TECHNICAL BASIS FOR 2000 g PuBe SEALED NEUTRON SOURCE MASS LIMIT

Background

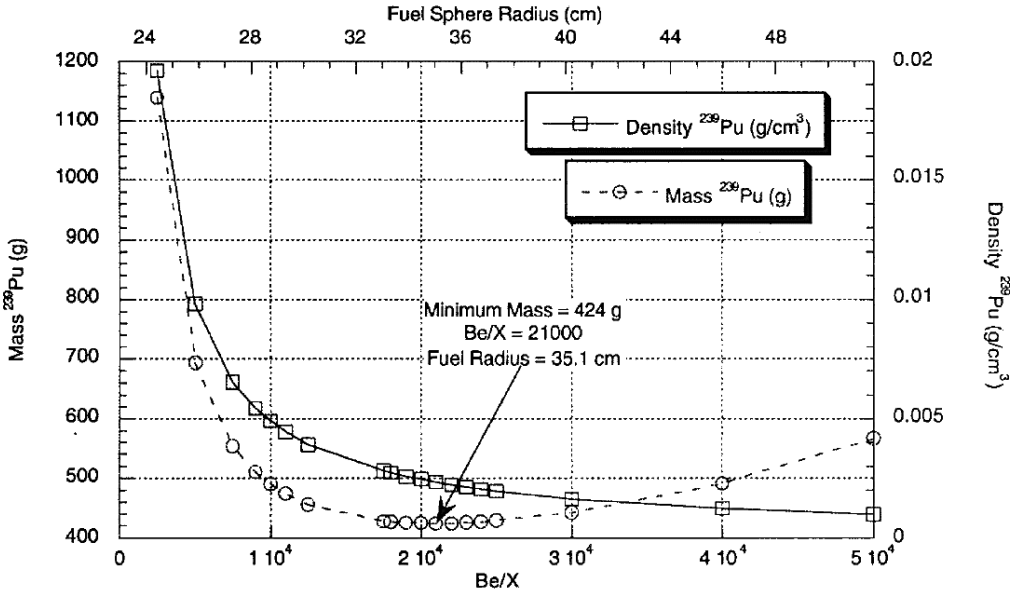
Plutonium (Pu) and Beryllium (Be) sealed sources are generally used as neutron sources. Such sources are widely used in research institutions for neutron activation and characterization, calibration of instruments and teaching activities. This report discusses the technical basis for allowing 2000 g of Pu-239 in the form of Pu-Be sealed sources to be considered as less than critical mass for future applications requesting an U.S. Nuclear Regulatory Commission (NRC) license. Guidance in NUREG 1556 Volume 17 (Ref. 1) and Regulatory Guide (RG)-10.3 (Ref. 2) states that up to 2000 g of Pu in sealed Pu-Be sources constitutes less than a critical mass. In addition, Title 10 of the *Code of Federal Regulations*, (CFR) Part 71.22 formerly had a mass limit of 2500 g of fissile material for such sources. According to NUREG/CR-5342 (Ref. 2), there is no known technical basis for the 2500 g limit. Therefore, as part of the development of an NRC critical mass facility guidance document, this report seeks to establish a technical basis.

Discussion

The minimum critical mass of Pu is commonly given as 450 g. This is based on a spherical geometry, with optimal moderation by light water, and full reflection by 30 cm of water. It is a well-known fact that moderators such as hydrocarbons, concrete, and especially “exotic” or “special” moderators such as heavy water and beryllium, can be more effective moderators than water, resulting in a smaller critical mass. In fact, the regulatory definition of a critical mass as stated in 10 CFR 70.4 calls for the critical mass to be reduced by half in such cases. However, in reality the minimum critical mass can vary widely depending on the isotopic, chemical, and physical characteristics of the material.

Beryllium is a more effective moderator than water primarily because of its significantly lower absorption cross section. Therefore, one would expect that an optimally moderated Pu-Be alloy would have a lower minimum critical mass than 450 g. It is therefore our conclusion that the mass limit of 2000 g Pu is not for the optimal Pu-Be system, but must be based on additional unstated assumptions. The guidance in NUREG 1556 Volume 17 should be more specific by stating the assumptions under which this mass limit is applicable.

