Licensing Issues Associated with PuO_2 and Mixed Oxide Powder Processes

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Abstract

The unique regulatory aspects of licensing processes involving low-moderated PuO_2 and mixed oxide (MOX) powders are described. This is based on the approach taken in the proposed MOX Fuel Fabrication Facility (MFFF) at the Savannah River Site, as well as the LaHague and MELOX facilities in France. Anticipated normal and upset conditions for these processes are compared to existing low-moderated benchmark experiments. To assist in comparing benchmarks to anticipated applications, the TSUNAMI sequences of the SCALE 5 code package were used. These sensitivity/uncertainty codes quantitatively measure the sensitivity of the system k-effective to the underlying cross section data, and compute an integral correlation coefficient for each experiment-application pair. The use of this code in addressing these issues is described.

Among the unique regulatory challenges for an MFFF are: the complexities of plutonium chemistry and isotopic composition, control of powder density for criticality control, and control of isotopic composition and homogeneity during the oxide blending process. The relative risks of the various process stages and anticipated conditions are discussed.

Once the highest-risk conditions were defined, the TSUNAMI codes were then used to compare them to the applicant's proposed benchmarks. The results of these codes were also confirmed using direct perturbation of system parameters. The application of these codes to address variations in powder density, moderator content, and isotopic nature of the plutonium is described. The application to determining a bounding fissile medium to represent miscellaneous plutonium compounds is described. The application of PuO_2-polystyrene and Pu-metal benchmarks to low-moderated PuO_2 applications is discussed. Areas where additional critical benchmarks would be helpful are then discussed.

MFFF Processes and Regulatory Issues

The introduction of plutonium into a historically uranium-based fuel raises several challenges in the area of nuclear criticality safety (NCS). The underlying physics concepts and basic phenomena are no different from those encountered in processes with low- or high-enriched uranium. However, there are important differences that need to be considered in the design of an MFFF.

Attachment

The use of MOX with a plutonium content in the range of 2-6wt% Pu/(U+Pu) can result in fuel that is substantially more reactive than typical (i.e., <5wt% ^{235}U) U.S. light-water reactor fuel. ^{239}Pu tends to have a much smaller minimum critical mass than ^{235}U. This is true even for plutonium nitrate vs. uranyl nitrate, despite a higher nitrogen-to-fissile ratio (Table 1). The larger volume and dimensional limits for plutonium nitrate indicate that optimal moderation for Pu tends to occur at a much higher H/X than for U—or equivalently, that a ^{239}Pu system exhibits a harder neutron spectrum than a ^{235}U at the same H/X. This becomes important in validation because, at moderation levels anticipated in MOX powder processes, there can be a strong component in the intermediate and fast neutron energy ranges.

Table 1. Single-Parameter Limits (from ANSI/ANS-8.1-1983)

	LEU (<5wt% ^{235}U)	$^{235}UO_2(NO_3)_2$	$^{239}Pu(NO_3)_4$
Mass	3.3 kg (7.3 lb)	0.78 (1.7 lb)	0.48 (1.1 lb)
Cyl. Diam.	42.7 cm (16.8 in)	14.4 cm (5.7 in)	15.4 cm (6.1 in)
Slab Thick.	23.4 cm (9.2 in)	4.9 (1.9 in)	5.5 (2.2 in)
Volume	111 l (29.3 gal)	6.2 l (1.6 gal)	7.3 (1.9 gal)

A second unique aspect is the complexity of the isotopic mixture. In a typical uranium-based fuel cycle facility, only the ^{235}U enrichment need be specified to fully characterize the fissile material. In an MFFF, both the plutonium and uranium isotopics must be specified, in addition to the ratio of plutonium to uranium. Incoming feed to the proposed U.S. MFFF would be limited to 90 – 95wt% ^{239}Pu, 5 – 9wt% ^{240}Pu, and < 1wt% ^{241}Pu. Depleted uranium would be limited to < 0.25wt% ^{235}U. PuO_2 would then be downblended to MOX powder in two stages, first in a master blend with 20wt% Pu content and then in a final blend with 2 – 6wt% Pu content. The master blend would be analyzed at 22wt% Pu content and the final blend at 6.3wt% Pu content. The applicant has proposed bounding assumptions for the isotopics of the incoming feed material. MOX isotopics would be controlled in the powder blend operation. A specified quantity of depleted UO_2 powder would be metered into the large geometry blend tank and then combined with a specified quantity of PuO_2 powder to produce MOX powder. The NRC staff considers this one of the key processes with regard to criticality safety in the entire MFFF, because the resultant isotopic mix is credited for the safety of all downstream processes. It is particularly important to obtain a highly uniform mix to avoid having "hot spots" with greater than allowed Pu content. This would be done by controlling the amount of moderator (both for reactivity and to prevent clumping), by mechanical stirring, and by subsequent homogenization of the powder. It is anticipated that in the blend tank there would be a significant contribution from the intermediate neutron energy range.

