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ABSTRACT

An alpha-contained pilot facility is being built at the Savannah River Laboratory (SRL) for research into the disassembly and decontamination of noncombustible, Transuranic (TRU) waste. The design and program objectives for the facility are presented along with the initial test results from laboratory scale decontamination experiments with Pu-238 and Cm-244.

INTRODUCTION

As part of an integrated program to dispose of TRU waste, a Disassembly and Decontamination Facility will be built at the Savannah River Plant (SRP) to process noncombustible waste. Cabinets, gloveboxes, and other large items of contaminated process equipment are currently stored retrievably on concrete pads beneath an earth cover in the SRP burial ground in compliance with DOE regulations. The waste is wrapped in plastic, boxed in sealed plywood containers, and in turn placed inside large, specially designed steel burial boxes which occupy substantial volumes on the concrete pads. Smaller waste items are stored in 55-gallon galvanized steel drums. Drums containing more than 0.5 Ci are in turn placed inside concrete culverts on the pad.

The plant facility will remove more than 99% of the initial surface activity from the waste by decontamination with chemical sprays. The radioactive solutions will be transferred to the SRP high level liquid waste system (HLW). HLW will be encapsulated in glass in the Defense Waste Processing Facility (DWPF), planned for operation in the late 1980's. The waste will be remotely disassembled and sectioned for volume reduction. Present design plans call for packaging and certifying the processed waste for shipment to the Waste Isolation Pilot Plant (WIPP) for geologic storage. Alternatively, additional decontamination

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techniques such as vibratory cleaning, electropolishing, or chemical etching would be used to decontaminate waste to less than 10 nCi per gram for disposal as low-level waste in the SRP burial ground.

The initial feed for the plant will consist of equipment removed after the renovation of a plutonium processing line which has produced hundreds of kilograms for Pu-238 and other TRU isotopes over 25 years. At a later date, the plant will process the TRU inventory already stored on pads and the waste in 55-gallon drums.

PILOT FACILITY

To support the design and operation of the proposed plant facility, a pilot scale facility is being built at SRL. It will implement, adapt, and evaluate available technology and demonstrate viable modes of operation. Contaminated gloveboxes generated by R & D programs at SRL will be used as feed in the research phase of its operation. The facility will later routinely dispose of TRU contaminated equipment retired from service at SRL.

The design, equipment, and instrumentation for the pilot facility have been chosen on the basis of previously demonstrated technology or operating experience at SRP and a number of sites [Pacific Northwest Laboratory (PNL), Los Alamos Scientific Laboratory (LASL) and others].

The pilot facility consists of a main cell 7.3 meters long, 2.7 meters wide, and 4.8 meters high with large, open areas for unpacking and disassembling waste and an inner room for initial decontamination. Figure 1 shows a plan view of the facility. The cell floor and frame are stainless steel with transparent wall panels of fire-retardant Lexan® (General Electric) polycarbonate. Three sides of the facility are in a regulated service area and have numerous gloveports at various levels for hands-on operations. The fourth wall is in a clean area where most process controls are located. Remote disassembly cutting operations and heavy lifting are performed with two pairs of Master/Slave Manipulators (MSM) and a bridge-mounted electromechanical type manipulator (PaR, Programmed and Remote System, Inc., model 3000), see Figure 2.

Feed is introduced into the cell through a walk-in airlock. Once through, air ventilation provides the primary contamination control barrier. Air leaving the cell is filtered through a dual-pac HEPA filter inside the cell and then passes through two more banks of external HEPA filters prior to entering the building exhaust. The dual-pac filter will be replaced remotely from inside the cell. The demister portion of the dual-pac filter minimizes clogging due to moisture. Opening packages, disassembling waste, and volume reduction will be accomplished with standard equipment frequently used in contained

environments which will be adapted for remote operation. Much of the effort in this area will be directed toward developing procedures and demonstrating an integrated remote operation.

