Log Number: 5.3CMH04
Topic Area Number: 5.3
Poster Presentation

Computational Physics and Engineering Division

Bounding Values for Low-Level-Waste Transport Exemptions and Disposal

C. M. Hopper, K. R. Elam, C. V. Parks, J. J. Lichtenwalter*
Oak Ridge National Laboratory,**
P.O. Box 2008
Oak Ridge, TN 37831-6370 USA

e-mail: fhh@ornl.gov
phone: (423) 576-8617
fax: (423) 576-3513

e-mail: k96@ornl.gov
phone: (423) 574-4538
fax: (423) 576-3513

e-mail: cvp@ornl.gov
phone: (423) 574-5280
fax: (423) 576-3513

e-mail: lichtenwalte@bechteljacobs.org
phone: (423) 241-1755
fax: (423) 576-8184

Submitted to the
ICNC'99
Sixth International Conference on Nuclear Criticality Safety,
September 20–24, 1999,
Palais des congrès . Versailles . France

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. DE-AC05-96OR22464. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

---

*Current Address: Bechtel Jacobs Company LLC, P. O. Box 4699, Bldg. K-1320, MS-7592, Oak Ridge, TN 37831-7592 USA.

**Managed by Lockheed Martin Energy Research Corporation under contract DE-AC05-96OR22464 for the U.S. Department of Energy.

BOUNDING VALUES FOR LOW-LEVEL-WASTE TRANSPORT EXEMPTIONS AND DISPOSAL

Calvin M. Hopper, Karla R. Elam, Cecil V. Parks, Jerry J. Lichtenwalter
Oak Ridge National Laboratory, USA

Abstract

Characterizations and bounding computational results determined by the Oak Ridge National Laboratory have been offered to the U.S. Nuclear Regulatory Commission as supporting technical bases for regulatory considerations in the packaging, transport, retrievable emplacement and disposal of radioactive low-level waste contaminated with fissile materials. The fissile materials included 100 wt % ^{235}U, 10 wt % ^{235}U in uranium, 100 wt % ^{233}U, 100 wt % ^{239}Pu, or plutonium as less than 76 wt % ^{239}Pu, more than 12 wt % ^{240}Pu, and less than 12 wt % ^{241}Pu. The considered waste matrixes included silicon dioxide, carbon, light water and polyethylene, heavy water, or beryllium with summary examinations of other potential matrixes. The limiting concentrations and geometries for these bounding conjectured low-level-waste matrixes are presented in this paper.

Introduction

Within the U.S. nuclear industry, there have been trends and initiatives in recent years to reduce and consolidate nonreactor nuclear facility operations and to decontaminate and decommission nonreactor nuclear facilities. These trends and initiatives require support activities such as fissile material contaminated low-level waste (LLW) packaging, transport, retrievable emplacement and disposal of radioactive LLW contaminated with fissile materials. All of these support activities require regulatory approval of the safety bases that demonstrates nuclear subcriticality for safety. This paper provides bounding values for consideration during reviews and approvals for operations and the development of regulatory guidance relative to LLW packaging, transport, retrievable emplacement and disposal of the LLW contaminated with fissile materials. The results reported in this paper are a compilation of past [1–4] and evolving [5] support work performed by the Oak Ridge National Laboratory for the U.S. Nuclear Regulatory Commission. This work was performed to address issues and regulatory needs or concerns [6] related to fissile-material-transportation exemptions and general licenses [6, 7] and a rule change petition [8], requesting a predisposal concentration limits for special nuclear material (SNM) in soil to amend the existing mass limits specified by regulations [9].

