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Abstract

A highly contaminated glove-box at LLNL containing plutonium was decontaminated using a strippable decontamination gel. 6 x 12 inch quadrants were mapped out on each of the surfaces. The gel was applied to various surfaces inside the glove-box and was allowed to cure. The radioactivity in each quadrant was measured using a LLNL Blue Alpha meter with a 1.5 inch standoff distance. The results showed decontamination factors of 130 and 210 on cast steel and Lexan® surfaces respectively after several applications. The gel also absorbed more than 91% of the radiation emitted from the surfaces during gel curing. The removed strippable film was analyzed by neutron multiplicity counting and gamma spectroscopy, yielding relative mass information and radioisotopic composition respectively.

Introduction

A glove-box commissioned in 1964 has been used at LLNL to cold roll plutonium metal. Historical information relating to the isotopic contents glove-box identifies weapons-grade plutonium (WG-Pu, see Table 1), followed by Pu-238. Mechanical and abrasive deposition of plutonium on some areas of the floor had occurred during operation. In 1994 programmatic operations within the box were no longer needed and the box was used to store samples of Plutonium-238. In 1996 a spill of Pu-238 occurred in the box resulting in a significant contamination of the interior of the glove-box. The Pu-238 contamination of the glove-box created significantly higher levels of activity and made decontamination much more difficult. Another commercially available strippable coating was used to stabilize the Pu-238 spill. The glove-box measures 54 inches wide x 93 inches long x 109 inches high. It is, constructed of a cast steel floor, aluminum walls, Lexan® windows and Hypalon® gloves and required decontamination. Previous unsuccessful decontamination efforts involved a commercially available strippable film coating. The goal of the decontamination was to reduce contamination levels to a point where the glove-box could be disposed of as low-level radioactive waste. Typically, a glove-box containing such activities and contamination might require 3 or 4 workers for 1 month to decontaminate using sandpaper and current commercially available strippable coatings.

Figure 1. Exterior and interior of glove-box. Note rolling mill inside glove-box was removed prior to decontamination and the floor of the glove-box was swept.

Cellular Bioengineering Inc (CBI) has developed a decontamination gel (Decon Gel 1101) that when cured allows efficient removal of contamination from surfaces in a strippable film that can be easily disposed. Decon Gel 1101 has been applied to a Pu-contaminated glove-box to determine its efficiency in removing contamination from several surfaces in a unique and highly contaminated environment.

Table 1. Typical isotopic composition of WG-Pu.

Isotope	Weight (%)	Weight Fraction	SA (Ci/g)	Mix SA (Ci/g)	Activity Fraction	Activity %
Pu-238	1.60E-02	1.60E-04	1.73E+01	2.77E-03	5.79E-03	5.79E-01
Pu-239	9.35E+01	9.35E-01	6.30E-02	5.89E-02	1.23E-01	1.23E+01
Pu-240	5.90E+00	5.90E-02	2.30E-01	1.36E-02	2.84E-02	2.84E+00
Pu-241	3.81E-01	3.81E-03	1.04E+02	3.96E-01	8.28E-01	8.28E+01
Pu-242	4.00E-02	4.00E-04	4.00E-03	1.60E-06	3.34E-06	3.34E-04
Am-241	2.01E-01	2.01E-03	3.47E+00	6.97E-03	1.46E-02	1.46E+00
Total	**1.00E+02**	**1.00E+00**	**1.25E+02**	**4.78E-01**	**1.00E+00**	**1.00E+02**
w/o Pu-241	*9.97E+01*	*9.97E-01*	*2.11E+01*	*8.22E-02*	*1.72E-01*	*1.72E+01*

Taken from: Device Assembly Facility (DAF) Glove-box Radioactive Waste Characterization, J. L. Domminick. LLNL Report. UCRL-ID-146615. 12/18/01.

