Materials of Criticality Safety Concern in Waste Packages

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ABSTRACT

10 CFR 71.55 requires in part that the fissile material package remain subcritical when considering "the most reactive credible configuration consistent with the chemical and physical form of the material." As waste drums and packages may contain unlimited types of materials, determination of the appropriately bounding moderator and reflector materials to ensure compliance with 71.55 requires a comprehensive analysis.

Such an analysis was performed to determine the materials or elements that produce the most reactive configuration with regards to both moderation and reflection of a Pu-239 system. The study was originally performed for the TRUPACT-II shipping package and thus the historical fissile mass limit for the package, 325 g Pu-239, was used [1]. Reactivity calculations were performed with the SCALE package to numerically assess the moderation or reflection merits of the materials [2]. Additional details and results are given in SAIC-1322-001 [3].

The development of payload controls utilizing process knowledge to determine the classification of special moderator and/or reflector materials and the associated fissile mass limit is also addressed.

WORK DESCRIPTION

Moderator Study

The moderation study evaluated numerous types of materials, including hydrocarbon based materials, inorganic elements, and hydrides. The hydrocarbon based materials examined included plastics, rubbers, lubricants, oils, and other important materials that have high hydrogen contents. The effectiveness of the hydrocarbon was evaluated by mixing the subject moderator with 325 g of Pu-239 forming a spherical mass and reflecting the sphere with 60.96 cm of water to ensure infinite reflection. SCALE calculations were performed for various H/Pu ratios to find the maximum reactivity.

Non-hydrogenous inorganic moderators and hydrides were also analyzed by either (1) adding inorganic materials to an optimized waste mixture containing 325 g of Pu-239 and polyethylene/water; or (2) mixing an inorganic hydride compound with 325 g of Pu-239. In both cases, the fissile mixture was reflected with 60.96 cm of water.

In (1), each non-fissile element in the periodic table was studied provided it was not a known neutron poison, was a solid at room temperature and had cross-section data available in SCALE. Compounds including BeO, SiO_2, MgO and concrete were also analyzed. The composition of the moderating mixture is 25% by volume polyethylene and 75% by volume water, as used in the TRUPACT-II Safety Analysis Report [1]. Calculations were performed with the inorganic material added at 1, 5, 10, 15, 20, 40, 60 and 80% by volume and the H/Pu ratio was varied at each point to determine the maximum reactivity.

In (2), those hydrides that form stable solid compounds and have large scattering cross-sections were investigated, including CaH_2, BaH_2, MgH_2, TiH_2, YH_2, and ZrH_2.

Reflector Study

To determine the reactivity effect of various reflectors, a fissile sphere containing 325 g of Pu-239, again moderated by a mixture of 25% by volume polyethylene and 75% by volume water, was reflected with a myriad of elements and compounds. A reflector thickness of 60.96 cm was used. The H/Pu ratio of the fissile mixture was varied to optimize the reactivity of the system. The reflector materials modeled included every element that is a solid at room temperature and has cross-section data available in SCALE plus numerous compounds including steels, gypsum, concrete, salt, and sand. The materials were modeled at their theoretical density, in a close-fitting spherical configuration around the fissile mixture.

In most cases, this configuration is not credible as the materials in the waste drum are not thick hemispherical shells that could closely reflect the most reactive geometry of a fissile sphere, so calculations were also performed with the materials at 70% of their theoretical density. Because water inleakage must be considered per 10CFR71, the 30% void volume was filled with water and the whole system was backed with 60.96 cm of water.

Payload Controls

The moderator and reflector study demonstrated that system reactivity was primarily a function of the volume fraction of moderating materials in the fissile region and reflecting materials surrounding the fissile region.

The volume fraction of the bounding moderating material (polyethylene) was determined by physical testing for the case where the waste was not machine compacted. The test consisted of determining the volume occupied by a given weight of 6-mil polyethylene sheeting that was periodically subjected to a 100 lb. overburden force to simulate manual loading operations. The resulting polyethylene volume fraction was given by the ratio of the theoretical solid volume occupied by the given weight of polyethylene to that measured by the test and was determined to be 13.36% [4]. Therefore, payload controls for the moderator were developed based on process knowledge of whether the waste was or was not machine compacted. A conservatively bounding packing fraction of 25% was utilized for non-machine compacted waste and 100% was utilized for waste that was machine compacted.