A third unique aspect is caused by the complexity of plutonium chemistry. Plutonium has up to six positive valence states and can form a large number of different chemical compounds. The valence state is dependent on process conditions (e.g., acidity, presence of oxidizing/reducing agents, temperature). Though the electronic configuration has essentially no effect on the nuclear properties of plutonium, it can affect both the process

flow and system reactivity. The efficiency of the purification cycle in aqueous polishing depends strongly on the Pu valence state; Pu(IV) forms much stronger complexes with tributyl phosphate(TBP) than Pu(III) does. The presence of Pu in the Pu(IV) oxidation state is required in the extraction, scrubbing, and stripping processes; the correct operation of these processes ensures there is very little plutonium left in the solution raffinate. In the stripping column, the plutonium is reduced to Pu(III) using hydroxylamine nitrate (HAN) and stripped from the solvent. The plutonium must then be oxidized back to Pu(IV) using nitrous oxide before being contacted with oxalic acid and precipitated as plutonium oxalate. In terms of reactivity, the valence state determines the chemical form (e.g., $Pu(III)(NO_3)_3$ versus slightly less reactive $Pu(IV)(NO_3)_4$). Because of this complex chemistry, a large variety of chemical forms must be considered in the criticality evaluation of the MFFF (PuO_2, PuO_2F_2, $Pu(NO_3)_3$, $Pu(NO_3)_4$, and $Pu(C_2O_4)_2$). In the U.S. MFFF design, PuO_2F_2 has been proposed as a bounding fissile medium; the validation of this material with existing benchmarks is treated below.

A fourth unique aspect is the important role that density plays in the criticality safety basis. The proposed U.S. MFFF contains processes that credit several different values of PuO_2 and MOX density, ranging from 3.5 – 7 g/cm^3 (218.8 – 437.6 lb/ft^3), as well as full theoretical density of 11.46 g/cm^3 (716.4 lb/ft^3) for incoming powder and sintered material. A PuO_2 density of 7 g/cm^3 (437.6 lb/ft^3) is recognized as being very conservative even for tapped powder. However, the method of preparation, process conditions, and any mechanical handling can all affect the density. Measurements have been conducted on PuO_2 powder prepared in several different chemical processes, resulting in densities of 1 – 2.3 g/cm^3 (62.5 – 143.8 lb/ft^3) (Ref. 1). The lowest reported densities were those for PuO_2 produced from oxalic precipitation, as in the proposed U.S. MFFF. The situation regarding the density of MOX powder is more indefinite. When powders with two different densities are combined, the total volume may be less than the sum of their individual volumes (i.e., powder blending may be non-volume additive). There appears to be little available data in the literature for the density of MOX powders with different isotopic mixes. The measurement of actual in-process powder density is therefore very important whenever less than theoretical powder density is credited for criticality safety. The effect of density on applicability of existing benchmarks is treated below.

Perhaps the most significant unique aspect associated with processing plutonium occurs in the area of computer code validation. This is not due to any inherent complexities associated with plutonium, but rather with benchmark experiments available in the literature. With regard to validation, the Code of Federal Regulations (CFR) includes the following requirement for criticality safety at NRC-regulated fuel facilities (10 CFR 70.61(d)):

> "…the risk of nuclear criticality accidents must be limited by assuring that under normal and credible abnormal conditions, all nuclear processes are subcritical, <u>including use of an approved margin of subcriticality for safety</u>…."

