EVALUATION OF THE SUM-OF-FRACTIONS METHODOLOGY FOR WATER AND POLYETHYLENE MODERATED SYSTEMS

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ABSTRACT

Sum-of-Fractions is a method intended to assure a subcritical margin for aqueous solutions and slurries of fissionable isotopes. The method indicates that a system is subcritical if the sum of the ratios of the mass of each isotope in a mixture to its individual minimum subcritical mass limit is less than or equal to one. The basis of the Sum-of-Fractions has historically been derived from allowances given in ANSI/ANS-8.15-1981. However, the allowance was removed in ANSI/ANS-8.15-2014 due to a lack of technical basis.

A methodology was developed to assess the validity of using the Sum-of-Fractions for water or polyethylene moderated systems for the following nuclides: ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{237}Np, ^{236}Pu, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{247}Cm, ^{249}Cf, *and* ^{251}Cf. *The methodology uses available benchmark data for mixtures of* ^{233}U, ^{235}U, *and* ^{239}Pu *to establish the calculational margin, and a mass limit reduction to establish the margin of subcriticality. Water or polyethylene moderated and reflected mixtures containing the nuclides are evaluated with SCALE 6.2.4.*

Including the calculational margin, subcritical mass limits for each nuclide were computed for optimally water or polyethylene moderated and fully reflected systems. These masses were used to create nuclide mixtures in which the sum of the mass to subcritical mass limit ratios is one. The various nuclide mixtures were modeled over a range of moderation and demonstrate the k_{eff} *does not exceed the calculational margin. For additional assurance of subcriticality, a significant mass reduction is applied to each computed minimum critical mass of the nuclides without adequate benchmark data consistent with the method in ANSI/ANS-8.15-2014.*

KEYWORDS

Criticality safety, Sum-of-Fractions, actinide, critical mass, moderation

1. INTRODUCTION

Methods for addressing multiple nuclides have been devised over the years to minimize efforts in criticality safety analysis. Some of these methods stem from the concept detailed in the American National Standards Institute (ANSI)/American Nuclear Society (ANS) ANSI/ANS-8.15-1981 section 5.2 statement "…the sum of the ratios of the mass of each fissile nuclide to its limit does not exceed unity…"[1]. In the context of the standard, the statement comes with caveats on the system composition and geometrical configuration, however, some of the methods derived extend the use of the sum of the ratios beyond what is permitted in the standard. Three methods of note that derive their basis from this statement are as follows:

- Rule-of-Fractions – Utilizes the ANS-8.15-1981 sum of ratios that applies to water moderated and water reflected systems.
- Fissile Gram Equivalent – A variant of the Rule-of-Fractions that is restricted to a specific moderated and reflected system.
- Sum-of-Fractions (SoF) – A variant of the Rule-of-Fractions that can be extended to a variety of moderated and reflected systems.

Sum-of-Fractions is a method to assure a subcritical margin for aqueous solutions and slurries of fissile and fissionable isotopes. Sum-of-Fractions suggests that, for mixtures containing one or more fissile or fissionable isotopes, if the sum of the ratios of the mass of each isotope in the mixture to its individual subcritical mass limit is less than or equal to one the mixture is assumed to maintain a subcritical margin. In equation form the method can be defined by:

$$\sum_i \frac{a_i}{A_i} \leq 1 \qquad (1)$$

Where a_i is the mass of isotope i present and A_i is the mass that corresponds to the minimum subcritical mass for isotope i.

The use of Rule-of-Fractions was permitted in ANSI/ANS-8.15-1981 for aqueous solutions and slurries reflected by water of unlimited thickness [1]. In part, this was allowed as the mass limits in ANSI/ANS-8.15-1981 were felt to have significant margins that would compensate for any potential anomalous results[1].

