RADIOLOGICAL CONTROL ASPECTS OF WIPP WASTE CHARACTERIZATION OPERATIONS AT ANL-W

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ABSTRACT

Argonne National Laboratory is characterizing and repackaging mixed hazardous contact-handled transuranic waste to be used in the Waste Isolation Pilot Plant Experimental Test Program. Currently, these activities are being accomplished in a hot cell and in an adjacent hot repair area in the Hot Fuel Examination Facility. While the wastes do not require remote handling, they contain significant amounts of transuranic nuclide. So the major radiation safety challenge is one of contamination control of alpha-emitters with little associated beta-gamma activity. The low-energy photons from ^{241}Am are of concern whenever workers are near or in contact with containers or contaminated surfaces. Two years of operational experience have demonstrated the effectiveness of our procedures in minimizing contamination and personnel radiation exposures. An extensive modification of the facility is underway to provide a dedicated area for this work; the new area is designed and equipped specifically for waste characterization and repackaging. In addition to increasing the efficiency and flexibility of examinations, it will reduce the potential for spreading contamination and exposure of personnel to ^{241}Am photons. This paper describes the radiological safety aspects of present and future operations including facility design, radiation monitoring, and personnel protection.

INTRODUCTION

Argonne National Laboratory (ANL) and EG&G-Idaho are collaborating on a project to characterize and repackage waste that is to be sent to the Waste Isolation Pilot Plant (WIPP) in New Mexico as part of the Department of Energy's Experimental Test Program. Characterization includes gas sampling from the headspace of the outer packaging container (i.e. 55-gallon drum) and inner containers (i.e. polybags), visually examining and weighing the waste contents, and categorizing the contents according to their potential for gas generation.

Evolution of gases, particularly flammable gases, are a major concern in the WIPP project. The drums contain materials such as plastics, cellulosics, metals, and ceramics that are representative of the entire CH-TRU inventory in terms of gas-generating potential. This year, we will add the capability to sample sludges with varying liquid content. Details of the facilities and operations during the first year of this project have been presented at Waste Management 92 (1).

OPERATIONS

Fifty-five gallon drums of selected mixed hazardous Contact Handled Transuranic Waste (CH-TRU) from the Radioactive Waste Management Complex (RWMC) operated by EG&G-Idaho are transported in the Transuranic Package Transporter-II (TRUPACT-II) to ANL's site at the Idaho National Engineering Laboratory. To date, all of this waste was generated at the defense-related facility known as the Rocky Flats Plant. Currently, the examinations are accomplished in the hot repair area and in an air-filled hot cell at the Hot Fuel Examination Facility (HFEF) at ANL. Drums are brought into the high bay area where they are prepared for transfer into the contaminated areas for examination. To initiate this work, individual drums are transferred into a shielded Hot Repair Area (HRA) that is designed to support hot-cell operations. Figure 1 shows the areas of HFEF presently involved in the characterization efforts. The first step of characterization is to take a gas sample through the carbon composite filter in the drum lid. Then the lid of the drum is removed and the plastic drum liner lid is cut off allowing another gas sample to be obtained from the headspace of the drum's inner plastic bag that holds the waste items.

After the gas samples have been obtained, the drum is lowered through a shielded hatch into an air-atmosphere hot cell called the Decontamination or Decon Cell. The drum is

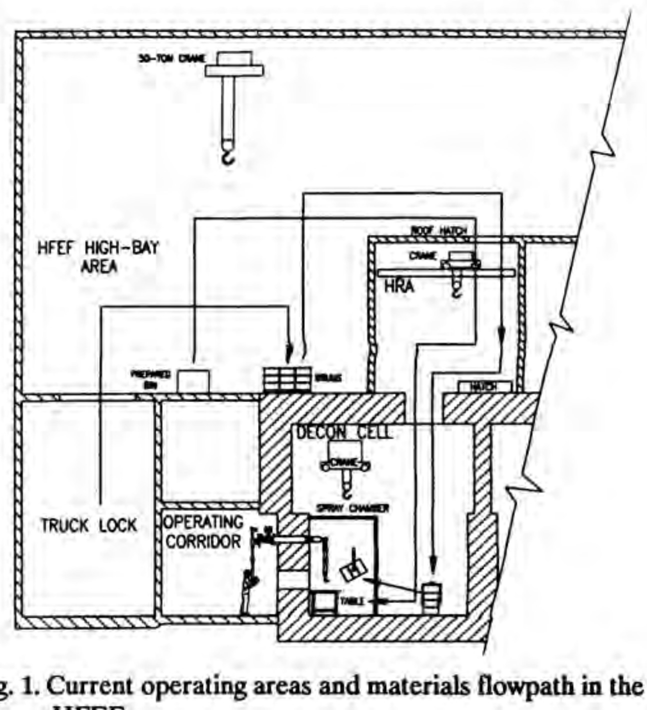

Fig. 1. Current operating areas and materials flowpath in the HFEF.