Ref. 3 contains the following diagram (Figure D-6 on p. 77):



This diagram shows the subcritical mass (at k_{eff} = 0.95) instead of the critical mass, but it has a minimum of 424 g Pu at Be/Pu = 21,000. Clearly, according to this figure, a mass limit of 2000 g must be for a system with a much lower Be/Pu ratio. Refs. 4 – 7 contain information on the composition of common Pu-Be sources. Most domestically available Pu-Be sources have a nominal Be/Pu ratio of 13 (ranging from 10.5 – 18.3 per Ref. 4), while some foreign sources have a nominal ratio of 30 (Ref. 7). While the curve shown above does not extend to Be/Pu ratios that low, such values will result in a much higher critical mass than at optimum.

With regard to the physical form, Refs. 4 – 7 indicate they consist of intermetallic mixtures of Pu and Be that have been pressed, sintered, and encapsulated, typically as cylindrical rods.

Calculation Model

NRC staff performed calculations to assess the subcritical mass limit (at k_{eff} = 0.95) for batches of Pu-Be sources. The following assumptions were used:

Characteristic	Value
Beryllium composition and density	Natural metal at 1.85 g/cc
Plutonium composition and density	^{239}Pu metal at 19.84 g/cc
Physico-chemical form	Homogeneous alloy
Be/Pu ratio	13 & 30
Geometry	Spherical
Interstitial moderation	0% and 26% water
Reflection	30 cm water

The material was modeled as Be and Pu metal sintered to theoretical density; when mixed together, volume additivity was assumed (*i.e.*, metal volume fractions summed to 1). The geometry consisted of a homogeneous sphere to represent batches of multiple sources. Although the batches would in reality consist of heterogeneous bundles of rods, they were smeared (homogenized) for simplicity of calculation. From experience, this is expected to have only a small effect on the results. The spheres were then surrounded by 30 cm of water. Two

ratios of Be/Pu were assumed, 13 and 30 to represent the two classes of sources described in Refs. 4 – 7.

While the model was homogenized, the sources are in reality heterogeneous, so it is possible the void space between them could be filled with water. Two sets of calculations were performed—one with no void space and one with the minimum void space between closely packed spheres. The minimum packing fraction for spheres of uniform size of 74 percent was used, leaving 26 percent void space. The "dry" model assumed the Pu-Be alloy at full theoretical density (using the theoretical density of the two metals mixed according to the volume additive rule). The "flooded" model reduced the alloy density to 74 percent, with 26 percent water density to represent flooding the void space between the rods; the alloy and water were smeared together in a homogeneous mixture as in the dry case.

The physical parameters associated with the four models are summarized below:

Var	Parameter	Case I	Case II	Case III	Case IV
A	Be/Pu	13	30	13	30
B	Alloy density	4.725 g/cc	3.224 g/cc	4.725 g/cc	3.224 g/cc
C	Internal Mod.	Dry	Dry	Flooded	Flooded
D	Vol frac Pu	0.1598	0.0764	0.1598	0.0764
E	Vol frac Be	0.8402	0.9236	0.8402	0.9236
F	wt% Pu	67.1%	47.0%	67.1%	47.0%
G	wt% Be	32.9%	53.0%	32.9%	53.0%
H	Alloy vol frac	1	1	0.74	0.74
I	Water vol frac	0	0	0.26	0.26
J	Eff. Pu density	3.170 g/cc	1.515 g/cc	2.346 g/cc	1.121 g/cc

Inputs to the calculation are Variables A and C. Let K = theoretical density of Pu (19.84 g/cc) and L = theoretical density of Be (1.85 g/cc). The remaining variables are computed thus:

Var	Formula
F	100% * 239/(239 + 9A)
G	100% - F
D	0.01 * (LF)/(K + 0.01*(L – K)F)
E	1 – D
B	KD + LE
H	1 for C = "Dry"; 0.74 for C = "Flooded"
I	1 - H
J	0.01 * BHF