However, due to limitations in determining if the limits produced using Sum-of-Fractions were safe/conservative, the allowance was removed in ANSI/ANS-8.15-2014 [2]. The current replacement is for each user to calculate their own subcritical limits for mixtures of materials, resulting in potentially repeated efforts among the criticality safety community. The work described in this analysis provides a technical basis and method for the criticality safety community and permits again the use of a commonly utilized method for generating limits for mixtures of nuclides. Bias, and uncertainty in bias consistent with ANSI/ANS-8.24-2017 [3] and subcritical mass reduction factors in line with ANSI/ANS-8.15.2014 Appendix C methodology [2] for special actinides is placed on the results to assure that application of the Sum-of-Fractions is subcritical.

The Sum-of Fractions extension is based on the idea that a minimum subcritical mass for a system can be calculated for a specific nuclide, i.e., ^{235}U, and the masses of other nuclides can be included in the rule-of fractions approach if the minimum subcritical masses for all the nuclides is based on a system that is more reactive than the specific system evaluated. An example would be to calculate the minimum subcritical mass of ^{235}U for a water moderated unreflected system, then using the minimum subcritical mass for water moderated water reflected systems for any of the other nuclides that might be mixed in the specific system being evaluated.

2. METHODOLOGY

The limitations in benchmark experiments prevent the direct determination of an upper subcritical limit (USL) for most actinides. To address this limitation an approach is devised that utilized benchmarks where available to develop a subcritical margin in conjunction with biases on the actinide masses. The available benchmarks provide evidence of the methods ability to handle mixtures of fissile isotopes via

[1] The Rule-of-Fractions assumes that using a mixture of actinides based on the minimum subcritical mass is less reactive than any individual actinide's minimum subcritical mass at optimum concentration. There have been reports that sometimes the mix can be slightly more reactive.

the calculational margin. The accuracies of the nuclides cross sections are then considered based on benchmark data availability.

First, a set of ^{233}U, ^{235}U, and ^{239}Pu mixtures are used to determine an appropriate bias and uncertainty in bias [This does not include the margin of subcriticality (MOS) for code and data uncertainties nor the MOS for validation applicability]. This is achieved by modeling mixes of the three isotopes near optimal moderation and full reflection. Sensitivity analyses are then performed on these mixes to provide the appropriate bias and bias uncertainty. Although the bias and uncertainty in bias originate from only a few isotopes, the intent of this is to demonstrate the code's ability to handle mixtures of fissile isotopes.

Secondly, the minimum subcritical mass for each actinide is computed for an optimally moderated and fully reflected system using SCALE 6.2.4 [4]. Calculations at various densities are performed in the optimally moderated realm to determine the mass at the desired eigenvalue. Following the calculations, a polynomial fit is utilized to determine the minimum subcritical mass. Benchmark data for the actinides of ANSI/ANS-8.15-2014 [2] are limited or nonexistent; therefore, the bias and bias uncertainty of the individual actinides cannot be evaluated. In lieu of this, the bias and bias uncertainty evaluated for the ^{233}U, ^{235}U, and ^{239}Pu mixture cases are applied when computing the minimum subcritical masses for the actinides. Given that these three isotopes are the most well studied isotopes out of the actinides, it would be expected that at least the bias and bias uncertainty of ^{233}U, ^{235}U, and ^{239}Pu mixtures would need to be applied to the remaining actinides.

Thirdly, mixtures containing an actinide at 1/6 their evaluated minimum subcritical mass and 5/6 the minimum subcritical mass of ^{233}U, ^{235}U, or ^{239}Pu are evaluated in the optimally moderated range. The resulting k-eff is assessed to determine if it falls below critical, including the bias and bias uncertainty of the ^{233}U, ^{235}U, or ^{239}Pu mixtures assessed. If it is above, the actinide minimum subcritical mass is reduced and the case is recomputed. This step checks for and corrects for anomalous mixtures of actinides.

