Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy.

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A Green's Function Approach for Determining Dose Rates for Small Gram Quantities in Shipping Packagings

Steven J. Nathan

Savannah River Nuclear Solutions: Savannah River Site, Aiken, SC, 29808, steven.nathan@srs.gov

INTRODUCTION

The Small Gram Quantity (SGQ) concept is based on the understanding that small amounts of hazardous materials, in this case radioactive materials (RAM), are significantly less hazardous than large amounts of the same materials. This paper describes a methodology designed to estimate an SGQ for several neutron and gamma emitting isotopes that can be shipped in a package in compliance with 10 CFR Part 71 external radiation level limits regulations.

DESCRIPTION OF THE ACTUAL WORK (HEADING A)

The Packaging Certification Program (PCP) Methodology for Determining Dose Rate for Small Gram Quantities in Shipping Packagings provides bounding shielding calculations that define mass limits compliant with 10 CFR 71.47 for a set of proposed SGQ isotopes. The approach is based on energy superposition with dose response calculated for a set of spectral groups for a baseline physical packaging configuration. The methodology includes using the MCNP radiation transport code to evaluate a family of neutron and photon spectral groups using the 9977 shipping package and its associated shielded containers as the base case. This results in a set of Green's Functions (multipliers) for "dose per particle" for each spectral group. For a given isotope, the source spectrum is folded with the Green's Functions to determine the total dose from the RAM in the container.

Analytic Methodology

The Monte Carlo N–Particle (MCNP) code package [1] was used for three dimensional Monte Carlo transport calculations to determine the radiation absorbed dose rates outside the package under Normal Conditions of Transport (NCT). The neutron transport calculations were performed as a series of calculations starting a unit source in each group using the BUGLE 47 group structure [2]. Similarly, photon transport calculations were performed as a series of calculations with a unit source in each group using a 77 group structure. Radiation source spectra and strengths were characterized using ORIGEN-S [3] and RASTA [4].

The 9977 is a single containment drum type package with a bolted flange closure and a 6 inch diameter right circular cylinder Containment Vessel (6CV) enclosed by LAST-A-FOAM® and Fiberfrax® insulation, aluminum load distribution fixtures, and a lid filled with TR-19® Block/Min K® 2000. Major materials of construction include stainless steel, polyurethane, and aluminum.

The drum consists of a stainless steel (SS) outer shell with a SS liner, aluminum load distribution fixtures and miscellaneous other hardware, as shown in Figure 1. The drum is modeled as a right circular cylinder, simplifying some of the components. The simplifications are conservative since they place the source material closer to the surface of the drum being analyzed.

The 6CV is modeled as a cylindrical main portion with a conical transition at the top, a short upper cylinder, a Cone-Seal Plug, and the Cone-Seal Nut. Some of the components of the 6CV that are simple to model (e.g., the conical transition at the top) are included exactly. Other more complex components are modeled as simpler shapes.

Three shielded containers were modeled:

- The SGQ-SC1 Lead Container
- The SGQ-SC2 Polyethylene, Container
- The SGQ-SC3 Tungsten Container

Representing the shielded containers as right circular cylinders simplifies the geometric modeling without impacting the calculated dose rates since the detectors are located on the source axis or mid-plane.

A set of thirty six source isotopes was proposed by personnel at Lawrence Livermore National Laboratory [5]. Thirty five of these isotopes (with the exception being the very short lived Pd-103), listed in Table 1, were analyzed to determine the neutron and photon source strengths. The 47 group structure used to calculate neutron spectra is shown in Table 3. The 77 group structure used to calculate photon spectra is shown in Table 4. The sources were computed as a function of decay time using ORIGEN-S and without decay using RASTA. Actinides were analyzed with varying amounts of beryllium to determine the effects of (□-n) interactions on the neutron source. The added beryllium is modeled as a percent of the actinide mass (e.g., ten percent added beryllium in Pu-238 is modeled as 1 gram of Pu-238 and 0.1 grams of beryllium).

For NCT, the package is considered to be intact and dose rates are calculated on contact at the side, bottom, and top of the package.