The determination of an appropriate "margin of subcriticality" for processes associated with MOX fuel manufacturing is not a trivial exercise. Traditionally, higher-enriched fuel facilities have been licensed with larger margins in k_{eff} than low-enriched facilities. Determination of an appropriate margin must take into account several considerations, including inherent risk of the various processes, the sensitivity of k_{eff} to changes in the underlying parameters, and the quality (i.e., number and similarity) of available critical benchmarks. This paper looks at each of these three considerations and applies them to operations in the proposed U.S. MFFF that is currently under NRC review.

MFFF Processes and Risk Perspective

In approaching criticality code validation, the proposed U.S. MFFF divides operations into five different area(s) of applicability (AOA(s)), as follows:

1. Plutonium nitrate solutions.
2. Mixed oxide fuel pellets, rods, and assemblies.
3. Plutonium oxide powder.
4. Mixed oxide powder.
5. Miscellaneous plutonium compounds.

A separate validation was performed, documented, and reviewed for each of the five AOAs above. The design of the proposed MFFF was based on Cogema's LaHague (aqueous polishing) and MELOX (MOX fuel fabrication process) facilities, with the exception that the U.S. plant would use weapons-grade plutonium (≤96wt% ^{239}Pu) as feed material instead of reactor-grade plutonium. This simplifies the plutonium isotopic vector, but also increases system reactivity and decreases any margin that may result from the fresh-fuel assumption for reactor-grade plutonium.

Based on the history of criticality accidents (Ref.2), the highest risk is expected to occur in the aqueous polishing process (covered by AOA(1) and AOA(5)). The lowest risk is expected to be in the areas handling finished MOX fuel (covered by AOA(2)) due to the comparatively low Pu assay and the fixed material form. This qualitative comparison is true in terms of the unmitigated risk—that is, based solely on the form of material and its potential to reach a critical state. Solutions, which have small critical masses and can readily conform to an unfavorable geometry, have relatively high unmitigated risk. MOX fuel, with a large critical mass and stable form, has relatively low unmitigated risk. Intermediate in risk between these are the PuO_2 and MOX powders, which are dry under normal conditions (with an assumed bounding moisture content of 1wt% H_2O), vary in assay from high to low Pu, and are conformable to an unfavorable geometry container.

The picture is somewhat different when mitigated risk is considered—the design ensures that plutonium nitrate solutions are passively controlled, relying mainly on fixed neutron absorbers and favorable geometry vessels. Powder operations, conversely, rely mainly on mass and moderation control, and are much more hands-on operations. Therefore, these operations are expected to be much more administratively controlled than aqueous

polishing. When all this is considered, PuO_2 and MOX powder processing operations may be the areas of highest overall mitigated risk.

These processes are also especially important due to their unique nature, reliance on isotopic blending, and the higher uncertainties associated with bulk powder systems (uncertainties in density, nuclear data in the intermediate energy range, and lack of benchmarks).

Parametric Evaluation of MFFF Processes

Ref. 3 contains a list of steps for validating calculational methods, and in particular for determining the method's AOA. The first step is the characterization of the key parameters associated with normal and credible abnormal conditions.

The parameters traditionally associated with validation include the moderator-to-fissile ratio (e.g., H/X for homogeneous mixtures), isotopic character (e.g., ^{239}Pu assay, ^{235}U enrichment, Pu/(U+Pu) content), and energy parameters (e.g., thermal fission fraction, average energy group, energy of average lethargy causing fission (EALF)). These continuous parameters are convenient for trending, and are among those thought to have the largest potential effect on system k_{eff}, and thus, on bias. The parameters to which k_{eff} has the highest sensitivity can be supposed to be the most likely to produce a trend in the bias (i.e., traditional trending parameters).

Anticipated design applications for powder systems include spheres (when relying on mass control) and other simple geometrical configurations of uniform PuO_2- or MOX-water systems (with 100, 22, and 6.3wt% Pu content). The powder may be assumed to be at full theoretical density (and thus dry) or at some reduced density. The system may have varyious dimensions depending on the size of batches, and may have various reflector conditions (from one-inch tight-fitting nominal reflection to full water reflection, or reflection by materials such as concrete, steel, or depleted uranium). The dimensions and reflector conditions primarily affect the neutron energy spectrum.

