ELECTROPOLISHING DECONTAMINATION SYSTEM FOR HIGH-LEVEL WASTE CANISTERS

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ABSTRACT

As part of a U.S. Department of Energy (DOE) project agreement with the Federal Ministry for Research & Technology (BMFT) in the Federal Republic of Germany (FRG), The Nuclear Waste Treatment Program at the Pacific Northwest Laboratory (PNL) is preparing 30 radioactive canisters containing borosilicate glass for use in high-level waste repository related tests at the Asse Salt Mine. After filling, the canisters will be welded closed and decontaminated in preparation for shipping to the FRG. Electropolishing was selected as the primary decontamination approach, and an electropolishing system with associated canister inspection equipment has been designed and fabricated for installation in a large hot cell. This remote electropolishing system, which is currently undergoing preliminary testing, is described in this report.

INTRODUCTION

The U.S. DOE has an agreement with the Federal Ministry for Research and Technology in the FRG to provide 30 canisters of radioactive waste for the German repository testing program. PNL is providing the vitrified waste in sealed stainless steel canisters for shipment to the FRG. The filled canisters will be inspected and decontaminated prior to shipment. The smearable surface radionuclides will not exceed 2000 d/m-100 cm^2 beta-gamma. Because transuranic radionuclides are not contained in the waste, no alpha contamination will be present. An electropolishing system was constructed for canister decontamination. This paper describes electropolishing decontamination principles, the FRG canister decontamination process, and the associated equipment. Radioactive decontamination operations will begin in the latter part of 1987.

ELECTROPOLISHING DECONTAMINATION PRINCIPLES

Electropolishing is an electrochemical process that has been used extensively by industry for many years to produce a smooth, polished surface on a variety of metals and alloys. Studies (1,2) conducted at PNL for the DOE have shown that electropolishing is also a rapid, versatile, and effective decontamination technique for radioactively contaminated metal surfaces. Mild steel, copper, aluminum, stainless steel, and highly alloyed corrosion-resistant metals have been successfully decontaminated using this technique. Electropolishing effectively removes a variety of radionuclides including plutonium, uranium, radium, cobalt, strontium, cesium, and americum, as well as baked-on and ground-in contamination, which has proved difficult to remove using other decontamination procedures.

Electropolishing is a complex electrochemical process whose application for decontamination is comparatively simple. For immersion electropolishing, the object to be decontaminated serves as the anode in an electrolytic cell. The passage of electric current produces anodic dissolution of the surface and, with the proper operating conditions, a progressive smoothing of the surface. Radioactive contamination on the surface or entrapped by surface imperfections is removed and released into the electrolyte by the surface dissolution process.

The amount of metal requiring removal to effect decontamination is a function of surface topography and depth of contamination, but it generally ranges from 5 to 50 μm for surfaces that are not heavily corroded or pitted. This corresponds to an electropolishing time of 3 to 30 min for a typical current density of 16 A/dm^2.[b] After electropolishing, the part is removed from the electrolyte and rinsed in hot water. The production of a smooth, polished surface on the electropolished part facilitates the removal of residual electrolyte by rinsing to leave a contamination-free surface.

Electropolishing can be used to prepolish the surfaces of items that are exposed to contamination in their normal service environment. A study (3) using specimens prepared at PNL have shown that electropolished surfaces become far less contaminated than other common surface finishes under equivalent service or exposure conditions, and the contamination that does adhere is more easily removed using conventional decontamination techniques. This beneficial effect of electropolishing is a direct result of the electrochemical removal of submicron-size surface imperfections that increase the effective surface area and serve as sites for the adsorption, entrapment, and retention of contamination.

Electropolishing was selected for the canister decontamination process based on its demonstrated ability to decontaminate surfaces with tightly adherent or even ground-in contamination to nondetectable levels. Also, since the decontaminated canisters could become recontaminated during the storage and handling operations prior to shipment, the electropolished surface would facilitate preshipment decontamination using a water rinse.

a Operated for the U.S. Department of Energy by Battelle Memorial Institute.
b English and metric units are used in this paper as appropriate.

