

Equivalence Relations for the 9972-9975 SARP (U)

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**KEYES A. NIEMER
ROBERT L. FROST**

OCTOBER 1994

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ABSTRACT

Equivalence relations required to determine mass limits for mixtures of nuclides for the Safety Analysis Report for Packaging (SARP) of the Savannah River Site 9972, 9973, 9974, and 9975 shipping casks were calculated. The systems analyzed included aqueous spheres, homogeneous metal spheres, and metal ball-and-shell configurations, all surrounded by an effectively infinite stainless steel or water reflector. Comparison of the equivalence calculations with the rule-of-fractions showed conservative agreement for aqueous solutions, both conservative and non-conservative agreement for the metal homogenous sphere systems, and non-conservative agreement for the majority of metal ball-and-shell systems. Equivalence factors for the aqueous solutions and homogeneous metal spheres were calculated. The equivalence factors for the non-conservative metal homogeneous sphere systems were adjusted so that they were conservative. No equivalence factors were calculated for the ball-and-shell systems since the SARP assumes that only homogeneous or uniformly distributed material will be shipped in the 9972-9975 shipping casks, and an unnecessarily conservative critical mass may result if the ball-and-shell configurations are included.

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INTRODUCTION

Equivalence relations based on the rule-of-fractions method have been used for some time in criticality analysis. These relations are useful because they allow determination of critically safe masses for a combination of fissile and/or fissionable nuclides based on the critically safe mass of a reference isotope. However, equivalence relations have not previously been applied to shipping cask criticality analysis. Previous applications of equivalence relations have been restricted to binary systems of isotopes. This report describes a study of equivalence relations for several higher order systems representative of fuel to be shipped in the 9972-9975 series of shipping casks. The equivalence relations developed here are used in the determination of critically safe masses of each fuel type.

The shipment of fissionable materials is regulated by the NRC, 10 CFR 71.¹ In past permit applications, relatively simple fuel content descriptions were used for various reasons:

1. In order to be conservative, no credit was taken for fission products when shipping spent nuclear fuel.
2. For ease of analysis, shipments within the weapons complex containing ^{235}U and/or ^{239}Pu with other actinide isotopes were analyzed simply as containing ^{235}U and/or ^{239}Pu .

In the current regulatory environment, it is not acceptable to ignore the other actinide isotopes; their contribution to the system reactivity must be known. The fuel to be shipped in the 9972-9975 series of shipping casks spans a wide range of actinide isotopes with sliding envelopes of content ranges. For instance, one envelope contains 0-95% ^{239}Pu , 0-50% ^{240}Pu , 0-2% ^{241}Pu , and 0-5% ^{242}Pu . This is interpreted as meaning the fuel could contain 50% ^{240}Pu or no ^{240}Pu , etc. There are 13 such content envelopes (only six are discussed in this report), and 4 different shipping casks to be analyzed. Direct calculations to find the most limiting cases would be a tremendously complex task. Instead, equivalence relations can be developed for the other isotopes using ^{235}U and ^{239}Pu as reference isotopes. The criticality analysis can then be performed for ^{235}U and ^{239}Pu only, and the safe fuel envelope masses can be determined from the equivalence relations. This report discusses the development of the appropriate equivalence relations for the 9972-9975 series of shipping casks.²

DISCUSSION

Fissile material to be shipped in the 9972-9975 series of shipping casks originates from several different sources. The material to be shipped can be delineated into six^a isotopic content envelopes as shown in Table 1. Equivalence relations were developed to determine safe mass limits for each of the content envelopes based on safe masses of the reference isotopes ^{235}U and ^{239}Pu .

^a There are actually 13 content envelopes, but only six have been considered to date.

Equivalence relations are based on the rule-of-fractions from American National Standard ANSI/ANS-8.15, Section 5.2, which states that the sum of the ratios of the mass of each fissile nuclide to its limit does not exceed unity for well-moderated, water-reflected systems.³ The rule can be extended to fast systems by adjusting the equivalence value as demonstrated in this report. The rule-of-fractions can be expressed as follows:

$$\sum_i \frac{m_i}{M_i} \leq 1.0 \quad (1)$$

where m_i is the mass of nuclide i , and M_i is the mass limit of nuclide i . Equation 1 can be rewritten using equivalence factors, EF_i , such that the mass of different nuclides in a mixture multiplied by an equivalence factor will be less than or equal to the mass limit of a reference nuclide, M :

$$\sum_i EF_i * m_i \leq M \quad (2)$$

where $EF_i = \frac{M}{M_i}$. (3)

For example, with two nuclides A and B , the rule-of-fractions is expressed as⁴

$$\frac{m_A}{M_A} + \frac{m_B}{M_B} \leq 1.0 \quad (4)$$

which can be rewritten as

$$m_A \frac{M_B}{M_A} + m_B \leq M_B. \quad (5)$$

Using an equivalence factor, EF_A , and B as the reference nuclide, equation 5 becomes

$$m_A EF_A + m_B \leq M_B \quad (6)$$

where $EF_A = \frac{M_B}{M_A}$. (7)

The implication of using an equivalence relation is that the multiplication factor is not increased when the mixture is formed.⁴

Previous studies⁴ have demonstrated that the rule-of-fractions is a good estimate of the reactivity of binary mixtures if they are well-moderated and water-reflected. For fast systems, a plot of k_{eff} versus concentration of one of the isotopes follows two possible scenarios: 1) all mixture k_{eff} 's

fall below the rule-of-fractions line (Figure 1), or 2) all mixture k_{eff} 's lie above the rule-of-fractions line (Figure 2). The rule-of-fractions is conservative with respect to the first scenario and non-conservative with respect to the second. In the latter case, the equivalence value can be increased until the highest point on the curve of k_{eff} versus concentration falls below the rule-of-fractions line, as demonstrated in Figure 2. This is the technique employed in the current work.