While difficult to quantify the weight percent of Pu-238 present in the box is believed to be well in excess of the normal weapons grade distribution due to a spill of Pu-238 in 1996.

Experimental Method

The contaminated surfaces to were mapped out in 6 x 12 inch quadrants and the original contamination levels were determined before decontamination using a Blue Alpha air proportional meter (model LEA751854018 Serial# 3098163), designed and manufactured at LLNL. The meter was calibrated before use and was set to 50% efficiency. The minimum detectable activity (MDA) of the instrument is approximately 50 counts per minute (cpm), equivalent to 100 disintegrations per minute (dpm) and 45 pico-Curies (pCi). The meter has a maximum detectable activity of 1 million cpm. To avoid saturation of the meter readings and to provide a consistent standoff distance, 1.5-inch legs were added to the bottom of the detector plate (see Figure 2). Measurements taken at less than a 1 inch stand off indicated contamination levels well in excess of 1,000,000 cpm.

Figure 2. LLNL Blue Alpha meter.

CBI's Decon Gel 1101 was prepared and applied with a trowel according to the manufacturer's recommendations (Figures 3 and 4). The gel was allowed to cure overnight and through-gel contamination readings were made to investigate the shielding effects of the cured gel. The cured gel was then removed from the surfaces (Figures 5 and 6), and surface readings again were made with the LLNL Blue Alpha meter. For the Lexan® window, this process was repeated once more, and twice more for the cast steel floor and aluminum siding. The decontamination was not repeated for the Hypalon® gloves.

Decontamination Factors (DFs) were calculated as a ratio of original measured alpha activity at a 1.5-inch stand-off distance to the measured activity after using Decon Gel 1101 at a 1.5-inch stand-off distance, averaged the measurements taken for each type of surface studied. These values are relative only to other measurements taken at the given stand-off distance and are not equal to the actual activity on the surface.

Figure 3. Glove-box floor (steel and aluminum) before and after application of Decon Gel 1101.

Figure 4. Glove-box window (Lexan®) and gloves (Hypalon®) during application of Decon Gel 1101.

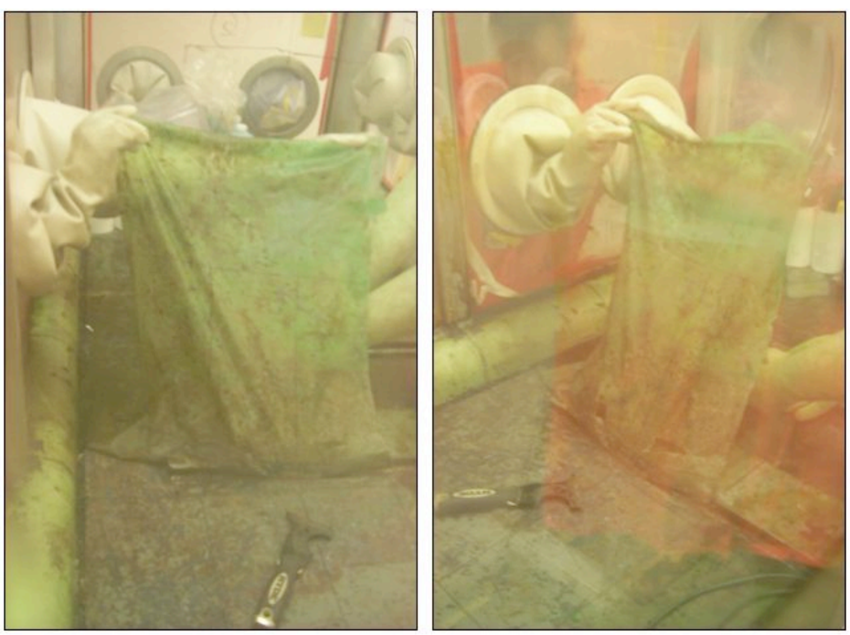

Figure 5. Removal of cured Decon Gel 1101 as a strippable film from cast steel glove-box floor.