Calculation Results

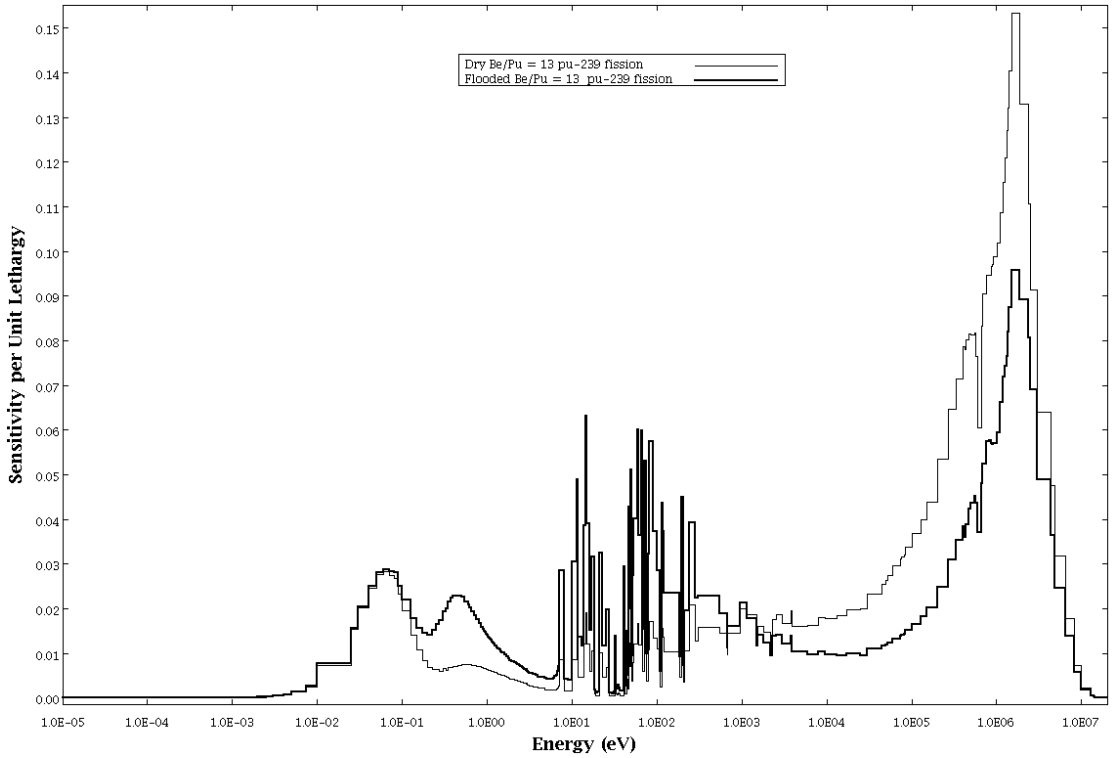
Once the parameters describing the mixture of Pu, Be, and, in Cases III and IV, water, were derived as above, spheres of this mixture surrounded by 30 cm of full-density water were modeled to determine k_{eff}. Due to the simplicity of the model, the 1-D discrete ordinates code XSDRN-PM (by means of the TSUNAMI-1D module) was used to perform the calculations. The radius of the Pu-Be region was varied till the calculated k_{eff} was equal to 0.95. The volume was then calculated and the mass of Pu determined by multiplying this volume by the effective Pu density (Variable J above). This is the "subcritical mass". The "safe mass" was taken as

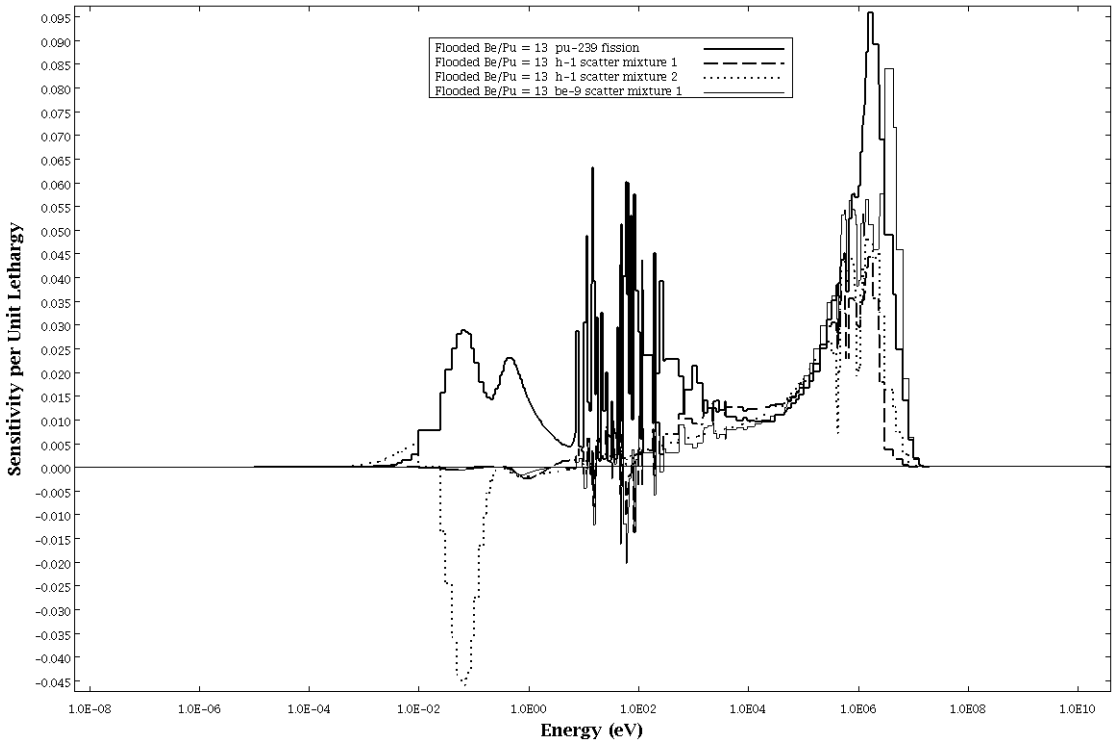
45 percent of the subcritical mass to account for double batching and the traditional additional margin. The calculated results are shown below:

