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Author(s):	Mahlen, Caden Lee Zhang, Ning
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(U) Parametric Study of ^{239}Pu and ^{235}U Mass Equivalency in Spherical Geometries

Signature and Date

Author	
Caden L. Mahlen	CADEN MAHLEN (Affiliate) Digitally signed by CADEN MAHLEN (Affiliate) Date: 2023.07.20 13:30:41 -06'00'
Co-Author	
Ning Zhang	NING ZHANG (Affiliate) Digitally signed by NING ZHANG (Affiliate) Date: 2023.07.20 13:32:14 -06'00'
Independent Reviewer	
Trevor N. Stewart	TREVOR STEWART (Affiliate) Digitally signed by TREVOR STEWART (Affiliate) Date: 2023.07.20 13:34:48 -06'00'
Quality Reviewer	
David K. Hayes	DAVID HAYES (Affiliate) Digitally signed by DAVID HAYES (Affiliate) Date: 2023.07.20 13:52:37 -06'00'

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1.0 SUMMARY

In this technical document, parametric studies were performed to determine if a bounding mass equivalency can be used to simplify multi-material models in MCNP6 for criticality evaluations. The studies explored the simplification of MCNP6 models by substituting materials, particularly ^{239}Pu for ^{235}U. The initial hypothesis for the material simplification is based on the subcritical mass limits from the ANSI/ANS- 8.1 standard which states that for water reflected metal spheres the limits are 20.1 kg ^{235}U and 5.0 kg ^{239}Pu [Ref. (2)]. Using those masses, it was hypothesized that a 20.1 kg sphere of ^{235}U and a 5.0 kg sphere of ^{239}Pu could be considered to have the same multiplication. Given that equivalency, 4kg ^{235}U could be modeled by 1kg ^{239}Pu (i.e., a mass ratio of 4:1). The parametric studies compare the materials ^{239}Pu and ^{235}U independently and in multi-material spherical models. Critical mass, critical thickness, critical mass ratio and critical thickness ratio values are presented as functions of inner radius. k_{eff} values are presented as a function of inner radius at different mass ratios and total masses. Results indicate that the conservative bounding mass equivalency is one. That is, if substituting ^{239}Pu for ^{235}U it should be done on a gram for gram basis.

This technical document was developed in accordance with NCS-AP-005 [Ref. (1)].

2.0 DESCRIPTION

This parametric study explored the simplification of MCNP6 models by substituting materials, particularly ^{239}Pu for ^{235}U. The initial hypothesis for the material simplification is based on the subcritical mass limits from the ANSI/ANS-8.1 standard which states that for water reflected metal spheres the limits are 20.1 kg ^{235}U and 5.0 kg ^{239}Pu [Ref. (2)]. Using those masses, it was hypothesized that a 20.1 kg sphere of ^{235}U and a 5.0 kg sphere of ^{239}Pu could be considered to have similar multiplication. The multiplication of these systems were computed to be 0.98738 ± 0.00016 for the water reflected uranium sphere and 0.97910 ± 0.00017 for the water reflected plutonium sphere. Given that equivalency, 4.0 kg ^{235}U could be modeled by 1.0 kg ^{239}Pu (i.e., a mass ratio of 4:1).

3.0 METHODOLOGY

Several MCNP6 models were developed to demonstrate this hypothesis. All models were spherical shells of varying inner radii. Studies were performed with models that were bare and models that had infinite (30 inches) water reflection. All models used pure ^{239}Pu and ^{235}U at the maximum theoretical densities, as stated in Table 2.

This study utilizes single-material and multi-material models to compare the complex system to the simplified system. The inner radius of the inner shell and the mass ratio were varied to examine different effects and to demonstrate the differences between the k_{eff} values in the simplified single-material model to the multi-material model. Both reflected and unreflected systems were modeled.

The primary study compares the multi-material model to the simplified model which only contains ^{239}Pu. These models varied the mass ratio of ^{235}U to ^{239}Pu. The multi-material model varied the mass of ^{235}U in the outer shell. The simplified model had the mass of ^{239}Pu in the multi-material model plus the conversion of ^{235}U to ^{239}Pu based on the mass ratio. These values are listed for each model in Table 1.

A secondary study compares shells of ^{235}U to shells of ^{239}Pu. The computed critical masses of these independent shells as a function of inner radius were used to derive the critical mass ratio as a function of inner radius. The purpose was to demonstrate the difference in behavior between ^{235}U and ^{239}Pu systems.

3.1.1 Single-material Models

For baseline analysis, single-material models were developed. The ^{239}Pu models were of particular importance as they were the basis for multi-material model comparison used to determine the effectiveness of the substitution hypothesis. A single-material model of ^{235}U was only used for the critical mass ratio study. The radii for the single-material models used in the critical mass study were determined by starting at 0.01 cm (for zero), then linearly interpolating from 2 cm to 50 cm with a step size of 2 cm.

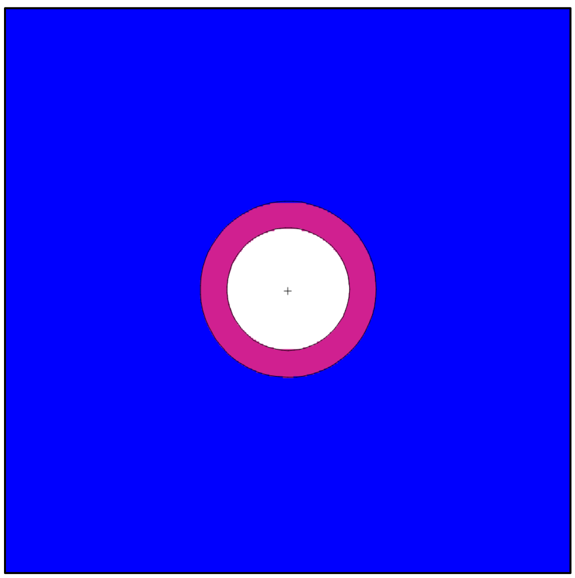

Figure 1. An example of a single-material model with infinite water reflection. The fissile material (either ^{239}Pu or ^{235}U) is modeled in magenta as a spherical shell and the water reflector is modeled in blue. Inside of the shell, in white, is modeled as void.

3.1.2 Multi-material Models

The multi-material models were comprised of two concentric shells. The inner radius of the inner shell was varied in all models. The inner radii for the multi-material models and the simplified single-material models were determined by logarithmically interpolating inclusively from 1 cm to 50 cm with 26 points. The inner shell was ^{239}Pu and the outer shell was ^{235}U. The purpose of these models was to evaluate the actual behavior of the materials together and compare that behavior to the equivalent ratio of ^{239}Pu modelled as a single material. The amount of ^{235}U in these models was determined by the mass ratio of that particular model, while the amount of ^{239}Pu was varied independently. All the mass values used in the models are listed in Table 1.

Table 1. Index of ^{239}Pu and ^{235}U masses in the multi-material systems for all mass ratios and the equivalent masses of ^{239}Pu in the simplified systems.