Table 1. Isotopic ranges of cases analyzed.^b

Isotope		Weight % of Isotopes					
		Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
²³⁵ U	Fissile	0	0	0-95	0	0	0-100
²³⁶ U	Fissionable	0	0	0	0	0	0-40
²³⁸ U	Fissionable	0	0	0-100	0	0	0-100
²³⁸ Pu	Fissionable	0	0	0	0-100	0	0
²³⁹ Pu	Fissile	0-95	0-95	0-100	0-40	0-95	0
²⁴⁰ Pu	Fissionable	0-50	0-50	0-50	0-13	0-50	0
²⁴¹ Pu	Fissile	0-2	0-2	0-2	0-1	0-2	0
²⁴² Pu	Fissionable	0-5	0-5	0-5	0-1.5	0-5	0
²⁴¹ Am	Fissionable	0-5	0-50	0	0	0-5	0

^b For each isotope, the maximum weight percent that could be contained in each packaging group is listed. Thus, the total percentages for each group sum to more than 100%. When finding the mass limits for a particular content group, the most reactive combination of isotopes that sum to 100% was used. The contents listed in this table are only those considered important for the criticality analysis. Several other fissionable isotopes are not listed because the maximum weight percentages of those isotopes result in extremely small isotopic masses. For example, content groups 1-5 allow a maximum of 1×10^{-4} wt. % of ²⁴³Am. The maximum mass limit for these content groupings is 4.4 kg, which results in an ²⁴³Am mass less than 0.44 g, an inconsequential quantity when compared to ²⁴³Am's critical mass of 54 kg (bare) and 36 kg (stainless steel reflected).

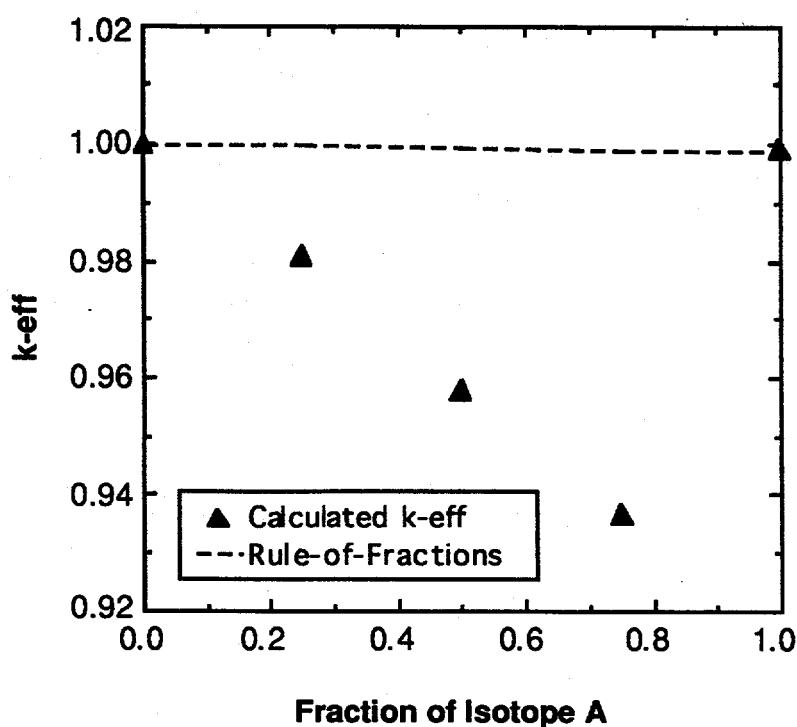


Figure 1. Example of conservative case with calculated k_{eff} 's falling below the rule-of-fractions line.

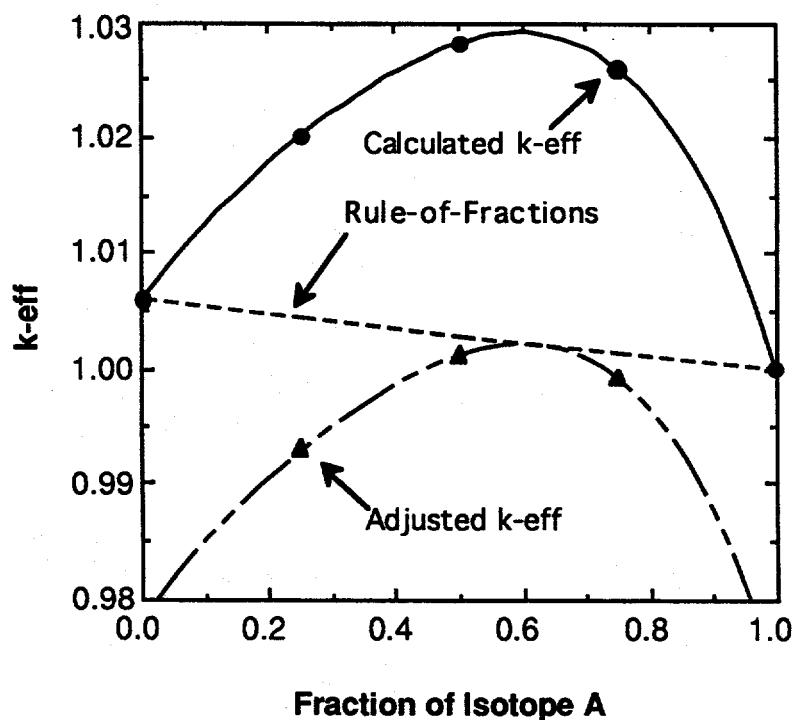


Figure 2. Example of non-conservative case with calculated k_{eff} 's lying above the rule-of-fractions line.