The volume fraction of the special reflector materials was divided into two cases; less than 1% and greater than or equal to 1%. The chemical composition of the waste on a weight percent basis is determined to be conservatively equivalent to volume percent and documented to establish whether the special reflector exists in the waste and whether it exceeds the threshold volume fraction.

Validation

The study provides a comparison of a Pu system reactivity for various moderating and reflecting materials but does not calculate subcritical limits. Thus an Upper Safety Limit determined by a validation study is not needed. The calculations used almost every element with cross-section data available in SCALE and it is recognized that the accuracy of the data for elements that are not commonly present in criticality safety validations is not well known.

RESULTS

The moderators resulting in a reactivity that exceeds that of the 25% polyethylene/75% water moderated system are given in Table I. The results indicate that the reactivity effect of most moderating materials encountered in the nuclear industry can be represented by high-density polyethylene. The only exceptions are a special silicone-based grease/lubricant that is more common in the automotive industry than the nuclear industry and butyl rubbers that are somewhat more reactive than polyethylene but not generally available in substantial quantities. The only inorganic materials that resulted in an increase in reactivity when added to an optimum fissile mixture moderated with water/polyethylene were beryllium and zirconium hydride. Zirconium hydride is not prevalent in nuclear waste either. Thus, with the exception of beryllium, modeling polyethylene in the representative fissile mixture at the maximum packing fraction expected for any moderating material will be conservative. Beryllium must be considered separately if present in the waste stream in significant quantities and thus is termed a "special moderator".

Table II lists the reflector materials that resulted in a higher reactivity than a 25% polyethylene/75% water reflected system. Of the materials evaluated, a beryllium reflector results in the maximum reactivity for the Pu-239 system. When the reflector packing fraction is reduced to 70%, the list of materials that increase reactivity above that of the poly/water mixture is reduced to those shown in Table III. These materials were termed "special reflectors". The packing fraction of some of these reflectors that resulted in a system reactivity equivalent to that of the polyethylene/water mixture was also calculated, as was the reflector thickness at theoretical density when backed by 60.96 cm of water, as given in Table IV.

Thus, the most reactive system would be moderated by polyethylene and reflected by beryllium. The fissile mass limit under these conditions, however, would be very restrictive for drums containing combustibles and similar loadings. Thus, three categories of limits are suggested.

Limit A would be applicable to non-machine compacted waste that contains less than or equal to 1% by weight quantities of special reflector materials. The calculational model would include a 25% by volume polyethylene and 75% by volume water moderator and reflector with 1% by volume beryllium optionally considered in the moderator and/or the reflector.

Limit B would be applicable to non-machine compacted waste that contains greater than 1% by weight quantities of special reflector materials. The reflector would be modeled as beryllium and the moderator would be the polyethylene/water mixture used for Limit A. The addition of beryllium, and zirconium hydride if expected in the waste, into the moderator at various volume fractions would need to be considered.

Limit C would be applicable to machine compacted waste that contains less than or equal to 1% by weight quantities of special reflector materials. For this limit, the moderator and reflector would be 100% polyethylene, unless a smaller packing fraction could be justified based on the compaction scheme utilized, with 1% by volume beryllium considered in either the moderator and/or the reflector. Special reflector materials in greater than 1% by weight quantities are necessarily restricted from shipment in machine compacted waste forms, or a fourth limit would need to be developed.

Table I. Organic Moderating Materials with Higher Reactivity than a Polyethylene/Water Moderated System

Moderator	**Chemical Formula**	**k+2σ**
Silicone Lubricant	$[(CH_3)_3SiOSi(CH_3)_3]n$	1.0089
Polyisobutylene	$[C_4H_8]n$	0.9880
Polyethylene	CH_2	0.9836
Paraffin	$[C_{25}H_{52}]n$	0.9824
Poly(Vinyl Butyral) (Vinyl Acetal)	$[C_4H_6CHOOC_3H_7]_n$	0.9727
Butyl Rubber	$[C_9H_{16}]n$	0.9669
ZrH_2	ZrH_2	0.9629
Polypropylene Oxide (PPO)	$[CH_2CHCH_3O]n$	0.9535
Reference 25% Poly/75% Water Mixture	25% CH_2, 75% H_2O	0.9370