Mass of ^{239}Pu in Multi-Material Model (kg)	Mass of ^{235}U in Multi-Material Model (kg)							Mass of ^{239}Pu in Simplified Single-material Model (kg)
Mass Ratio	1	1.5	2	2.5	3	3.5	4	N/A
0.25	0.25	0.375	0.5	0.625	0.75	0.875	1	0.5
0.5	0.5	0.75	1	1.25	1.5	1.75	2	1
0.75	0.75	1.125	1.5	1.875	2.25	2.625	3	1.5
1	1	1.5	2	2.5	3	3.5	4	2
2	2	3	4	5	6	7	8	4
4	4	6	8	10	12	14	16	8
8	8	12	16	20	24	28	32	16
12	12	18	24	30	36	42	48	24
16	16	24	32	40	48	56	64	32

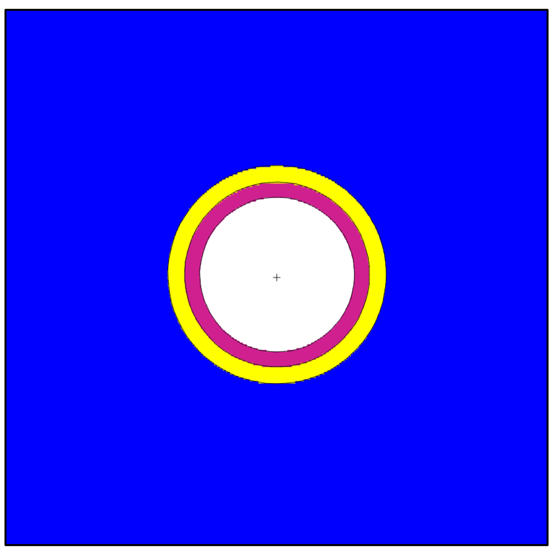

Figure 2. An example of a multi-material model with infinite water reflection. The inner shell, modeled in magenta, is ^{239}Pu and the outer shell, modeled in yellow, is ^{235}U. The water reflector is modeled in blue. Inside of the inner shell, in white, is modeled as void.

3.2 Materials

The materials considered in this study are listed in Table 2. A total of 3 materials were modeled. The definitions of the materials in this study were referenced directly from NCS-TECH-18-024, R1 [Ref. (6)].

Table 2. Index of Materials

Fissile Material	Reflector Material
Plutonium 239 100 wt% 19.84 g/cc Uranium 235 100 wt% 18.95 g/cc	Water (H_2O) 100 wt% 1 g/cc

3.3 Analytical Method

MCNP6 calculations were performed using ENDF B/VII cross sections sets on the high-performance computing cluster Blowfish, with code and validation information as stated in NCS-TECH-21-001, NCS-TECH-18-014 and NCS-TECH-18-015 [Ref (5) (3) (4)]. The models were run using at least 50,000 neutrons per cycle, skipping 100 cycles, with at least 500 active cycles.

A PERL script, Worm Solver, was used to iteratively compute the critical masses of the models in the critical mass ratio study. The raw results were subsequently curve fitted in Microsoft Excel to a third order polynomial in order to interpolate for the critical mass.

Effective neutron multiplication factor (k_{eff}) computation results are reported as:

$$k_{calc} + 2\sigma$$

4.0 ANALYSIS

There were two separate studies performed to analyze the critical behaviors of ^{239}Pu and ^{235}U in single-material and multi-material systems.

4.1 Single-material Critical Mass Ratio Study

In the single-material study, two separate models were developed and compared. Shells of ^{235}U at various inner radii was compared to shells of ^{239}Pu at various inner radii. At each radius, the critical mass was determined for each material. These critical masses, reflected and unreflected, as a function of inner radius are shown in Figure 3. The critical masses of the ^{235}U shells were divided by the critical masses of the ^{239}Pu shells to achieve a critical mass ratio as a function of inner radius, shown in Figure 4.

The ratio at an inner radius of near 0 cm (essentially a sphere) in the reflected systems approaches 4, which is similar to the expected value from the ANSI/ANS-8.1 single-parameter subcritical mass limit ratios. However, as the inner radius increased, the ratio values changed significantly, decreasingly rapidly then flattening out at larger inner radii. This result conflicted with the hypothesis of a four-to-one mass equivalency and suggested that such a ratio cannot be applied to all systems. The mass ratio values were observed to have an asymptotic behavior, suggesting that at increasingly large inner radii the mass ratio is no longer dependent on inner radius.

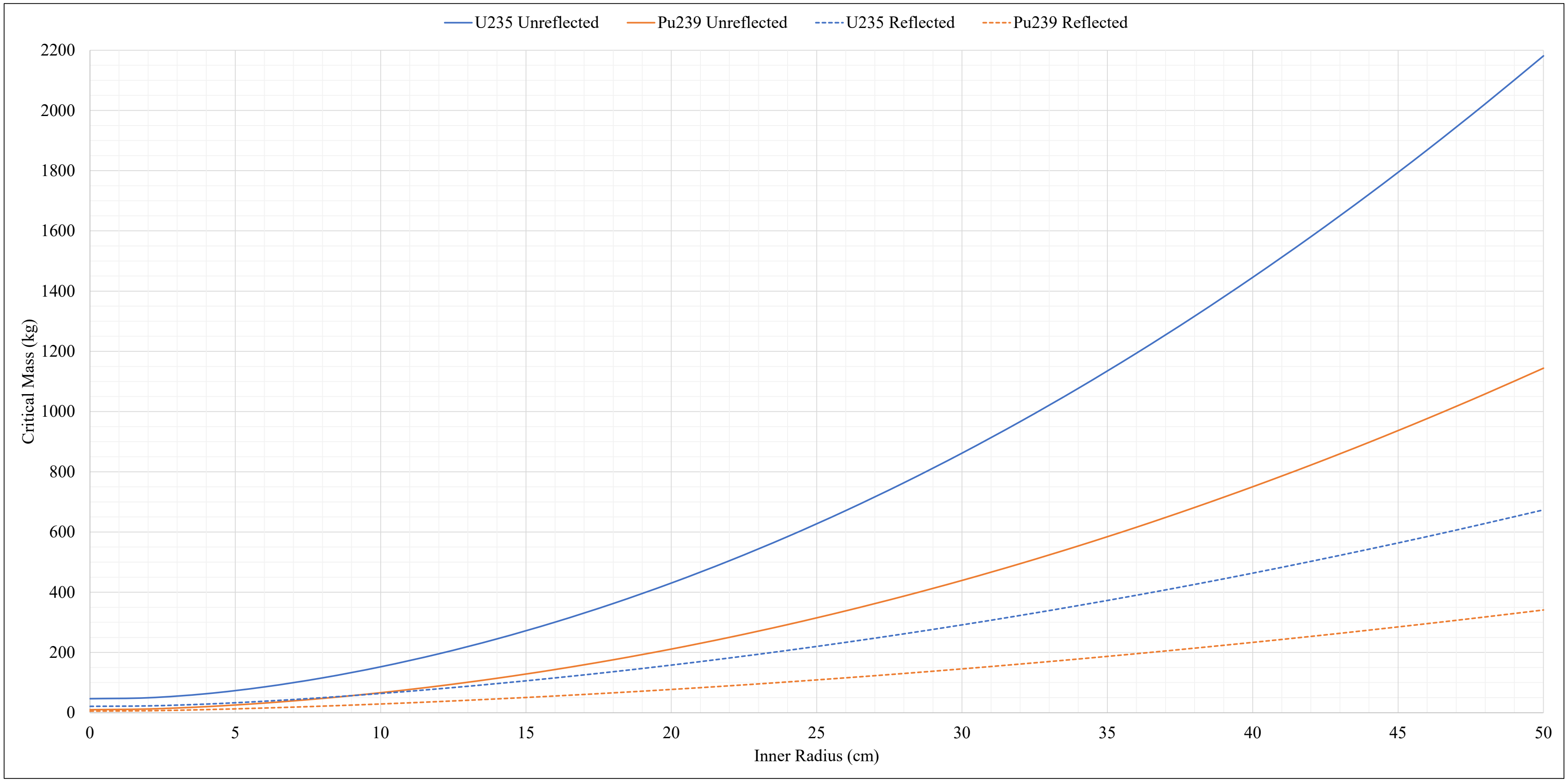

Figure 3. The critical mass of ^{235}U and ^{239}Pu shells as a function of inner radius for reflected and unreflected systems.