then moved into a decontamination Spray Chamber within the Decon Cell. Inside the Spray Chamber, Master-slave manipulators are used to remove the inner containers (i.e., plastic bags, fiberpaks) from the drum and to place them onto an examination table.

The steel drum and empty drum liner are sent back up to the HRA; from here, they will be monitored for contamination and returned to the RWMC. The contents of the inner containers are then weighed, gas sampled if required, inspected, and described by a voice-over narrative on a color video tape. The use of this spray chamber, which can be decontaminated, reduces the probability of contaminating the rest of the facility. The waste materials are then placed in the test bin which can hold the contents of 4 to 6 drums. The filled bins are decontaminated, if necessary, and placed in a standard waste box and shipped back to the RWMC in the TRUPACT-II. When WIPP opens, EG&G will ship the loaded bins to New Mexico where they will be used in specific performance assessment tests.

SOURCE DESCRIPTION

Previous work conducted in HFEF involved mostly spent nuclear fuel with significant beta-gamma activities from mixed fission products. Even though the gamma radiations required the use of heavily shielded containers and hot cells, the ease of beta and gamma measurement greatly simplifies contamination control. The WIPP waste characterization project introduces alpha sources with little or no associated beta-gamma activity into the facility. Typical radionuclide inventories from the Rocky Flats Plant packaged in the 55-gallon waste drums and in a filled bin are given in Table I (2). Only ^{241}Am emits photons with enough energy and intensity to be of concern as a source external to the body.

TABLE I
Average Radioactivity Level In The Waste

Nuclide	Contribution	Activity in Curies	
		One Drum	One Bin
Pu-238	10.6%	0.257	1.540
Pu-239	5.7%	0.140	0.840
Pu-240	1.4%	0.034	0.206
Pu-241	44.3%	1.080	6.480
Am-241	38.0%	0.926	5.560
Totals	100.0%	2.437	14.626

In addition to radiological concerns, personnel must be protected from the nonradioactive hazardous materials that may be present in the waste content of the drums. Table II lists the organic compounds and inorganics potentially present in drums of waste (3). Techniques that prevent contact with or inhalation of radionuclides are also effective against toxic and carcinogenic materials. Since these materials are mixed with the radionuclides, chemical contamination can be located readily by radiation surveys.

TABLE II
Hazardous Materials Potentially Present in Waste to be Characterized

Organic Compounds	
Acetone	
Benzene	
Bromoform	
1-Butanol	
1-Butanone	
Carbontetrachloride	
Chlorobenzene	
Chloroform	
Cyclohexane	
1,1-Dichloroethane	
1,2-Dichloroethane	
1,1-Dichloroethene	
cis-1,2-Dichloroethene	
Ethylbenzene	
Ethylether	
Methanol	
4-Methyl-2-pentanone	
Methylenechlorine	
1,1,2,2-Tetrachloroethane	
Tetrachloroethene	
Toluene	
1,1,1-Trichloroethane	
Trichloroethene	
1,1,2-Trichloro-1,2,2-Trifluoroethane	
1,3,5-Trimethylbenzene	
1,2,4-Trimethylbenzene	
m-Xylene	
o-Xylene	
p-Xylene	
Inorganic Compounds	**Toxic Materials**
Argon	Lead
Carbon Monoxide	Cadmium
Carbon Dioxide	Mercury
Hydrogen	Beryllium
Nitrogen	Lithium
Oxygen	Asbestos
Methane	
Ethane	
Propane	

RADIOLOGICAL CONTROLS

The HFEF is uniquely suited to safely handle the materials involved in the waste characterization project. It is built and equipped as an examination facility for irradiated objects and is equipped with ancillary support areas designed for contamination control and to minimize radiation exposure (4,5). The HRA is located on the third floor or high bay of HFEF above the Decon Cell as shown in Fig. 1.