Limiting critical concentrations and geometries are provided for specific fissionable nuclides (i.e., 100 or 10 wt % ^{235}U in uranium, 100 wt % ^{233}U, 100 wt % ^{239} Pu, and plutonium as #76 wt% ^{239}Pu, $12 wt % ^{240}Pu, #12 wt % ^{241}Pu) as contaminating conjectured LLW matrixes comprised of silicon dioxide (SiO_2), carbon (C), light water (H_2O), heavy water (D_2O), or beryllium (Be). Also provided are the results of broad-ranging computational comparisons among natural elements and some compounds substituting as the waste matrix. Compounds that were considered included calcium fluoride (CaF_2), calcium carbonate ($CaCO_3$), magnesium fluoride (MgF_2), magnesium carbonate

($MgCO_3$), magnesium oxide (MgO), calcium-magnesium carbonate ($CaMg(CO_3)_2$), beryllium oxide (BeO), and carbon tetrafluoride (CF_4). The systems were modeled as infinite-homogeneous media, SiO_2-reflected infinite-planar arrays of optimally water-moderated cylindrical units, and SiO_2-reflected infinite-planes, cylinders and spheres.

Even though the computational results were derived for specific issues related to general licenses and exemptions for transportation and for specific issues related to LLW emplacement and disposal, the results can be generalized and may be used for estimating trends or confirming expectations for fissile materials safety. In particular, the results of these broad-ranging computational studies may be useful to the nuclear criticality safety community for bounding operations involving LLW contaminated with fissile material by referring to the determined subcritical envelopes for nuclear criticality safety.

Evaluated Computational Models

The evaluated computational models were specific to the assessments requested for regulatory concerns or guidance. Two general categories of criticality evaluations were considered: transportation and LLW emplacement and disposal.

Transportation computational models were based upon regulatory specifications found in references 6 and 7 for general licenses and exemptions for packaging and transportation of fissile material: The typical nuclear criticality safety evaluation for general-license materials, therefore, was for five times the intended shipment under normal transport conditions and twice the intended shipment under hypothetical accident conditions. However, the models used for the nuclear criticality evaluations for fissile-material-transportation exemptions included broad extensions of permissible materials to infinite media.

Low-level-waste computational models were based upon permissible licensed disposal limits [10, 11] and hypothetical waste matrixes in finite and infinite media configurations.

The fissile-material contaminants of the waste matrixes included 100 wt % ^{235}U, 10 wt % ^{235}U in uraniu

Specifications for exemptions are provided in Ref. 6, under §71.10 and §71.53. The above specifications provide general licenses or exemptions for

- any material with less than 70 Bq/g of matrix;
- less than Type A quantities, certain low-specific-activity (LSA) material with acceptable radiation limits, and limited quantities of special-form Pu or Am from all requirements, except U.S. Department of Transportation (DOT) compliance and shipment-by-air requirements, provided there is no fissile material or the fissile-material exemptions of §71.53 are satisfied,
- packages with 15 g or less of fissile material, §71.53(a)(1);
- select homogeneous solutions [hydrogenous, §71.53(a)(2) and uranyl nitrate, §71.53(c)] or mixtures of fissile material with limits specified on combinations of mass, ^{235}U enrichment, hydrogen content, and/or solution concentration;
- packages that can be demonstrated to have less than 5 g of fissile material in any 10-L volume, §71.53(a)(3);
- packages containing uranium enriched in ^{235}U to a maximum of 1 wt%, §71.53(d); and
- packages with less than 1 kg of plutonium, of which not more than 20% by mass may consist of ^{239}Pu, ^{241}Pu, or any combination of these radionuclides, §71.53(d).

The geometries considered for the transportation evaluations were comprised of finite geometries or volumes of homogeneous or containerized fissile materials for "typical" shipments qualifying under the general-license specifications. Materials qualifying for broad material exemptions were modeled as infinite media.

Geometries

Four types of system geometries were considered for the transportation evaluations. These systems were (1) infinite homogeneous media, (2) homogeneous finite spheres (584-m^3 volume) reflected with 30.48-cm-thick water, (3) minimum critical masses of the hypothetical materials as 30.48-cm-thick, water-reflected spheres, and (4) 30.48-cm-thick water-reflected arrays (approximately 584-m^3 total array volume) of packages. The arrays were comprised of the hypothetical material mixtures contained within geometries typical of 110-gal drums that are constructed from two joined 55-gal drums. The volume of 584 m^3 was selected to represent the maximum volume of five U.S. public highway transportation vehicles (i.e., each vehicle consisting of two tandem trailers pulled by a single tractor). Evaluations were performed for noncontainerized and for containerized fissile materials. For evaluations with containerized fissile material, the assumed containers were 20-gauge 316-stainless steel DOT-17E drums. These drums were selected because of the minimum quantity of iron (a neutron-absorbing material) that would be represented in the computational models.