FRG CANISTER DECONTAMINATION PROCESS DESCRIPTION

The FRG canisters are cylindrical and constructed of stainless steel similar to 316L. The canisters are 300 mm in diameter by 1200 mm high with a wall thickness of 8 mm. The canister is closed by welding on a lid that has a 43-mm-high pintle attached for handling. The canister has a 12-mm-high skirt on the bottom with a 40-mm-high x 70-mm-diameter dimple in the middle for standing canisters upright and stacking them on top of each other. The glass fill volume in the canisters is 60 liters. The radionuclide content, heat generation, and radiation from the canisters are summarized in Table I. The degree of canister surface contamination has not been characterized.

TABLE I

Canister Radionuclide Characteristics

Number of Canisters	Cs-137 kCi/ Canister	Sr-90 kCi/ Canister	Heat Generation Rate W/Can	Dose Rate R/h
10	210	100	1680	5×10^5
10	40	220	1680	8×10^4
10	210	160	2065	5×10^5

The FRG canisters are being filled with molten glass at 1150°C. During filling, the canisters are contained in a closed turntable, which moves the canister beneath the melter. The canister is contained in a sleeve during filling to minimize external radionuclide contamination. After glass cooling and solidification, the canister is removed from the turntable and later the sleeve. A helium leak source will be placed in the filled canister; the lid will be placed on the canister and seal welded shut. The canister will then undergo preliminary decontamination by water spraying prior to movement into the air lock and A-Cell for characterization and final decontamination as shown in Fig. 1.

The filled canister will be moved into an air lock for a dose rate reading with a fixed monitor. The canister will then be placed in the helium leak check vessel. The canister will be leak checked by evacuating the chamber and checking the chamber atmosphere for helium. The system detects leak rates down to 10^{-7} atm/cc-s. If the test fails, the canister will be returned back to the cell containing the melter. On passing the leak check, the canister will be moved into the electropolishing tank located in another hot cell.

The electropolishing tank (EPT) will contain 85 wt% phosphoric acid at 120°C. Air flow through the electropolishing tank (EPT) will be maintained whenever current is flowing to the electropolish system to assure a noncombustible concentration of hydrogen ($\leq$4.1 vol%) and contamination control. A vacuum relative to the cell will be maintained on the EPT and rinse/soak tank (RST) to prevent fugitive aerosol contamination of the cell. The tank tops will be open to the cell to permit canister handling with the cell crane.

The canister will be electropolished in segments by passing 500 amps of direct current at approximately 12 volts for 15 min between the canister and an adjacent cathode. The top of the canister and the pintle will be electropolished first using the ring cathode shown in Figure 1 with the canister sitting on an electrode in the EPT to provide the necessary electrical contact. The canister will be then lifted approximately 1 in. and the bottom of the canister will be electropolished using the same electrode in the bottom of the EPT as the cathode. The grapple that supports the canister also provides the electrical contact. A pump recirculation system pumps electrolyte into the cavity at the canister bottom to displace air and provide good solution contact with the canister. To electropolish the canister walls, the canister will be alternately raised 6 in.; then 500 amperes of current at 12 vdc will be passed between the canister and the cathode ring with the anode grapple providing the electrical contact.

Electropolishing conditions and electrolyte temperature (120°C) have been chosen so that about 1 mil of stainless steel will be removed with minimal thermal shock to the glass. During electropolishing, solution will be circulated through the tank via a pump recirculation system using two eductors in the tank bottom to increase the flow rate. The circulation enhances the electropolishing process. A filter in the recirculation system removes solids and sediment from the electrolyte.

As the canister is removed from solution using the anode grapple, it will be sprayed with water to remove residual phosphoric acid and will then be allowed to air dry above the tank. The dried canister will be moved into the RST and released on the tank bottom. In the RST, as the canister is raised, the bottom will be sprayed with water as are the side and top when the canister is raised through a spray ring. The canister will be allowed to dry suspended in the RST. The cleaned canister will then be ready for inspection and testing.

The canister will be smear tested with a pad on a fixture held by a manipulator. All surfaces--top, sides, bottom--will be smeared. The fixture with pad is transferred through the cell wall using a pass-through tray. The pad will be counted to ensure smearable contamination has been reduced to prescribed levels. The canister will be weighed using a strain gage that will be placed between the crane hook and canister grapple. The canister will be inserted in a full-length pipe gage to assure, by not binding on placement/removal, that the canister has not deformed. On canister removal from the gage, the canister will be scanned by a gamma energy detector inserted in the cell wall to obtain a radiation profile. A thermocouple will be pressure contacted at points over the canister length to obtain a temperature profile. The canister will then be placed in a water-cooled storage pod until ready for shipment.