The neutron source was modeled as a cylinder 2.5 cm in diameter by 4 cm tall. The source cylinders were run initially as void and then as reduced density plutonium oxide without shielded containers to determine the effects of self-shielding. The gamma source was modeled as a cylinder 1 cm in diameter by 2 cm tall. The source cylinders were run initially as void and then as reduced density (1 g/cc) iron without shielded containers to determine the effects of self-shielding.

Table 1 Typical Sealed Source Isotopes

Nuclide	Half Life (days)	E-max (MeV)	
		Gamma	Alpha
Ac-227	7.95E+03	0.242	4.953
Am-241	1.58E+05	0.060	5.49
Am-241/Be	1.58E+05	0.060	5.49
Am-243	2.69E+06	0.075	5.28
Am-243/Be	2.69E+06	0.075	5.28
Cd-109	4.63E+02	0.088	0
Cf-252	9.66E+02	0.160	6.12
Cm-244	6.61E+03	0.153	5.81
Cm-244/Be	6.61E+03	0.153	5.81
Cm-248	1.27E+08	-	5.078
Co-60	1.93E+03	1.333	0
Cs-137	1.10E+04	0.662*	0
Eu-152	4.93E+03	1.410	0
Fe-59	4.45E+01	1.292	0
Gd-153	2.42E+02	0.103	0
Hf-181	4.24E+01	0.482	0
Ho-166m	4.38E+05	0.951	0
Ir-192	7.38E+01	0.612	0
Mn-54	3.12E+02	0.835	0
Np-237	7.82E+08	0.415*	4.79
Np-237/Be	7.82E+08	0.415*	4.79
Pb-210	8.15E+03	0.047	0
Pm-147	9.58E+02	0.121	0
Po-210	1.38E+02	-	5.304
Pu-238	3.20E+04	0.014	5.5
Pu-238/Be	3.20E+04	0.014	5.5
Pu-239	8.80E+06	0.014	5.16
Pu-239/Be	8.80E+06	0.014	5.16
Pu-240	2.40E+06	0.014	5.168
Pu-240/Be	2.40E+06	0.014	5.168
Pu-241	5.26E+03	0.060*	4.896
Pu-241/Be	5.26E+03	0.060*	4.896
Pu-242	1.37E+08	0.014	4.9
Pu-242/Be	1.37E+08	0.014	4.9
Ra-226	5.84E+05	0.186	4.78
Ru-106	3.73E+02	0.622*	0
Sc-46	8.38E+01	1.120	0
Se-75	1.20E+02	0.401	0
Sm-145	3.40E+02	0.061	0
Sr-90	1.05E+04	0.018*	0
Tm-170	1.29E+02	0.084	0
Yb-169	3.20E+01	0.198	0
Zn-65	2.44E+02	1.116	0
Zr-95	6.40E+01	0.757	0

* Radiation from daughter isotopes

RESULTS

The neutron and photon sources were calculated using both ORIGEN-S and RASTA. The response from a unit source in each neutron and photon group was calculated using MCNP5 with each unshielded and shielded container configuration. Effects of self-shielding on both neutron and photon response were evaluated by including either plutonium oxide or iron in the source region for the case with no shielded container.

For the cases of actinides mixed with light elements, beryllium is the bounding light element. The added beryllium (10 to 90 percent of the actinide mass) in the cases studied represents between 9 and 47 percent concentration of the total mixture mass. For beryllium concentrations larger than 50 percent, the increase in the neutron source term and dose rate tend to increase at a much lower rate than at concentrations lower than 50%. The intimately mixed actinide-beryllium form used in these models is very conservative and thus the limits presented in this report are practical bounds on the mass that can be safely shipped.

The calculated dose rate from one gram of each isotope was then used to determin the maximum amount of a single isotope that could be shipped in the Model 9977 Package (or packagings having the same or larger external dimensions as well as similar structural materials) and have the external radiation level within the regulatory dose limits at the surface of the package. The estimates of the mass limits presented would also serve as conservative limits for both the Models 9975 and 9978 packages. If a package contains a mixture of isotopes, the acceptability for shipment can be determined by a sum of fractions approach.

