2027 Transfer Function Method in Estimating External Radiation Levels

Shivakumar Sitaraman
Lawrence Livermore National Laboratory
Livermore, CA 94550, USA

Brian Anderson
Lawrence Livermore National Laboratory
Livermore, CA 94550, USA

Abstract

The methodology of transfer functions (TF) is based on calculating the contribution to the dose rates generated by a radiation source from each of the discrete energy groups that comprise the spectrum. This set of functions generated can be folded into any source spectrum with the same discrete energy group structure for similar geometry and material properties to estimate the final dose rates of interest, thus avoiding the need for repeated calculations. This approach was used to estimate allowed mass limits for several actinides in pure form or combined with light elements, as well as a set of gamma sources, valid for a set of modern transportation packages used by the US Department of Energy. However, in several instances this approach failed since the estimated masses were non-conservative because subcritical multiplication that enhanced the neutron contribution to the dose rate, was not accounted for in many of the actinides. In other instances, self-shielding was more dominant than subcritical multiplication thus making the estimates too conservative. In other applications the TF method works well. In the case of dose rates outside spent fuel assemblies the TF can be useful in eliminating the need for repeated full radiation transport calculations. This paper will address the issue of the applicability of the TF method with examples and provide a cautionary note on the use of this method.

Introduction

The specific contents approved for shipment in a Type B radioactive material transportation package have historically been descriptions of discrete items, or groupings of well-defined similar items, in the package safety basis documentation [i.e., Safety Analysis Report for Packaging (SARP)]. As new radioactive items were identified that needed to be shipped, an addendum to the safety basis was needed to add these new items. To develop, review and approve numerous addenda over time as more contents need to be added to the safety basis is both expensive and time consuming. Of late, there has been an increasing need at the DOE/NNSA complex to transport a wide range of radioactive materials, including sources and special nuclear materials and other actinides with light element impurities. It was thus in the interest of the community to develop a broad set of contents for both neutron and gamma emitting materials compliant with regulations pertaining to external radiation limits that can be transported in modern shipping containers. This effort involved a large set of both actinides in pure form or combined with varying amounts of light element impurities as well as several gamma sources. This led to the need for a way to minimize the number of calculations that

would otherwise be required for such a large set of contents.

The proposal was to adopt a transfer function (TF) method where a set of functions could be defined for a specific energy group structure that could be used repeatedly for folding in an actual source spectrum and summing over the energy groups to obtain a desired quantity such as a dose rate at a specific location. The method involved starting a single source particle in each energy group and estimating its contribution to the dose rate at specified locations. An identical energy group structure is used to obtain a source spectrum based on a specific mass, e.g. 1g, and multiplying the source term in each group by the corresponding transfer function and summing over all energies. This would then give the dose rate at a specific location on the basis of 1g used to derive the source. This quantity could then be scaled up to determine a mass that would be compliant with the external radiation limits prescribed by the regulations.

An early study established a set of mass limits for actinides in pure form and with beryllium impurity as well as for gamma sources [1]. This methodology was adopted to establish an expanded content envelope containing actinides combined with several light elements in proportions ranging from 0.1% to 90% by mass in the mixture. A modern transportation packaging used by the USDOE, Model 9977, was used as the basis of developing this set since its use would be bounding for Models 9975 and 9978. Mass limits were also established for a set of gamma sources. In all cases design decay heat limits were taken into account in developing the content envelope. The results of this study were published in 2012 [2]. However, upon examining a few random cases from this study it was determined that one very important aspect was potentially neglected in using this method- subcritical multiplication was ignored when dealing with these actinide isotopes. Further study revealed that this was indeed the case and several of the actinide as well as gamma source mass limits were non conservative. This paper will present some results from the study that was done to remedy the issues resulting from the neglect of subcritical multiplication and address the limitations of the TF method.

However, there are instances where this method can be successfully used to minimize the number of separate calculations that would otherwise be needed. A case in point is the estimation of the dose rate outside a spent fuel assembly and the paper will present details from this study.