ELECTROPOLISHING EQUIPMENT DESCRIPTION

The electropolishing decontamination process is performed in A-Cell of the Radiochemical Engineering Cells in the 324 Building. These cells consist of four large operating hot cells surrounding an air lock cell. A-Cell has overall dimensions of 2.8 m x 6.4 m x 10.4 m (9.25 ft x 21 ft x 34 ft) high. The cell and air lock can be serviced by two remotely operated 5-ton cranes. One crane is dedicated to A-Cell operations. A-Cell has operating/service galleries on the first, second, and third floor levels. Two oil-filled lead glass windows are provided for in-cell viewing, one at the first floor level and another at the second floor level. Two master-slave manipulators can be made available at each window. The shielding walls of A-Cell are 1.4 m (54 in.) thick normal density concrete.

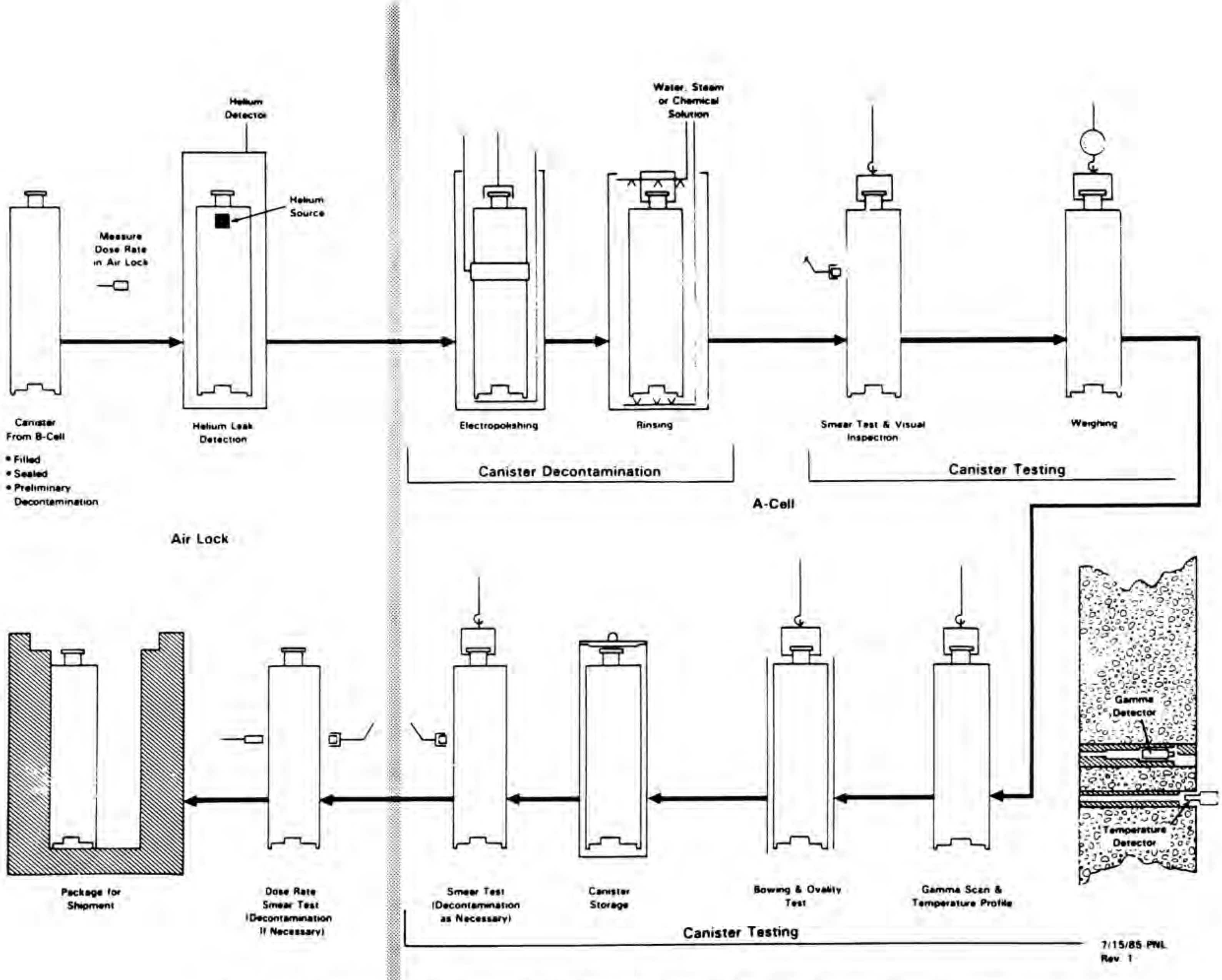

Fig. 1. Decontamination, Characterization, and Storage Facility Process Flow Diagram.