Air is drawn from the high bay into the HRA; it then passes around a shielded hatch into the Decon Cell. Air leaving this cell passes through a single stage High Efficiency Particulate-Air (HEPA) filter and is exhausted to the atmosphere through the building double HEPA filtration train. No increases in gross alpha or beta-gamma particulate activity have been detected by the stack monitoring system in over the two years of waste characterization work performed at HFEF.

The only operation that requires possible contact with unsealed waste packages is conducted in the HRA. Specifically, it is the puncturing of the drum's inner plastic bag to obtain a headspace gas sample. After this procedure has been accomplished, the drums are lowered through the hatch and moved into the Decon Cell Spray Chamber. Since no unpackaged waste (i.e., waste removed from the inner containers packaged in the drums) is handled in the HRA, the probability of contaminating the facility is minimized. Radiological smear tests of surfaces in the vicinity where work on the WIPP waste has been conducted have demonstrated the success of this handling sequence; alpha contamination levels in the HRA have averaged less then 100 dpm per 100 cm^2 so far. All personnel leaving the high bay areas dedicated to this project must pass through portal monitors that scan the entire body surface and are sensitive to both alpha and beta-gamma emitters.

Personnel working in the HRA are dressed in three layers of anti-contamination clothing which consist of green scrub underalls, yellow cloth anti-Cs, and outer TYVEK coveralls which are pressurized and supplied with breathing air through a hose. This method of respiratory protection provides body cooling, positive pressure protection and is much more comfortable than using full-face respirator masks. Figure 2 shows workers surveying an open drum for alpha contamination prior to obtaining the gas sample from the drum's inner plastic bag containing the waste.

Only the photons from ^{241}Am represents an external radiation exposure hazard. The actual ^{241}Am content of a given drum depends on the initial content of ^{241}Pu and the elapsed time since the plutonium was chemically separated from other elements. As this time increases, the ^{241}Am activity increases because its half-life (432.2 years) is longer than that of its parent, ^{241}Pu (14.4 years). The decay scheme of ^{241}Pu down to the long lived (2,140,000 years) ^{237}Np is shown below.

$$^{241}\text{Pu} \dashrightarrow {}^{241}\text{Am} \dashrightarrow {}^{237}\text{Np}$$

Dose rates from waste containing this nuclide vary markedly depending on the materials in the drum and how they have been packaged. The self-shielding of the waste and the attenuation of the drums and drum liners are effective in reducing the external radiation levels during most of the operations. Typical measured photon radiation levels on the outside surfaces of the drums used in the characterization program to date range from 1 mr/hr to about 80 mr/hr. These rates are of concern only during those operations in the high bay that involve drum storage and drum preparation and in the HRA that require workers to be in close proximity to the sources.

If the effects of self-shielding are lost, radiation levels can increase by orders of magnitude. Dose equivalent rates from ^{241}Am can be significant if only air separates the source and the detector and whenever self-shielding effects are small. Contamination that is spread on surfaces has very little self shielding. Dose equivalent rates from unshielded area and point sources of one Curie of ^{241}Am as a function of distance are shown in Fig. 3. Its 59.5 KeV gamma ray dominates the radiation field; while it is readily attenuated by thin layers of metal, low density hydrogenous materials like plastic are ineffective shields.

As of January 1993, the waste characterization work has not caused any noticeable increase in measured personnel exposure even though workers are in close contact with the waste drums during some operations in the high bay and the HRA. External radiation sources are monitored by the standard INEL four-element thermoluminencent dosimeter (TLD) packet, self-reading dosimeters. Finger TLD rings and alarm dosimeters were worn on the outside of the anti-contamination clothing to give warning in case of a sudden increase in dose rate; they can be seen in Fig. 2. The use of TLD rings and alarm dosimeters were only used in the first few entries into the HRA. However, based on a time study conducted by the ANL-West Health Physics Section and operational experience, the TLD rings and alarm dosimeters are no longer used.

Fig. 2. Surveying an open drum in the HRA.