Content Envelope Development for Modern Packagings

Model used for Calculations

The Model 9977 Packaging was used as the basis for the development of the updated content envelope. The actinides- ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu+^{241}Am, ^{242}Pu, ^{244}Cm, ^{243}Am, ^{237}Np, ^{248}Cm, and ^{252}Cf- in pure form or in combination with eleven common light elements – Be, B, F, Li, Na, Mg, Al, C, O, Cl, and Si- were included in the study. ORIGEN-S [3] was used to develop source terms and

the radiation transport Monte Carlo code, MCNP [4], was used to obtain dose rates. Starting with the mass values presented in Ref. 2, the masses were determined by a few repeated calculations. The limiting dose rates were at the surface of the package where a maximum of 2 mSv (200 mrem) per hour is permitted per the regulations [5] [6]. The masses were determined such that the limiting dose rates were between 1.85 and 1.95 mSv/h (185 and 195 mrem/h) giving an average margin of 5% to the regulatory limit. This packaging was also authorized to have shielded inserts of polyethylene for the neutron sources and lead or tungsten for the gamma sources. The contents could also be held within the packaging in an engineered container that was treated as the unshielded case for the study. The package decay heat limit for the engineered container and tungsten shield was 19W, with the lead and polyethylene decay heat limits being 6W and 3W, respectively. The physical source was modeled as pure actinide to maximize any subcritical multiplication while the actual source spectrum had the correct combination of actinide and light element in proportion ranging from 0.1% to 90% by mass of the light element. The source was placed at the bottom of the containment vessel for the unshielded case (see Figure 1) and on the bottom of the shielded container and up against its wall to minimize the distance to the package surface (see Figure 1). All of these model features added some conservatism to the final set of derived mass limits. The neutron source included both the contribution from the (α, n) reaction with light elements as well as the spontaneous fission source.

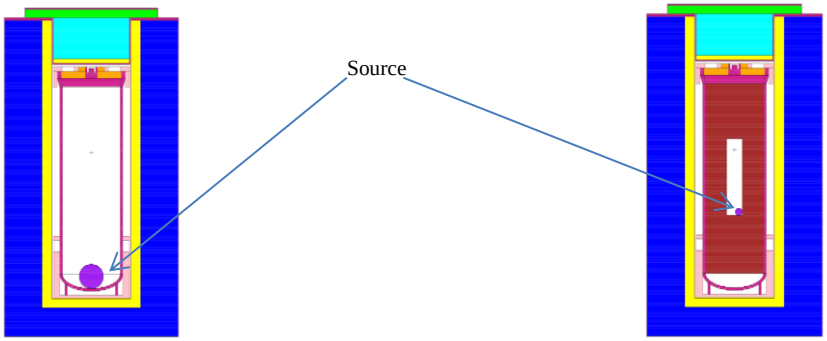

Figure 1 MCNP Model

Actinide Contents

Since the intent of the paper is emphasize the issues related to the applicability of the TF method, only the change of the limits from the previous set using the TF method will be presented for selected actinides. The full set of mass limits and the changes from the previous study can be found in RAMPAC [7]. The ratios that are less than one indicate that the TF method had produced non-conservative results. Table 1 shows the results for ^{238}Pu in an unshielded form and Table 2 presents the results for ^{244}Cm in the polyethylene shielded container. The green shaded values indicate limits due to design decay heat that were taken into account previously and thus show no change for the weaker (α, n) sources. It can be seen that for the stronger (α, n) sources combined with

^{238}Pu, the old values obtained using the TF method were non-conservative by up to 33%. In the case of ^{244}Cm many combinations were limited by the 3W decay heat limit of the polyethylene shield though here too the non-conservatisms were as much as 17%.

