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Alternate Airborne Release Fraction Determination for Hazardous Waste Management Storage Repository Hazard Categorization at the Lawrence Livermore National Laboratory

Gregg P. Brumburgh

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Gregg P. Brumburgh
Lawrence Livermore National Laboratory
Livermore, CA

Introduction

Hazardous Waste Management (HWM) facilities are used in the handling and processing of solid and liquid radioactive, hazardous, mixed, and medical wastes generated at Lawrence Livermore National Laboratory (LLNL). Wastes may be treated or stored in one of the HWM facility units prior to shipment off site for treatment or disposal. Planned facilities such as the Decontamination and Waste Treatment Facility (DWTF) and the Building 280 Container Storage Unit are expected to handle similar waste streams.

A hazard classification was performed in each facility safety analysis report (SAR) according to DOE Standard 1027-92, *Hazard Categorization and Accident Analysis Techniques for Compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports*[1]. The general methodology practiced by HWM to determine alternate airborne release fractions (ARFs) in those SARs was based upon a beyond evaluation basis earthquake accident scenario characterized by the release of the largest amount of respirable, airborne radioactive material. The alternate ARF was calculated using a three-factor formula consisting of the fraction of failed waste containers, fraction of material released from failed waste containers, and the fraction of material entrained to the environment. Recently, in deliberation with DOE-Oakland representatives, HWM decided to modify this methodology. In place of the current detailed analysis, a more straightforward process was proposed based upon material form, credible accident environments, and empirical data. This paper will discuss the methodology and derivation of ARFs specific to HWM treatment and storage facilities that are alternative to those presented in DOE-STD-1027-92.

Background

In hazard categorizations conducted at HWM to date, waste inventories were initially compared to the threshold limits provided in Table A-1 of DOE-STD-1027-92. These comparisons usually resulted in an initial hazard determination of Hazard Category 2 because a few of the isotopes in the inventories routinely surpassed their Hazard Category 2 (HC-2) curie threshold quantities (TQs). Further, the sum of the ratios of the total inventory of nuclides to their respective TQs exceeded 1.0.

In accordance with DOE-STD-1027-92, if the alternate ARFs can be shown to be significantly different from the values used in the standard, based on physical and chemical form and available dispersive energy sources, the threshold inventory values for HC-2 (in Table A.1 of the standard) may be divided by the ratio of the maximum potential ARFs to standard ARFs. Mathematically, the equation is

$$TQ_{facility} = \frac{TQ_{1027}}{ARF_{facility}/ARF_{1027}}$$

The DOE ARFs were developed assuming radionuclides in forms typically much more dispersible than those that may be encountered in archetypal DOE Environmental Management (EM) waste treatment, storage, and disposal facilities and activities. Consequently, the DOE ARFs are overly conservative for application with HWM storage operations. Section 3.1.2 of DOE-STD-1027 provides for use of facility/segment-specific ARFs, based on actual physical and chemical form and available dispersal energy, when actual ARFs can be shown to be substantially different from the DOE values.

HWM Operations

The HWM storage units are housed in a variety of facilities, ranging from reinforced-concrete buildings to polyester tents, all differing in size and construction styles. Individual storage units and their applicable structures, systems, and components also vary with respect to Performance Category (PC) design levels, and preventive and mitigative design features.

Radioactive wastes are stored in solid, liquid, and sludge form. By far, the majority (and the most hazardous) of the stored waste is made up of transuranic (TRU) mixed waste, which consists primarily of solid materials on which surface contamination may be found (e.g., wiping tissues, paper, plastic, chemistry glassware, ceramics, and metal) and small quantities of solidified liquids and sludges contaminated with radionuclides and hazardous constituents. Except for a very small amount, all TRU material is contained in U.S. Department of Transportation (DOT) Type-A drums and stored six drums per pallet maximum, no more than two pallets in height. All TRU drums are equipped with carbon composite filters to prevent the buildup of explosive gases. Inside each Type-A 55-gal. drum in which TRU waste is stored, is a rigid plastic liner and polyethylene bags to provide further protection. Other radioactive waste material (e.g., low-level waste) is not required to be stored in this manner.

Alternate ARF Methodology & Derivation

The methodology to characterize the alternate ARF consists of three steps:

1. Determine the disposition of the material at risk (MAR) with respect to quantity, material form, location, and containment characteristics;

2. Consider the energy sources or bounding accident scenarios assumed to influence the ARFs; and,

3. Utilize DOE-HDBK-3010-94, *Airborne Release Fractions/Rates and Respirable Fractions for Nonreactor Nuclear Facilities*[2], empirical data.

Step 1 - MAR Disposition

An accident at an HWM storage facility can involve any and all of the radionuclides present as waste. For step 1, then, the MAR must be based on the maximum possible storage capacity. The HWM facilities are segmented for inventory management purposes so the MAR must be established for each. The MAR disposition was determined by waste group type. A waste group is characterized by a specific material form and the container in which the waste is stored. Due to the limited material forms and kinds of containers present at HWM facilities, each can be divided into two categories: solid waste versus non-solid waste for the former, and steel, Type-A containers versus all others for the latter. Table 1 provides the requirements for each waste group. Note: tritium is always assumed to be in oxide form and treated as a gas for safety analysis purposes. Consequently, the ARF for tritium is not adjusted.