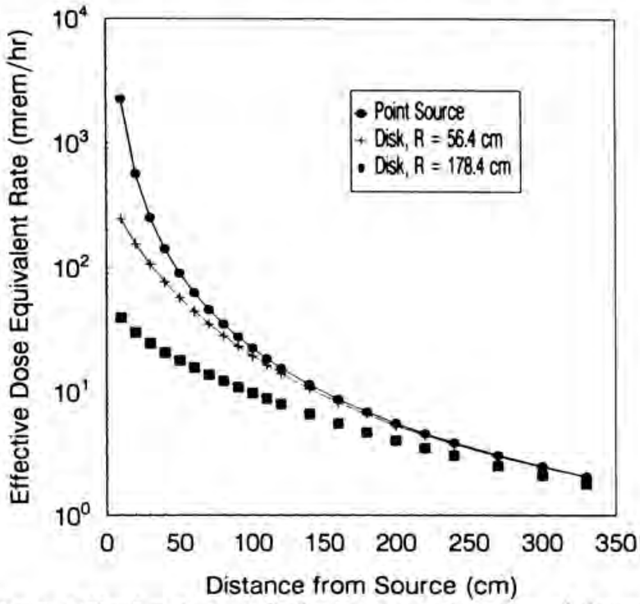

Fig. 3. Effective dose equivalent rate as a function of distance from one Curie ^{241}Am point and area sources.

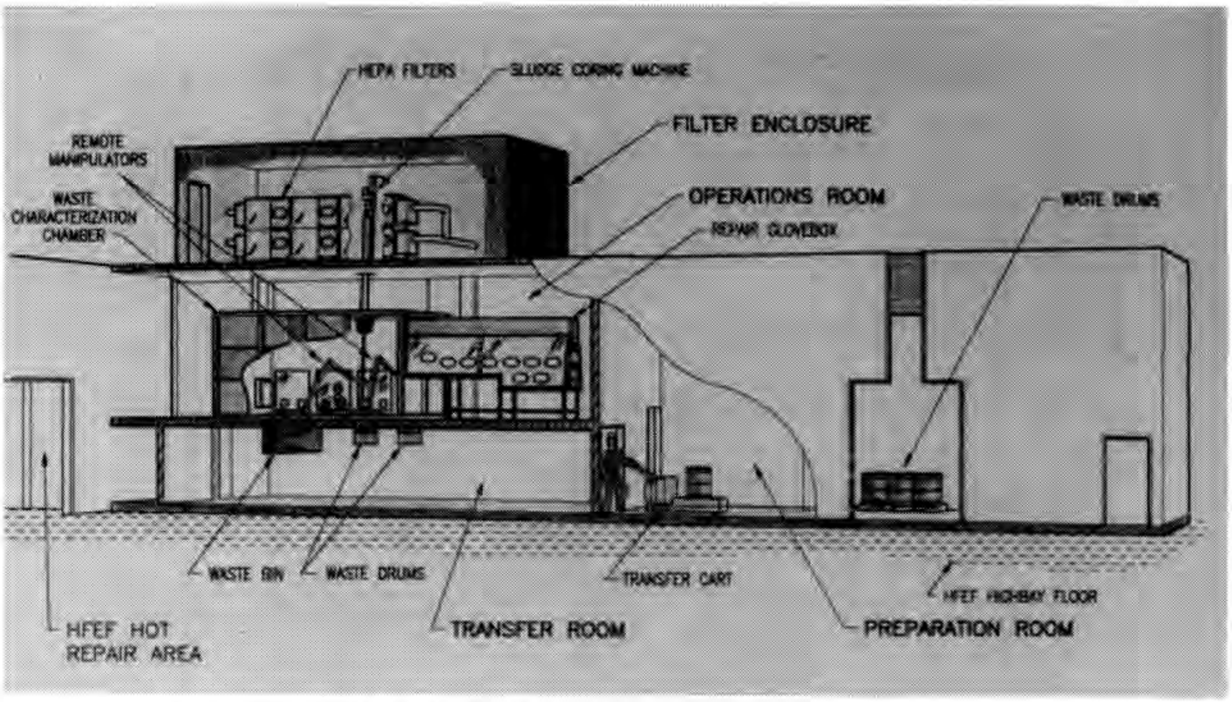

Fig. 4. The WIPP waste characterization chamber.

Outside of the hot cell and HRA, active continuous air monitors (CAMs) measure real-time concentrations of both alpha and beta-gamma emitters in the respirable air. Another facility air sampling system is passive rather than active; a central vacuum station draws air through filter assemblies strategically located throughout the HFEF. Twice a week, these filters are removed and counted for airborne activity; a three day delay between filter removal and counting allows the naturally occurring radon and thoron daughters to decay. This passive system is sensitive to better than one percent of a derived air concentration for any alpha emitting nuclide (6). As of February 1993, no increase in airborne activity that could be associated with waste characterization operations has been detected at any of the sampling locations.