Materials

Five different material matrixes were considered for the general-license and exemption evaluations. They included (1) light water or polyethylene, (2) silicon dioxide – light water, (3) carbon – light water, (4) beryllium – light water, and (5) heavy water. The individually evaluated fissile materials that were homogeneously blended with the material matrixes included ^{235}U, ^{233}U, and ^{239}Pu metals. Mixtures of the material matrix and fissile materials with water or polyethylene were assumed to be at full density, whereas material matrixes of silicon dioxide, carbon, beryllium, and heavy water were assumed to be at 60% of their theoretical densities, thereby permitting the introduction of various fissile material and water fractions into the remaining 40% of the matrix volume.

Low-Level Retrievable Waste Emplacement and Disposal

The approach for developing low-level retrievable waste emplacement and disposal guidance [5] was to use information from previous studies [1–4] for developing nuclear criticality safety evaluations specific to typical and projected operational practices in the United States for the emplacement and disposal of LLW. The guidance evolved through graduated evaluations reported in references:

- 1 for very general and conservative surface density screening criteria that were based on optimum water-moderated and variable unit geometries comprised of 100 wt % ^{235}U, 10 wt % ^{235}U + 90 wt % ^{238}U, or 76 wt % ^{239}Pu + 12 wt % ^{240}Pu + 12 wt % ^{241}Pu,
- 2 for LLW matrixes (i.e., "nominal soil" or SiO_2 and water) contaminated with 100 wt % ^{235}U,
- 3 for LLW matrixes (i.e., SiO_2 and water) contaminated with 10 wt % ^{235}U, and finally
- 5 for LLW matrixes (i.e., silicon, or other substituted element, dioxide or compounds moderated with water) contaminated with 100 wt % ^{235}U, 10 wt % ^{235}U + 90 wt % ^{238}U, 100 wt % ^{239}Pu, and ^{233}U.

Reference 5 provides the bases and concentration limits for low-level retrievable waste emplacement and disposal guidance. It provides consideration for emplacement techniques for:

- blended LLW and soil (potentially "infinite" in dimensions) into unrestricted burial depths,
- LLW in variable-depth burial trenches, and
- LLW within stacked concrete vaults for burial.

The concentration limits are provided in terms of fissile-material mass-per-unit area for blended LLW and soil or in terms of fissile-material mass per mass of LLW, thereby avoiding general concerns about variable volumes of waste at differing densities. Fissile-material mass limits are provided for containerized LLW.

Geometries

The primary geometries considered for the modeling of the LLW were infinite media, infinite planes of variable thicknesses and spheres. Computational model results for optimum light-water-moderated limiting fissile-material areal densities in unrestricted burial depths of LLW were taken from Ref. 1, which used a historic surface density safety evaluation technique [12, 13].

Computational model results were taken from Ref. 2 and 3 for infinite media, infinite planes of variable thicknesses, and spheres of LLW contaminated with fissile materials having variable light-water moderation. All computational models were augmented with calculations to provide completeness of LLW fissile-material contaminants (i.e., 100 wt % ^{235}U, 10 wt % ^{235}U + 90 wt % ^{238}U, ^{233}U, ^{239}Pu, and plutonium containing 76 wt % ^{239}Pu + 12 wt % ^{240}Pu + ^{241}Pu). Reflector regions for LLW infinite planes of variable thickness and spheres were assumed to be thick SiO_2, having identical SiO_2 and light-water moderation densities as the considered LLW. Except for 10-wt % ^{235}U-contaminated LLW, vaulted units of LLW were modeled as 3.05-m (120-in.) -thick infinite planes of dry (i.e., no water moderation for maximum reactivity) LLW sandwiched between two 0.203-m (8-in.) infinite planes of concrete. Maximum reactivity for the 10 wt % ^{235}U-contaminated LLW vaulted waste was determined to occur with light-water moderation at about 0.03 g H_2O/cm^3. Single-level or multiple-stacked vault units in contact were reflected with thick dry SiO_2.