A-Cell "plug-in" modular groupings of process equipment are attached to the shielding walls with rack-mounted shield plugs containing process piping and wiring. Connections are made manually on the "cold" side of the shielding walls. The sleeves terminating in the galleries are for "cold" connections (i.e., steam, water, air, electrical supplies), except for some heat exchanger steam condensate and cooling water effluents.

Electropolishing System

The major electropolishing system assemblies are shown in Fig. 2 and 3 and subsequently described.

Electropolish tank (EPT) - The electropolish tank contains the canister electropolishing system. The EPT is 13 ft 5 in. high with an outside diameter of 4 ft 5 in. The EPT can hold up to 1000 gal of solution. The outer wall is constructed of 304L stainless steel (SS). The inner wall and internal piping are constructed of Hastelloy® C-276 for hot phosphoric acid corrosion resistance. There is about 4 in. of space between the inner wall and the outer wall containing calcium silicate thermal insulation, a 304L SS cooling coil, and a 304L SS steam jacket for temperature control.

There are three thermocouples (TCs) to monitor solution temperatures. Three 1/2-in. Hastelloy C-276 pipes lined with ceramic extend 12 ft, 8 ft, and 4 ft from the EPT top flange just inside the EPT inner wall. The open pipe ends allow TC immersion in solution. To monitor the solution, specific gravity (SPG) and weight factor (WF) dip tube air bubblers are installed in the EPT. The tubes are constructed of Hastelloy C-276 1/2-in. pipe lined with ceramic on the outside. The WF monitor is located 1-1/2 in. from the bottom of the EPT and the SPG monitor is located 10 in. above the WF monitor.

The EPT has a 3-ft-diameter spray ring located 1.5 ft from the top flange of the vessel. The spray ring is constructed of 1-in. Hastelloy C-276 pipe and has 16 equally spaced 1/4-in. holes. The 1/2-in. 304L SS pipe chemical add line is used to add solutions. There are two solution circulators (ejectors) on the tank bottom to provide tank contents agitation. There are also facilities for solution sampling and unloading

Rinse/soak tank (RST) - After electropolishing, a canister is transported to the RST to rinse off residual phosphoric acid with a steam-water spray. The RST can be utilized as a canister immersion tank.