Table 1 Change from TF Method-^{238}Pu Bare

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
0.10%	0.77	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
0.50%	0.69	0.76	1.00	0.93	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1%	0.67	0.72	1.00	0.80	1.00	1.00	1.00	1.00	1.00	1.00	1.00
5%	0.66	0.67	0.77	0.70	1.00	1.00	0.87	0.82	1.00	1.00	1.00
10%	0.66	0.67	0.73	0.68	1.00	0.93	0.78	0.75	1.00	1.00	1.00
30%	0.66	0.66	0.69	0.67	1.00	0.94	0.72	0.70	1.00	1.00	1.00
50%	0.66	0.66	0.68	0.67	1.00	0.75	0.70	0.71	1.00	1.00	1.00
70%	0.66	0.66	0.67	0.67	1.00	0.74	0.70	0.69	1.00	1.00	1.00
90%	0.66	0.66	0.67	0.67	1.00	0.73	0.69	0.69	1.00	1.00	0.92

Table 2 Change from TF Method-^{244}Cm Shielded

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
0.10%	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
0.50%	0.87	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1%	0.83	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
5%	0.83	0.83	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
10%	0.83	0.83	1.00	0.94	1.00	1.00	1.00	1.00	1.00	1.00	1.00
30%	0.83	0.83	1.00	0.89	1.00	1.00	1.00	1.00	1.00	1.00	1.00
50%	0.83	0.83	1.00	0.86	1.00	1.00	1.00	1.00	1.00	1.00	1.00
70%	0.83	0.83	1.00	0.89	1.00	1.00	1.00	1.00	1.00	1.00	1.00
90%	0.83	0.83	1.00	0.89	1.00	1.00	1.00	1.00	1.00	1.00	1.00

Conversely, some of the TF mass limits were too conservative thus unnecessarily penalizing the shipper. This can be seen in Table 3 where for the high levels of fluorine impurity in ^{241}Am+^{241}Pu, the previous limits were too limiting though for Be and B, the old masses were still for the most part non-conservative.

Table 3 Change from TF Method-^{241}Am+^{241}Pu Shielded

Impurity Content	Be	B	Li	F	C	Al	Mg	Na	Si	O	Cl
0.10%	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
0.50%	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
1%	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
5%	0.90	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
10%	0.88	1.03	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
30%	0.83	0.93	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
50%	0.84	0.93	1.00	1.11	1.00	1.00	1.00	1.00	1.00	1.00	1.00
70%	0.84	0.91	1.00	1.06	1.00	1.00	1.00	1.00	1.00	1.00	1.00
90%	0.83	0.91	1.00	1.05	1.00	1.00	1.00	1.00	1.00	1.00	1.00

Overall, for each of the bare actinides evaluated, about 45% were too high or too low with 10%

unchanged mainly due to decay heat limits. In the shielded form, almost 71% were unchanged mainly due to the lower decay heat limitations with remaining limits too high. Some of the shielded masses were limited by the size of the cavity in the polyethylene container, e.g. pure ^{242}Pu. In conclusion, the mass limits that were not limited by decay heat were either non-conservative or too conservative depending on whether subcritical multiplication or self-shielding was the dominant factor.

Gamma Contents

A set of twenty four gamma sources were included in the study. The previous TF-based study used a small voided vial as the source volume. The current study uses a sphere that in many instances is very small. Table 4 presents the change in the allowable mass from the TF method generated set. Two thirds of the unshielded gamma mass limits were non-conservative and about a quarter of the mass were too conservative. Isotopes with high and mid-energy gammas were responsible for the non-conservatism. With the lead shield insert about 46% were non-conservative with some of the mid-energy gamma sources becoming a too conservative. All the low energy sources that were too conservative in the unshielded case were now limited by decay heat. With the more effective tungsten shield, 33% of the masses were non-conservative with a few more sources becoming too conservative compared to the lead shield. In the case of the gamma sources, self-shielding increased as the mass and volume of the source sphere increased leading to either over conservatism compared to the TF generated masses or decay heat limitations. Four of the heavy gamma sources are also neutron emitters via the (α, n) reaction with light elements. This impact on the mass limits for ^{227}Ac, ^{210}Pb, ^{210}Po, and ^{226}Ra are being evaluated.