Materials

The fissile materials modeled for determining limiting areal densities were 100 wt % ^{235}U, 10 wt % ^{235}U + 90 wt % ^{238}U, ^{233}U, ^{239}Pu, and plutonium containing 76 wt % ^{239}Pu + 12 wt % ^{240}Pu + ^{241}Pu as variably moderated with light water. The applied surface density evaluation technique assumed a planar array of units with fixed masses of fissile material per unit (i.e., 350 g ^{235}U, 250 g ^{233}U and 225 g ^{239}Pu or 256 g plutonium as 76 wt % ^{239}Pu + 12 wt % ^{240}Pu + 12 wt % ^{241}Pu). Because the array units were comprised of fixed masses of fissile material with varying, yet full-density, light-water moderation, the unit volumes were variable. Variable unit height-to-diameter ratios were examined to ensure that fissile-material lumping effects were optimized for the optimum degree of light-water moderation.

The LLW matrixes were assumed to have a fixed 1.6 g SiO_2/cm^3 density (i.e., 60% of the 2.7-g SiO_2/cm^3 theoretical density) plus variable densities of individually considered fissile materials unless substituted with other elements or compounds. Silicon dioxide (SiO_2) was used as the surrogate for LLW in order to simplify the calculations and to provide a conservative estimate of the critically safe concentration of SNM in LLW. Silicon has a very low-neutron-capture cross section, and silicon dioxide (sand) is often a major constituent in the soil or backfill materials used at LLW facilities. In actual waste disposal environments, neutron absorbers, such as iron, calcium, and sodium, would be expected to be present in the waste, thus making the SNM waste less likely to cause an inadvertent criticality. To ensure that the use of silicon dioxide as a surrogate for LLW would give conservative results, other elements were substituted for the silicon on an atom-for-atom basis in a series of computer calculations. The baseline model was an infinite dry system containing ^{235}U at a density of 0.00141 g/cm^3 and silicon dioxide at a density of 1.6 g/cm^3 which had a calculated k_{inf}, of 0.95. Calculations using all other available elements resulted in lower values of k_{inf} with the following exceptions: helium, beryllium, carbon (graphite), fluorine, magnesium, lead and bismuth. Helium, fluorine, magnesium, and bismuth are not expected to be dominant materials in disposal sites, relative to silicon. In particular, fluorine is generally found in chemical compounds with elements that are stronger absorbers of neutrons, such as calcium. Using the same assumptions described above (i.e., 1.6 g of compound/cm^3), CaF_2 with ^{235}U at a density of 0.00141 g/cm^3 had a calculated k_{inf} of 0.53, much lower than that of silicon dioxide. However, the substitution of MgF_2 for SiO_2 has a calculated k_{inf} of 1.30, which is greater than that of silicon dioxide. Magnesium fluoride is not a common compound in soil but it may be present in the waste matrix itself.

Computational Methods

Criticality calculations were performed using the SCALE [14] computer code system, version 4.4, which includes XSDRNPM and KENO V.a using the 238-group ENDF/B-V neutron cross-section library. The calculations were performed on the Oak Ridge National Laboratory Computational Physics and Engineering Division Nuclear Engineering Applications section workstations CA37 and CA38. The BONAMI, NITAWL, XSDRNPM, KENO V.a, and cross-section data set creation dates were BONAMI—1/12/99; NITAWL—9/18/98; XSDRN—5/6/99; KENO V.a—7/31/98; and scale.rev07.xn238—6/22/98.

As previously discussed, much of the data were derived from previously published studies (Refs. 1–4). The data were augmented by using methods outlined in those studies, and calculating data for fissile isotopes that were not previously evaluated, such as ^{233}U.

