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Electrolytic decontamination of conductive materials

George M. Campbell, Timothy O. Nelson* and John L. Parker
Nuclear Materials Division, Mail Stop E510, Los Alamos National Laboratory, Los Alamos, NM 87545 (USA)

Richard H. Getty*, Tom R. Hergert, Kirk A. Lindahl and Larry G. Peppers
MST, Building 881, EG&G/Rocky Flats, Inc., P.O. Box 464, Golden, CO 80402-0464 (USA)

Abstract

Using the electrolytic method, we have demonstrated removal of Pu and Am 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 from >1 000 000 counts per minute (cpm) down to levels ranging from 1500 to 250 cpm using the electrolytic method.

More recently, the electrolytic work has continued at Los Alamos National Laboratory as a joint project with EG&G/Rocky Flats. Impressively, electrolytic decontamination of Pu/Am from U surfaces (10 cm^2 per side) shows decreases in swipable contamination from 500 000–1 500 000 disintegrations per minute (dpm) down to 0–2 dpm. Moreover, the solid waste product of the electrolytic method is reduced in volume by more than 50 times compared with the liquid waste produced by the previous U decontamination method — a hot concentrated acid spray leach process.

1. Introduction

Because radioactive waste issues are foremost in the actinide industry, better methods to decontaminate conductive materials are of interest to all who work with actinides. Electrolytic techniques can be used for decontamination or waste treatment to replace existing processes, and benefits include the following: waste minimization (including process waste reduction), recategorization (high level to low level), reduced exposure (ALARA — as low as reasonably achievable), 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. By applying a low d.c. 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 occurs. Typical electrolytes being studied are nonhazardous inert salts such as sodium nitrate or sodium chloride.

In general, we have shown decontamination of two types of conductive materials, stainless steel and Oak Ridge enriched uranium (oralloy). The stainless steel electrolytic studies were oriented toward glove box decontamination in an effort to show reductions from transuranic (TRU)-high level waste to low level waste, which could save at least $100 000 per discarded glove box. Moreover, a method to remove highly active radioactive contamination, such as americium, could significantly reduce personnel exposures during daily operations. Swipable contamination can also be removed using electrolytic methods, which will reduce labor-intensive cleaning, and hence personnel exposure.

The need to remove Pu and Am contamination from oralloy comes from limited vault storage space and limits on the amount of oralloy that 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 that produced >11 liters of liquid waste per decontaminated part. In addition, this leach process was not always successful in decontaminating parts with a single pass, which led to more waste

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*Authors to whom correspondence should be addressed.

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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.

2. Experimental details

Electrolytic decontamination experiments were conducted in two fashions. First, the work done at Rocky Flats used a portable handheld 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 V, 100 A battery charger was attached to the stainless steel to be decontaminated. Solutions of electrolyte were 40% sodium nitrate in water. To concentrate efforts 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 handheld fixture were made over the fixed contaminated area.

The second set of experiments conducted at Los Alamos National Laboratory (LANL) used an EG&G Model 273 power controller for the H-cell electrochemistry experiments. These experiments were done as follows: cyclic voltammetry, 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 about 85 cm^2. Swipable contamination from oralloy was first swiped, then the swipe counted with a Ludlum probe. If contamination was less than 100 counts per minute (cpm), the swipe was counted using a scintillation counter. The Eberline alpha scintillation counters were calibrated twice daily; 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. Also, 2 dpm is approximately equal to 1 cpm.)

3. Results

3.1. 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.).

TABLE 1. Summary of electrolytic decontamination (EDC) results on stainless steel. (Units of activity are counts per minute (cpm) per 85 cm^2)

Test No.	Initial fixed activity	Final fixed activity after EDC
1	3000	<250[a]
2	150000	<250[a]
3	>1000000	1500
4	10000	<250
5	1000–100000[b]	<250–1000[b]

[a] Probe limit.
[b] Area dependent.

As noted previously, these tests were done to determine removal of fixed contamination. Contamination levels before and after EDC are summarized in Table 1. Since the probe lower limit was 250 cpm, the actual fixed counts were <250 cpm. Since the probe upper limit was 1 000 000 cpm, the initial fixed contamination value for test (3) was >1 000 000 cpm. The removal of contamination from this wide variety of geometries demonstrates the effectiveness of this decontamination method.

3.2. Oralloy decontamination results

ELOYD tests were done on several oralloy pieces that had surface areas of approximately 10 cm^2 per side. Total applied voltage, anode to cathode, was 3.5 V and current draw was approximately 1 A. Constant voltage between anode and reference electrode, Corning Ag/AgCl, was varied between 1.0 and 2.0 V with similar results. Dissolution rates of metal were approximately 10 mg min^{-1}. 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.

4. Discussion

The stainless steel electrolytic decontamination was successful. Low level wastes can be associated with surface fixed values <50 000 cpm; thus, the three tests that started above 50 000 cpm were decontaminated to low level waste. Since numerous geometries were electrolytically decontaminated, the general applicability of the decontamination process was also shown. Future work will look at electrolytic decontamination of glove boxes with an in situ device being developed at LANL.

For oralloy piece decontamination, a linear interpolation results in final values ranging from 0 to 20 dpm/100 cm^2 (total alpha), which is at or below the removable Pu contamination limit of 20 dpm/100 cm^2. Therefore we met our objective to decontaminate oralloy. Typical oralloy decontamination using sodium nitrate electrolyte solution forms a precipitate that can easily be separated from the liquid; i.e. the electrolyte liquid is used over and over, and can also be regenerated by the addition of more sodium nitrate. Therefore, the waste product produced by the electrolytic method is in the form of a solid of substantially less volume than that produced by the acid spray leach process. A modest estimate is a reduction in waste volume by a factor of 50; moreover, future studies should enable an even lower volume of waste product to be achieved.

During the oralloy experiments, we have demonstrated selectivity to remove material by a variety of means in an effort to reduce the amount of waste. For oralloy decontamination, the outer surface comprises U and Pu oxides that readily dissolve before the U metal. Additionally, preferential etching of a contaminated area can be achieved by using solution turbulence or differences in distance between the cathode and the conductive material to be decontaminated.

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