Table 1. Waste Group Material Form and Container Type.

Waste Group	Material Form	Container Type
1	Solid, combustible	Type-A* or Equivalent
2	Solid, combustible	Non-Type-A Equivalent
3	Non-solid, non-combustible	Type-A* or Equivalent
4	Non-solid, non-combustible	Non-Type-A Equivalent

*Defined as steel containers which meet or exceed Type-A requirements outlined in 49 CFR 173.465

Waste group 1 is defined as solid, combustible radioactive waste stored in steel containers meeting or exceeding Type-A packaging requirements outlined in 49 CFR 173.465[3]. Waste group 2 is defined as the inventory of all solid, combustible radionuclide wastes not in waste group 1. Essentially, this is solid radioactive waste stored in non-Type-A equivalent containers. Waste group 3 is defined as non-solid radioactive waste stored in steel containers meeting or exceeding Type-A packaging requirements outlined in 49 CFR 173.465. Waste group 4 is defined as the inventory of all non-solid radionuclide wastes not in waste group 3. Again, this is non-solid radioactive waste stored in non-Type-A equivalent containers.

Step 2 - Bounding Accident Scenarios

For step 2, fire and spill accidents were used to address the alternative ARF derivation because they have been shown to be representative of and to bound all HWM storage facility accidents as determined by hazard and accident analysis[4]. Explosions, criticality, and most natural phenomena hazards were screened out due to type of material, location, a lack of energy sources, or a combination thereof. It is recognized that the accident stressor that can generate the largest release fraction associated with this waste material is thermal (fire).

For a release of solid material, both fire and spill accidents are applicable. Release mechanisms for spill accidents include free-fall, impact, and aerodynamic entrainment (suspension). The free-fall release mechanism was selected over impaction stress and aerodynamic entrainment release mechanisms for numerous reasons. First, the bounding accident for a release of solid material is anticipated to be initiated by a design basis earthquake. Due to the design of the facilities in which solid waste is stored (i.e., PC-2 qualified structures or polyester tents) little or no impacting of the waste containers is expected from falling debris. Next, the use of steel, Type-A drums for TRU waste storage and steel drums for all other solid radioactive waste reduces the probability of drum failure due to any debris impact. Lastly, an accident causing the release of contaminated material from a free-fall spill is expected to encompass a much larger population of waste containers than from an impact accident.

For a release of all aqueous solutions, two release mechanisms were considered: free-fall and explosive release via venting of pressurized liquids. All other release mechanisms were eliminated for the same reasons as outlined for solid waste accidents.

Step 3 - Application of Empirical Database

After the MAR has been characterized with respect to form and containment, and the bounding accidents have been selected, then a database can be employed to determine an alternate ARF. The DOE Handbook 3010-94 was the technical basis for establishing the alternative ARFs for all release scenarios.

As discussed earlier, solid waste at HWM facilities is predominately represented by TRU radionuclides as surface contamination associated with lightweight combustible materials having high surface-area-to-mass ratios (e.g., paper, wipes, gloves, cloth, personal protective equipment) situated inside robust containers. The most likely sequence of events leading to a free-fall spill of solid waste material from a container initiated by an earthquake is the toppling of stacked drums from a height of about 1 meter onto a hard, unyielding surface. From Handbook 3010, the bounding ARF for packaged, combustible solids under impact conditions is 1×10^{-3}. The impact ARF was derived by using the ARF for failed drums, which came from tests on powder in open pails. The pail-scenario ARF of 1×10^{-3} is derived from two test data points for impacts in unconfined powder and assumes the drums failed and ejected a major portion of their contents. Those experiments, however, do not closely model the anticipated accident scenarios for solid waste at HWM facilities. The majority of these materials would generate little force during impact with surfaces. From Handbook 3010, “for materials with high surface area to mass ratios, no significant suspension is expected for free-fall spill from typical working heights (~1 - 1.5 m)...for such materials, no significant suspension of surface contamination is postulated.” Hence, the ARF values for these accident types are deemed negligible as compared to the fire ARF described below.

The fire ARF for solid waste in Handbook 3010 was derived by using the ARF from fire experimental data of single cardboard cartons, entirely filled with contaminated combustible materials, that were allowed to burn under natural convection until self-extinguished[5]. The 18-in. × 18-in. × 24-in.-high single cartons were placed on a grille-like holder and situated in a 10-ft-diameter × 10-ft-high stainless-steel vessel. Balled-milled depleted uranium dioxide powder, air-dried uranium nitrate hexahydrate (UNH) solution, and UNH solutions were used individually and together as the contaminants. The measured ARFs ranged from 3×10^{-5} to 5×10^{-4}, and had a median value of 8×10^{-5}, with an average ARF of 1×10^{-4}.