Results

Guidance resulting from the above-referenced computational evaluations and safety analyses provides bounding values for LLW transport exemptions and disposal.

Guidance for General Licenses and Exemptions for Transportation

Recommended fissile-material mass-limit results for general licenses in transportation are provided in Table 1. Likewise, the recommended fissile-material mass-limit results for general licenses in transportation of packages containing ^{235}U of known enrichments is provided in Table 2. Recommended fissile-material mass and fissile-to-nonfissile-mass ratios for fissile-exempt-mass ratios are provided in Table 3. These mass ratios were recommended to preclude limits on the accumulation of packages and provide improved accountability over current volume-related or concentration criteria. Detailed information on the technical basis for the recommendations of Tables 1–3 is provided in Ref. 4.

Table 1 — Mass limits for general-license packages containing mixed quantities of fissile material or ^{235}U of unknown enrichment

Fissile material	Fissile-material mass (g) mixed with moderating substances having an average hydrogen density less than or equal to H_2O	Fissile-material mass (g) mixed with moderating substances having an average hydrogen density greater than H_2O[a]
Uranium-235 (*X*)……	60	38
Uranium-233 (*Y*)……	43	27
Plutonium-239 or Plutonium-241 (Z)……	37	24

[a] For mixtures of moderating substances: if more than 10% of the moderating substance has an average hydrogen density greater than H_2O, then the lower mass limits shall be used.

Source: Ref. 6.

Table 2 — Mass limits for general-license packages containing ^{235}U of known enrichment

Uranium enrichment in wt % of ^{235}U not exceeding	Permissible maximum grams of ^{235}U per package (X)
24	60
20	63
15	67
11	72
10	76
9.5	78
9	81
8.5	82
8	85
7.5	88
7	90
6.5	93
6	97
5.5	102
5	108
4.5	114
4	120
3.5	132
3	150
2.5	180
2	246
1.5	408
1.35	480
1	1,020
0.92	1,800

Table 3 — Proposed fissile-exempt-mass ratios to replace current mass and concentration limits

Package fissile-material limit	Ratio: Fissile-to-nonfissile
15 g	1:200
350 g	1:2000
350 g	1:200[a]

[a] Packaging required to satisfy standards for normal conditions of transport.
Source: Ref. 6.

Low-Level-Waste Retrievable Emplacement and Disposal

Recommended LLW emplacement and disposal limits derived by extracting bounding results of numerous waste matrix combinations for (1) unrestricted emplacement depths, (2) restricted emplacement depths, and (3) concrete vaulted emplacements are provided in Tables 4–6 respectively. Specific discussion and technical basis for the limits in Tables 4–6 are provided in Ref. 5.

Table 4 — Limiting areal density limits for unrestricted depths of LLW emplacements

SNM	Operational limit[a]
100% enriched uranium	94 g U/ft^2
10% enriched uranium	1740 g U/ft^2
Uranium-233	82 g U/ft^2
Plutonium-239	52 g Pu/ft^2
Isotopic mixture of plutonium (76 wt % plutonium-239, 12 wt % plutonium-240, and 12 wt % plutonium-241)	67 g Pu/ft^2

[a] The areal density in kg/m^2 can be obtained by multiplying the g/ft^2 values by 0.010763.

Table 5 — Limiting fissile-material concentrations for restricted depths of LLW emplacements

SNM	Limit for up to 10-ft emplacement	Limit for up to 20-ft emplacement	Limit for greater than 20-ft emplacement
100% enriched uranium	8.395E-4 gU/g	7.059E-4 gU/g	6.888E-4 gU/g
10% uranium	1.209E-2 gU/g	1.066E-2 gU/g	1.040E-2 gU/g
Uranium-233	6.277E-4 gU/g	5.359E-4 gU/g	5.166E-4 gU/g
Plutonium-239	3.838E-4 gPu/g	3.053E-4 gPu/g	2.942E-4 gPu/g
Isotopic mixture of plutonium (76 wt % plutonium-239, 12 wt % plutonium-240, and 12 wt % plutonium-241)	5.690E-4 gPu/g	4.452E-4 gPu/g	4.149E-4 gPu/g