Chisels, pry bars, and a hot-wire cutter will be used to uncrate the waste and remove plastic overpacking. An impact wrench will aid in the removal of gloves, glass, and other unboltable items. Appendages will be severed by a hacksaw or bandsaw prior to decontamination. A variety of tools (power hacksaws, Skil® (Skil Corp.) saws, drills, impact wrenches, etc.) will be adapted for remote operation along with a plasma arc cutting torch.

The decontamination room is separated from the main cell area by a roll-up door to contain the spread of contamination during pressurized water and chemical spray treatment. It has its own set of HEPA filters and a separate drain and liquid waste hold tank. The inner surfaces of the Lexan® panels in the decontamination room are coated with sheets of transparent Teflon® (Du Pont) film to prevent corrosive chemical attack by chemical sprays which may reduce visability.

In the decontamination procedure, based on previous work by Crawford[1], a pressurized water wash will be followed by chemical sprays to remove oxide film and gross amounts of loose contamination from the metal surfaces of the interior of the box. Acidic permanganate has previously proven to be an effective decon reagent in bench scale tests at SRL and on production scale at both SRP and Allied Chemical at Idaho Falls.[2] It will be used in combination with oxalic acid and pressure washing. Solutions will be collected, sampled, and pacified, if required prior to disposal to the building's high-level drain system.

Construction of the pilot facility is approximately 85% complete with the installation of major process equipment under way (the PaR manipulator). During an initial "cold" phase of operation scheduled for Summer 1982 several uncontaminated gloveboxes will be processed to check out material handling techniques, as well as disassembly equipment, such as a remote standoff and cutting guide for the plasma arc torch. Hot operation of the pilot facility is planned for Fall 1982. Ten gloveboxes from a recently retired PuO_2 powder line in the Actinide Materials Facility (AMF) have been placed in temporary storage. About a kilogram of PuO_2 (85% Pu-238) had been handled in these boxes over a period of years. At a later date, contaminated equipment and drummed waste from the SRP process lines will be treated. A bagout port and procedure for packaging waste in a WIPP approved shipping box will also be developed.

TRU MONITORING INSTRUMENTATION

Passive neutron counting and gamma-ray spectrometry will be available for monitoring and assay of TRU waste at the pilot facility. Technical aspects of many of the available passive techniques for waste assay of plutonium with weapons or reactor grade isotopics have been reviewed. Modification and/or addition to these techniques will be required to account for the presence of significant amounts of other TRU isotopes such as Pu-238, Am-241, Am-243, Cm-244, and others in the SRL/SRP waste.

Low-density combustible wastes will be assayed by low-energy photon (LEP) counting on an existing barrel scanner. A multichannel analyzer - minicomputer system will be used in conjunction with a LEP detector for quantitative analyses of liquid samples and isotopic analysis. A glovebox at the rear of the pilot facility is being adopted for gammaray counting of liquid samples. The gamma-ray spectrometry system has been used to analyze coupons and liquid samples from lab experiments. The detection limit for plutonium is approximately 10^5 dpm, depending upon the isotopic composition.

A passive neutron counter using ^3He detector tubes imbedded in a slab of polyethylene moderator was developed to monitor packages prior to introduction into the facility. Six of the slab counters were arranged to form a well, and evaluated for the assay of noncombustible waste. The detection limits of the well counter for some TRU isotopes in 5-gallon containers by total neutron counting were approximately 700 mg for weapon's grade plutonium oxide, 4 mg for Pu-238 (85% 238) oxide, and 0.006 mg for Cm-244 oxide. Limits were determined by the neutron background in the counting area. Detector efficiency was approximately 12%. A shift register controller will be used for discriminating between random neutron events from alpha, n reactions on low atomic number elements, and the time correlated events from spontaneous fissions.

BENCH SCALE EXPERIMENTS

In preparation for the "hot" operation of the pilot facility, a number of lab scale experiments on decontamination have been completed. Their objectives were to evaluate a modified spray cleaning procedure and to determine the decontamination factors (DFs) that could be achieved by a multistep process which included the added decontamination by vibratory cleaning and chemical etching. Etching tests compared the effectiveness of an oxalic acid/hydrogen peroxide mixture, both acidic and alkaline permanganate and Ce^{+4} in dilute nitric acid. DFs were measured for various techniques on both contaminated samples from process line equipment and short-term, "synthetically" contaminated samples.