Figure 6. Removal of cured Decon Gel 1101 as a strippable film from Lexan® glove-box windows.

Results

Contamination readings measured using an LLNL Blue Alpha meter at a 1.5-inch standoff distance before decontamination, after application and after removal are detailed in Table 2. Initial contamination levels on the glove-box floor were on average 37,000 cpm (74,000 dpm, 33 nCi, SD=15%) for the cast steel horizontal floor and 28,000 cpm (56,000 dpm, 25 nCi, SD=7%) for the aluminum siding. Initial contamination levels on the Lexan window were on average 27,000 cpm (54,000 dpm, 24 nCi, SD=4%) and 56,000 cpm (112,000 dpm, 50 nCi, SD=32%) on the Hypalon gloves.

After the gel had cured, the film barrier provided on average above 91% shielding from the measured radiation over all surfaces studied. This is not surprising given that the majority of the radioactivity is alpha radiation and the measurements are made using an air proportional alpha counter. However, the ability of the gel to form an impermeable film provides extra protection to the worker from re-suspension and extremity dose.

After one application and removal of Decon Gel 1101, the activity measured on the floor was reduced by 57% (SD=7%) and on the Lexan window by 37% (SD=8%). The Lexan window was subject to a second application and removal of Decon Gel 1101, resulting in an overall observed 99.5% (SD=0.1%) removal of all measured radioactivity. Similarly, the glove-box floor was subject to a second and third application and removal of Decon Gel 1101, resulting in an overall 99.4% (SD=0.3%) removal of all measured radioactivity. After two and three applications of Decon Gel 1101, measured radioactivity was reduced to less than 200 cpm (400 dpm, 0.18 nCi) in almost all cases. Given the highly contaminated nature of the surfaces within the glove-box, this decontamination efficiency, given the high contaminated environmental, is considered excellent. Relative, averaged DF values are shown below each surface in Table 2.

Table 2. Measured Radioactivity Levels for Each Quadrant.

Aluminum Wall	**Measured Radioactivity, cpm**						
Location	*Initial*	*Thru Gel*	*Shielding*	*1st Decon*	*Initial Efficiency*	*Last Decon*	*Total Efficiency*
	cpm	cpm	%	cpm	%	cpm	%
A2	27000	3000	89	NA	NA	120	100
A3	28000	3000	89	NA	NA	120	100
B1	28000	3000	89	NA	NA	140	100
B5	28000	3000	89	NA	NA	100	100
C1	28000	4000	86	NA	NA	160	99
C5	26000	3000	88	NA	NA	200	99
D1	28000	2000	93	NA	NA	220	99
D5	26000	4000	85	NA	NA	220	99
E1	30000	3000	90	NA	NA	240	99
E5	28000	3000	89	NA	NA	160	99
F1	32000	4000	88	NA	NA	NA	NA
F5	32000	3000	91	NA	NA	140	100
Average	*28417*	*3167*	*89*	*NA*	*NA*	*165*	*99*
2 Sig Fig Ave	*28000*	*3200*	*89*	*NA*	*NA*	*170*	*99*
SD	*1975*	*577*	*2*	*NA*	*NA*	*47*	*0.2*
RSD	*7*	*18*	*2*	*NA*	*NA*	*29*	*0.2*