Table 6 — Limiting fissile material concentrations for concrete vaulted emplacements

SNM	Limit for 3 × 10-ft vaults as described
100% enriched uranium	8.709E-4 gU/g
10% enriched uranium	1.208E-2 gU/g
Uranium-233	6.307E-4 gU/g
Plutonium-239	3.938E-4 gPu/g
Isotopic mixture of plutonium (76 wt % plutonium-239, 12 wt % plutonium-240, and 12 wt % plutonium-241)	6.562E-4 gPu/g

Summary

Recent studies for the U.S. NRC have provided an opportunity to develop potential criteria (e.g., elemental compositions, relative concentrations, and fissile mass quantities) that would allow transport and disposal of fissile material without a case-specific criticality safety assessment. In the course of the studies hundreds of criticality analyses were performed to develop multiparameter trending curves that would ensure that bounding values were established and recommended. These values and criteria are currently being considered by the U.S. NRC staff for potential use in regulatory practice. To help expedite such studies in the future, the U.S. Department of Energy is supporting ORNL to utilize automated search techniques to help establish bounding values for criticality safety assessments.

Acknowledgements

The work reported in this paper was supported by the U.S. NRC Office of Nuclear Material Safety and Safeguards. The authors are appreciative of the technical guidance and helpful insights provided by the following NRC staff: P. G. Brochman and E. P. Easton, related to the transportation studies and T. E. Harris and D. Damon, related to the LLW disposal studies.

References

1. C. M. Hopper, R. H. Odegaarden, C. V. Parks, P. B. Fox, *Criticality Safety Criteria for License Review of Low-Level Waste Facilities*, NUREG/CR-6284 (ORNL/TM-12845), U.S. Nuclear Regulatory Commission, Oak Ridge National Lab., November 1994.

2. L. E. Toran, C. M. Hopper, M. T. Naney, C. V. Parks, J. F. McCarthy, B. L. Broadhead (ORNL), V. A. Colten-Bradley (U.S. NRC), *The Potential for Criticality Following Disposal of Uranium at Low-Level Waste Facilities, Volume 1: Uranium Blended with Soil*, NUREG/CR-6505, Vol. 1 (ORNL/TM-13323/V1), U.S. Nuclear Regulatory Commission, Oak Ridge National Lab., June 1997.

3. L. E. Toran, C. M. Hopper, C. V. Parks (ORNL), V. A. Colten-Bradley (U.S. NRC), *The*

9. United States *Title 10 of the* ***Code of Federal Regulations*** *Part 150*, Office of the Federal Register National Archives and Records Administration, X.

10. Utah Department of Environmental Quality, Division of Radiation Control, Radioactive Material License, UT 2300249 to Envirocare of Utah, Inc. Supplementary sheet, 1995.

11. United States Nuclear Regulatory Commission Radioactive Material License No. 12-13536-01 issued to Chem-Nuclear Systems, Inc., Barnwell, South Carolina, Low-Level Waste Facility.

12. Hugh C. Paxton, *Correlations of Experimental and Theoretical Critical Data Comparative Reliability Safety Factors for Criticality Control*, LA-2537-MS, Los Alamos Scientific Lab., March 1961.

13. R. L. Stevenson and R. H. Odegaarden, "Studies of Surface Density Spacing Criteria Using KENO Calculations," originally prepared circa 1970 by the Division of Materials Licensing, U.S. Atomic Energy Commission. Available as Appendix A of the Safety Evaluation Report for the Barnwell Low-Level-Waste Site, License No. 12-13536-01, Amendment 23, U.S. Nuclear Regulatory Commission (April 1991).

14. *SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluations*, NUREG/CR-0200, Rev. 5 (ORNL/NUREG/CSD-2/R5), Vols. I, II, and III, March 1997. Available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-545.