Two stainless steel shelves were removed from the powder handling areas in the AMF. They had not been previously decontaminated but had been coated with several layers of the Clearcoat® (Oakite Products, Inc.) paint. Six stainless steel coupons (3 cm x 3 cm x 0.8 cm) were exposed to the Pu-238 powder in AMF for three weeks and for the purposes of this study termed "synthetically" contaminated. The synthetic coupons had levels of 10^9 dpm/100 cm^2 Pu-238 and 10^5 dpm/100 cm^2 Am-241. A third shelf was removed from another SRL glovebox in which approximately 0.5 grams (40 Ci) of Cm-244 oxalate had recently been precipitated and calcined.

The three shelves and a blank piece of stainless steel sheet were spray decontaminated by dipping sequentially in dilute caustic to remove the Clearcoat®, acidic permanganate, oxalic acid, and finally dilute nitric acid. Between dips, the shelves were spray cleaned with water at 300-500 psi. DFs were measured by counting the shelves before and after treatment both in a passive neutron well and with a gamma-ray probe. DFs of >85, 280, and >90 were measured by neutron counting (the first and third shelves had no net signal above background after treatment). The gamma probe measured DFs of 200, 400, and 200 respectively for the shelves. Table 1 gives the results from gamma-ray and alpha spectrometry of the liquid waste streams.

The three shelves were again coated with Clearcoat® and cut into small pieces with a hand-held shear. The areas of highest activity in the center of the shelves were sheared into 5 cm x 5 cm coupons suitable for counting on a low-energy photon detector with a 20 cm^2 active area coupled to a multichannel analyzer. The coupons and the synthetic samples which had not been spray cleaned were treated for 6 to 8 hours by a vibratory cleaner with a 1-ft^3 capacity using hardened steel ball media and dilute caustic spray of about 4 liters per hour. The transferable activity was measured after processing by disc smearing and alpha counting. DFs were calculated from the before and after gamma-counting of the coupons. DFs for the shelf pieces ranged from 5 to 25, and for the synthetic coupons from 20 to 50. Transferable activity after vibratory cleaning was generally less than 10^4 dpm/100 cm^2 and greatly facilitated subsequent handling and processing. Most of the samples had contamination levels above 10 nCi/gram (approximately 10^6 dpm/100 cm^2) after the vibratory cleaning.

In the first etching tests it was found that acidic permanganate was considerably more effective than either the alkaline reagent or the oxalic acid/hydrogen peroxide mixture. DFs for the acid permanganate ranged from 50 to 2000 for Pu-238. The larger DFs were obtained by eight hours of treatment at 60 degrees Centigrade with fresh permanganate after four hours, and hourly dipping of the samples in warm oxalic acid to remove the manganese dioxide surface film. Figure 3 shows a plot of the activity levels versus time for four AMF coupons

treated by this method. An estimated 0.2 to 0.3 mil of surface was removed by the treatment and final activity levels were below 10 nCi/gram. Repeated treatments did not reduce the level below 10^3 - 10^4 dpm/100 cm^2. The acidic permanganate reagent was not found to be effective for more than about four to eight hours at 60 degrees Centigrade, due to decomposition of the reagent and recontamination of the surfaces. The Pu-238 contamination remaining on the AMF coupons was found to be very tenacious, resisting repeated attacks with Ce^{+4} in nitric acid which removed several mils or more of surface. Recontamination of the coupon surfaces was also observed with the Ce^{+4}. The synthetic coupons were decontaminated to essentially background levels by this treatment (less than approximately 50 dpm/100 cm^2), while the AMF shelf samples had measurable activities up to 10^3 dpm/100 cm^2.