METHODOLOGY

Equivalence relations were calculated using the LAW 44-energy group cross section library, developed at Oak Ridge National Laboratory⁵ and based on ENDF/B-V and VI data. The LAW 44-energy group library was validated at SRS for relevant systems and isotopes in the SARP work.⁶⁻⁹ For this equivalence analysis, the cross sections were processed with the AJAX, BONAMI, NITAWL, and ALPO modules of the AMPX-77¹⁰ modular code system to create an ANISN library. In thermal systems where resonance overlap was a concern, the ROLAIDS module was used in place of the BONAMI and NITAWL modules for resonance processing. The ANISN library was used by the ONEDANT module of TWODANT-SYS¹¹ to compute eigenvalues for each mixture. Equivalence factors for these mixtures were calculated such that the rule-of-fractions guaranteed a conservative result.

Table 2 lists the isotopes and type of systems analyzed. Equivalence factors based on binary mixtures were calculated and are expected to hold for higher order systems. The first step in calculating equivalence factors for different mixtures was to compute the critical mass of each isotope. One-dimensional spherical models of the metal and aqueous systems were prepared. The models consisted of a fissile core (either metal or aqueous) surrounded by a 20-cm thick reflector (a water reflector for one set of equivalence factors and a stainless steel reflector for a second set of equivalence factors). The critical mass of each individual isotope was computed using a dimension search to vary the radius of the fissile core while keeping the thickness of the reflector region constant. A constant k_{eff} was used in the ONEDANT dimension search. For ^{235}U and ^{239}Pu , the bias adjusted k_{eff} from previous validation work was used in the search.⁶⁻⁸ For the other isotopes, a k_{eff} of 1.0 was used in the search since no validation data were available.

After calculating the critical mass of each isotope, binary mixtures were formed based on equation 4. Mixtures were formed using fractions of the isotopes' critical mass such that the sum of the fractions equaled 1.0:

$$f_A + f_B = 1.0 \quad (8)$$

where

$$f_A = \frac{m_A}{M_A} \quad (9)$$

and

$$f_B = \frac{m_B}{M_B} \quad (10)$$

Note that f_A and f_B are not weight fractions of the isotopes in the mixture; they are the fraction of the critical mass of isotopes A and B.

Five combinations of f_A and f_B were formed for the aqueous and metal homogeneous systems (Table 3). Two endpoints were formed consisting of 100% of the critical mass of each isotope (isotopes A and B), and three intermediate mixtures were formed consisting of: 1) 25% of the

critical mass of isotope A ($f_A=0.25$) and 75% of the critical mass of isotope B ($f_B=0.75$), 2) 50% of the critical mass of isotope A ($f_A=0.50$) and 50% of the critical mass of isotope B ($f_B=0.50$), and finally 3) 75% of the critical mass of isotope A ($f_A=0.75$) and 25% of the critical mass of the isotope B ($f_B=0.25$). Four combinations of f_A and f_B were formed for the metal ball-and-shell systems (Table 4). Two endpoints were formed consisting of 100% of the critical mass of each isotope. One intermediate point was formed consisting of 50% of the critical mass of isotope A ($f_A=0.50$) as the ball and 50% of the critical mass of isotope B ($f_B=0.50$) as the shell. A second intermediate point was similarly formed using isotope B ($f_B=0.50$) as the ball and isotope A ($f_A=0.50$) as the shell. The mass, m , of each isotope in the mixture was calculated using equations 9 and 10. The radius of the mixture and atomic number density of the isotopes were then calculated from m as shown in Appendix A.

All mixtures were modeled as either aqueous spheres, homogeneous metal spheres, or metal ball-and-shell configurations. The aqueous systems were surrounded by a 20-cm thick water reflector, and the metal systems were surrounded by either a 20-cm thick stainless steel (type 304) or water reflector. The LAW cross sections were processed with the appropriate AMPX-77 modules to generate an ANISN library for each mixture. The ANISN library was used by ONEDANT to compute eigenvalues for each mixture using S_8 quadrature and P_3 Legendre scattering, which was consistent with previous validation work.⁶⁻⁹

Since the rule-of-fractions is defined for aqueous solutions and slurries, it had to be validated for the metal systems. Therefore, the computed k_{eff} for each mixture was compared to the expected rule-of-fractions value. If the computed k_{eff} was less than or equal to the rule-of-fractions value, then the rule-of-fractions was conservative for that system (Figure 1), and the equivalence factor was calculated using equation 7. However, if the computed k_{eff} was greater than the rule-of-fractions value (Figure 2), then the mass limit for the nonreference nuclide was reduced until the maximum computed k_{eff} was less than the rule-of-fractions value. For this case, the equivalence factor for the mixture was computed as follows:

$$EF_A = \frac{M_B}{M'_A}, \quad (11)$$

where M'_A is the reduced mass limit for the nonreference nuclide.

Table 2. Isotopes and type of systems analyzed.

Isotopes	Type of System
^{235}U - ^{239}Pu	Aqueous Sphere ^c Metal Homogeneous Sphere Metal Ball-and-Shell
^{235}U - ^{240}Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
^{239}Pu - ^{238}Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
^{239}Pu - ^{240}Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
^{239}Pu - $^{241}\text{Pu}^{\text{d}}$	Aqueous Sphere ^c Metal Homogeneous Sphere
^{239}Pu - $^{242}\text{Pu}^{\text{d}}$	Metal Homogeneous Sphere
^{239}Pu - ^{241}Am	Metal Homogeneous Sphere Metal Ball-and-Shell

^c The aqueous sphere system was only analyzed for cases which could contain a mixture of two fissile nuclides. All the other cases contain mixtures of fissile and fissionable nuclides. Aqueous mixtures of fissile and fissionable nuclides will be less reactive than an aqueous solution of the fissile nuclide alone.

^d These mixtures were analyzed after all the other mixtures were analyzed; based on the previous analyses, it was determined that no equivalence factors would be calculated for the ball-and-shell systems since the SARP assumes that only homogeneous or uniformly distributed material will be shipped in the 9972-9975 shipping casks, and an unnecessarily conservative critical mass may result if the ball-and-shell configurations are included. Thus no ball-and-shell systems were analyzed for these mixtures.

Table 3. Percentage of critical mass of isotopes in aqueous and metal homogeneous mixtures.

