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DECONTAMINATION OF
PLUTONIUM-CONTAMINATED GLOVEBOXES

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ABSTRACT

In connection with the Argonne National Laboratory efforts to reduce potential hazards in the event a plutonium use facility is hit by a tornado, a decontamination experiment was carried out to establish the lowest practicable limits of loose contamination in an operating glovebox, and to determine the relative merits of solvent wiping and vacuum cleaning as methods of decontamination. The results showed that a single wiping of the heavily contaminated test glovebox with Calgon Hel-Cat, Myco Tiara, or Pennwalt 2187 solvent for a short period of time removes >95% of the loose contamination originally present, with a resultant contamination level of 10^6 - 10^7 dpm/100 cm². Subsequent wipings had little effect on removing the remaining (<5%) contamination. Vacuum cleaning was ineffective as the sole decontamination method; however, this cleaning method is recommended for removing loose plutonium in crevices and other hard-to-wipe areas. These results, although limited by the narrow scope of the experiment, offer the possibility of decreased decontamination costs for gloveboxes compared to the standard technique, which requires successive wipings until the smears are essentially clean.

I. INTRODUCTION

In January 1973, a Task Force* was appointed at Argonne National Laboratory to provide action recommendations to improve the safety and security posture of the Laboratory's plutonium** use facilities. Initial efforts of the Task Force addressed the problem of minimizing any release to the environment from the effects of tornadoes. After inspection of selected plutonium use facilities and preparation of safety-evaluation reports, a recurring recommendation, applicable on a Laboratory-wide basis, was made as follows:

"The quantity of loose contamination in operating gloveboxes should be maintained at the lowest practicable limit."

*The Task Force consisted of H. A. Flaughner, PO, and R. L. Jarry, PRO, as Staff; and G. A. Bennett, OOS, as Project Coordinator.

**The term "plutonium" includes all transuranium elements.

On April 24, 1973, G. A. Bennett, C. A. DeLorenzo, R. L. Jarry, L. R. Kelman, G. T. Lonergan, and W. J. Tyrrell met to develop an approach for accomplishing this objective. At this meeting, the suggestion was made that vacuum cleaning might be a suitable technique for removing loose, finely divided contamination more rapidly and at less cost than conventional solvent cleaning. In consideration of this suggestion, discussion established the following:

1. Argonne has not kept records from which to estimate the resultant level of loose contamination remaining in a glovebox after either vacuuming or solvent cleaning.
2. The only vacuuming experience of Plant Operations (Reclamation) on gloveboxes at Argonne goes back at least 15 years. At that time, inexpensive, disposable vacuum cleaners were used.
3. The design, construction materials, and use of a glovebox could vastly change its capacity for decontamination. The large amount of equipment contained in the gloveboxes in the plutonium Fuel Fabrication Facility in Building 350 makes them particularly inaccessible for decontamination. The level of contamination in these boxes is estimated at 10^9 - 10^{12} dpm/100 cm².
4. A vacuum cleaner for use with alpha-contaminated gloveboxes should be designed so that only a hose will become contaminated. The vacuum cleaner should have a small nozzle for reaching into crevices.
5. Fixed contamination does not necessarily stay fixed, but, through oxidation of the surface layer containing the contamination, is released as a surface "smut."

It was decided that an experiment should be undertaken to estimate the "practicable" degree of decontamination possible in a "typical" glovebox by vacuum cleaning and by solvent decontamination. This report describes this experiment.

II. EXPERIENCE WITH VACUUM CLEANING AT OTHER LABORATORIES

Before the Argonne experiment was devised, a survey was made of the experience of other laboratories in the use of vacuum cleaners for removal of alpha contamination.

Rocky Flats is using modified Hoover canister machines with an absolute filter of their own design attached to the exhaust for plutonium-contamination removal.

ORNL is using vacuum cleaners with ²³³U gloveboxes only where water is not permitted.

LASL formerly used modified home-type vacuum cleaners with a HEPA filter attached to the exhaust for plutonium-contamination removal. A vacuum of their own design, which employs a cyclone separator with the exhaust passing through a HEPA Model 50 filter, is now used for cleaning surfaces contaminated with uranium and graphite dust. This unit is made in their own shops. Most decontamination is currently done by solvent wiping.

New Brunswick Laboratory formerly used a specially designed Lucite filtering chamber packed with fiber glass wool for its plutonium wet-chemistry gloveboxes. This unit was connected into the glovebox vacuum-pump system. The laboratory also used a Rex-Air vacuum cleaner without additional filtering.