Among the most important parameters are those characterizing the fissile medium. PuO_2 and MOX powders will be assumed to have a moisture content of 1wt% H_2O (corresponding to an H/X of ~0.3) under normal conditions. Some water moderation should always be assumed, due to the hygroscopic nature of this material. The energy spectra of these systems are expected to vary widely, depending on the total mass, dimensions, and reflection. Mass will vary from effectively infinite for the blend tank down to a few kg for the incoming PuO_2 containers. The spectra of individual design applications and benchmarks were characterized by specifying the value of EALF. However, it should be recognized that the spectrum of a low-moderated, reflected system typically covers the entire range from very fast to thermal energies, as shown in Fig. 1. Systems with different values of EALF can still have similar neutron spectra. Experiment NSE55T5-07 has an EALF = 43.5 eV (5.15×10^{-18} ft-lb), while PMF016-01 has an EALF = 11.7 keV (1.38×10^{-15} ft-lb), even though there is considerable overlap in much of the spectra.

Figure 1. Fission Spectra for Typical Pu-Metal and MOX-Polystyrene Benchmark

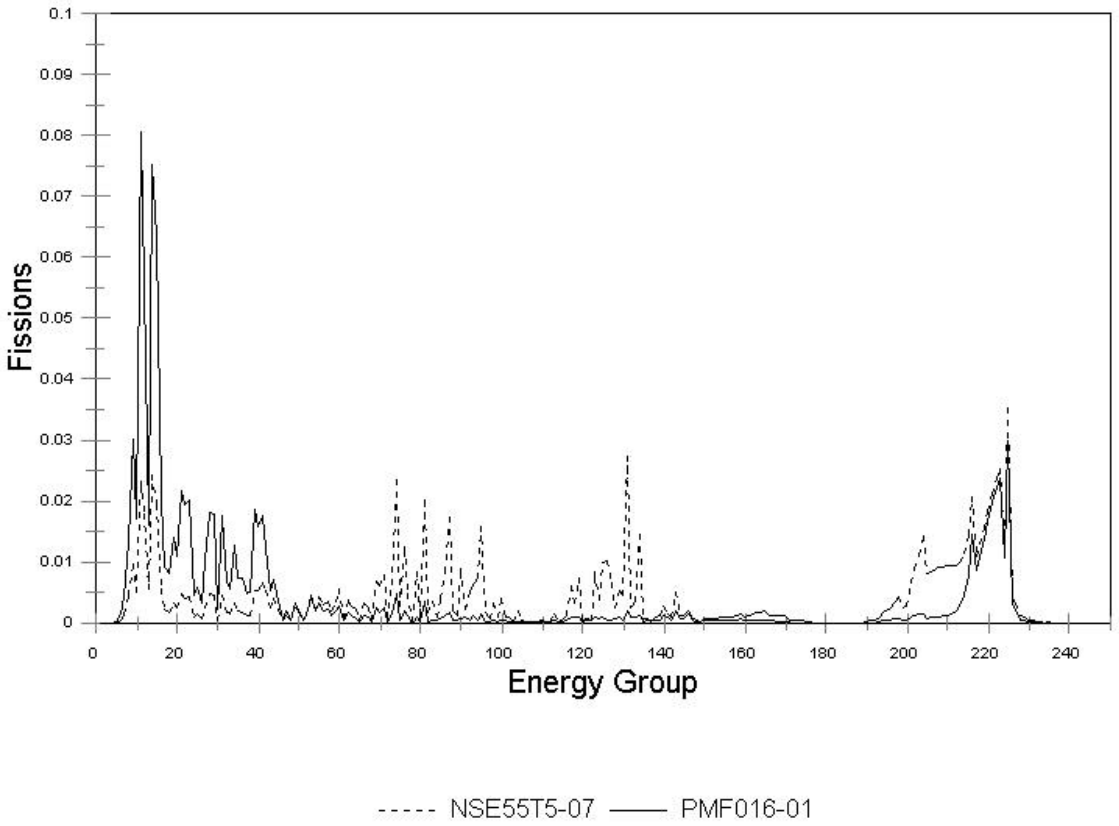


Table 2 below shows some of the more important parameters associated with MFFF processes under both normal and credible abnormal conditions. Under abnormal conditions, a broader range of parameters may be encountered, such as upsets involving the moderation of dry powders, which will account for the highest value of H/X.