Mixture	f_A Fraction of Isotope A Critical Mass	f_B Fraction of Isotope B Critical Mass
1 (endpoint)	0	1.00
2 (intermediate)	.25	.75
3 (intermediate)	.50	.50
4 (intermediate)	.75	.25
5 (endpoint)	1.00	0

Table 4. Percentage of critical mass of isotopes in metal ball-and-shell mixtures.

Mixture	Description	f_A Fraction of Isotope A Critical Mass	f_B Fraction of Isotope B Critical Mass
1 (endpoint)	Isotope A is Ball	0	1.00
2 (intermediate)	Isotope A is Ball / Isotope B is Shell	.50	.50
3 (intermediate)	Isotope B is Ball/ Isotope A is Shell	.50	.50
4 (endpoint)	Isotope B is Ball	1.00	0

RESULTS

Equivalence relations for five isotopic content ranges of nuclides were calculated. The ranges correspond to Cases 1-5 in Table 1. Case 6 was dismissed since it contained only uranium, and the limits for these isotopes could be obtained from references for natural and enriched uranium.¹² The systems analyzed included aqueous spheres, homogeneous metal spheres, and metal ball-and-shell configurations, all surrounded by an effectively infinite stainless steel or water reflector.

The fission spectrum, χ , has a significant effect on the computed k_{eff} for the metal systems. A composite χ for each mixture was produced by weighting the χ 's for each isotope based on a fission rate, $v\Sigma_f\phi$, weighting. For each mixture, ONEDANT was first executed to calculate the $v\Sigma_f\phi$ for each isotope using the χ for one of the isotopes in the mixture. The computed $v\Sigma_f\phi$'s were then normalized and used to weight the χ 's for each isotope to form a composite χ for the mixture. ONEDANT was then re-executed with the composite χ , and the $v\Sigma_f\phi$'s calculated with the composite χ were verified to ensure that the weighting had not changed with the composite χ .

Tables 5 and 6 list the critical radii and the critical masses for the isotopes considered. These values were used in computing the k_{eff} 's and rule-of-fractions values for all the cases using both a water and stainless steel reflector. Appendix B lists the calculated k_{eff} 's, the rule-of-fractions values, and the differences between the k_{eff} and rule-of-fractions values for all mixtures with a H_2O reflector; and Appendix C lists the same information for mixtures with a stainless steel reflector. Comparison of the computed k_{eff} 's with the rule-of-fractions values showed conservative agreement for aqueous solutions; both conservative and non-conservative agreement for the metal homogenous sphere systems, which was not surprising since the rule-of-fractions is defined for aqueous solutions or slurries,³ and non-conservative agreement for the majority of the metal ball-and-shell systems.

For the conservative aqueous solutions and conservative homogeneous metal systems, the equivalence factors were calculated using equation 7. For the non-conservative homogeneous metal systems, the mass limit of the nonreference nuclide was reduced until the maximum computed k_{eff} was less than the rule-of-fractions value, and the equivalence factors were calculated using equation 11.

Table 7 and 8 list the mass limits and the equivalence factors for the cases with aqueous spheres, and Tables 9 and 10 list the mass limits and the equivalence factors for the cases with homogenous metal spheres.

As stated above, the majority of cases with metal ball-and-shell configurations were found to be non-conservative. The rule-of-fractions grossly underpredicted k_{eff} for ball-and-shell configurations when a plutonium isotope formed the ball part of the system. The ability of TWODANT and the LAW 44-energy group library to accurately calculate eigenvalues for these systems was verified by modeling critical experiments.^{8,9} In fact, the critical experiments showed that the critical mass of a metal plutonium ball - uranium shell system is less than the critical

mass of a uranium or plutonium system alone.^{8,9} Thus the results observed in this work are consistent with experiments. In order for a ball-and-shell configuration to be credible, the fissile fuel must be very heterogeneous and must consist of large pieces of which at least one is a concave surface. Three types of solid fuel are to be shipped in the 9972-9975 shipping casks: loose oxide powders, ash, and metal. Only the metal is a concern for a ball-and-shell configuration. Of the six content envelopes listed in Table 1, only cases 5 and 6 represent materials in metallic form. Case 6 contains only uranium isotopes and can be dismissed, but case 5 is a potential concern since it contains ^{239}Pu , ^{240}Pu , and ^{241}Am . However, the fuel corresponding to case 5 consists of metal pieces (about 2.2 kg each), two of which will be loaded into a single shipping cask. The pieces have a uniform isotopic distribution and are not concave. Thus, the ball-and-shell configuration is not relevant for the 9972-9975 shipping casks, and no equivalence factors for ball-and-shell configurations were calculated for this work. Note that if ball-and-shell configurations were considered, unnecessarily conservative critical masses would result. Potential users of the equivalence factors listed in this report are advised to confirm that ball-and-shell configurations are not relevant to their process. Revised equivalence factors are required if ball-and-shell systems (or systems that approximate them, such as two adjacent plates each of different isotopic composition) are considered.

Table 5. Critical radii and masses for isotopes in aqueous mixture with 20-cm H₂O reflector.^e

Isotope	Critical Radii (cm)	Critical Mass (g)
²³⁵ U ^f	15.07	774.68
²³⁹ Pu ^f	15.93	508.82
²⁴¹ Pu ^f	12.98	275.01

Table 6. Calculated critical radii and masses for isotopes in metal systems with either a 20-cm H₂O reflector or a 20-cm type 304 stainless steel reflector.