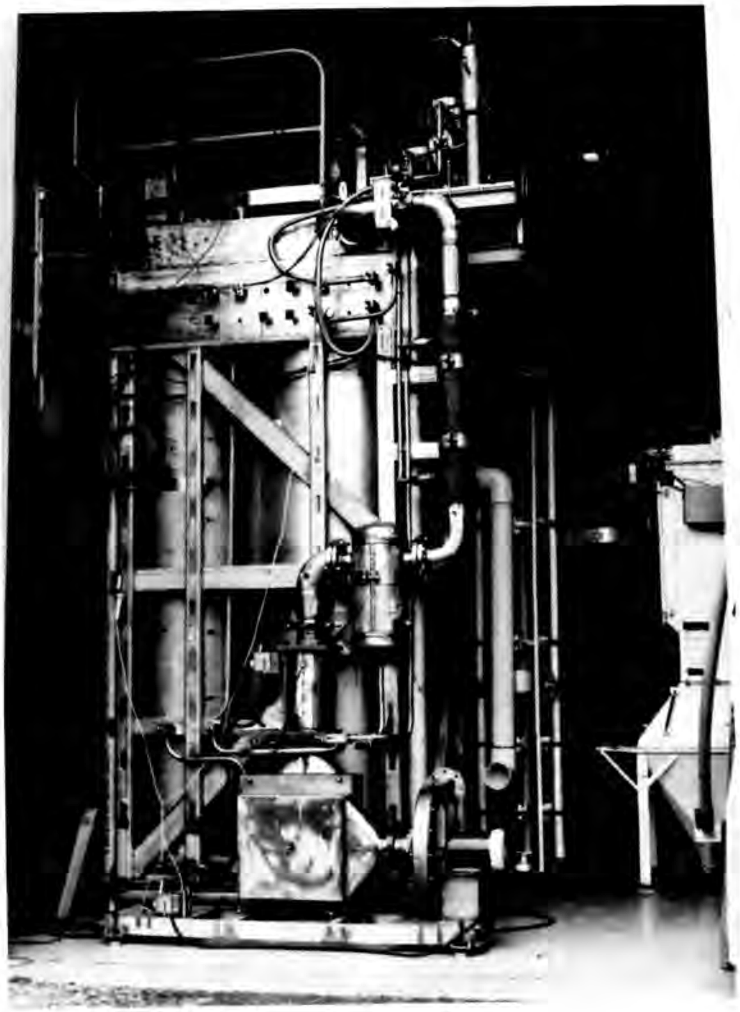

Fig. 2. Electropolish Tank.

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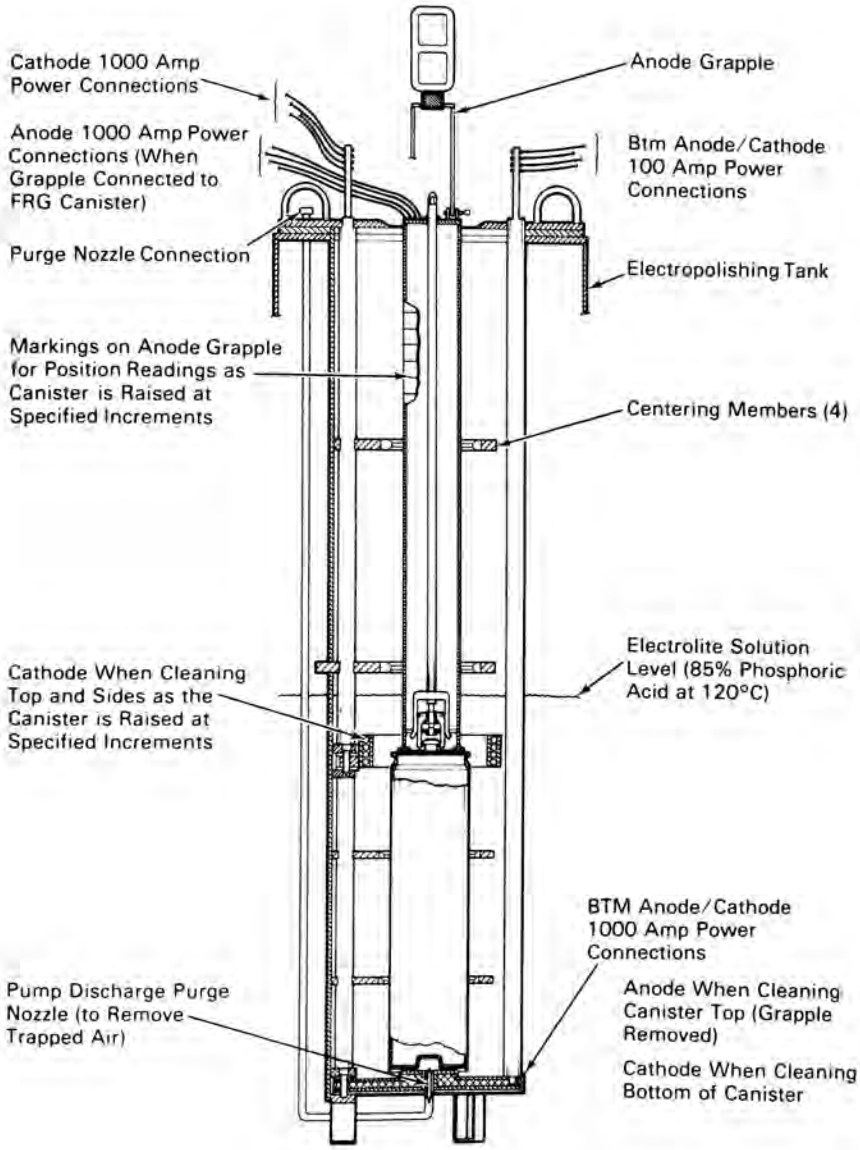

Fig. 3. Cathode Cage/Anode Grapple.