Interferences from the radioactive daughter nuclide of ^{226}Ra and ^{228}Ra influence the ability of the active alpha-sensitive CAMs to detect airborne transuranic contaminants in the facility. Since they are so effective at causing high dose commitments, the CAMs are set to alarm whenever extremely low levels of airborne alpha-emitting nuclide are detected. Naturally occurring radionuclides present in construction materials and from nearby thorium deposits frequently cause alarms during atmospheric inversion conditions or if the ventilation rate in HFEF is reduced.

A goal of this project is to procure instrumentation that minimizes any interferences from radon and thoron daughters. Beta particles from ^{241}Pu and photons from ^{241}Am increase the value of the beta-gamma sensitive CAMs in detecting the presence of man-made actinides. The passive air sampling system and an extensive program of obtaining radiological smears from surfaces provide data that allow early detection of contamination or any loss of containment in spite of the interferences from radon and thoron daughters. Low-energy gamma spectrometry is used to identify specific radionuclides on wipes and filter media.

FACILITY MODIFICATIONS

To allow more detailed examinations, and to increase the efficiency of the characterization and repackaging processes, a large (4.9 by 2.4 by 2.4 meters high) chamber containing two robotic arms is being installed. This waste characterization chamber (WCC) and its supporting facilities are scheduled to be in operation by the end of summer of 1993. An isometric sketch of the WCC is shown in Fig. 4. The potential for worker radiation doses from gas sampling operations and drum preparation will be eliminated; it will also reduce the probability of contaminating other areas of HFEF. The activation of the WCC will allow the presently used HRA and Decon Cell areas to return to their original mission of supporting advanced reactor development programs, but will remain available for special case handling of WIPP waste.

To provide shielding against the ^{241}Am photons, the WCC is constructed of 0.475 cm (7 gauge) stainless steel sheet over a steel frame. Table III lists the thicknesses of five shield materials required to reduce ^{241}Am dose equivalent rates by factors of ten (7). Note that buildup in lower atomic number materials causes the initial ten-value-layer to be greater than succeeding ten-value-layers. Wherever there would be a direct line of sight between surfaces where contaminated objects are placed and normally occupied areas, two layers of the stainless steel sheet are used for added protection. Since only 0.27 cm of steel is required to reduce the dose rate sources by a factor of ten, this double thickness (0.95 cm) is sufficient to reduce the photon dose rate from ^{241}Am by more than a factor of 3300.

Windows constructed of sheets of leaded glass, LEXAN™, and laminated safety glass allow unrestricted views by the operators of the interior. Most of the attenuation of photons is provided by 0.79 cm of RD-50 lead glass which is equivalent to 0.15 cm of lead sheeting (8). A 1.27 cm thick sheet of LEXAN™ polycarbonate plastic is sealed to the steel

TABLE III
Thicknesses Necessary to Reduce Dose Equivalent Rates from ^{241}Am Photons by a Factor of Ten

Material	Density (g/cc)	Thicknesses of Materials Initial (cm)	Suceeding (cm)
Plastic	1.05	29.0	14.0
Glass	2.50	5.9	4.9
Aluminum	2.70	4.8	4.1
Steel	7.87	0.27	0.27
Lead	11.35	0.043	0.043

body of the WCC. A 0.95 cm thick laminated safety glass plate is mechanically fastened to the inside of the chamber; this plate may be changed using the robot arms. The attenuation provided by these windows is equivalent to about 0.87 cm of steel. Television cameras are mounted on the East and West walls near the roof and on the ceiling of the WCC to provide additional visibility to the operators. Even though most of the work will be accomplished using the robot arms, glove ports are provided through the four front windows to allow access on the operating side of the chamber. When not in use, these glove ports are covered with steel shields and locked. The top of the WCC is constructed of one sheet of 7 gauge steel because occupancy above the WCC is limited and the radiation sources should be concentrated near the bottom of the chamber.

Windows in the roof of the chamber allow the use of external fluorescent lighting; interior light fixtures are not used because of the problems associated with maintaining any system in a contaminated area. As much as possible, the interior surfaces have been made smooth and free of sharp bends to minimize trapping of particulate contamination. Two drum ports and one bin port located in the floor of the WCC are designed so that polyethylene bags can be used to safely interface the drums or bin to the WCC with minimal effort and efficient contamination control. A radio-frequency heat sealer will be used to seal the bags when the containers are ready to be disconnected from the chamber. A single stage of HEPA filtration is located on the air inlets and outlets of the chamber; charcoal absorption is also provided on the outlet stream to capture any volatile organic compounds that may have been released from the waste matrix during characterization activities. Filtered exhaust from the chamber will be sent through the double HEPA filter trains already installed in the HFEF.