Isotope	Critical Radii (cm)		Critical Mass (kg)	
	H ₂ O Reflector	Stainless Steel Reflector	H ₂ O Reflector	Stainless Steel Reflector
²³⁵ U	6.49	6.13	21.52	18.15
²³⁸ Pu	4.47	3.86	7.41	4.77
²³⁹ Pu	4.03	3.82	5.45	4.62
²⁴⁰ Pu	7.18	6.07	30.84	18.72
²⁴¹ Pu	4.07	3.96	5.67	5.22
²⁴² Pu	9.43	7.92	70.53	41.77
²⁴¹ Am	11.58	9.83	89.13	54.47

Table 7. Mass limits and equivalence factors for aqueous mixtures with ²³⁵U as the reference nuclide.^g

Isotope	Mass Limit (g)	Equivalence Factor
	H ₂ O ^h Reflector	H ₂ O ^h Reflector
²³⁵ U	774.68	1.00
²³⁹ Pu	508.82	1.53

^eThe aqueous sphere was only analyzed with a H₂O reflector because the critical values calculated for an aqueous solution with an H₂O reflector are more conservative than those calculated with a stainless steel reflector.

^f²³⁵U critical radius and mass based on ²³⁵U concentration of 54 g/l, which corresponds to the minimum critical mass for homogenous water-moderated ²³⁵U spheres.¹³ ²³⁹Pu and ²⁴¹Pu critical radii and masses based on Pu concentration of 30 g/l, which corresponds to the minimum critical mass for homogenous water-moderated Pu spheres.¹³

^g No limits or equivalence factors were developed for the ball-and-shell geometries.

^h A 20-cm thick water reflector.

Table 8. Mass limits and equivalence factors for aqueous mixtures with ^{239}Pu as the reference nuclide.^g

Isotope	Mass Limit (g)	Equivalence Factor
	H_2O^h Reflector	H_2O^h Reflector
^{239}Pu	508.82	1.00
^{241}Pu	275.01	1.86

Table 9. Mass limits and equivalence factors for metal homogeneous mixtures with ^{235}U as the reference nuclide.^g

Isotope	Mass Limit (kg)		Equivalence Factor	
	H_2O^h Reflector	SS ⁱ Reflector	H_2O^h Reflector	SS ⁱ Reflector
^{235}U	21.52	18.15	1.00	1.00
^{239}Pu	5.45	4.62	3.95	3.93
^{240}Pu	20.00	14.40	1.08	1.27

Table 10. Mass limits and equivalence factors for metal homogeneous mixtures with ^{239}Pu as the reference nuclide.^g

Isotope	Mass Limit (kg)		Equivalence Factor	
	H_2O^h Reflector	SS ⁱ Reflector	H_2O^h Reflector	SS ⁱ Reflector
^{238}Pu	6.10	4.58	0.90	1.01
^{239}Pu	5.45	4.62	1.00	1.00
^{240}Pu	29.70	18.56	0.19	0.25
^{241}Pu	5.45	5.22	1.00	0.89
^{242}Pu	51.00	41.77	0.11	0.12
^{241}Am	89.13	54.47	0.07	0.09

ⁱ A 20-cm thick type 304 stainless steel reflector, density equals 7.92 g/cm^3 , 19 wt% Ni, 2 wt% Mn, 69.5% Fe, and 9.5 wt% Cr.¹⁴

CONCLUDING REMARKS

Equivalence relations for seven binary combinations of nuclides were calculated. The systems analyzed included aqueous spheres, homogeneous metal spheres, and metal ball-and-shell configurations, all surrounded by an effectively infinite stainless steel or water reflector. Comparison of the equivalence calculations with the rule-of-fractions showed conservative agreement for aqueous solutions, and both conservative and non-conservative agreement for the metal homogenous sphere systems. The majority of cases with metal ball-and-shell configurations were found to be non-conservative. Equivalence factors for the aqueous solutions and homogeneous metal spheres were calculated. The equivalence factors for the non-conservative metal homogeneous sphere systems were adjusted so that the equivalence factors were conservative. No equivalence factors were calculated for the ball-and-shell systems since the SARP assumes that only homogeneous or uniformly distributed material will be shipped in the 9972-9975 shipping casks, and an unnecessarily conservative critical mass may result if the ball-and-shell configurations are included. Confirmation of these equivalence factors for higher order systems of isotopes is underway.

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APPENDIX A: ATOMIC NUMBER DENSITIES

$$\text{Atomic number density } N = \frac{.60221 * \frac{\text{gm}}{\text{cm}^3}}{A} [\text{atm / b-cm}]$$

where A is the atomic mass.

Aqueous Solutions

$$\text{Mass of nuclide } i = m_i = f_i * M_i [\text{g}],$$

where M_i is the critical mass of nuclide i , and f_i is the fraction of critical mass.

$$\text{Volume of nuclide } i = V_i = \frac{m_i}{CS_i} [\text{l}],$$

where CS_i is the critical concentration of nuclide i .

$$\text{Volume of mixture} = V_{\text{tot}} = \sum_i V_i [\text{l}].$$

$$\text{Radius} = R = \left(\frac{3V_{\text{tot}} * 1000}{4\pi} \right)^{\frac{1}{3}} [\text{cm}].$$

$$\text{Concentration of nuclide } i = C_i = \frac{m_i}{V_{\text{tot}}} [\text{g / cm}^3].$$

$$\text{Concentration of H}_2\text{O} = C_{\text{H}_2\text{O}} = \left(1 - \sum_i \frac{C_i}{\rho_i} \right) \rho_{\text{H}_2\text{O}} [\text{g / cm}^3],$$

where ρ is the density.

$$\text{Atomic number density of nuclide } i = N_i = \frac{.60221 * C_i}{A_i} [\text{atm / b-cm}].$$

$$\text{Atomic number density of O} = N_{\text{O}} = N_{\text{H}_2\text{O}} = \frac{.60221 * C_{\text{H}_2\text{O}}}{A_{\text{H}_2\text{O}}} [\text{atm / b-cm}].$$

$$\text{Atomic number density of H} = N_{\text{H}} = 2N_{\text{H}_2\text{O}} [\text{atm / b-cm}].$$