Finally, minimum subcritical masses verified in the previous step are taken and safety margin mass reduction factors are applied based on the confidence of the cross-section accuracy. This considers the lack of experimental data as well as reservations associated with the uncertainties of the actinide cross sections. For actinides that do not have available experimental critical benchmarks, a mass penalty is included on the minimum subcritical mass to assure subcriticality; consistent with the approach used in ANSI/ANS-8.15-2014 as described in Appendix C [2].

3. VALIDATION

The validation approach considers mixtures of ^{233}U, ^{235}U, and ^{239}Pu in a range of water moderated systems with a thick water reflector. A validation study was performed with each mixture and concentration and a complete description of the validation methodology can be found in a companion paper by Oak Ridge National Laboratory [5]. Not all mixtures had sufficient benchmark experiments to determine a bias and uncertainty in bias; those that have sufficient data showed that a calculated k_{eff} of less than 0.98 are subcritical. There was sufficient coverage of the mixtures to interpolate that all mixtures were covered. This demonstrates that code can calculate water moderated systems with multiple fissile nuclides.

The same validation approach was taken with mixtures of ^{233}U, ^{235}U, and ^{239}Pu in polyethylene with a thick polyethylene reflector. There were less applicable benchmark experiments that covered the polyethylene mixtures than with the water. Mixtures that had sufficient data showed that a calculated k_{eff} of less the 0.965 are subcritical. There is sufficient coverage to the mixture to interpolate that all the mixtures are covered.

This validation demonstrates that the code can be used with mixtures of well validated fissile nuclides and the other nuclides that do not have benchmark experiments provided an adequate additional margin is included.

The validation applicability for the Sum-of-Fractions method is determined based on the range of the energy of average lethargy causing fission (EALF) for the cases used in the validation study. The EALF range is 0.035 – 0.090 eV for the water moderated and reflected systems and 0.034 – 0.082 eV for the polyethylene moderated and reflected systems.

4. MINIMUM CRITICAL MASS

Minimum critical masses were computed utilizing 10000 neutrons per generation for 200 active generations and 50 skipped generations. Masses were computed for an eigenvalue of 0.9775 ± 0.0025 for water moderated and reflected systems and 0.9625 ± 0.0025 for polyethylene moderated and reflected systems. Minimum subcritical masses for individual fissile isotopes were estimated based off a polynomial fit to data over the data range of densities. These masses are used to generate minimum subcritical mass mixtures with the Sum-of-Fractions. Values presented are rounded down to the nearest 1000 grams for isotopes with subcritical masses that exceed 10000 grams, 100 grams for isotopes with subcritical masses that exceed 1000 grams, 50 grams for isotopes subcritical masses that exceed 100 grams. The isotopes with subcritical masses below 100 grams are rounded down to the nearest gram. The resulting minimum subcritical masses are presented in Table I.

Table I. Subcritical masses for water and polyethylene moderated and reflected systems

Nuclide	Water Mass (grams)	Polyethylene Mass (grams)
U-233	500	250
U-235	700	400
Pu-239	450	250
U-232	3300	2900
U-234	100000	90000
Np-237	47000	44000
Pu-236	1000	600
Pu-238	6300	5800
Pu-240	31000	29000
Pu-241	250	100
Pu-242	59000	55000
Am-241	56000	52000
Am-242m	21	11
Am-243	120000	108000
Cm-242	9800	9000
Cm-243	200	100
Cm-244	20000	19000
Cm-245	58	33
Cm-246	65000	60000
Cm-247	1100	650
Cf-249	58	33
Cf-251	27	15

5. MIXTURE EVALUATION

With the critical masses of Table I, a set of mixtures are evaluated consisting of five-sixths of the minimum subcritical mass of either ^{233}U, ^{235}U, and ^{239}Pu and one-sixth the minimum subcritical mass of one of the nuclides from ANSI/ANS-8.15-2014 [2]. Calculations were performed where the fissionable mass of the system is set such that the mass remained constant and moderation is adjusted to find the peak reactivity. Results of the calculations are provided in Table II and indicate that the peak $k_{eff}+2\sigma$ does not exceed the calculational margins for the water or polyethylene moderated and reflected systems, respectively.