In summary, the spray treatment with acidic permanganate dip gave DFs of greater than 10^2 and generates less secondary waste than the alkaline reagent used by Crawford. Vibratory cleaning combined with chemical etching in acid permanganate decontaminated samples to less than 10 nCi per gram. The vibratory cleaner safely processed samples as high as 5 x 10^9 dpm/100 cm^2 in a hot hood. Fixed activity levels remained high, but samples could be handled with little or no transferable contamination. Significant amounts of the Pu-238 activity appeared to be imbedded in the steel. Comparison of alpha probe surface measurement with LEP counting data and analyses of decon solutions showed that alpha probing results were consistently low by factors of ten or more. Samples from the Pu-238 process line material were much more difficult to decontaminate than coupons exposed to the activity for short periods of time.

REFERENCES

1. J. H. Crawford. Decontamination of TRU Glove Boxes. USDOE Report DP-1473, E. I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, SC (March 1978).

2. J. E. Johnson and A. G. Westra. ICPP Decontamination Manual. USDOE Report ICP-1172, Allied Chemical Corp., Idaho National Engineering Laboratory, Idaho Falls, Idaho (January 1979).

TABLE 1. Spray Decon - Activity of Waste Streams

Step #	Description	Activity Collected (mCi) Pu-238	Cm-244	Percent of Total Collected Pu-238	Cm-244
1	Caustic soln. from Clearcoat removal	2.9	6.8	5.8	10
	Solids	4.0	1.0	7.8	1.5
		6.9	7.8	13.6	11.5
2	Pressure wash	0.14	0.95	0.28	1.4
3	Acidic Permanganate	41	59	80	86
4	Pressure wash	0.22	0.21	0.44	0.33
5	Oxalic Acid	2.1	0.23	4.1	0.34
6	Pressure wash	0.81	0.15	1.6	0.23
7	Final rinse	0.0015		<0.1	
TOTAL		51.2	68.3		

Average activity of liquid waste stream approximately 2×10^6 dpm/ml

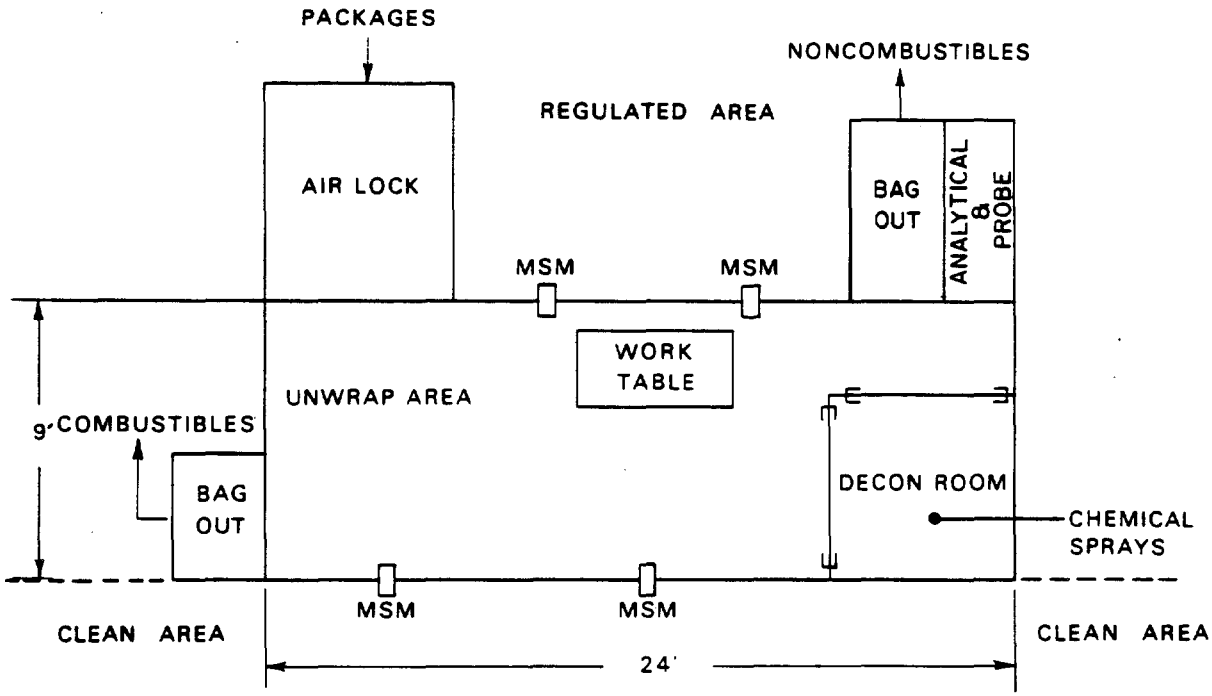

FIGURE 1. Cell Plan View.