Metal Systems**Homogeneous metal spheres**Mass of nuclide $i = m_i = f_i * M_i$ [g],where M_i is the critical mass of nuclide i , and f_i is the fraction of critical mass.Volume of nuclide $i = V_i = \frac{m_i}{\rho_i}$ [cm³],where ρ_i is the density of nuclide i .Volume of mixture = $V_{tot} = \sum_i V_i$ [cm³].Radius of homogeneous sphere = $R = \left(\frac{3V_{tot}}{4\pi} \right)^{\frac{1}{3}}$ [cm].Concentration of nuclide $i = C_i = \frac{m_i}{V_{tot}}$ [g / cm³].Atomic number density of nuclide $i = N_i = \frac{.60221 * C_i}{A_i}$ [atm / b - cm]**Metal ball-and-shells**Mass of nuclide $i = m_i = f_i * M_i$ [g],where M_i is the critical mass of nuclide i , and f_i is the fraction of critical mass.Volume of nuclide $i = V_i = \frac{m_i}{\rho_i}$ [cm³],where ρ_i is the density of nuclide i .Radius of ball $b = R_b = \left(\frac{3V_b}{4\pi} \right)^{\frac{1}{3}}$ [cm].Outer radius of shell $s = R_s = \left(\frac{3 \sum_i V_i}{4\pi} \right)^{\frac{1}{3}}$ [cm].Atomic number density of nuclide $i = N_i = \frac{.60221 * \rho_i}{A_i}$ [atm / b - cm].

Water Reflector

$$N_O = N_{H_2O} = \frac{.60221 * \rho_{H_2O}}{A_{H_2O}} \text{ [atm / b - cm]}$$

$$N_H = 2N_{H_2O} \text{ [atm / b - cm].}$$

Stainless Steel Reflector

Type 304L stainless steel reflector, density = 7.92 g/cm³, 19 wt% Ni, 2 wt% Mn, 69.5% Fe, and 9.5 wt% Cr.¹⁴

$$N_i = \frac{.60221 * \text{wt\%} * \rho_{ss}}{A_i} \text{ [atm / b - cm].}$$

All atomic number densities used in this analysis are listed in reference 2.

APPENDIX B: DATA FOR MIXTURES WITH H₂O REFLECTOR

Table B-1 lists the isotopes and type of systems initially analyzed with a 20-cm H₂O reflector. These mixtures were analyzed using a constant density for all the isotopes of an element, no density adjustments were performed for isotopic variations. Tables B-2 and B-3 list the critical radii and masses for isotopes listed in Table B-1. Tables B-4 to B-8 list the calculated k_{eff} 's, the rule-of-fractions (ROF) values, and the differences between the k_{eff} and ROF values for mixtures of the isotopes listed in Table B-1. Table B-9 and B-10 list the critical radii and masses calculated with isotopic density adjustments. In addition, the isotopes ²⁴¹Pu and ²⁴²Pu were added to the list of isotopes to be analyzed. Tables B-11 and B-12 list the calculated k_{eff} 's, the rule-of-fractions (ROF) values, and the differences between the k_{eff} and ROF values for mixtures of ²³⁹Pu-²⁴¹Pu and ²³⁹Pu-²⁴²Pu. Equivalence factors listed in Tables 7-10 of the report were based on the isotopic density adjusted critical masses.

Table B-1. Isotopes and type of systems analyzed with a 20-cm H₂O reflector.

Isotopes	Type of System
²³⁵ U- ²³⁹ Pu	Aqueous Sphere ^{b1} Metal Homogeneous Sphere Metal Ball-and-Shell
²³⁵ U- ²⁴⁰ Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
²³⁹ Pu- ²³⁸ Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
²³⁹ Pu- ²⁴⁰ Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
²³⁹ Pu- ²⁴¹ Am	Metal Homogeneous Sphere Metal Ball-and-Shell

^{b1} The aqueous sphere system was only analyzed for cases which could contain a mixture of two fissile nuclides. All the other cases contain mixtures of fissile and fissionable nuclides. Aqueous mixtures of fissile and fissionable nuclides will be less reactive than an aqueous solution of the fissile nuclide alone.

Table B-2. Critical radii and masses for isotopes in aqueous mixture with 20-cm H₂O reflector.

Isotope	Critical Radii (cm)	Critical Mass (g)
²³⁵ Ub ^{b2}	15.07	774.68
²³⁹ Pu ^{b3}	15.93	508.82

Table B-3. Critical radii and masses for isotopes in metal systems with a 20-cm H₂O reflector.^{b4}

Isotope	Critical Radii (cm)	Critical Mass (kg)
²³⁵ U	6.48	21.52
²³⁸ Pu	4.46	7.38
²³⁹ Pu	4.03	5.46
²⁴⁰ Pu	7.22	31.22
²⁴¹ Am	11.58	89.13

^{b2} ²³⁵U critical radius and mass based on ²³⁵U concentration of 54 g/l, which corresponds to the minimum critical mass for homogenous water-moderated ²³⁵U spheres.¹³

^{b3} ²³⁹Pu critical radius and mass based on ²³⁹Pu concentration of 30 g/l, which corresponds to the minimum critical mass for homogenous water-moderated Pu spheres.¹³

^{b4} No density adjustments were performed for isotopes.

Table B-4. Computed k_{eff} and rule-of-fractions (ROF) values for ^{235}U - ^{239}Pu mixtures with H_2O reflector.

	235U	239Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Aqueous Solutions	0%	100%	1.0069		
	25%	75%	1.0046	1.0046	0.0000
	50%	50%	1.0022	1.0023	-0.0001
	75%	25%	0.9998	0.9999	-0.0001
	100%	0%	0.9976		
Homogeneous Metals	0%	100%	1.0059		
	25%	75%	0.9692	1.0052	-0.0360
	50%	50%	0.9726	1.0044	-0.0318
	75%	25%	0.9861	1.0037	-0.0176
	100%	0%	1.0029		
U Ball - Pu Shell	50%	50%	0.9402	1.0044	-0.0642
Pu Ball - U Shell	50%	50%	1.0483	1.0044	0.0439

Table B-5 Computed k_{eff} and rule-of-fractions (ROF) values for ^{235}U - ^{240}Pu mixtures with H_2O reflector.