The results demonstrate that for the mixtures containing only fissile isotopes, the reactivity of the system approaches the calculational margin as the moderator conditions needed for peak reactivity for the individual isotopes are close to the optimal moderator density of the mixture. However, when one is fissile and the other fissible[2], the difference in the peak reactivity and the calculational margin is greater due to the large differences in the moderator conditions needed for the individual isotope to be at its peak reactivity.

Table II. Peak keff+2σ for water and polyethylene moderated and reflected systems

	Water			Polyethylene		
Actinides	**U-233**	**U-235**	**Pu-239**	**U-233**	**U-235**	**Pu-239**
U-232	0.94460	0.95325	0.95936	0.91788	0.95464	0.95785
U-234	0.69193	0.68034	0.68686	0.65213	0.65001	0.65339
Np-237	0.68804	0.67331	0.68838	0.65988	0.65371	0.66329
Pu-236	0.97117	0.97020	0.97235	0.93390	0.96216	0.96080
Pu-238	0.66346	0.61320	0.66999	0.63505	0.60340	0.63979
Pu-240	0.68379	0.65930	0.67622	0.65067	0.63757	0.64615
Pu-241	0.97272	0.97275	0.97387	0.92016	0.95183	0.95114
Pu-242	0.75434	0.73505	0.74328	0.71013	0.70779	0.71319
Am-241	0.66221	0.65124	0.66054	0.64177	0.63613	0.64149
Am-242m	0.96940	0.96659	0.96620	0.92844	0.95487	0.95351
Am-243	0.68449	0.68054	0.68890	0.65719	0.65839	0.66441
Cm-242	0.86693	0.88840	0.90021	0.79658	0.85772	0.87052
Cm-243	0.97070	0.96670	0.96969	0.92501	0.95282	0.95283
Cm-244	0.78007	0.81321	0.83216	0.71151	0.75483	0.77501
Cm-245	0.96671	0.96500	0.96487	0.92613	0.95339	0.95102
Cm-246	0.86514	0.88055	0.88915	0.80447	0.85556	0.86265
Cm-247	0.97597	0.97401	0.97720	0.93719	0.96479	0.96445
Cf-249	0.96783	0.96444	0.96513	0.92713	0.95415	0.95145
Cf-251	0.97240	0.96958	0.97093	0.93252	0.96118	0.95740

6. MARGIN OF SUBCRITICALITY

To address the lack of benchmarks and uncertainties in cross sections for the ANSI/ANS-8.15 nuclides, a mass penalty is applied to the computed subcritical masses via a subcritical factor consistent with Appendix C of ANSI/ANS-8.15-2014 [2]. This subcritical factor is extended to polyethylene moderated and reflected systems. The subcritical masses computed are then compared to the subcritical masses in ANSI/ANS-8.15-2014 [2] and the lower minimum critical mass for each actinide is selected as the subcritical mass for use with the Sum-of-Fractions. Table III and Table IV provide the resulting masses.

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[2] Fissible refers to a nuclide that cannot support a slow-neutron chain reaction but is only capable of a fast neutron chain reaction, provided that the effective fast-neutron production cross section exceeds the effective fast neutron removal cross section [6].