While those experiments are representative of the basic fire scenario for HWM storage facilities, many differences exist between the conditions of the experiments and those of standard operations at HWM storage facilities. The differences (noted below) are significant enough to warrant a selection of an ARF less than the bounding ARF derived in those experiments:

- Burning of the contents of a single drum from internal ignition is assumed not to propagate to adjacent drums.[4] For a large fire to threaten even a portion of the total inventory in a storage facility, an external fire/heat source is needed. Facility design features (e.g., steel construction) and operating procedures (e.g., low fire-loading) for TRU waste storage reduce this possibility.

- As opposed to using cardboard boxes, the HWM uses closed metal drums fitted with filters that provide pressure relief and are less prone to the type of catastrophic failure that would allow combustion and ejection of contaminated, combustible material directly to the ambient atmosphere. Furthermore, because the combustible vapors generated are continuously vented, a fire will more likely be limited to the drums directly affected by the external fire.

- Rather than being stored in a single box, TRU waste material is stored in steel drums stacked two high; thus, there are many metal surfaces around and over the suspended materials vented from the drums. "The presence of more substantive material surrounding the contaminated, combustible waste would limit the availability of oxygen and force particles generated in the interior of the mass to pass through the ash/residue formed prior to the release."[2]

- Plateout in the immediate vicinity of the fire was considered a generic phenomena in those experiments. Considering that many more deposition surfaces exist around the stored waste in HWM facilities, more plateout is expected.

Based upon these differences, an ARF of 5×10^{-5} was selected as a more representative hazard categorization value for waste group 1. Though this value is approximately one order of magnitude less than the bounding value determined in the DOE Handbook's experiments, it does fall between the experiment's lower bound value and its median value.

For waste group 2, a more conservative ARF is considered when compared to waste group 1 as conditions between the two with regard to containerization and storage construction (e.g., polyester tents versus steel and concrete) may exist. These variations increase the likelihood that energy sources available in an accident involving waste group 2 have the potential to affect more of the MAR and yield a greater consequence. Therefore, for waste group 2, the alternate ARF is determined to be the average ARF value, or 1.0×10^{-4}, for fire experimental data presented in the DOE Handbook.

The non-solid waste located at HWM facilities is dominated by non-combustible, aqueous solutions. Both free-fall and explosive release via venting of pressurized liquids mechanisms were investigated. The initiator for the accident is, again, the design basis earthquake. A maximum fall distance of 3 m was selected for the free-fall spill accident based upon aqueous tank/container construction and configuration. DOE Handbook 3010 distinguishes between general aqueous solutions, concentrated heavy metal solutions, and slurries. As HWM facilities possess each kind, the most conservative solution (aqueous solution, density ~ 1.0 g/cm^3) was chosen. Handbook 3010 provides a median ARF for an aqueous solution of 4 x 10^{-5} and a bounding ARF of 2 x 10^{-4}.

An analogous model in Handbook 3010 for an explosive venting of pressurized liquid in an HWM aqueous tank is a low pressure (0.35 MPa_g or less) release. This release mechanism has a bounding ARF of 5 x 10^{-5} for all aqueous solutions. As this is comparable to the aqueous free-fall median ARF value but less representative of the conditions at the HWM facilities, the former values will be adopted. Thus, the aqueous median ARF was designated for waste group 3 and the bounding ARF for waste group 4. Waste group 4 was given a more conservative ARF due to the differences between the robustness of the two waste group's tank/containers. Table 2 provides the ARFs for each HWM identified waste group.

Table 2. Waste Group Airborne Release Fractions (ARFs).

Waste Group	Airborne Release Fraction (ARF)
1	5 x 10^{-5}
2	1 x 10^{-4}
3	4 x 10^{-5}
4	2 x 10^{-4}

Conclusions

The airborne release fractions (ARFs) furnished in DOE-STD-1027-92 are deemed much too conservative for the physical and chemical form and available dispersal energy associated with HWM stored waste material. To derive alternate ARFs, a 3-step methodology was employed which consisted of determining the disposition of the material at risk with respect to quantity, material form, location, and containment characteristics; considering the energy sources or bounding accident scenarios assumed to influence the ARFs; and, utilizing empirical data from DOE-HDBK-3010-94.

Four general waste groups were identified for all HWM facilities. These waste groups were binned by material form and container characteristics. Of the two representative accidents investigated for HWM storage facilities (i.e., fire and free-fall spill), the fire scenario generated the largest release fraction. In comparison, no significant suspension of surface contamination was expected from a free-fall spill accident of solid waste material. It was determined that the experimental bounding value of 5 x 10^{-4} documented in Handbook 3010 was still too conservative due to significant differences between the experimental conditions and conditions present at HWM storage facilities (e.g., steel, Type-A drums with filters in which waste material is stored, benefits of storage unit construction, plateout of airborne material on surrounding surfaces). For aqueous release mechanisms, the median and bounding case experimental ARF values were considered appropriate.

In summary, it was determined that the alternative ARFs assigned for categorizing the inherent hazards of stored waste are unique for each waste group. The HWM facility alternate ARF values range from 1/2 to 1/20 of the ARF values provided in DOE-STD-1027-92.

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Technical Information Department • **Lawrence Livermore National Laboratory**
University of California • Livermore, California 94551

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