	235U	240Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	1.0000		
	25%	75%	1.0503	1.0007	0.0496
	50%	50%	1.0543	1.0015	0.0529
	75%	25%	1.0360	1.0022	0.0338
	100%	0%	1.0029		
U Ball - Pu Shell	50%	50%	0.9624	1.0015	-0.0391
Pu Ball - U Shell	50%	50%	1.0958	1.0015	0.0943

Table B-6. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{238}Pu mixtures with H_2O reflector.

	^{239}Pu	^{238}Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	0.9999		
	25%	75%	1.0261	1.0014	0.0247
	50%	50%	1.0282	1.0028	0.0254
	75%	25%	1.0201	1.0043	0.0158
	100%	0%	1.0058		
239 Ball - 238 Shell	50%	50%	0.9701	1.0028	-0.0327
238 Ball - 239 Shell	50%	50%	1.0568	1.0028	0.0540

Table B-7. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{240}Pu mixtures with H_2O reflector.

	^{239}Pu	^{240}Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	1.0000		
	25%	75%	1.0080	1.0014	0.0066
	50%	50%	1.0071	1.0029	0.0042
	75%	25%	1.0044	1.0043	0.0001
	100%	0%	1.0058		
239 Ball - 240 Shell	50%	50%	1.0480	1.0029	0.0451
240 Ball - 239 Shell	50%	50%	1.0196	1.0029	0.0167

Table B-8. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{241}Am mixtures with H_2O reflector.

	^{239}Pu	^{241}Am	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	1.0000		
	25%	75%	0.9724	1.0014	-0.0290
	50%	50%	0.9404	1.0029	-0.0625
	75%	25%	0.9060	1.0043	-0.0983
	100%	0%	1.0058		
Pu Ball - Am Shell	50%	50%	1.0273	1.0029	0.0244
Am Ball - Pu Shell	50%	50%	1.0022	1.0029	-0.0007

Table B-9. Critical radii and masses for isotopes in aqueous mixture with 20-cm H₂O reflector.

Isotope	Critical Radii (cm)	Critical Mass (g)
²³⁵ Ub ²	15.07	774.68
²³⁹ Pu ^{b3}	15.93	508.82
²⁴¹ Pu ^{b5}	12.98	275.01

Table B-10. Calculated critical radii and masses for isotopes in metal systems with a 20-cm H₂O reflector.

Isotope	Critical Radii (cm)	Critical Mass (kg)
²³⁵ U	6.49	21.52
²³⁸ Pu	4.47	7.41
²³⁹ Pu	4.03	5.45
²⁴⁰ Pu	7.18	30.84
²⁴¹ Pu	4.07	5.67
²⁴² Pu	9.43	70.53
²⁴¹ Am	11.58	89.13

^{b5} ²⁴¹Pu critical radius and mass based on Pu concentration of 30 g/l, which corresponds to the minimum critical mass for homogenous water-moderated Pu spheres.¹³

Table B-11. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{241}Pu mixtures with H_2O reflector.

	239Pu	241Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Aqueous Solutions	0%	100%	1.0098		
	25%	75%	1.0087	1.0104	-0.0017
	50%	50%	1.0090	1.0110	-0.0020
	75%	25%	1.0103	1.0116	-0.0013
	100%	0%	1.0122		
Homogeneous Metals	0%	100%	0.9994		
	25%	75%	1.0019	1.0011	0.0008
	50%	50%	1.0042	1.0028	0.0014
	75%	25%	1.0065	1.0044	0.0021
	100%	0%	1.0061		

Table B-12. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{242}Pu mixtures with H_2O reflector.

	239Pu	242Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	1.0001		
	25%	75%	1.0509	1.0016	0.0493
	50%	50%	1.0489	1.0031	0.0458
	75%	25%	1.0260	1.0046	0.0214
	100%	0%	1.0061		

APPENDIX C: DATA FOR MIXTURES WITH STAINLESS STEEL REFLECTOR

Table C-1 lists the isotopes and type of systems initially analyzed with a 20-cm stainless steel reflector. These mixtures were analyzed using a constant density for all the isotopes of an element, no density adjustments were performed for isotopic variation. Tables C-2 lists the critical radii and masses for isotopes listed in Table C-1. Note that two critical values are listed for ^{239}Pu and ^{240}Pu . The differences are due to the inclusion of NITAWL resonance calculations for the isotopes (Fe, Ni, Cr, Mn) in the stainless steel reflector region. These resonance calculations for the isotopes in the stainless steel increased the computed k_{eff} for the systems, thus reducing the critical mass of the systems. Computations for mixtures of ^{235}U - ^{239}Pu (Table C-3) and ^{235}U - ^{240}Pu (Table C-4) did not include resonance calculations for the stainless steel; while mixtures of ^{239}Pu - ^{238}Pu (Table C-5), ^{239}Pu - ^{240}Pu (Table C-6), and ^{239}Pu - ^{241}Am (Table C-7) did include resonance calculations for the stainless steel. All mixtures included resonance calculations for the fissile and fissionable isotopes. Tables C-3 to C-7 list the calculated k_{eff} 's, the rule-of-fractions (ROF) values, and the differences between the k_{eff} and ROF values for mixtures of the isotopes listed in Table C-1. Table C-8 lists the critical radii and masses calculated with isotopic density adjustments. In addition, the isotopes ^{241}Pu and ^{242}Pu were added to the list of isotopes to be analyzed. Tables C-9 and C-10 list the calculated k_{eff} 's, the rule-of-fractions (ROF) values, and the differences between the k_{eff} and ROF values for mixtures of ^{239}Pu - ^{241}Pu and ^{239}Pu - ^{242}Pu . Equivalence factors listed in Tables 9 and 10 of the report were based on the isotopic density adjusted critical masses and included NITAWL resonance calculations for the isotopes in the stainless steel reflector.