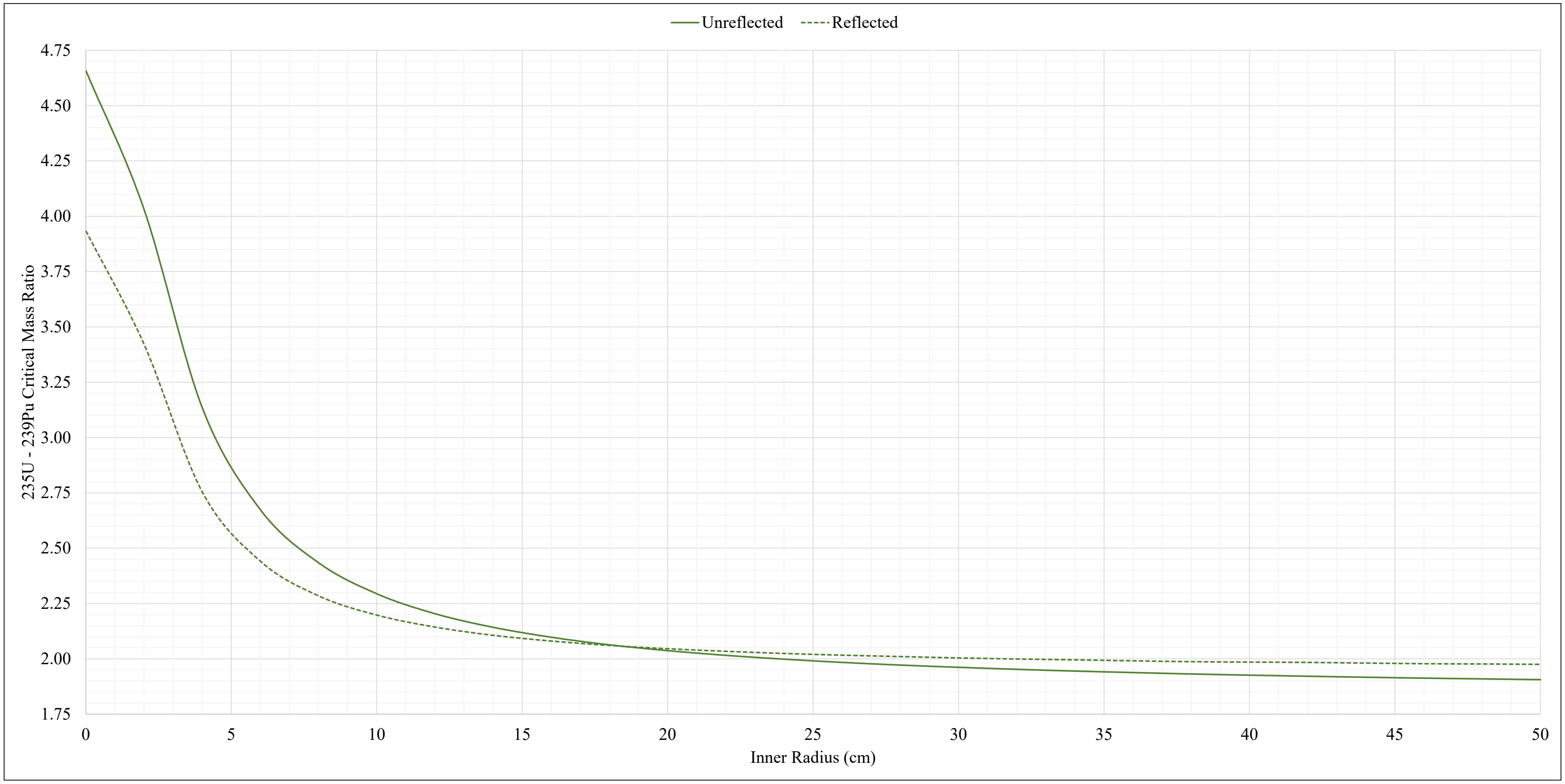

Figure 4. The critical mass ratio of ^{235}U shells to ^{239}Pu shells as a function of inner radius for reflected and unreflected systems.

4.1.1 Critical Thickness Analysis

To supplement the analysis of these shells, the critical thickness for each shell was calculated from the respective critical mass results as a function of inner radius using known geometric formulas and the material densities. These calculations are shown in Figure 5. The critical thickness ratio was calculated by dividing the critical thickness of the ^{235}U shells by the critical thickness of the ^{239}Pu shells and is shown in Figure 6. The behavior of the critical thickness curves in Figure 5 were asymptotic, which suggests that there was some critical thickness of a shell that would be critical regardless of inner radius at increasingly large inner radii. The system at increasingly large inner radii approaches infinite slab thickness to sustain critical state. Figure 6 demonstrated that the ratio of the critical thicknesses between ^{239}Pu and ^{235}U seemed to become constant because the curves were also asymptotic.