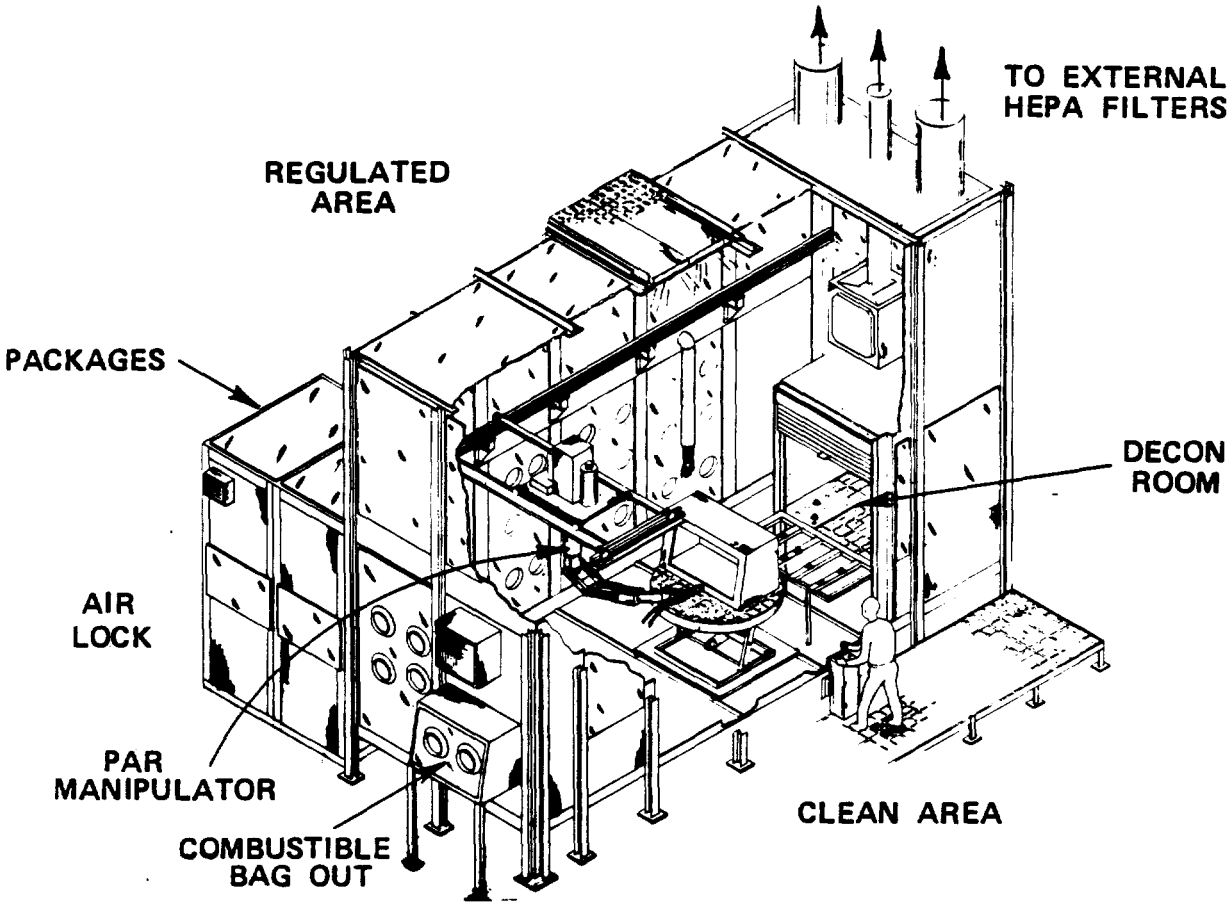

FIGURE 2. Alpha D and D Pilot Facility.