Table III. Subcritical masses in water moderated and reflected systems

Nuclide	Computed Mass (g)	factors	Computed w/ factors (g)	ANSI 8.15 (g)	SoF Mass (g)
U-233	500	1	500	--	500
U-235	700	1	700	--	700
Pu-239	450	1	450	--	450
U-232	3300	0.5	1650	1000	1000
U-234	100000	0.5	50000	59000	50000
Np-237	47000	0.7	32900	35000	32900
Pu-236	1000	0.5	500	600	500
Pu-238	6300	0.7	4410	5100	4410
Pu-240	31000	0.7	21700	20000	20000
Pu-241	250	0.7	175	185	175
Pu-242	59000	0.7	41300	55000	41300
Am-241	56000	0.5	28000	24000	24000
Am-242m	21	0.5	10.5	11	10.5
Am-243	120000	0.5	60000	65000	60000
Cm-242	9800	0.5	4900	6000	4900
Cm-243	200	0.5	100	90	90
Cm-244	20000	0.5	10000	11000	10000
Cm-245	58	0.5	29	23	23
Cm-246	65000	0.5	32500	16000	16000
Cm-247	1100	0.5	550	500	500
Cf-249	58	0.5	29	10	10
Cf-251	27	0.5	13.5	5	5

Table IV. Subcritical masses in polyethylene moderated and reflected systems

Nuclide	Computed Mass (g)	factors	Computed w/ factors (g)	ANSI 8.15 (g)	SoF Mass (g)
U-233	250	1	250	--	250
U-235	400	1	400	--	400
Pu-239	250	1	250	--	250
U-232	2900	0.5	1450	1000	1000
U-234	90000	0.5	45000	59000	45000
Np-237	44000	0.7	30800	35000	30800
Pu-236	600	0.5	300	600	300
Pu-238	5800	0.7	4060	5100	4060
Pu-240	29000	0.7	20300	20000	20000
Pu-241	100	0.7	70	185	70
Pu-242	55000	0.7	38500	55000	38500
Am-241	52000	0.5	26000	24000	24000
Am-242m	11	0.5	5.5	11	5.5
Am-243	108000	0.5	54000	65000	54000
Cm-242	9000	0.5	4500	6000	4500
Cm-243	100	0.5	50	90	50
Cm-244	19000	0.5	9500	11000	9500
Cm-245	33	0.5	16.5	23	16.5
Cm-246	60000	0.5	30000	16000	16000
Cm-247	650	0.5	325	500	325
Cf-249	33	0.5	16.5	10	10
Cf-251	15	0.5	7.5	5	5

7. APPLICATION

The Sum-of-Fractions method is applied to a nuclide mixture similar to irradiated targets used for californium production (Table V) [7]. The weight fraction of each isotope in the mixture is used in conjunction with the Sum-of-Fractions subcritical masses to determine the mixture subcritical mass of 768 grams. Utilization of the ANSI/ANS-8.15-2014 [2] would restrict the subcritical mass of the mixture to the most limiting nuclide subcritical mass, which is the ^{251}Cf mass limit of 5 grams.

Table V. Example of subcritical masses determined by the SoF method for californium production

Nuclide	Weight Percent	Nuclide Masses (g) at SoF = 1
pu-238	0.060	0.461
pu-239	0.036	0.276
pu-240	33.581	257.902
pu-241	0.121	0.929
pu-242	5.023	38.577
am-241	0.591	4.539
am-243	3.829	29.407
cm-244	10.914	83.820
cm-245	2.563	19.684
cm-246	40.955	314.534
cm-247	2.261	17.364
cf-249	0.054	0.415
cf-251	0.012	0.092

To demonstrate the 768 gram limits applicability, the mixture mass reactivity is evaluated for various moderations at a mass of 1977 grams. The mass represents the computed subcritical masses, which do not include the subcritical factors. As shown in Fig. 1, the peak reactivity is 0.47716 (k_{eff}+2σ = 0.47818) in the unmoderated region of the reactivity curve. Assessing the mixture with fissible nuclides removed results in an increase in reactivity in the moderated region and results in a peak k_{eff} of 0.94968 (k_{eff}+2σ = 0.95140) as shown in Fig. 2. For both methods of assessment, the peak k_{eff} falls below the calculational margin of 0.980. The validation applicability is also provided to show the moderation ranges covered by the validation. If a process/operation does not fall under the domain of the validation applicability, it would need to be expanded via additional validations.