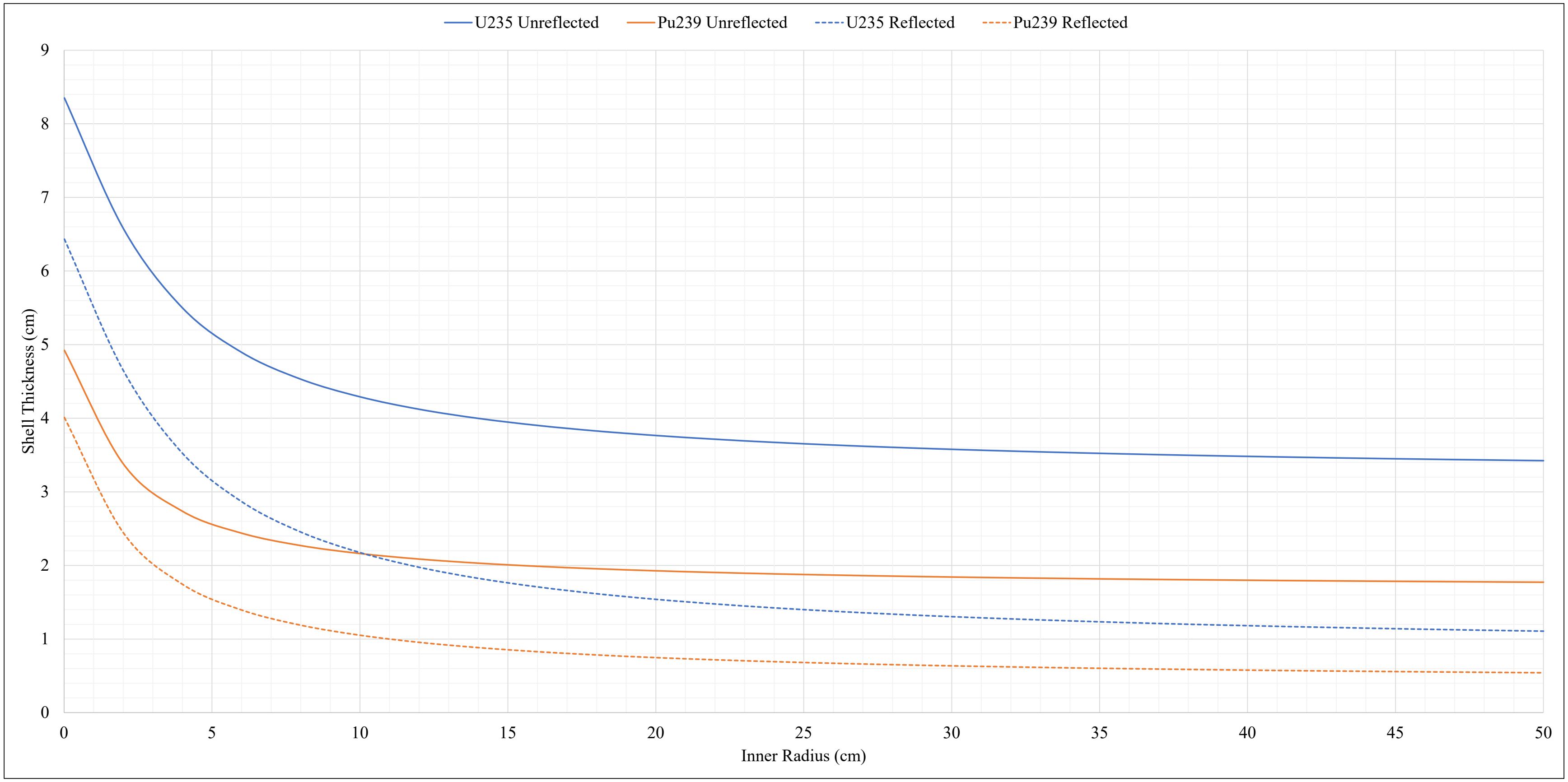

Figure 5. The critical thickness of ^{235}U and ^{239}Pu shells as a function of inner radius for reflected and unreflected systems.

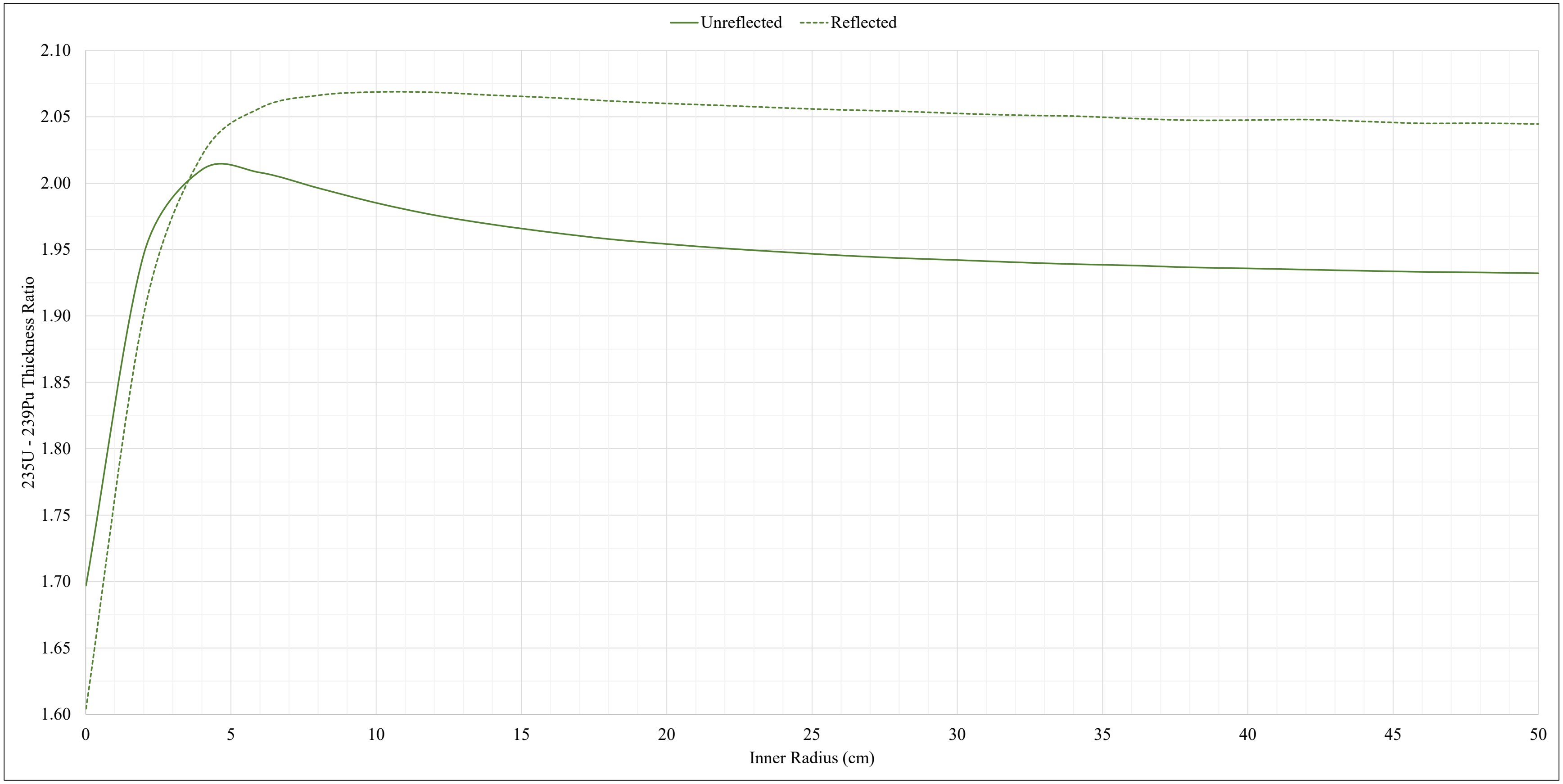

Figure 6. The critical thickness ratio of ^{235}U to ^{239}Pu shells as a function of inner radius for reflected and unreflected systems.

4.2 Multi-material Mass Ratio Study

In the multi-material mass ratio study, several mass ratios were evaluated. Each multi-material model was compared to the equivalent single-material model at varying inner radii. Only reflected systems were considered in this study to be conservative and due to the instability of the models at extreme cases (i.e., large radii) no unreflected models were analyzed. The mass of the simplified single-material model was the sum of the ^{235}U mass in the multi-material model divided by the mass ratio and the ^{239}Pu mass in the multi-material model, as provided in Table 1. Results of the comparison shown in Figure 7 through Figure 13.