Table 2. Parameters Associated with Powder Handling Operations

	PuO_2 Powder AOA(3)	MOX Powder AOA(4)
Material Form	PuO_2-water mixture	MOX-water mixture
Geometric Form	Parallelepipeds Arrays of cylinders Spheres	Parallelepipeds Spheres
Isotopics	96wt% ^{239}Pu 0.3wt% ^{235}U 100wt% Pu/(U+Pu)	96wt% ^{239}Pu 0.3wt%^{235}U 6.3, 22wt% Pu/(U+Pu)
Density	3.5 – 11.46 g/cm^3	5.5 g/cm^3 (343.8 lb/ft^3)

	(218.8 – 716.4 lb/ft^3)	
H/X	0 – 16.5	1.1 – 1.6
EALF	3 eV – 266 keV ($3.55\text{x}10^{-19}$ – $3.14\text{x}10^{-14}$ ft-lb)	0.28 eV – 850 eV ($3.31\text{x}10^{-20}$ – $1.00\text{x}10^{-16}$ ft-lb)
Reflector	Water, Cd, Concrete	Water

A challenge is posed by the fact that there are few, if any, experiments similar to anticipated design applications for AOA(3) and AOA(4). Applications will consist largely of homogeneneous masses of uncontained, slightly moderated PuO_2- and MOX-water mixtures. Experiments drawn from the "International Handbook of Evaluated Criticality Safety Benchmark Experiments" (Ref. 4) were evaluated for their suitability for use in validating PuO_2 and MOX powder calculations (Ref. 5 and 6). These experiments can be lumped into two broad categories: (1) Pu-metal benchmarks, with an H/Pu = 0; and (2) low-moderated PuO_2- or MOX-polystyrene compacts. The geometric shape, density, physical form, chemical composition, moderation, and isotopic character of these benchmarks can vary significantly from those of anticipated design applications. Application of the traditional method of relying on qualitative comparison of benchmarks to design applications does not yield enough applicable benchmarks to enable calculation without the use of a statistical margin of up to several percent in k_{eff}.

Therefore, it was necessary to develop other means to determine whether benchmarks that do not appear applicable on the surface do have sufficiently similar neutron physics to be useful for validation. This was done by developing a set of "screening criteria" that can be applied to a set of candidate benchmark experiments. The first decision faced by an analyst is which of the system parameters (including those in Table 2) are the most likely to affect the bias. Table 2.3 of Ref. 3 contains parametric criteria for determining the AOA, though this reference states that these are based on the consensus of a number of criticality experts and are therefore conservative.

Ref. 5 and 6 used Oak Ridge's TSUNAMI sensitivity/uncertainty codes to compare a set of benchmark experiments directly to hypothetical design applications (uniform spheres of PuO_2- and MOX-water mixtures with various reflector conditions). This code provides powerful insights into the neutronic behavior of fissile systems, but is still under development. One caveat is that care must be taken to select an appropriate cutoff value of c_k. The NRC staff experience has been that cases with a $c_k \geq 0.95$ show a high degree of similarity in terms of traditional comparison, while some differences were noted for cases with c_k between 0.90 and 0.95. A working definition has evolved that agreement between systems with $c_k > 0.90$ is "good" and agreement between systems with $c_k > 0.8$ is "marginal."

In the present study, the TSUNAMI code was used to investigate the relative importance of different system parameters to aid in the development of a set of screening criteria, rather than to directly compare benchmarks to design applications. Once a hypothetical design has been defined, the key parameters can be varied to investigate the effect of

