor reactor pressure control. The hulls jammed again at the entrance port and caused interference at the rotating bellows seal, opening up a gap in the seal. Erratic HF flow indications were generated and assumed to be caused by condensation of gas in the flowmeter. The gas supply line and control equipment was insulated to help retain more heat.

High reactor temperatures were still encountered. For the first time, the influence of something other than water vapor on temperature was suspected.

TEST #5

No hulls were admitted to the reactor chamber because reactor pressure could not be adequately controlled and HF flow indications were very erratic.

TEST #6

Reactor pressure was extremely difficult to stabilize. HF flow could not be maintained at rates sufficient for effective descaling. Continued high temperature problems were encountered. It was during this run that hydrogen gas was recognized to be the main cause for pressure and temperature problems. Hydrogen would cause the reactor pressure to fluctuate wildly during its evolution and would also react with the hulls to create zirconium hydrides. The reaction between HF and ZrH_{2x} then caused significant heat generation. It also became apparent that inadequate mixing of HF and argon allowed pockets of undiluted HF to react with the hulls, contributing to uneven reaction rates and undesirable side reactions.

TEST #7

Mixing of HF and argon gases in a tubing tee was tried for the first time during this test. The modification slightly improved reactor pressure

control, but high temperature problems continued (enough heat was produced in the feed chute region alone to melt the PTFE bellows seal). The thermocouple to the feed chute region was contacting the outside diameter of the reactor drum and registered 654°C at its peak. However, the Inconel feed chute appeared to be over 700°C as indicated by its orange color. The PTFE bellows were replaced with stainless steel bellows.

The motorized HF flow control valve leaked gas from the steam seal during this test, causing damage to various electronic controls on the valve. The entire valve was replaced. Continuing problems were experienced with maintaining sufficient HF flow rates for effective descaling.

One marked hull was retrievable from the reactor charge. The hull was somewhat representative of the 13% of the hulls that were cleaned, losing approximately 770 mg/dm² during treatment. The clean hulls that were separated and melted probably lost even more, judging solely on appearance. After several pounds of hulls went through the reactor, a jam-up occurred. Upon later inspection, large crystals of ZrF₄ were found surrounding the hulls and lining the feed chute (Figure 11). The powder acts as a binder and causes the hulls to agglomerate, blocking the opening to the reactor chamber and causing mechanical interference between the rotating and stationary feed chutes. The interference causes hull crushing and misalignment of the front rotary seal.

The cleanest hulls from this test (1.27 kg) were separated by hand to provide feed for the Inductoslag furnace.

TEST #8

The modification made to remove off-gas from the mouth of the reactor seemed to prevent excessive temperatures in the feed chute (maximum was 660°C). However, insufficient total gas flow still allowed exothermic reactions to occur as evidenced by the 729°C temperature reached near the hull entrance.

Erratic HF flow indications and off-gas filter clogging caused a shutdown of the test 27 min after it began. Reactor pressure could not be maintained

below control limits even when pure argon was used to eliminate the possibility of hydrogen production. Later inspection revealed sufficient ZrF_4 powder in the off-gas filter to cause a flow restriction.

The wide variation in weight losses or gains of identified hulls can be explained by the short time (20 min) the hulls were washed. Crystalline deposits of ZrF_4 were very evident on hulls 3 and 5. These deposits would probably be removed by the dilute aqueous stripping solution if given enough time. Hulls 1, 2 and 4 had no deposits; in fact, 1 and 4 experienced partial removal of the black oxide layer on both inside and outside surfaces. Hull 2 appeared to be unchanged from its original condition. The hulls were not fed into the reactor in any known sequence, so it is not possible to infer anything about their location at any particular time during the test.

The cleanest hulls from this test (1.33 kg) were separated by hand and combined with those from Test #7 to provide feed for the Inductoslag furnace.

CONCLUSIONS

1. Insufficient time was available to solve process control and equipment problems. However, the process itself is a viable method for decontaminating Zircaloy-4 cladding hulls.
2. The prototype cladding hull decontamination system can descale 95% of cladding hull surfaces providing all operating parameters are under control.
3. Process control cannot be maintained for periods longer than 10 min because of undersized and underdeveloped materials handling equipment.
4. An improved method for removing gases produced during the descaling operation is needed to prevent undesirable reactions between the Zircaloy-4 cladding, hydrogen gas, and HF gas.
5. Treatment of autoclaved Zircaloy-4 hulls 1/2 to 1 hr at $550^{\circ}\text{C} \pm 50^{\circ}\text{C}$ in a 15% HF/85% Ar atmosphere did allow subsequent aqueous removal of the oxide surface layer.
6. In view of the relatively trouble-free operation of the laboratory-scale equipment,⁽¹⁾ consideration should be given to scaling up the batch process.
7. To enable continuous operation of the HF reactor and to prevent hull bridging, a hull feeding device with an airlock should be used at the reactor entrance.
8. In order to detect the presence (or absence) of hulls at various points in the equipment, a gas-operated proximity sensor could be employed. Such sensors are capable of detecting the presence of an object at ranges up to 1/8 in. In the absence of a hull, argon would flow freely from the sensor, and no output signal would be generated. If a hull were present within the sensing range, the normal argon flow would be deflected, resulting in an output signal. These pressure signals would be converted to electrical signals which would operate warning lights or other desirable apparatus.

9. It would be advantageous to use a larger HF gas supply line that is well insulated and evenly heated. A fairly high watt-density (10 W/ft) heat tape should be spirally wound around the entire supply manifold. A better method of heating and cooling the HF gas cylinders should be implemented, such as immersing the cylinders in a water bath.
10. An efficient method of mixing the relatively large quantities of HF and argon gases would aid in controlling undesirable reactions between Zircaloy-4 and hydrogen. In addition, an HF flow control valve which has an externally adjustable stem packing would prevent destruction of electronic controls in the event of gas leakage.
11. Another option for the airlock valve system between the HF reactor and the washer/rinser is to use butterfly valves. There are commercially available types which may prove to be more trouble free than ball valves. The thinner profile of butterfly valves would be advantageous because so little space is available on the equipment. A Viton liner might be acceptable as indicated by the good short-term performance of the Viton O-rings in the vacuum gate valves which were originally used. A PTFE liner would better resist the HF atmosphere but may not be resilient enough for a good seal at low seating forces.
12. The programmable sequencer could be used as an alarm rack with the purchase of threshold modules. Consolidation of other switching and interlock systems into the controller would also be advantageous. The system was added late in the project, and many functions were not integrated into its control. For even greater utility, the sequencer should be replaced by a system capable of both sequential and process loop control.
13. The entire off-gas system tubing should be made larger for more effective removal of hydrogen and excess HF gas. The ball valve which was used to control reactor pressure should be changed to a throttling-type valve for better control over reactor pressure. In addition, a centrifugal or cyclone filter would be better suited to remove ZrF_4 powder from the off-gas. The powder would then settle into a container and not clog the filter medium as it does with the current type of filter.

14. The rotating seals at both ends of the HF reactor currently employ PTFE bellows as a main component. The front bellows melted during Test #7 when the hydrogen reaction caused temperatures in excess of 700°C. The Teflon bellows were replaced with stainless steel but Inconel would be even better due to its improved resistance to HF at high temperatures.

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