The results of the models were complex and multivariable dependent. Conservative results, meaning when the multiplication of the simplified model was greater than the multiplication of the multi-material model, changed at different mass ratios, at different radii, and at different total mass amounts. There were points on some pairs of related curves where the model inverts between conservative and nonconservative. In the models that started conservative, the point when it became nonconservative shifted to larger inner radii with more total mass present. The conservatism in the small inner radius region became larger at lower mass ratios and stayed conservative at greater inner radii. In the lower total mass systems, the simplified model fluctuated between conservative and nonconservative as a function of inner radius.

The models with a mass ratio of greater than 2.5 generally did not produce conservative results. For low total mass systems, conservative values were only observed at low mass ratios at low inner radii. Generally, the more total mass present in the system, the smaller the inner radius, and the smaller the mass ratio, the more conservative the model was.

The results of this study showed that there are many variables that affect the behaviors of these systems and there are other effects that may not be accounted for in this analysis. In summary, the conservative bounding mass equivalency is one. That is, if substituting ^{239}Pu for ^{235}U it should be done on a gram for gram basis. As always, professional judgement should be used when applying the results of these studies.

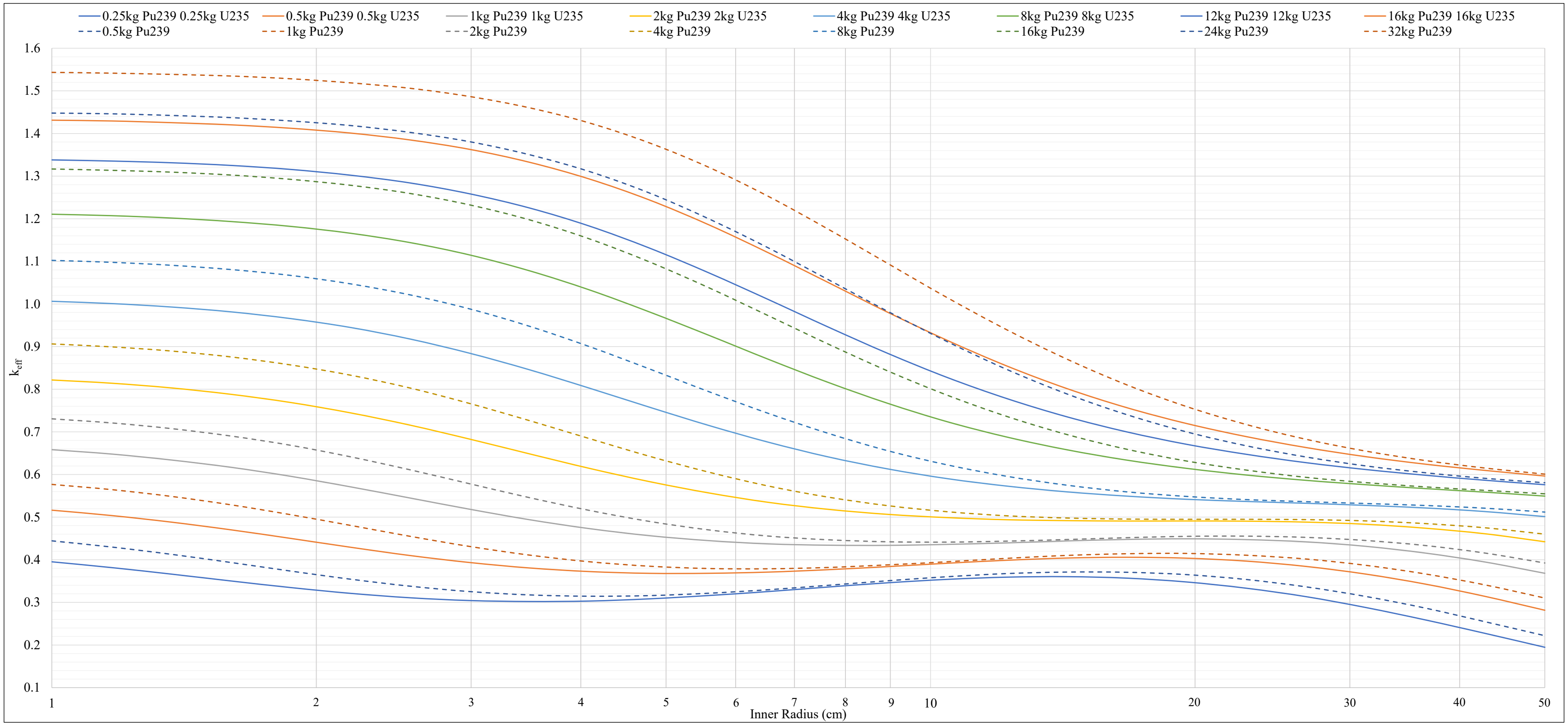

Figure 7. k_{eff} values of reflected single and multi-material systems as a function of inner radius logarithmically interpolated from 1 cm to 50 cm with a mass ratio of 1.0. The dotted lines represent the simplified single-material models, and the solid lines represent the multi-material models.