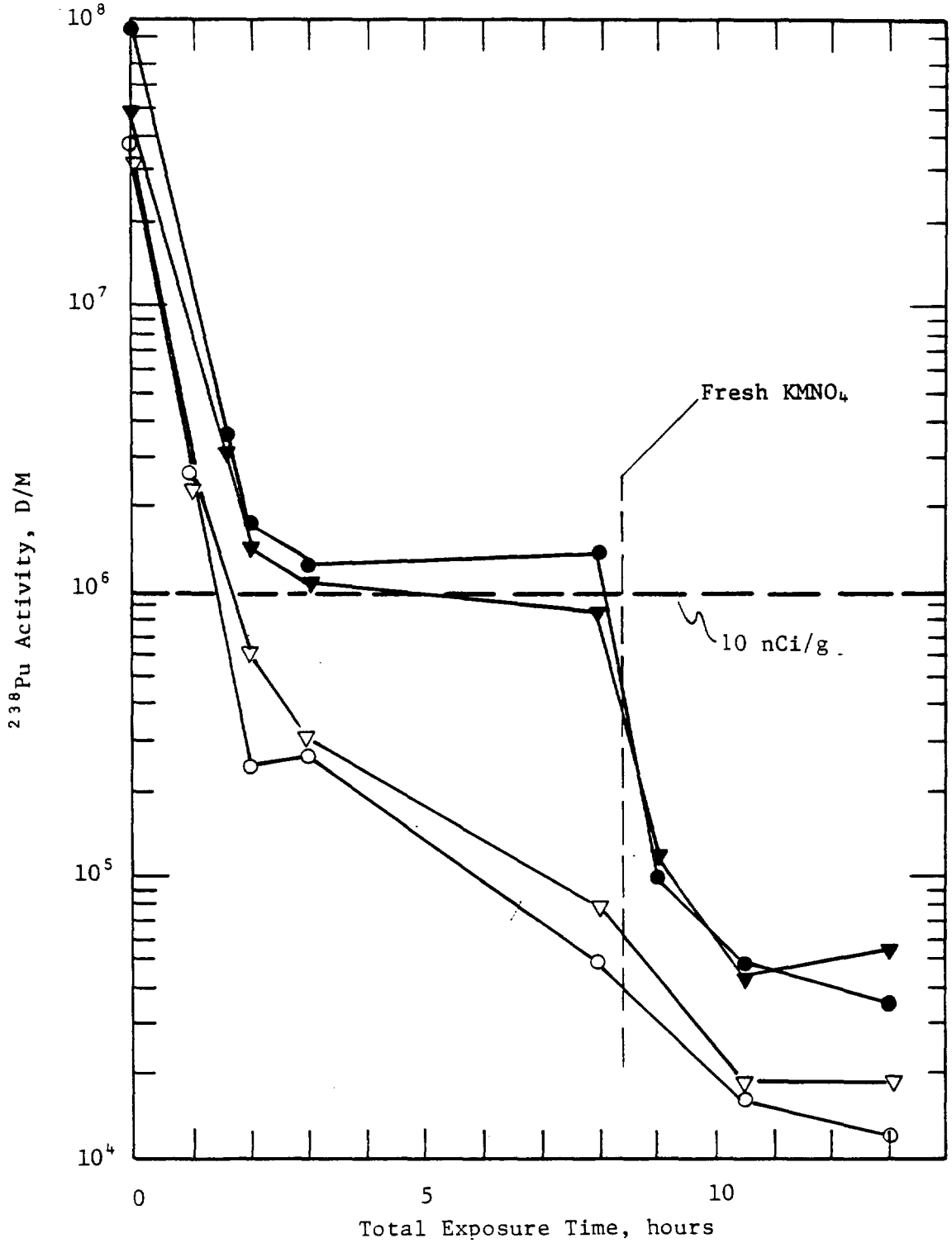

FIGURE 3. Decontamination of AMF Coupons Treated Batchwise in Acid $KMnO_4$ at 60°C.*

* Data points are at times when coupons were removed from solution, cleaned with hot oxalic acid and assayed by LEP counting.