Case	Description Be/Pu; Mod	Subcritical Radius (cm)	Calc k_{eff}	Subcritical Mass (g Pu)	Safe Mass (g Pu)
I	13; Dry	10.68	0.9507	16,178	7,280
II	30; Dry	14.00	0.9499	17,417	7,838
III	13; Flooded	10.33	0.9506	10,833	4,875
IV	30; Flooded	12.17	0.9502	8,466	3,810

Interpretation of Calculation Results

The results demonstrate that both the subcritical mass and the safe mass are much larger than the mystery limit of 2000 g Pu, though the safe mass limit for Case IV is approaching this limit. Since the staff calculations have been very conservative with respect to geometry, reflection, density, and plutonium isotopics, it is hypothesized that the 2000 g limit was intended to be very conservative, or may be based on a Be/Pu ratio somewhat higher than 30. It is recommended therefore that reasonable limits be placed in the guidance on the Be/Pu ratio. This seems to be the most important parameter determining the critical mass. This is supported below.





The assumption of whether the sources are immersed in water, and if so by how much, is not very important is borne out by the first figure above. This is the sensitivity profile generated by TSUNAMI (roughly proportional to the neutron flux times the differential cross section). The profile above is for ^{239}Pu fission, for Cases I and III, and shows that flooding slightly softens the spectrum but not by very much. (The spectrum is harder than expected because we're a long way from the optimum Be/Pu, at ~21,000. For practitioners used to hydrogen, a highly thermalized system would be expected with so much beryllium around. The fact that it is not is because beryllium is 9 times heavier than hydrogen and about 1/10 as good a scatterer, so much more of it is needed to thermalize the spectrum.)

This is further explained in the second figure above, which is for Case III. The heavy solid line is again ^{239}Pu fission for reference. The dashed line represents hydrogen scattering in the Pu-Be-water core. The dotted line represents hydrogen scattering in the thick reflector. The light solid line represents beryllium scattering. (Scattering is what causes moderation, which has the dominant effect on the spectra.) This shows that when fast neutrons are first emitted, the internal water, the reflector, and the beryllium all contribute roughly equally to moderation. But at thermal energies, both the beryllium and the internal water have ceased to be important and the reflector is producing almost all of the moderation.[1] There is so much other moderation going on that adding some water to the core has little effect on the spectrum. While it does have an effect on the critical mass, that is likely due to the reduced density to account for the

[1] Note that the sensitivity coefficient is negative, meaning that increasing the density of the thick reflector causes a *decrease* in reactivity. That is counterintuitive since the system is on the under moderated side of the curve in the figure above and increasing the reflector seldom causes the reactivity to decrease, but criticality safety can be counterintuitive. What appears to be happening is that the negative peak perfectly overlaps the thermal hydrogen absorption peak, so presumably many of the neutrons are being scattered forward into the reflector and subsequently being absorbed by the hydrogen instead of being bounced back into the core. Basically, hydrogen absorption is stronger here than fission.