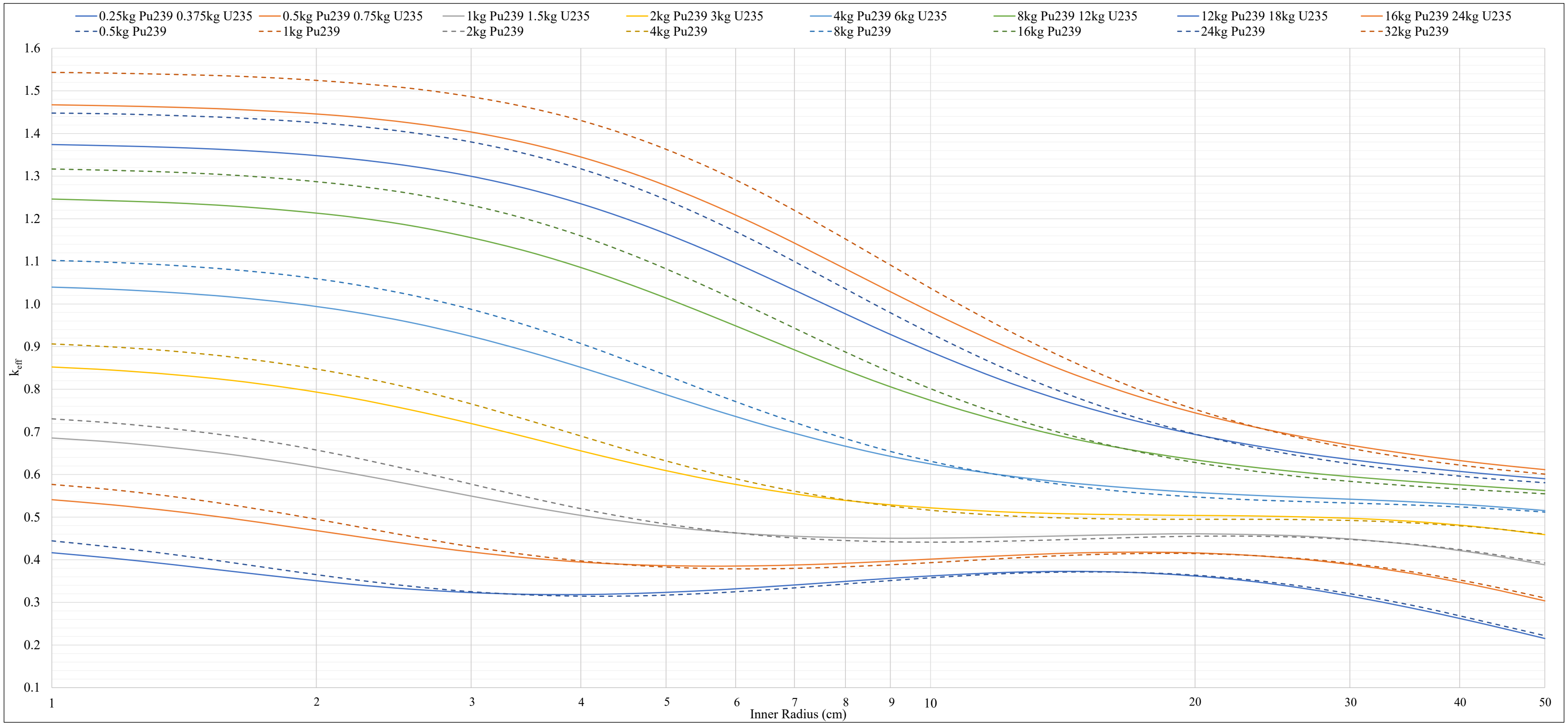

Figure 8. k_{eff} values of reflected single and multi-material systems as a function of inner radius logarithmically interpolated from 1 cm to 50 cm with a mass ratio of 1.5. The dotted lines represent the simplified single-material models, and the solid lines represent the multi-material models.

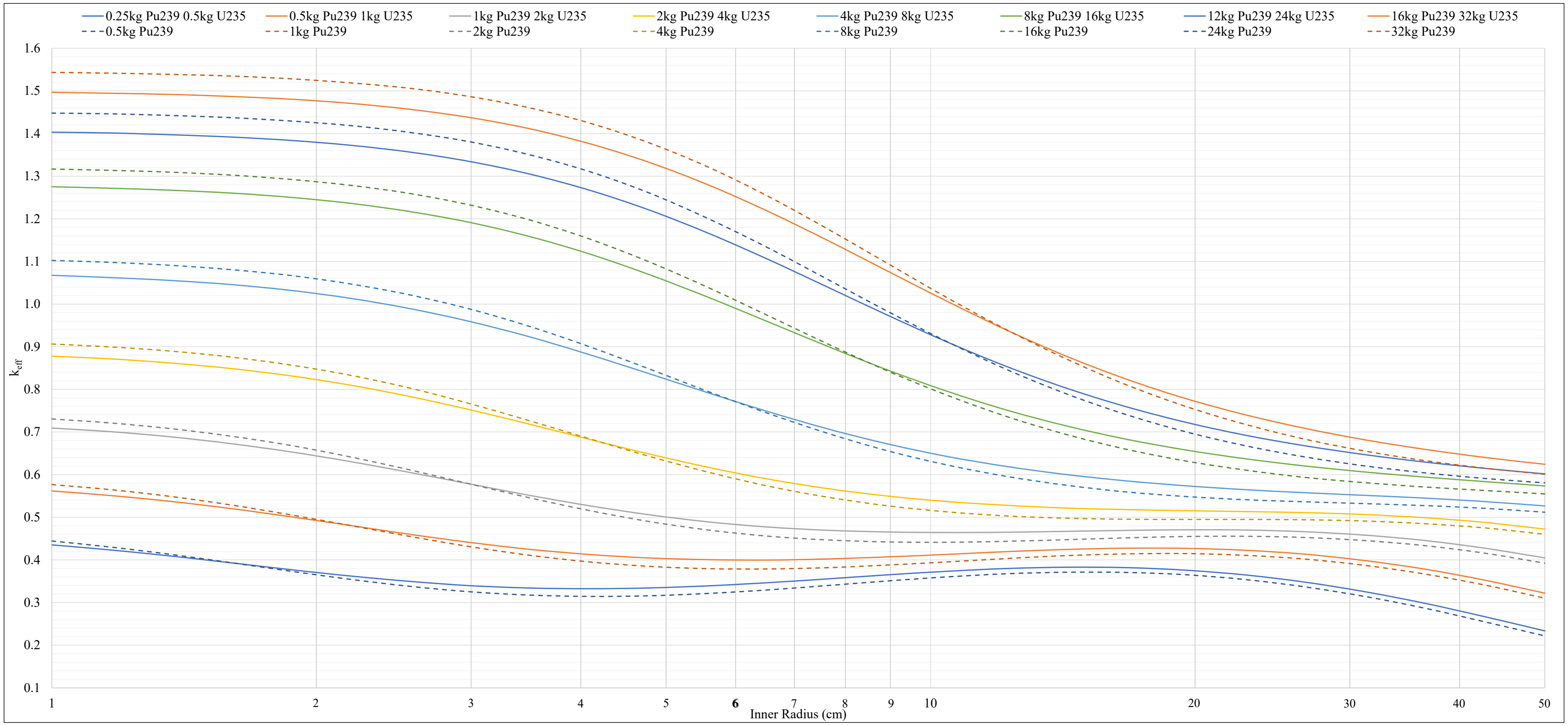

Figure 9. k_{eff} values of reflected single and multi-material systems as a function of inner radius logarithmically interpolated from 1 cm to 50 cm with a mass ratio of 2.0. The dotted lines represent the simplified single-material models, and the solid lines represent the multi-material models.

