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TITLE: ELECTROLYTIC DECONTAMINATION OF CONDUCTIVE MATERIALS

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ABSTRACT

Using the electrolytic method, we have demonstrated removal of Pu from contaminated conductive material. At EG&G Rocky Flats, we electrolytically decontaminated stainless steel. Results from this work show removal of fixed contamination, including the following geometries: planar, large radius, bolt holes, glove ports, and protruding studs. More specifically, fixed contamination was reduced from levels ranging $> 1,000,000$ counts per minute (cpm) down to levels ranging from 1,500 to < 250 cpm with the electrolytic method.

More recently, the electrolytic work has continued at LANL as a joint project with EG&G. Impressively, electrolytic decontamination experiments on removal of Pu from oralloy coupons have shown decreases in swipable contamination that initially ranged from 500,000 to 1,500,000 disintegrations per minute (dpm) down to 0-2 dpm.

INTRODUCTION

Because radioactive waste issues are foremost in the actinide industry, better methods to decontaminate conductive materials is of interest to all who work with actinides. Electrolytic techniques can be used as a decontamination or a waste treatment method to replace existing processes, and benefits include the following: waste minimization-including process waste reduction, recategorization (TRU to Low Level), reduced exposure-ALARA, and easier compliance with policies and procedures.

Decontamination of surfaces using electrolytic methods has an established technology base in electropolishing. Additionally, previous work at Battelle Pacific Northwest Laboratories demonstrated that actinide contaminants can be removed from metal surfaces using electrolytic techniques.[1] Other work at EG&G Rocky Flats has also been done to study the electrolytic decontamination method.[2]

Electrolytic decontamination is relatively easy to implement experimentally. By applying a low DC voltage from an anode to a cathode through an electrolyte solution, an electrochemical reaction can be induced. By proper selection of the applied voltage, selective anodic dissolving of the contaminant can occur. Typical electrolytes are relatively inert salts such as sodium nitrate or sodium chloride.

In general, we have shown decontamination of two types of conductive materials, stainless steel and uranium-oralloy. The stainless steel electrolytic studies were oriented toward glove box decontamination in an effort to show reductions from TRU waste levels to low level waste, which could save $\geq \$100,000$ per discarded glove box. Moreover, a method to remove highly radioactive contamination, such as Americium, could significantly reduce personnel exposures. Swipable contamination can also be removed using electrolytic methods, which will reduce labor intensive cleaning-personnel exposure.

The need to decontaminate oralloy comes from limited vault storage space and limits on the amount of oralloy facilities can hold. Since decontaminated oralloy can be

sent to Oak Ridge for storage or processing, these needs can be met if an effective oralloy decontamination process can be developed. The previous standard method to decontaminate oralloy used hot concentrated nitric acid as a spray leach process, which produced >11 liters of liquid waste per decontaminated part. In addition, this leach process was only successful about half of the time in decontaminating parts with a single pass, which led to more waste generated and more personnel exposure for additional passes through the process. Therefore, electrolytic oralloy decontamination (ELOYD) is being studied in an effort to circumvent the large amount of waste produced and overall ineffectiveness of the acid/spray leach process.

EXPERIMENTAL DETAILS

Electrolytic decontamination experiments were conducted in two fashions. First, the work done at Rocky Flats used a portable hand held type of apparatus that contained a medium, (nylon pad, cloth, sponge, etc.) to support the electrolyte. The cathode of the power supply was attached to this medium. The anode of the 6 or 12 volt, 100 amp battery charger was attached to the stainless steel that was decontaminated. Solutions of electrolyte were 40% sodium nitrate in water. To concentrate on removal of fixed contamination, swipable contamination was removed prior to the electrodecontamination experiment. Typical tests were performed for short time periods; i.e., single passes of the hand held fixture were made over the fixed contaminated area.

The second set of experiments conducted at LANL used an EG&G Model 273 power supply to control the H-cell type of electrochemistry experiments. These experiments were done as follows: cyclic voltametry, constant voltage-variable current, or constant current-variable voltage. Electrolyte solutions were 200 g of sodium nitrate per liter of deionized water.