Cast Steel	**Measured Radioactivity, cpm**						
Location	*Initial*	*Thru Gel*	*Shielding*	*1st Decon*	*Initial Efficiency*	*Last Decon*	*Total Efficiency*
	cpm	cpm	%	cpm	%	cpm	%
B2	34000	4000	88	18000	47	200	99
B3	34000	3000	91	18000	47	220	99
B4	38000	2000	95	15000	61	140	100
C2	50000	3000	94	16000	68	520	99
C3	42000	1000	98	14000	67	320	99
C4	34000	2000	94	16000	53	180	99
D2	42000	1000	98	16000	62	700	98
D3	32000	3000	91	15000	53	180	99
D4	30000	2000	93	15000	50	140	100
E2	40000	3000	93	15000	63	400	99
E3	32000	3000	91	14000	56	140	100
E4	38000	2000	95	16000	58	160	100
Average	*37167*	*2417*	*93*	*15667*	*57*	*275*	*99*
2 Sig Fig Ave	*37000*	*2400*	*93*	*16000*	*57*	*280*	*99*
SD	*5686*	*900*	*3*	*1303*	*7*	*179*	*0.4*
RSD, %	*15*	*37*	*3*	*8*	*13*	*65*	*0.4*
Average Decontamination Factor (DF): 2 after first application, 57 after second and third application combined, 130 total including all three applications.							

Continued - Table 2. Measured Radioactivity Levels for Each Quadrant.

Lexan Window	**Measured Radioactivity, cpm**						
Location	*Initial*	*Thru Gel*	*Shielding*	*1st Decon*	*Initial Efficiency*	*Last Decon*	*Total Efficiency*
	cpm	cpm	%	cpm	%	cpm	%
WA3	27000	3000	89	20000	26	100	100
WA4	27000	2000	93	17000	37	120	100
WA5	24000	2000	92	17000	29	140	99
WB1	26000	1000	96	17000	35	110	100
WB2	28000	3000	89	18000	36	120	100
WB5	27000	2000	93	19000	30	120	100
WC1	26000	1000	96	14000	46	180	99
WC2	28000	1000	96	13000	54	120	100
WC5	28000	3000	89	20000	29	140	100
WD1	28000	1000	96	18000	36	180	99
WD2	27000	1000	96	16000	41	120	100
WD5	26000	3000	88	15000	42	120	100
Average	*26833*	*1917*	*93*	*17000*	*37*	*131*	*100*
2 Sig Fig Ave	*27000*	*1900*	*93*	*17000*	*37*	*130*	*100*
SD	*1193*	*900*	*3*	*2216*	*8*	*25*	*0.1*
RSD, %	*4*	*47*	*4*	*13*	*22*	*19*	*0.1*
Average Decontamination Factor (DF): 2 after first application, 130 after second application, 210 total after two applications.							

Hypalon Gloves	**Measured Radioactivity, cpm**						
Location	*Initial*	*Thru Gel*	*Shielding*	*1st Decon*	*Initial Efficiency*	*Last Decon*	*Total Efficiency*
	cpm	cpm	%	cpm	%	cpm	%
GA2	40000	3000	93	NA	NA	NA	NA
GB3	45000	3000	93	NA	NA	NA	NA
GC3	60000	3000	95	NA	NA	NA	NA
GD3	80000	4000	95	NA	NA	NA	NA
Average	*56250*	*3250*	*94*	*NA*	*NA*	*NA*	*NA*
2 Sig Fig Ave	*56000*	*3300*	*94*	*NA*	*NA*	*NA*	*NA*
SD	*17970*	*500*	*1*	*NA*	*NA*	*NA*	*NA*
RSD, %	*32*	*15*	*1*	*NA*	*NA*	*NA*	*NA*

Notes and caveats: Decontamination efficiencies and factors are all calculated for a 1.5-inch stand-off distance, i.e. relative to each other, not to the actual activity at the surface. Only alpha particle measurements were included. Since measurements were only recorded to 2 significant figures, the calculated average activity at each location is corrected to 2 significant figures. Therefore, DF values are reported to 2 significant figures. Significant contamination remains on the glove-box based on measurements at closer distances. Decontamination of the Hypalon® gloves was stopped after the first application. Significant difficulty in removing Decon Gel 1101 was encountered after the first application.