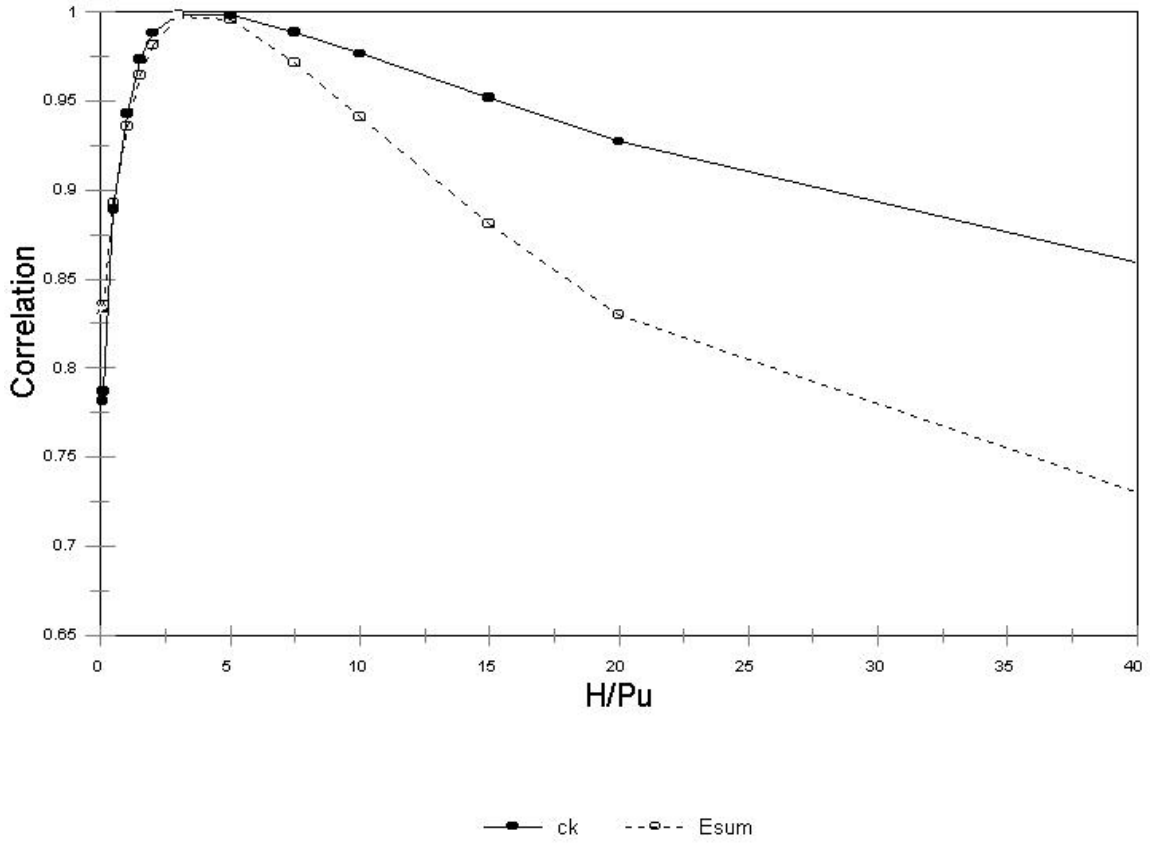
parametric changes on the sensitivity and correlation coefficients. This results in a curve showing the value of a sensitivity or correlation coefficient as a function of the parameter. Implicit in this approach is the idea that if k_{eff} is relatively insensitive to a given parameter, then the correlation coefficient will be close to unity and will not exhibit much variation. If k_{eff} is highly sensitive to a given parameter, then the correlation coefficient will exhibit a strong dependency on this parameter and will drop off rapidly. In addition to giving information on the relative importance of various parameters, this also enables an analyst to develop a set of screening criteria without reference to specific benchmarks. Generally the more sensitive k_{eff} is to a given parameter, the narrower the screening range, and the higher the desired confidence (i.e., the higher the c_k cutoff), the narrower the range. The results of several sensitivity studies showing the effect of parameter variations on c_k are shown in the following sections.

Plutonium Powder Systems

Figure 2 shows the result of a sensitivity study in which a uniform PuO_2-water sphere was modeled with varying levels of moderator, in order to determine an appropriate screening criterion in H/X. Early investigations showed that the material properties (isotopic composition, moderation, absorbers) had a much more significant effect on c_k than did system geometry. Therefore, the comparisons were done on spherical models using the TSUNAMI-1D code, which relies on the XSDRNPM one-dimensional discrete ordinates code, instead of modeling a realistic process explicitly. Uniform spheres of the fissile medium were used in most cases, although cylinders or slabs were also modeled and showed very similar results.