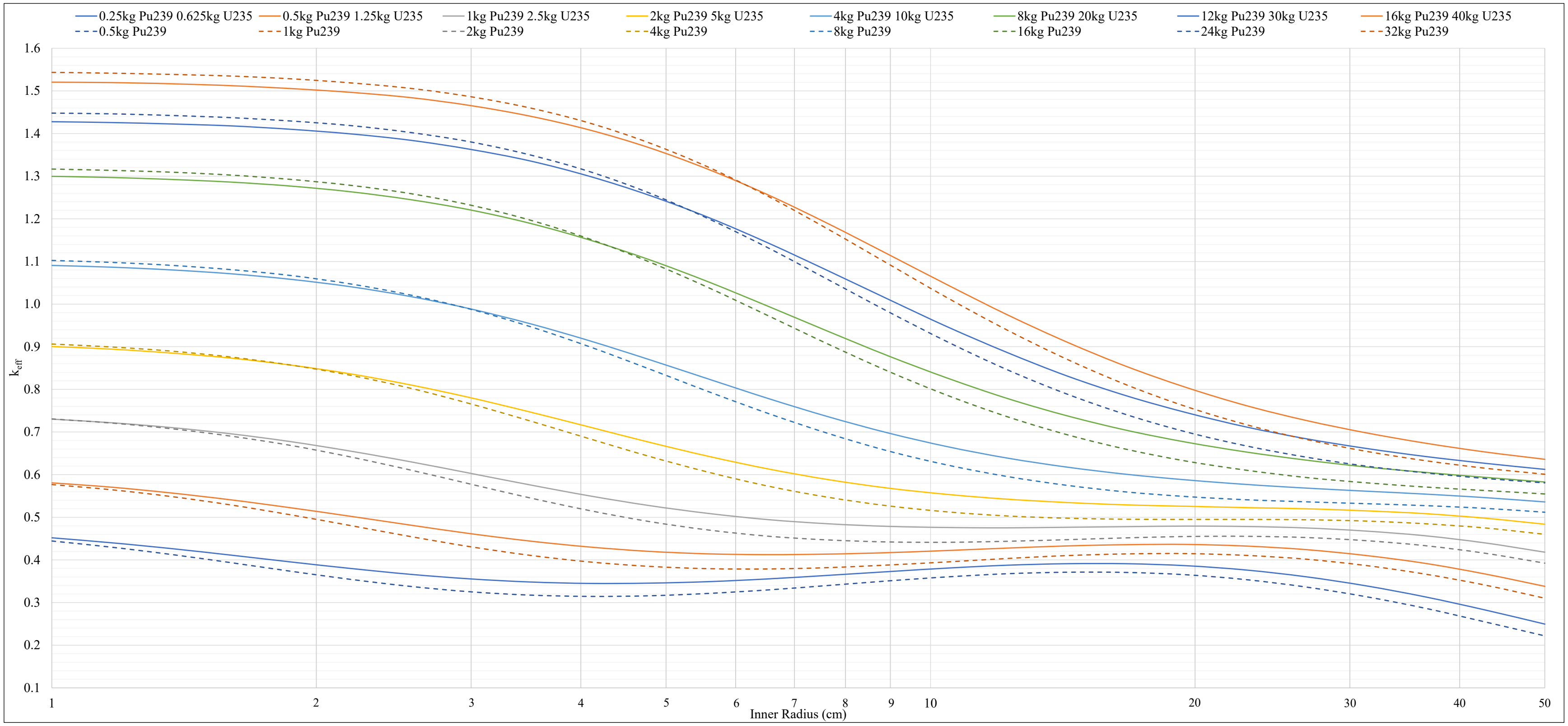

Figure 10. k_{eff} values of reflected single and multi-material systems as a function of inner radius logarithmically interpolated from 1 cm to 50 cm with a mass ratio of 2.5. The dotted lines represent the simplified single-material models, and the solid lines represent the multi-material models.

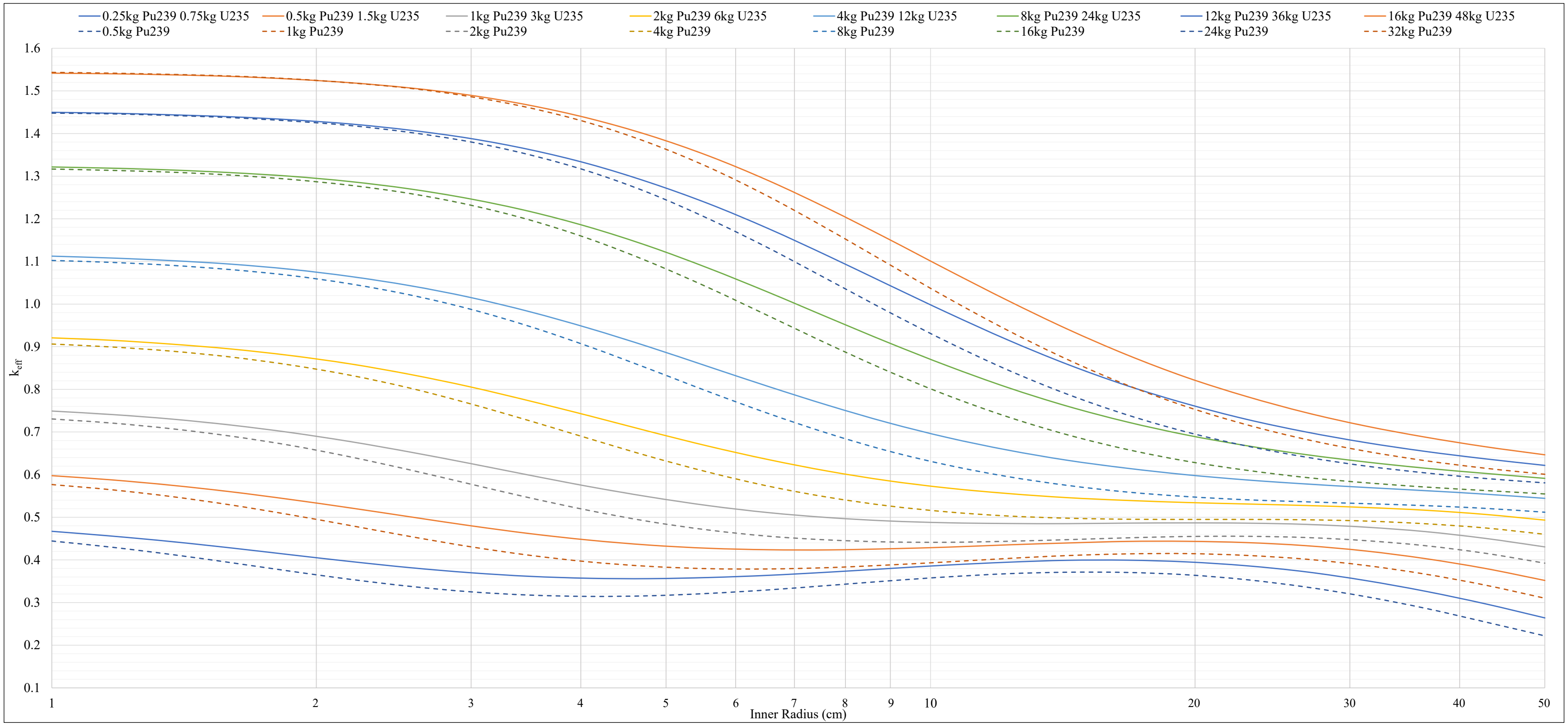

Figure 11. k_{eff} values of reflected single and multi-material systems as a function of inner radius logarithmically interpolated from 1 cm to 50 cm with a mass ratio of 3.0. The dotted lines represent the simplified single-material models, and the solid lines represent the multi-material models.