The WCC will be installed in a new enclosure that is being constructed in the High Bay specifically for the waste characterization project. Separate rooms for operations, drum and bin preparations, and drum and bin transfers are designed to simplify contamination control; a dedicated health physics office with gamma spectroscopy capability is located at the main entrance of the enclosure. Twenty sampling stations for the high-sensitivity, passive facility air sampling system are placed throughout the area. Four alpha-sensitive and four beta-gamma-sensitive CAMs are deployed to give active coverage of the rooms.

Full-body portal monitors sensitive to both alpha and beta-gamma radiations must be used when exiting the enclosure. These units are shielded with 1.9 cm of steel to reduce their background count rates. Hand and foot monitors are used within the area to serve as initial screening for personnel moving from one room to another. Alpha-sensitive hand monitors are installed near the glove ports; they are mounted vertically to reduce the probability of contaminating their thin windows.

RADIOLOGICAL CONSEQUENCES OF ACCIDENTS

Operations within the HFEF have been reviewed to determine the probability of accidents that could disperse the waste to the environment. Most of the potential accidents involve dropping a filled bin or a fire in the waste materials. Table IV presents the estimates of dose rates to an on-site worker located 280 meters from HFEF and to a member of the public who remains at the nearest site boundary 5000 meters away for the duration of the accident (9). Dose commitments are almost entirely from the inhalation pathway with ^{241}Am being the dominant contributor. The calculations were performed using the RSAC-4 program (10). The annual probability classifications correspond to the criteria given by Elder, et al (11). The accident scenarios were developed for the present mode of operations; no additional events have been identified for use of the WCC.

Hydrogen explosions are incredible because the waste drums are vented through a carbon composite filter for at least two months before they are transported to HFEF. Experiments have demonstrated that hydrogen gas diffuses through these filters within a few hours of its formation (12).

Even though criticality accidents are judged to be incredible, active criticality monitors are required because more than 350 grams of plutonium could be present in the High Bay of HFEF at one time (13). In addition to the active gamma-sensitive units, passive nuclear accident dosimeter packages are located around the High Bay. These contain activation foils and TLDs that can provide information on the intensity and energy spectra of neutrons and photons released in a criticality event.

CONCLUSION

All activities associated with the characterization of waste in the HFEF have been reviewed to assure that radiation exposures are as low as reasonably achievable. The principal efforts are spent in minimizing contamination levels in all parts of HFEF and maintaining containment of alpha-emitters to reduce the probability of internal sources.

To date, the procedures have been successful. Analyses of potential accidents indicate low probabilities of occurrence and minimal radiological consequences to on-site personnel as well as to the general public. Currently, external radiation levels are controlled by the use of a massive hot cell for sorting and packaging waste. A new examination chamber has been designed to shield the photons from ^{241}Am while providing better access to the contents of the drums and more efficient examination and repackaging of the items.

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TABLE IV
Potential Accidents for Waste Characterization Operations

Accident Description	Probability Category	Annual Probability	Dose Equivalent (millirem)	
			On-Site	Off-Site
Fire in the High Bay Storage Area	Unlikely	10^{-2} to 10^{-4}	8.45	0.55
Waste Bin Dropped on the High Bay Floor	Unlikely	10^{-2} to 10^{-4}	2.53	0.16
Fire and Explosion when a Drum is Opened	Unlikely	10^{-2} to 10^{-4}	$1x10^{-4}$	$1x10^{-5}$
Fire in a Bin While in the Spray Chamber	Unlikely	10^{-2} to 10^{-4}	$1x10^{-3}$	$2x10^{-5}$
Fire in a Bin While in the Glovebox Area	Unlikely	10^{-2} to 10^{-4}	$8x10^{-4}$	$1x10^{-5}$
Partial Collapse of the HRA in a Seismic Event	Unlikely	10^{-2} to 10^{-4}	2.53	0.16
Loss of Exhaust Blowers and Decon Cell Exhaust	Unlikely	10^{-2} to 10^{-4}	0	0
Waste Bin Dropped in the Truck Lock	Extremely Unlikely	10^{-4} to 10^{-6}	10.5	0.25
Hydrogen Explosion	Incredible	$< 10^{-6}$	N/A	N/A
Accidental Criticality	Incredible	$< 10^{-8}$	N/A	N/A

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