Table C-1. Isotopes and type of systems analyzed with a 20-cm type 304 stainless steel reflector.

Isotopes	Type of System
^{235}U - ^{239}Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
^{235}U - ^{240}Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
^{239}Pu - ^{238}Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
^{239}Pu - ^{240}Pu	Metal Homogeneous Sphere Metal Ball-and-Shell
^{239}Pu - ^{241}Am	Metal Homogeneous Sphere Metal Ball-and-Shell

Table C-2. Critical radii and masses for isotopes in metal systems with a 20-cm type 304 stainless steel reflector.^{c1}

Isotope	Critical Radii (cm)	Critical Mass (kg)
²³⁵ U	6.21	18.92
²³⁸ Pu	3.85	4.75
²³⁹ Pu ^{c2}	3.81/3.86	4.62/4.77
²⁴⁰ Pu ^{c2}	6.10/6.13	18.89/19.13
²⁴¹ Am	9.83	54.47

Table C-3. Computed k_{eff} and rule-of-fractions (ROF) values for ²³⁵U-²³⁹Pu mixtures with stainless steel reflector.

	235U	239Pu	k_{eff}	ROF	k_{eff} -ROF
Homogeneous Metals	0%	100%	0.9994		
	25%	75%	0.9609	1.0010	-0.0401
	50%	50%	0.9650	1.0025	-0.0375
	75%	25%	0.9851	1.0041	-0.0190
	100%	0%	1.0056		
U Ball - Pu Shell	50%	50%	0.9285	1.0025	-0.0740
Pu Ball - U Shell	50%	50%	1.0494	1.0025	0.0469

Table C-4. Computed k_{eff} and rule-of-fractions (ROF) values for ²³⁵U-²⁴⁰Pu mixtures with stainless steel reflector.

	235U	240Pu	k_{eff}	ROF	k_{eff} -ROF
Homogeneous Metals	0%	100%	1.0000		
	25%	75%	1.0328	1.0014	0.0314
	50%	50%	1.0374	1.0028	0.0346
	75%	25%	1.0267	1.0042	0.0225
	100%	0%	1.0056		
U Ball - Pu Shell	50%	50%	1.0158	1.0028	0.0130
Pu Ball - U Shell	50%	50%	1.0373	1.0028	0.0345

^{c1} No density adjustments were performed for isotopes.^{c2} Note that two critical values are listed for ²³⁹Pu and ²⁴⁰Pu with a stainless steel reflector. The differences are due to the inclusion of NITAWL resonance calculations for the isotopes (Fe, Ni, Cr, Mn) in the stainless steel reflector region. Computations for mixtures of ²³⁵U-²³⁹Pu and ²³⁵U-²⁴⁰Pu did not include resonance calculations for the stainless steel, while mixtures of ²³⁹Pu-²³⁸Pu, ²³⁹Pu-²⁴⁰Pu, and ²³⁹Pu-²⁴¹Am did include resonance calculations for the stainless steel. All mixtures included resonance calculations for the fissile and fissionable isotopes.

Table C-5. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{238}Pu mixtures with stainless steel reflector.

	^{239}Pu	^{238}Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	0.9999		
	25%	75%	1.0039	0.9997	0.0042
	50%	50%	1.0049	0.9995	0.0054
	75%	25%	1.0032	0.9993	0.0039
	100%	0%	0.9991		
239 Ball - 238 Shell	50%	50%	1.0000	0.9995	0.0005
238 Ball - 239 Shell	50%	50%	1.0064	0.9995	0.0069

Table C-6. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{240}Pu mixtures with stainless steel reflector.

	^{239}Pu	^{240}Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	0.9998		
	25%	75%	1.0008	0.9998	0.0010
	50%	50%	0.9997	0.9996	0.0001
	75%	25%	0.9975	0.9993	-0.0019
	100%	0%	0.9991		
239 Ball - 240 Shell	50%	50%	1.0695	0.9996	0.0699
240 Ball - 239 Shell	50%	50%	0.9501	0.9996	-0.0495

Table C-7. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{241}Am mixtures with stainless steel reflector.

	^{239}Pu	^{241}Am	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	1.0000		
	25%	75%	0.9807	0.9998	-0.0191
	50%	50%	0.9578	0.9996	-0.0418
	75%	25%	0.9364	0.9993	-0.0629
	100%	0%	0.9991		
Pu Ball - Am Shell	50%	50%	1.0466	0.9996	0.0470
Am Ball - Pu Shell	50%	50%	0.9248	0.9996	-0.0748

Table C-8. Calculated critical radii and masses for isotopes in metal systems with a 20-cm type 304 stainless steel reflector.

Isotope	Critical Radii (cm)	Critical Mass (kg)
^{235}U	6.13	18.15
^{238}Pu	3.86	4.77
^{239}Pu	3.82	4.62
^{240}Pu	6.07	18.72
^{241}Pu	3.96	5.22
^{242}Pu	7.92	41.77
^{241}Am	9.83	54.47

Table C-9. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{241}Pu mixtures with stainless steel reflector.

	^{239}Pu	^{241}Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	1.0001		
	25%	75%	0.9999	1.0001	-0.0002
	50%	50%	0.9998	1.0001	-0.0003
	75%	25%	0.9998	1.0001	-0.0003
	100%	0%	1.0001		

Table C-10. Computed k_{eff} and rule-of-fractions (ROF) values for ^{239}Pu - ^{242}Pu mixtures with stainless steel reflector.

	^{239}Pu	^{242}Pu	k_{eff}	ROF	$k_{\text{eff}}\text{-ROF}$
Homogeneous Metals	0%	100%	1.0001		
	25%	75%	0.9968	1.0001	-0.0033
	50%	50%	0.9908	1.0001	-0.0093
	75%	25%	0.9837	1.0002	-0.0165
	100%	0%	1.0002		