Neutron Multiplicity Measurements

During the removal of the Decon Gel after the first application, the peeled gel was placed into cans and assayed by neutron multiplicity counting and gamma spectroscopy. Quantification was based on neutron measurements assuming a weapons grade plutonium distribution. The results are shown in Table 3. Actual isotopic composition was measured by gamma spectroscopy and shows 99.8 and 152 keV peaks from Pu-238, a 129 keV peak from Pu-239, a 59 keV peak from Am-241 (daughter of Pu-241). The relative height of the 152 and 129 keV peaks indicates the Pu-238 content is higher than in normal weapons-grade plutonium, which is consistent with the historical activities in the glove-box (a Pu-238 spill in 1996). If a normal weapons grade isotopic distribution is used to analyze the neutron flux, Pu-239 will be overestimated because a lot of the neutrons are actually from spontaneous fission of Pu-238.

Table 3. Estimated Mass of Pu Removed by Initial Use of Decon Gel 1101.

Surface	**Quantity**
Floor	0.067 ± 0.046 grams
Extruded Aluminum Frame	0.177 ± 0.038 grams
Lexan® Windows	0.034 ± 0.021 grams
Hypalon® Gloves	0.002 ± 0.022 grams

Note: It is believed that the extruded aluminum frame contained higher quantities of plutonium because of a gasket area that may have entrained radioactive material.

Operational Perspectives

The Nuclear Materials Processing and Technology Program personnel conducting the decontamination activities reported that the material had a good workability and allowed sufficient working time before drying. In general the material adhered to the sides of the glove-box without a excessive amount of dripping being observed. The Decon Gel 1101 was able to penetrate crevices and was easily removed from all surfaces with the exception of the Hypalon® gloves. While a hand application method was used for the study personnel commented that spray application might be advantageous for future applications.

Several questions were raised by CBI regarding the documented use and potential future applications of Decon Gel 1101.

Q1. Can you please list positive and negative traits of Decon Gel (DG)? Consider both ergonomics and efficacy.
A1. Positive: DG appeared to penetrate tight and difficult to reach places, and remove more contamination per application; it met all ES&H requirements for use in a nuclear facility; it was applied well and set in a reasonable time. Negative: DG could use a stronger color indicator so as to identify an edge more easily and begin peeling. However, using tape as an "edge-starter" was not implemented.

Q2. How did your experience of DG compare to other strippable coatings and gels? Consider both ergonomics and efficacy, be as descriptive as possible.
A2. DG appeared to remove more substantial contamination (as witnessed by the lower meter readings and the activity contained in the removed layer. However, in a difficult working environment such as a glove-box, the coating was difficult to remove (a factor associated with constricted access). CBI questioned whether the workers had applied a thick enough coat, or perhaps another issue such just the normal clumsiness of working

through double gloves etc. Working in a glovebox is always difficult. The Decon Gel adhered so well to surfaces that extra care needed to factored into determining how best to start a peel. We believe after working with the material for awhile the workers would have grown accustom to its characteristics and been able to overcome any removal difficulties.

Q3. Are there other jobs coming up that you would consider using DG on? If so, can you describe?
A3. During this fiscal year it appears there is not any additional field applications. However, during next fiscal year, it is expected that ventilation ducts will require decontamination. These materials will present unusual geometries and difficult to reach locations. Additionally, we believe DG may be used as a fixative in the event of a spill. In such a case, it would be useful to have DG continually on-hand. DG would also be useful in tasks requiring size reduction of contaminated objects, preventing or reducing dispersal or resuspension of radioactivity. In this case, research would be needed to determine if DG could be saw-cut without degrading, or without compromising the saw operation. There may also be opportunities to use DG in future glove-box and equipment decontamination during the next fiscal year.

Q4. Also if so, are any of those jobs amenable to a field documentation study such as you just did?
A4. Decontamination of additional glove-boxes, equipment and ventilation ductwork would be amenable to field studies, as would size-reduction work.