Fixed contamination on stainless steel was counted with a Ludlum Model 139 Survey meter-alpha probe with a surface area of $\sim 85 \text{ cm}^2$. Swipable contamination from OY was first swiped then the swipe counted with a Ludlum probe. If contamination was less than 100 counts per minute, the swipe was counted using a scintillation counter. The Eberline alpha scintillation counters were calibrated twice daily; and results are corrected for the counter response and reported in disintegrations per minute (dpm). (It should be noted that these counters do not distinguish between uranium alpha and plutonium alpha.)

RESULTS

Stainless Steel Decontamination Results

The tests to determine electrodecontamination (EDC) for removal of Pu-239, Pu-238, and Am-241 from stainless steel at EG&G/Rocky Flats were the following:

- 1.) outside surface of a 304 stainless steel storage tank (large radius surface);
- 2.) sight gauge bracket (horizontal planar surface) attached to 1;
- 3.) 1000 series carbon steel sight gauge bracket (complex surface geometry) with multiple bolt holes, threaded U-bolts, and corresponding nuts;
- 4.) 304 stainless steel flange (vertical planar surface, 2 feet by 2 feet) on the end of a glove box with bolt holes around perimeter;
- 5.) 304 stainless steel exterior surface of a glove box line (mixture of complex and simple surface geometries-vertical planar, protruding studs, glove ports, etc.).

As noted previously, these tests were done to determine removal of fixed contamination. Before and after contamination levels are summarized in Table 1. Since the probe lower limit was 250 cpm, the actual fixed counts were $< 250 \text{ cpm}$. Since the probe upper limit was 1,000,000 cpm, the initial fixed contamination value for 3 was $> 1,000,000 \text{ cpm}$. The

Table 1. Summary of electrolytic decontamination results on stainless steel. (Units of activity are counts per minute (cpm) per 85 cm².)

<u>Test #</u>	<u>Initial Fixed Activity</u>	<u>Final Fixed Activity after EDC</u>
1	3,000	<250 (Probe limit)
2	150,000	<250
3	>1,000,000	1,500
4	10,000	<250
5	1,000 to 100,000 (area dependent)	<250 to 1,000 depending on area

removal of contamination from this wide variety of geometries demonstrates the effectiveness of this decontamination method.

Oralloy Decontamination Results

Electrolytic oralloy decontamination (ELOVD) tests were done on several OY coupons that had surface areas of ~10 cm² per side. Total applied voltage, anode to cathode, was 3.5 volts and current draw was about 1 amp. Constant voltage between anode and reference electrode-Corning Ag/AgCl was varied between 1.0 and 2.0 volts with similar results. Dissolution rates of metal were approximately 10 mg/min. Initial swipable counts ranged from 500,000 to 1,500,000 dpm per side. Final swipable counts ranged from 0 to 2 dpm per side.

DISCUSSION

The stainless steel electrolytic decontamination was successful. Although the total amount of Pu holdup is not known for these tests, TRU level wastes can be associated with 50,000 cpm; thus, three of the five tests showed a lowering of the decontamination limit to low level waste. Since numerous geometries were electrolytically decontaminated, the general applicability of the decontamination process was shown. Therefore, electrolytic decontamination of glove boxes should be good. Future work will look at complete

electrolytic decontamination of a glove box with an in situ device being developed at LANL.

For OY coupon decontamination, a linear interpolation results in final values ranging from 0-20 dpm/100 cm², which is at or below the removable Pu contamination limit of 20 dpm/100 cm². Therefore, we met our objective to decontaminate OY. Typical OY decontamination using sodium nitrate electrolyte solution forms a precipitate that can easily be separated from the liquid. Therefore, the waste produced by the electrolytic method is in the form of a solid that is substantially less volume than the acid spray leach process. A modest estimate is a factor of ten reduction in waste volume; however, future studies should enable an even lower volume of waste to be achieved.

During the OY experiments, we have demonstrated selectivity to remove material by a variety of means in an effort to reduce the amount of waste. For OY decon, the outer surface is U and Pu oxides that readily dissolve before the metal. Additionally, preferential etching can be achieved by using solution turbulence or using differences in distance between the cathode and the conductive material to be decontaminated. This preferential etching enables decontamination of the area of contamination on the surface, which also reduces the amount of waste produced.

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