Table 4 Change from TF Method-Gamma Sources

Source	No Shield new/old	Lead new/old	Tungsten new/old	Source	No Shield new/old	Lead new/old	Tungsten new/old
Ac-227	1.01E+00	1.00E+00	1.00E+00	Pm-147	1.15E+01	1.00E+00	1.00E+00
Cd-109	9.37E+00	1.00E+00	1.00E+00	Po-210	1.00E+00	1.00E+00	1.00E+00
Co-60	7.47E-01	9.33E-01	9.25E-01	Ra-226	7.97E-01	9.58E-01	9.76E-01
Cs-137	8.04E-01	9.25E-01	1.32E+00	Ru-106	6.43E-01	6.63E-01	7.31E-01
Eu-152	7.75E-01	8.75E-01	9.87E-01	Sc-46	7.52E-01	9.69E-01	9.02E-01
Fe-59	7.37E-01	9.56E-01	9.36E-01	Se-75	9.24E-01	1.00E+00	1.00E+00
Gd-153	2.49E+00	1.00E+00	1.00E+00	Sm-145	1.15E+01	1.00E+00	1.00E+00
Hf-181	7.84E-01	1.43E+00	6.95E+00	Sr-90	3.43E-01	3.85E-01	3.96E-01
Ho-166m	8.20E-01	4.11E+02	7.69E+02	Tm-170	1.74E+00	1.00E+00	1.00E+00
Ir-192	8.26E-01	1.09E+00	1.53E+00	Yb-169	1.00E+00	1.00E+00	1.00E+00
Mn-54	7.57E-01	9.74E-01	1.03E+00	Zn-65	7.51E-01	9.02E-01	9.52E-01
Pb-210	6.11E-01	1.00E+00	1.00E+00	Zr-95	7.76E-01	9.38E-01	1.07E+00

no change
conservative
non-conservative

Dose Rates outside Spent Nuclear Fuel

The TF method was applied to determine the dose rates outside spent nuclear fuel assemblies. Two main characteristics of the spent fuel assembly is that the dose rate is dominated by gamma radiation that is being emitted by a fixed fuel matrix with a fixed high Z, high density material. The goal here is to estimate the dose rate rather than adhere to any regulatory or other limits. After a period of about 2 years, the main source of gammas is ^{137}Cs, a fission product with a long half-life and a fission yield that is essentially the same whether it is produced from ^{235}U or ^{239}Pu fissions. The gamma source term increases linearly with burnup. Though the neutron contribution to the dose rate is extremely small, this component of the total dose rate was also calculated to determine if the effects of burnup and cooling time will have an impact on the validity of the TF method.

The calculations were performed by generating source terms in a twenty group structure for gammas and a 47-group structure for neutron using ORIGEN-S. Source spectra were obtained at four different burnups ranging from 25 GWd/t to 37 GWd/t based on a spent fuel assembly from an operating reactor. The cooling times ranged from 5 to 40 years. Transfer functions were obtained based on a 33 GWd/t burnup at a cooling time of 20 years. Full calculations were performed for each cooling time at a burnup of 33 GWd/t as well as for the different burnups at 20 years cooling time with appropriate isotopic composition of the fuel material. Source terms were based on one fuel pin and scaled up to represent the whole assembly. Dose rates shown are at 15 cm from assembly.

Table 5 Spent Fuel Assembly Dose Rates: Varying Burnups

Total Dose Rate				Neutron Dose Rate			
Burnup (GWd/t)	TF Method (Sv/h)	Full Calculation (Sv/h)	Ratio	Burnup (GWd/t)	TF Method (Sv/h)	Full Calculation (Sv/h)	Ratio
24.9	1.38E+03	1.38E+03	1.00E+00	24.9	8.13E-03	9.18E-03	8.90E-01
29	1.61E+03	1.62E+03	1.00E+00	29	1.60E-02	1.71E-02	9.40E-01
33.2	1.85E+03	1.86E+03	9.90E-01	33.2	2.90E-02	2.91E-02	9.90E-01
37.3	2.09E+03	2.10E+03	1.00E+00	37.3	4.87E-02	4.68E-02	1.04E+00