The RST is constructed of 1/2-in. 304L SS plate. Its height is 13 ft 5 in. with an outside diameter of 2 ft, which can hold up to 276 gal of liquid.

The RST has two types of spray systems. One nozzle is located 9 in. from the bottom and rinses the canister bottom. The upper spray system, located in the tank top, is a ring constructed of 1 in. 304L SS pipe with a diameter of 1 ft 7-3/8 in. and contains thirty-two 1/8-in.-diameter holes. The canister is rinsed with a steam-water mixture as it is lifted out of the RST. The RST is also provided with a heating/cooling coil for temperature control. Similar to the EPT, the RST is equipped with TC, WF, SPG, samplers, a chemical add line, and an empty outline. There is an air sparge ring in the RST bottom. It is constructed of 304L SS schedule 40 pipe with a diameter of 1 ft 7-3/8 in. containing thirty-two 1/8-in.-diameter holes. The air sparger promotes solution mixing when the RST is used as an immersion tank.

Vessel vent system (VVS) - The VVS provides an air sweep into the EPT and RST to prevent release of contaminated gas or aerosol into the cell. The air also dilutes hydrogen gas produced by water hydrolysis during electropolishing. The VVS consists of a condenser, entrainment separator, heater, filter, and blower. It is supported by a remotely replaceable rack constructed of 304L SS plate and mounted on the EPT rack.

About 450 scfm of air is cooled in the condenser from 52°C (125°F) to about 32°C (90°F). The condenser removes water vapor from the EPT and RST air. The condenser is constructed of 316L SS. The gas and condensate flows to the entrainment separator from the condenser for removal of water drops and aerosols. The entrainment separator is constructed of 304L SS.

The gas then flows to the electric heater for heating at least 5 F above the dew point to protect the filter from moisture. The electric heater is constructed of 304L SS. The heated gas flows to the filter for particulate removal. The filter element is replaceable.

The blower consists of a fan and a motor. All fan parts are constructed of type 304L SS, and the fan motor shaft is constructed of 400 series SS. The blower has a capacity to move 450 scfm of air at 21.5 in. of H_2O pressure drop. The unit is a direct-drive, 5-hp, 3600-rpm motor with a lifetime lubricated ball bearing.

Recirculation system - Radioactive particles and sediment in the solution, which are generated during the electropolishing process, are removed by the recirculation system. The recirculation system is composed of a pump, filter, flowmeter, and various jumpers and adapters. The recirculation system is attached to the electropolish rack by dowel pins on the top of the electropolish rack.

The pump is constructed of Hastelloy C-276. The pump is equipped with a 2-hp motor. It has permanently lubricated ball bearings with radiation-resistant grease. The pump has a capacity of 22 gpm. The liquid flows through a magnetic flowmeter followed by a filter assembly. Each of the three filter elements are constructed of Hastelloy C-276. The filter elements have 100-micron pore size. To monitor the pressure drop through the filter assembly, pressure is measured before and after the filter. When the filter becomes clogged it can be backflushed.

Cathode cage - The cathode cage functions as a working electrode and a counter electrode during electropolishing, depending on the electrode polarity. The cathode cage is about 10 ft high and 2 ft in diameter (Fig. 3). The cathode cage top supports the cage internals from the tank top. The top is constructed of 304L SS plate. To this top layer, two bent pipes with diameters of 3/4 in. are attached to serve as bails for handling. There is a 1-in. ceramic plate attached to the top as electrical insulation. There are four ceramic guides in the cathode cage to ensure the canister is positioned correctly within the cage.

Electric current is conducted by two 1-in.-diameter copper rods, which also provide cage structural support. The copper bars have three layers of protective outer coverings, 1-3/8 in. ceramic tubing as electrical insulator, 1-1/2 in. Hastelloy C-276 to protect the copper from acid attack, and 2-1/2 in. ceramic tubing as electrical insulator. One copper bar is 5 ft 10 in. long and provides power to the middle electrode. The other copper bar is 10 ft 6 in. long and provides power to the bottom electrode.