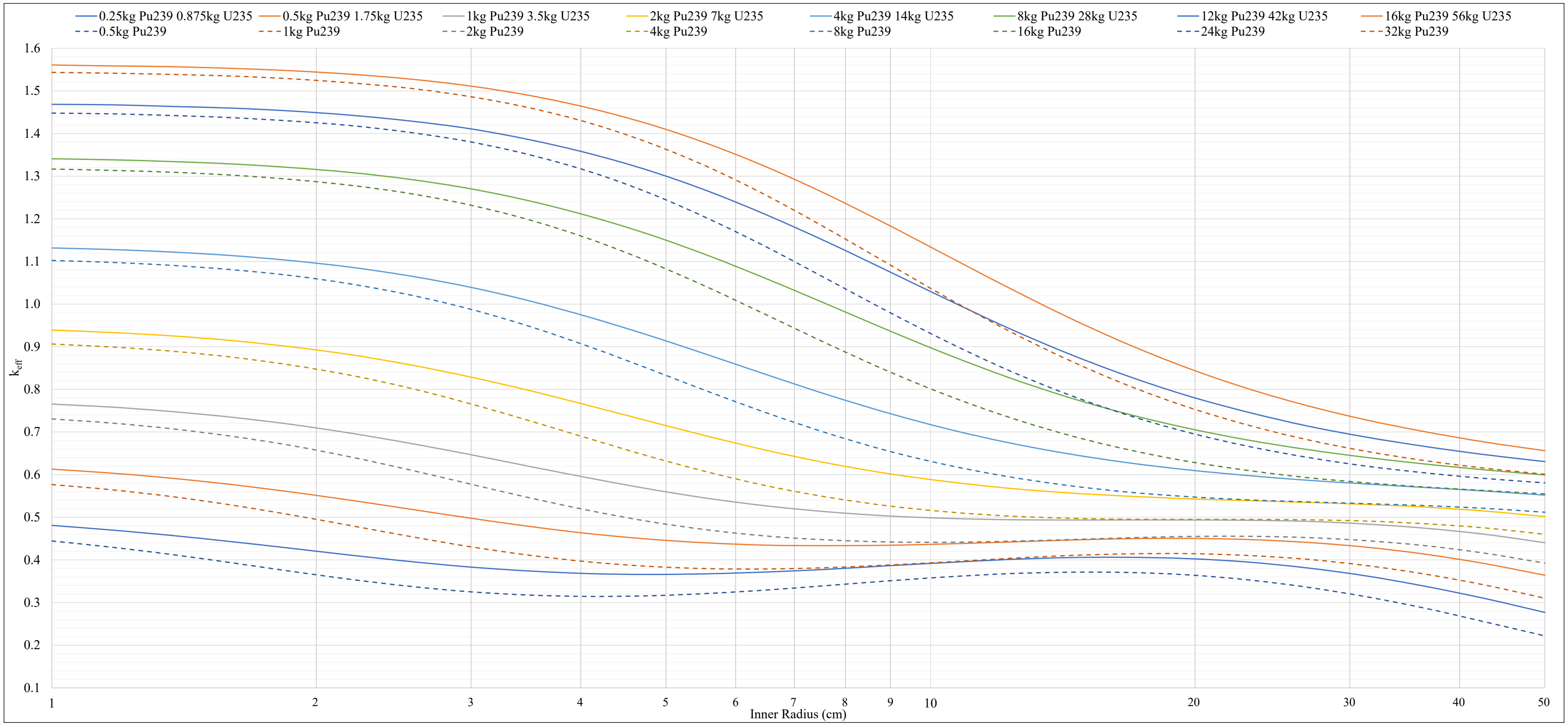

Figure 12. k_{eff} values of reflected single and multi-material systems as a function of inner radius logarithmically interpolated from 1 cm to 50 cm with a mass ratio of 3.5. The dotted lines represent the simplified single-material models, and the solid lines represent the multi-material models.

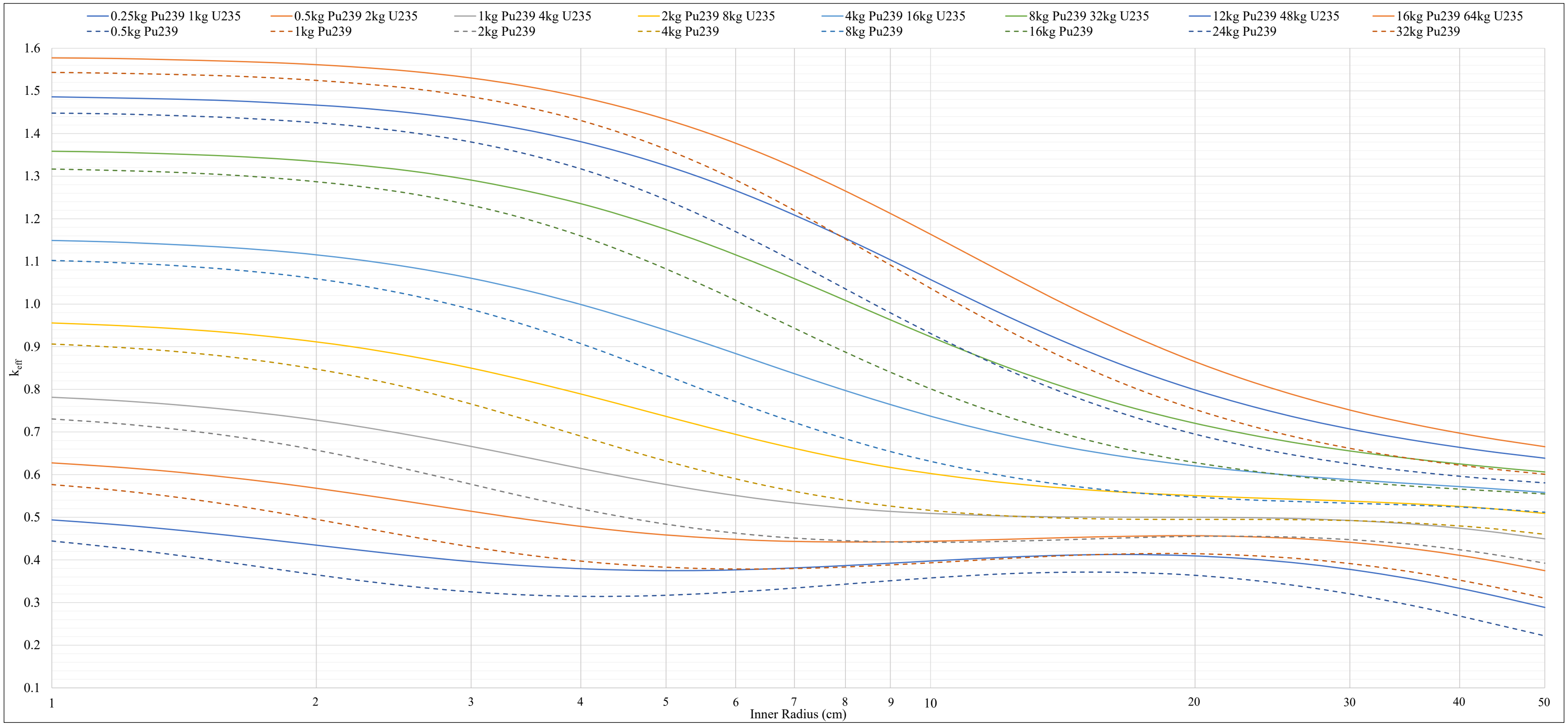

Figure 13. k_{eff} values of reflected single and multi-material systems as a function of inner radius logarithmically interpolated from 1 cm to 50 cm with a mass ratio of 4.0. The dotted lines represent the simplified single-material models, and the solid lines represent the multi-material models.

5.0 CONCLUSION

The initial hypothesis of a four-to-one mass ratio equivalency for multi-material systems including ^{235}U and ^{239}Pu was modeled and it was demonstrated that it is not conservative in most cases. There were factors, other than material properties, that affected the behaviors of the results.

Higher total mass systems were demonstrated to be more conservative, and the conservativism extended to larger radii compared to lower total mass systems. Lower mass ratios were demonstrated to be more conservative. The general trend was observed to be greater conservatism at lower mass ratios, at lower radius and with higher total mass. To be conservative, ^{239}Pu should be substituted for ^{235}U on a gram for gram (1:1) basis, as demonstrated by Figure 7.

Depending on the applicable system, an analyst should take note of the conservative trends in this model and the geometric effects of the system. An analyst should use professional judgement when applying any mass equivalency.

6.0 REFERENCES

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2. **ANSI/ANS-8.1-2014, (R2018).** *Nuclear Criticality Safety in Operations with Fissionable Material Outside Reactors.*
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