Table II. Reflecting Materials with Higher Reactivity than a Polyethylene/Water Reflected System

Reflector	**k+2σ**	**Reflector**	**k+2σ**
Be	1.1903	Cu	0.9908
BeO	1.1900	Fe	0.9845
C	1.1237	V	0.9781
D_2O	1.0787	Cr	0.9711
MgO	1.0489	SiO_2	0.9710
U (Natural)	1.0431	Oak Ridge Concrete	0.9648
U (0.6% ^{235}U)	1.0367	Mo	0.9560
Pb	1.0319	Co	0.9533
Ni	1.0272	Mn	0.9518
Inconel	1.0233	Nb	0.9507
U (0.3% U-235)	1.0132	Gypsum	0.9476
Stainless Steel 304	1.0091	Sn	0.9446
Zr	1.0006	CH_2	0.9396
Bi	0.9959	Reference 25/75 Poly/Water Mixture	0.9370

Table III. Reflecting Materials with Higher Reactivity than a Polyethylene/Water Reflected System when Modeled at 70% Packing Fraction

Reflector	**k+2σ**	**Reflector**	**k+2σ**
Be	1.0626	Pb	0.9696
BeO	1.0583	Bi	0.9621
U (Natural)	1.0203	Zr	0.9610
C	1.0197	Oak Ridge Concrete	0.9519
U (0.6% ^{235}U)	1.0140	SiO_2 (Wet Sand)	0.9512
D_2O	0.9966	Gypsum	0.9430
MgO	0.9913	Ni	0.9397
U (0.3% ^{235}U)	0.9778	Reference 25% Poly/75% Water Mixture	0.9370

Table IV. Parameters for Special Reflectors to Yield Reactivity Equivalent to 25% Poly/75% Water Reflected System

Reflector	**Equivalent Thickness at 70% Packing Fraction (in)**	**Equivalent Packing Fraction (nearest %) at 24 in. thickness**
Be	0.12	7
BeO	0.16	7
U (Natural)	0.10	1
C	0.25	9
U (0.6% ^{235}U)	0.18	1
D_2O	0.27	14
MgO	0.33	15
U (0.3% ^{235}U)	0.73	5
Pb	0.72	21
Bi	0.86	26
Zr	0.87	24
Oak Ridge Concrete	1.52	34
SiO_2	1.65	33
Gypsum	6.97	N/A
Ni	24.0	70

CONCLUSION

The results of the study indicates that the reactivity state of a fissile waste mixture containing 325 g of Pu-239 is governed by a complex interrelationship between the nuclear characteristics and the moderating and/or reflecting material present, their geometric relationship and concentrations, and neutron energy spectrum. As long as polyethylene is modeled at a packing fraction that exceeds the packing fraction of any hydrocarbon based or hydride materials expected in the payload, it will represent a bounding reactivity state. The only inorganic material that results in an increase in reactivity when added to an optimum fissile mixture moderated with water/polyethylene is beryllium. Special reflector materials including beryllium, beryllium oxide, carbon, heavy water, magnesium oxide and depleted uranium at enrichments ≥0.3 wt% U-235 must be either limited in quantity or in a form that would not allow these materials to tightly surround an optimally moderated fissile mixture; otherwise reduced fissile limits are necessary to ensure the subcriticality of the system.

REFERENCES

1. NRC Docket 71-9218, *TRUPACT-II Safety Analysis Report*, Rev. 21, Washington TRU Solutions LLC, Carlsbad, NM, May 2005.
2. Oak Ridge National Laboratory (ORNL), *SCALE 4.4: Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation for Workstations and Personal Computers*, ORNL/NUREG/CSD-2/R6, March 2000.
3. G.W. Neeley, S.L. Larson, R.J. Green, *Reactivity Effects of Moderator and Reflector Materials on Finite Plutonium Systems*, SAIC-1322-001, Rev. 1, SAIC, Oak Ridge, Tennessee, May 2004.
4. Washington TRU Solutions LLC, *Test Plan to Determine the TRU Waste Polyethylene Packing Fraction*, WP 08-PT.09 Rev. 0, Carlsbad, New Mexico, June 2003.