When electropolishing the canister top, the middle electrode functions as the cathode and the bottom electrode functions as the anode. Both electrodes are constructed with Hastelloy C-276 and are filled with copper. The bottom electrode has two 8-in.-diameter strips of tantalum imbedded on its top to ensure proper electrical contact with the canister bottom. The bottom electrode serves as the cathode and the middle electrode is not utilized when electropolishing the canister bottom. In this configuration, the anode grapple is used as an anode. The canister bottom has a cavity that can entrap gases produced during electropolishing. Recirculated acid is sprayed into the cavity via the canister bottom spray to displace air and gases. The canister bottom spray is fed by a 12-ft Hastelloy pipe with 1/2-in. diameter that is covered with 1-3/8-in. ceramic tube. When electropolishing the wall of a canister, the middle electrode functions as the cathode and the bottom electrode is not utilized. The anode grapple again serves as the anode.

Anode grapple - When electropolishing the canister bottom and side, the anode grapple provides the electrical contact. The anode grapple is used to raise the canister during the canister wall polishing and to transfer the canister from the EPT to the RST for rinsing. The anode grapple is about 7 ft high with a diameter of about 10 in. (Fig. 3). The bottom 3 ft of the anode grapple is constructed of Hastelloy C-276; the rest is constructed of 304L SS. Tantalum is used to enhance electrical contact in the two places where the anode grapple makes contact with the canister.

The clamp of the anode grapple is used to latch on to a canister and can lift a canister weighing up to 1500 lb. Prior to electropolishing the canister bottom and side, a torque of 200 ft-lb is applied to the clamp to close the hook-like clamp around the canister lid pintle. As the clamp tightens, the tantalum ring pad makes a good electrical contact with the canister lid.

After electropolishing the canister, the outside of the anode grapple is sprayed by the EPT spray ring with a steam-water mixture while the inside is rinsed by the anode grapple spray ring with steam. The anode grapple spray ring is located inside the anode grapple.

Four cable connectors located on the anode grapple top complete the electrical circuit for canister electropolishing. A pivot bail on the anode grapple is used to for transport.

Power rack - The power rack provides the circuitry from the power supply to the electropolishing system. It is constructed of SS pipe and structural members. The power rack contains 20 power cables that provide power to five circuits. Each circuit has four power cables that provide a total of 1000 amps of current. Two circuits lead to the cathode cage, one circuit leads to the anode grapple, and two other circuits are the spares.

The power rack is about 12 ft high and is supported at two places. The electropolish rack provides a groove made out of angle iron to fit the lip on the bottom of the power rack. The power rack top is supported by the plug support attached to the wall. The power rack plugs into the wall plug and the bottom rests on the electropolish rack. The electrical plugs are similar in construction to the service plugs of the electropolish rack.

Electropolish rack - The electropolish rack contains and supports the electropolishing tank, rinse soak tank, vessel vent system, recirculation system, power rack, various plugs, and manipulator faces. The electropolish rack is fabricated from SS pipe and structural members. Its dimensions are about 16 ft high × 8 ft 10 in. long × 4 ft × 11 in. wide (includes plug). The electropolish rack is equipped with two leveling pads on the bottom. These two pads can be adjusted from the top of the electropolish rack.

There are three service plugs on the electropolish rack. Each plug weighs about 650 lb and is filled with lead to shield radiation. There are radiation bends in each service line. One plug provides most of the services such as steam, cooling water, and instrument air to the EPT. Another plug provides most of the services to the RST. Both plugs are constructed of SS plate and are 26-1/2 in. long. The third plug provides an outlet for cooling water, condensate, and jet discharges from both EPT and the RST. The jacket of the outlet plug is constructed of SS plate and is 27 in. long.

Two manipulator faces are on the electropolish rack. The upper manipulator face (UMF) is located about 12 ft 6 in. from the floor on the electropolish rack. The UMF brings service to equipment that are separate from the electropolish rack. The lower manipulator face (LMF) is located about 1 ft 6 in. from the floor. The LMF is used to discharge condensate from the steam jacket of the EPT, cooling water from the VVS, and the liquid from the separator drain.

FUTURE PLANS

Prior to installation of the remote canister electropolishing system in the A-Cell facility, the system will undergo a series of design verification tests to confirm the operability and maintainability of the process equipment and establish the necessary control parameters for effective, uniform canister decontamination. These tests are currently underway. Between July and September 1987, the system will be installed in the hot cell. The actual FRG canister decontamination activities will begin in the 4th quarter of 1987.

ACKNOWLEDGMENT

The authors wish to acknowledge Jaiduk Jo's contribution to this paper. Much of the information in this paper is based on internal reports written by him.

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