Table 6 Spent Fuel Assembly Dose Rates: Varying Cooling Times

Cooling Time (years)	TF Method (Sv/h)	Full Calculation (Sv/h)	Ratio	Cooling Time (years)	TF Method (Sv/h)	Full Calculation (Sv/h)	Ratio
5	4.89E+03	4.90E+03	1.00E+00	5	4.95E-02	5.12E-02	9.70E-01
10	2.75E+03	2.76E+03	1.00E+00	10	4.13E-02	4.25E-02	9.70E-01
15	2.16E+03	2.17E+03	9.90E-01	15	3.45E-02	3.50E-02	9.90E-01
20	1.85E+03	1.86E+03	9.90E-01	20	2.90E-02	2.91E-02	9.90E-01
30	1.43E+03	1.43E+03	1.00E+00	30	2.05E-02	2.04E-02	1.00E+00
40	1.12E+03	1.13E+03	9.90E-01	40	1.47E-02	1.45E-02	1.00E+00

Tables 5 and 6 show the comparisons of the dose rates from the TF method and full calculations for varying burnup and cooling time, respectively. The total dose rates, dominated by gammas, are the same regardless of the method in both tables. The negligible neutron contribution is also shown in these tables. In Table 5, the neutron dose rate shows differences as the burnup changes from the reference burnup used in the TF calculations. The isotopics are different at these burnups and this impacts the dose rate due to subcritical multiplication. In the case of the neutron dose rates with varying cooling times shown in Table 6, the difference between the two methods is not very significant since the main fissile material quantities at the fixed burnup, except for the small amount of ^{241}Pu, change negligibly over this span of cooling times. Once again, it is clear that the neutron portion of the dose rate can show variation between the TF method and the full calculations in a multiplying medium like spent fuel.

Conclusions

The transfer function method, more often than not, fails when it is applied to determining allowable amounts (mass or activity) of radioactive material in shipping packages. Use of the method can lead to material limits that produce non-compliant levels of external radiation. Conversely the results of the TF method could be overly conservative, unnecessarily penalizing the shipper by limiting the amount of material that can be shipped. The issue in question that determines which of these holds true depends on whether subcritical multiplication or self-shielding is more dominant.

In other cases such as for estimating dose rates outside spent fuel assemblies, and potentially spent fuel shipping casks, the method works well when the dose rate is dominated by the gamma contribution.

While the TF method can be a powerful tool in saving time and effort by reducing the number of calculations required to estimate dose rates or other quantities of interest, it should be used with caution to ensure that the accuracy of the needed quantity is not compromised.

Acknowledgments

The authors wish to thank the Office of Packaging and Transportation-EM33 of the USDOE for their support of this project.

This work was performed under the auspices of the US Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344.

References

1. S.J. Nathan, J.M. Risner, and S. Sitaraman, “Packaging Certification Program Methodology for Determining Dose Rates for Small Gram Quantities in Shipping Packages,” PCP-2011-0001, DOE Packaging Certification Program, August 2011.

2. S. Sitaraman, S. Kim, and B. Anderson, “Functional Bounding Content Envelope for Type B Radioactive Material Transportation Packages,” 53rd Annual Meeting of the INMM, Orlando, Florida, July 2012.

3. ORNL/TM-2005/39, Version 6.1, ORIGEN-S: Scale System Module to Calculate Fuel Depletion, Actinide Transmutation, Fission Product Buildup and Decay, and Associated Radiation Source Terms, ORNL, June 2011.

4. T. Goorley, et al., "Initial MCNP6 Release Overview", Nuclear Technology, 180, pp 298-315 (Dec 2012).

5. Packaging and Transportation of Radioactive Material, Code of Federal Regulations, Title 10, Part 71, US Nuclear Regulatory Commission, Washington, DC, (Federal Register 2004).

6. Regulations for the Safe Transport of Radioactive Materials, SSR-6, International Atomic Energy Agency, 2012 Edition.

7. **http://rampac.energy.gov/reference/knowledge/whitepapers/default.aspx**