In addition to the above information, the Chemical Engineering Division at Argonne used a vacuum cleaner in October 1972 for reducing the amount of spread alpha contamination. Overhead ductwork and other horizontal surfaces were vacuumed prior to spray cleaning with Calgon Hel-Cat. The resultant reduction in activity levels due to vacuum cleaning ranged from almost NCD (no contamination detectable) to slight improvement.

Summarizing this experience, vacuuming is effective for removing large-particulate contamination; however, it is not effective for glovebox decontamination.

III. EXPERIMENTAL PROCEDURE

An aluminum glovebox in which dry-fuel fabrication operations on plutonium-containing fuels had been performed for a number of years was selected for the decontamination experiment. This glovebox was considered typical and offered available horizontal surfaces that were not overly crowded with equipment.

The floor of the glovebox was brushed with a paint brush to (1) reproduce the conditions after a typical cleanup and (2) remove as much loose activity as possible so that initial readings would be within the limits of the instrument used.

Seven 9 x 12-in. areas were taped to provide standard areas in which to work, and to delineate one area from another. One area was for each of six solutions to be tested, and one for vacuuming.

Six solutions, with a maximum total volume of <500 ml, the maximum allowable under the Criticality Hazards Control Statement for a single glovebox, were introduced into the box. The solutions selected were the following:

1. Pennwalt 2187. Acidic (HF) liquid, $\text{pH} < 1$; used as an aluminum cleaner and deoxidizer.
2. Myco Tiara. Low-sudsing detergent; $\text{pH} = 11.1$ (concentrate), used in automatic scrubbing machines.
3. Calgon Hel-Cat. Basic liquid, $\text{pH} = 11.2$ (1% solution); general-purpose cleaner containing detergents, buffering salts, and water softeners.
4. Acetone.
5. Ethyl alcohol.
6. Sodium citrate (0.05M).

A household-tank-type vacuum cleaner (Electro-Lux), fitted with a HEPA filter on the exhaust, was also introduced into the system.

A proportional alpha counter (PAC-3G) was used for all measurements. The instrument was checked for operability, new batteries were installed, and the instrument was calibrated for 10% efficiency. The probe was masked so that the active area was reduced from 61 to 0.555 cm^2 . The 10% probe efficiency made readings of 10^6 dpm/61 cm^2 possible, and the reduced probe area allowed the readings to be equated to a maximum of 1.75×10^8 dpm/100 cm^2 for surface-area or smear measurements. The cable was sealed through a pouch placed on a gloveport so that the electronic portion of the instrument was external, the probe being internal to the glovebox.

The method used for the experiment was to measure the initial activity in a 4 x 4-in. area, smear the area, read the smear and the remaining activity directly, decontaminate for 2 min, and repeat. Nine readings were taken for each area, and the average was used in the tabulation.

After the third decontamination with the first solution, a background on the PAC-3G (assumed to be contamination of the probe) was noted. Although it was felt at that time that further effort would have yielded better results, each area was limited to three decontaminations for each solution in the hope that the entire experiment could be conducted using the same instrument for as consistent results as possible. The assumptions were made that the glovebox floor had uniform activity within each 4 x 4-in. area and that the instrument scales were linear.

IV. RESULTS

The results are summarized in Table I, which indicates the percent of the total initial counts removed after the initial, second, and third wipings or vacuum cleaning. This table shows:

1. By far the greatest decontamination occurs during the initial wiping.
2. Hel-Cat, Myco Tiara, and Pennwalt 2187 are all effective for removing the specific material deposited in the test glovebox. Each of these solvents removed >95% of the loose contamination with a single wiping. Within the accuracy of the data, successive wipings after the first accomplished little, if any, further decontamination.
3. The other solvents--0.05M sodium citrate, ethyl alcohol, and acetone--and vacuum cleaning are ineffective in decontamination.
4. The resultant contamination level, after a single wiping with the effective solvents, was 10^6 - 10^7 dpm/100 cm².