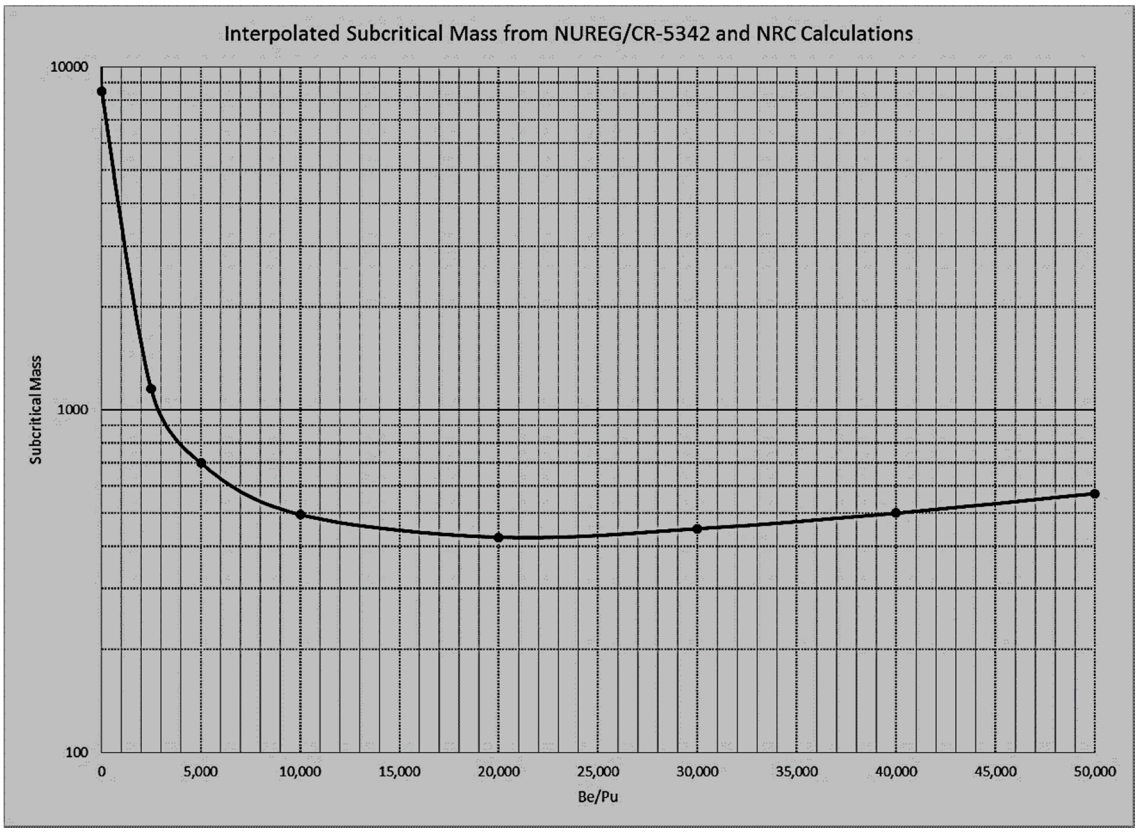
void space caused by heterogeneity in Case III relative to Case I and correspondingly greater leakage into the reflector.

Regardless, k_{eff} and this critical mass seem relatively insensitive to anything except the Be/Pu ratio, so this is probably the only parameter that needs to be specified.

Conclusion and Recommendation

The 2000 g Pu limit is much greater that the safe mass for an optimal system, and therefore it was derived based on assumptions that are not stated in NUREG 1556 Vol. 17. Therefore, it is recommended that some limits be placed on the Be/Pu ratio. The 2000 g limit is conservative by a wide margin for ratios normally encountered in the industry, but given the amount of margin looser bounds can be placed in the ratio without compromising the safety margin. This is particularly important given some uncertainty as to the exact assumptions used in the calculations supporting Ref. 3. Approximating the values off the figure above, and adding the lowest results for Be/Pu = 13 (Case III) and 30 (Case 40) results in the graph below.

Based on this graph, it is therefore recommended that this be used as a technical basis for the 2000 g Pu limit including a restriction of Be/Pu ≤ 1000.



References

1. NUREG 1556 Volume 17, “Consolidated Guidance About Materials Licenses,” November 2000.

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3. NUREG/CR-5342, “Assessment and Recommendations for Fissile-Material Packaging Exemptions and General Licenses within 10 CFR Part 71,” July 1998.

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