Q5. Would you recommend purchasing the product for the LLNL system, and if so, what is approval process and/or who would be contact?
A5. Yes, we would recommend purchasing the product for LLNL use. DG is already approved for use in the nuclear facility. Pricing information would be required for cost-effectiveness review. LLNL would also like more information on the shelf-life of DG in regard to the amounts that might be ordered. The workers watched the instructional video before the application, which was extremely helpful. I think the one tip and/or trick that could be emphasized would be the starting of a peel. Even though the film emphasized the tape method our workers choose not to use it. It turned out that it might have been the best way to go in certain areas. Nooks and crannies were another area that was difficult to get large sheets started. Perhaps including a glovebox application in the video would be helpful. CBI guarantees the shelf-life of DG for 1 year and a price list was provided.

Q6. What do you see as the widest use-potential for DG? What would be 3 top job types you might see within DOE (e.g. glove box decon)?
A6. Immediate use would be for decontamination of equipment and glove-boxes. In the future, use in spill response, fixative and pre-use protective coating applications might provide the widest use potential. The latter uses would require study or information relating to self-life, applied-life (can it be left to age for x months or y years?), criticality safety (i.e. neutron absorbing or reflecting properties), and degradation over time due to radiation or environmental factors.

Q7. You had previously said that normal decon of such a box would take 3-4 men a month to do using sandpaper etc. Can you please comment on how much ADDITIONAL work needs to be done to the box you decon'ed to get it down to the same level as you would observe with 3-4 men with sandpaper? We'd like to use this information to answer how much time and effort was saved by using DG.
A7. This is a difficult question to answer because there were areas of the glove-box that had previously been treated with another strippable coating. This coating was used as a fixative and was in place for 11 years. Removal of the coating took considerable effort. However, in the opinion of the Subject Matter Expert (SME) assigned to this operation, DG was more effective in removing contamination than other strippable coatings. If not for the problems associated with removing old strippable coating, DG would have contributed to a timely decontamination of the system.

Recommendations for Future Work

It is recommended that the future studies focus on several aspects of understanding the application, behavior and applied-life of Decon Gel 1101. This work should be performed using more controlled laboratory testing (in areas less contaminated with less background radiation). Specifically, we recommend studies that provide information regarding the following:

- Criticality effects – does DG absorb or reflect neutrons, and how would this impact the use in neutron flux environments;
- Radiation damage – does DG suffer radiation damage, including physical and chemical breakdown if exposed to alpha or beta radiation for extended periods of time if used as a fixative;
- Environmental damage – similarly, does DG suffer damage through exposure to heat, water, humidity or sunlight for long periods of time if used as a fixative;
- Shelf-life information – can DG be stored for years at a time and kept on hand for spill response;
- Cutting effects – does DG allow saw-cutting without film breakdown or compromising sawing operations;

CBI also reports previous studies have identified differing DFs for steel and Plexiglas when actinides are deposited specifically in nitric or hydrochloric acids. Under more controlled laboratory conditions, these observations can be studied more carefully. The use of nitric or hydrochloric acid will not only impact the speciation of the radionuclide, but will also affect the migration of the radionuclide into the surface, and the physical and chemical characteristics of the surface. Specifically, we recommend:

- A concise review of the behavior (speciation, precipitation, sorption and colloid formation) of the actinides of interest in nitric and hydrochloric acids applied to steel, Plexiglas and concrete;
- Bench-top laboratory experiments to investigate the efficiency of DG in removing actinides from controlled test materials in a low-background environment;
- Study of the chemistry and speciation occurring at the interface between the surface, actinide and gel.

These results may also be supported by using chemical-thermodynamic modeling, yielding information on the chemical speciation of the radionuclide in a given environment, and the chemical reactions that occur between the radionuclide, the counter-ion, the surface and the gel.

This future work would allow a better understanding of the previously obtained results, and possibly provide avenues to further improve the efficiency of Decon Gel 1101 in a variety of physical and chemical matrices, and in a variety of applications.

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