TABLE 1. Cumulative Percent Loose Contamination Removed after Each Successive Wipe

Solvent:	Calgon Hel-Cat		Myco Tiara		Pennwalt 2187		Sodium Citrate (0.05M)		Ethyl Alcohol		Acetone		Vacuum ^b	
Percent Removed after First Wipe ^a :	98 Based on		97 Based on		>96 Based on		>76 Based on		67 Based on		>59 Based on		≥0 Based on	
	Direct Read- ings	Smear Read- ings	Direct Read- ings	Smear Read- ings	Direct Read- ings	Smear Read- ings	Direct Read- ings	Smear Read- ings	Direct Read- ings	Smear Read- ings	Direct Read- ings	Smear Read- ings	Direct Read- ings	Smear Read- ings
Cumulative Percent Removed after Second Wipe	97.4	98.5	94	98.2	>95	>96	>79	>81	78	80	>66	>59	≥0	≥41
Cumulative Percent Removed after Third Wipe	97.6	98.7	79	98.4	>97	>98	>87	>89	79	90	>67	>61	≥0	≥30

^aBased on direct readings.^bVacuums substituted for wipings.

V. SUPPLEMENTARY EXPERIMENT

A short supplementary experiment was carried out to determine the rate at which "fixed" contamination migrates to the surface of the test glovebox. Immediately after the experiment, one test area was recleaned, the contamination level was measured, and the area was covered with plastic. The same area was measured two days later and four months later. The results are as follows:

Initial reading: 1.9×10^5 dpm/100 cm² (smear).

Reading after two days: 1.9×10^5 dpm/100 cm² (i.e., no measurable change).

Reading after four months: 6.8×10^5 dpm/100 cm².

These data indicate a very slow release of fixed activity to the surface of the aluminum glovebox.

Contamination on top of the plastic, which had been kept in the glovebox during the experiment, increased in the first two-day period from 5.4×10^5 to 2.3×10^6 dpm/100 cm². This increase can be attributed to turbulence due to the glovebox ventilation system and the presence of suspended particulates from the vacuuming operation.

VI. DISCUSSION

The major amount of decontamination is accomplished during the first wipedown. This result suggests that, depending upon the purpose of decontamination, considerable savings in time and costs could be realized by the use of a modified technique, which consists of a single wiping, as compared to the conventional technique, which continues wiping until the smears become essentially clean.

Based upon these results, a satisfactory decontamination procedure for gloveboxes contaminated with solid plutonium or its compounds would be as follows:

1. Brush loose material from smooth surfaces, and, where deemed appropriate, remove loose contamination from crevices by vacuuming.
2. Decontaminate only once, with an effective solvent such as Calgon Hel-Cat, Myco Tiara, or Pennwalt 2187.
3. Spray the resulting surface with a film (such as Turco Contam-Affix) to aid fixation of the remaining material. (The data indicate that this step could be omitted with little deleterious effect.)

These conclusions are based on the results for a limited experiment using a single heavily contaminated aluminum glovebox in which dry-plutonium operations had been carried out. However, the "loose" contamination should be removed more easily from either a stainless steel glovebox or one in which wet-chemistry operations had been carried out than from the test glovebox. This is because, due to the difference in oxidation rates, the release of "fixed" activity to the surface of a stainless steel box should be even slower than the slow release found in the test glovebox. Also, depending upon the nature of the glovebox surface, the contamination occurring in a glovebox in which wet-chemistry operations had been carried out should be "fixed" more firmly than the contamination in the test glovebox. Consequently, with respect to tornadoes, a stainless steel glovebox, or one in which wet-chemistry operations had been carried out, should pose less of a problem than the test glovebox.

An attempt was made to estimate whether the contamination level of 10^6 - 10^7 dpm/100 cm², which results after a single wiping, could be considered "safe," i.e., whether this level of loose contamination is sufficiently low so that, in case of a direct tornado hit, the resultant release of contamination would not exceed the specified dose guidelines* to an individual at the Laboratory's boundaries. The problem in making such an estimate is that there is presently no valid model for transport of particulate material in a tornado. Since the single limiting dose of small particles to the critical organ,

*Dose guidelines contained in "Minimum Design Criteria for New Plutonium Facilities," August 8, 1973 (intended to be incorporated into AECM Appendix 6301 as Part II.H).

the lung, is so extremely small,* conservative practice dictates that, until such a model is available, precautions be taken in the storage of gloveboxes decontaminated by this technique.

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*Harry Howe (OHS) has shown that the amount of material contained in one 40 x 40 x 40-in. glovebox module at a ^{239}Pu contamination level of 10^6 dpm/100 cm² would constitute 1250 limiting dosages of 10- μ particles or ~4000 limiting dosages of 1- μ particles.

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