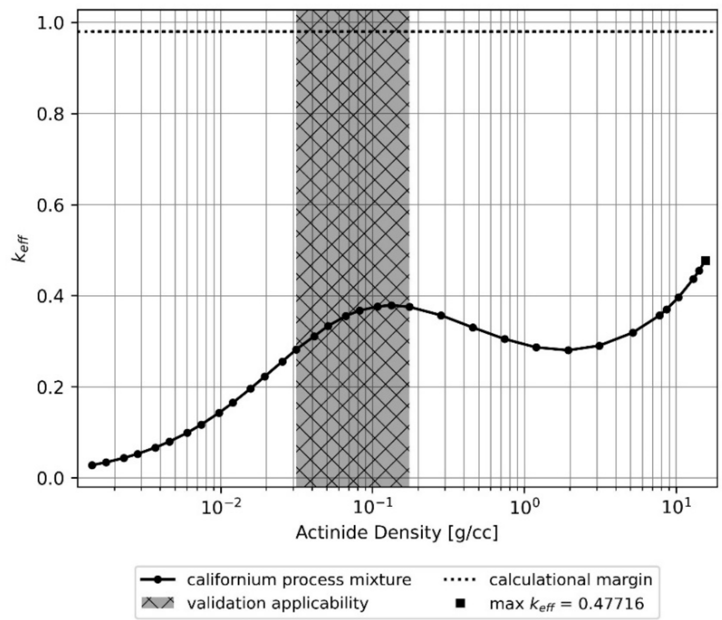

Figure 1. Reactivity of californium process mixture at SoF of one for various moderations; fissile and fissible nuclides

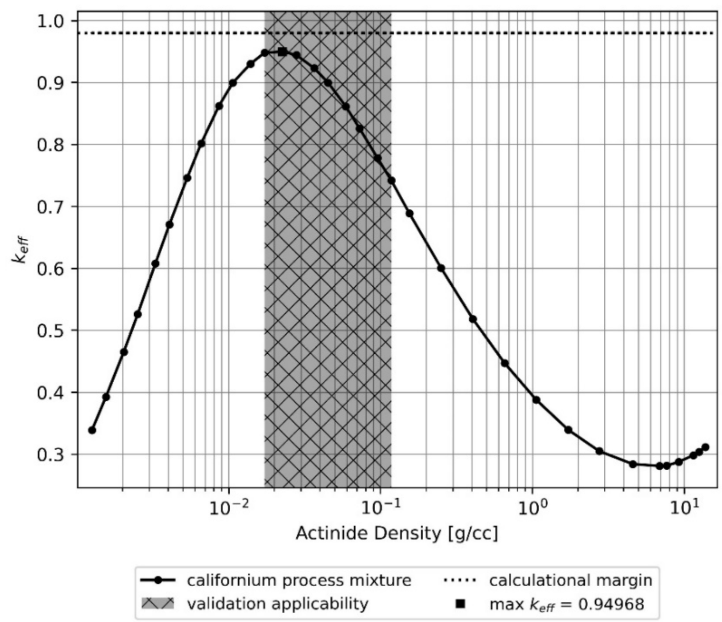

Figure 2. Reactivity of californium process mixture at SoF of one for various moderations; fissile only nuclides

8. CONCLUSIONS

A Sum-of-Fractions method was developed in this work for homogenous water and polyethylene moderated and reflected systems. The method addressed uncertainties with the models as well as cross section libraries. This is accomplished through a calculational margin derived through sensitivity and uncertainty analyses using SCALE/TSUNAMI [4] and a margin of safety through the use of subcritical factors that are consistent with Appendix C of ANSI/ANS-8.15-2014 [2]. Selection of the final subcritical masses for the nuclides was based on the minimum critical mass between the evaluated values and the ANSI/ANS-8.15-2014 [2] values for water moderated and reflected systems.

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