It should be noted that the SGQ masses presented in this report represent limits that would comply with the external radiation limits under 10CFR Part 71. They do not necessarily bound lower limits that may be required to comply with other factors such as heat load of the package.

REFERENCES

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Table 2 Allowed Mass

Isotope	Allowed Mass of Isotope (g)	
	No Shielded Container	SC1 Lead Container
Ac-227	4.1E-04	5.3E-01
Am-241	2.2E+01	
Am-243	3.2E-01	
Cd-109	5.7E-03	Unlimited*
Cf-252	1.9E-06	
Cm-244	3.9E-01	
Cm-248	1.1E-01	
Co-60	3.3E-06	1.2E-04
Cs-137	1.6E-04	1.1E-01
Eu-152	4.4E-05	2.7E-03
Fe-59	1.6E-07	6.0E-06
Gd-153	1.4E-04	Unlimited*
Hf-181	9.1E-07	1.5E-02
Ho-166m	2.6E-03	7.2E-01
Ir-192	1.0E-06	6.8E-03
Mn-54	1.3E-06	2.0E-04
Np-237	3.2E+01	
Pb-210	2.8E-02	3.0E+02
Pm-147	2.7E-01	Unlimited*
Po-210	2.2E-01	4.0E+01
Pu-238	1.1E+02	
Pu-239	3.5E+03	
Pu-240	3.0E+03	
Pu-241	2.3E+01	
Pu-242	2.5E+03	
Ra-226	5.3E-03	2.3E-01
Ru-106	9.3E-06	2.3E-03
Sc-46	1.3E-07	8.7E-06
Se-75	1.5E-06	1.6E+00
Sm-145	1.4E-02	2.3E+03
Sr-90	2.9E-03	1.4E+00
Tm-170	2.7E-04	7.3E+00
Yb-169	2.1E-06	9.8E+00
Zn-65	1.9E-06	9.5E-05
Zr-95	4.6E-07	1.3E-04

* Unlimited denotes that the allowed mass of the isotope is restricted only by the capacity of the container.

Table 2 Allowed Mass (Continued)

Isotope	Allowed Mass of Isotope (g)	
	SC2 Polyethylene Container	SC3 Tungsten Container
Ac-227	1.4E-03	2.7E+00
Am-241	1.2E+02	
Am-243	1.2E+00	
Cd-109	2.8E-02	Unlimited*
Cf-252	8.6E-06	
Cm-244	1.8E+00	
Cm-248	5.3E-01	
Co-60	9.6E-06	4.5E-04
Cs-137	5.0E-04	6.6E-01
Eu-152	1.3E-04	1.0E-02
Fe-59	4.6E-07	2.2E-05
Gd-153	6.4E-04	Unlimited*
Hf-181	3.0E-06	3.7E-02
Ho-166m	8.3E-03	3.7E+00
Ir-192	3.4E-06	4.1E-02
Mn-54	3.9E-06	1.1E-03
Np-237	2.0E+02	
Pb-210	1.0E-01	1.7E+03
Pm-147	1.2E+00	Unlimited*
Po-210	6.5E-01	2.1E+02
Pu-238	4.7E+02	
Pu-239	1.2E+04	
Pu-240	1.4E+04	
Pu-241	1.2E+02	
Pu-242	1.2E+04	
Ra-226	1.6E-02	7.3E-01
Ru-106	2.9E-05	8.5E-03
Sc-46	3.8E-07	3.7E-05
Se-75	5.6E-06	2.5E+01
Sm-145	7.6E-02	5.5E+03
Sr-90	1.0E-02	5.8E+00
Tm-170	1.0E-03	4.4E+01
Yb-169	8.1E-06	6.3E+01
Zn-65	5.5E-06	3.9E-04
Zr-95	1.4E-06	7.7E-04

* Unlimited denotes that the allowed mass of the isotope is restricted only by the capacity of the container.