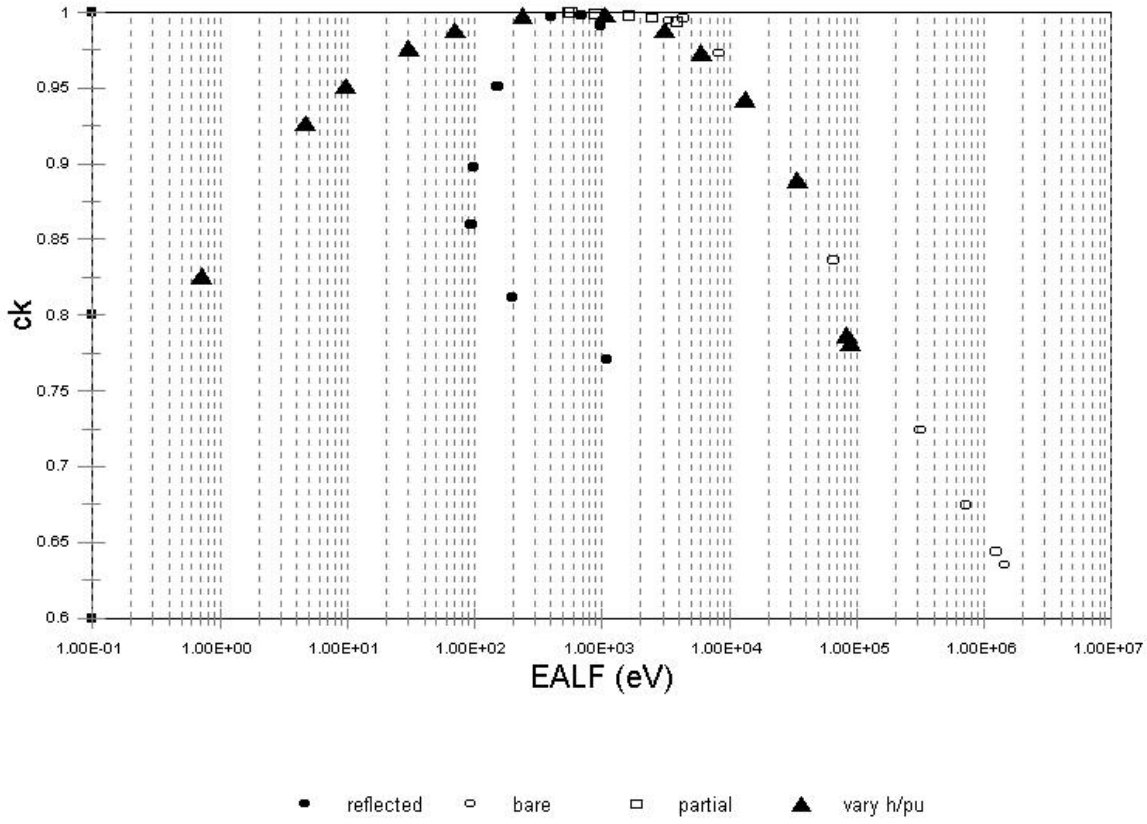
The reference case chosen consisted of a fully-reflected PuO_2-water sphere with H/Pu = 3.79 and EALF = 544 eV (6.43×10^{-17} ft-lb). These values were chosen because this was near the middle of the parametric range defining AOA(3) at an earlier stage in the proposed U.S. MFFF design. The range over which c_k exceeded 0.9 was $0.9 \leq H/Pu \leq 13.5$. The range over which c_k exceeded 0.8 was $0.6 \leq H/Pu \leq 28$. The strong dependence of c_k on H/Pu is not unexpected, because most of the sensitivity in the PuO_2-water cases is consistently due to ^{239}Pu and ^{1}H.

Figure 2. C_k dependence on H/Pu ratio (H/Pu = 3.79 Reference Case)



Because there is a strong correlation between H/Pu and EALF, additional sensitivity studies were performed with spheres having different diameters and reflection conditions, but with H/Pu = 3.79. (EALF for the hypothetical spherical cases ranged from 89 keV (1.05×10^{-14} ft-lb) (at H/Pu = 0.05) down to 0.72 eV (8.51×10^{-20} ft-lb) (at H/Pu = 50). The value of c_k was then graphed as a function of EALF, as shown in Fig. 3. Also included on this graph are the results of the above study, in which H/Pu was varied from 0.05 to 50 with full water reflection.

Figure 3. C_k as a function of EALF (EALF = 544 eV (6.43×10^{-17} ft-lb) Reference Case)



The similarity between the curves when H/Pu was varied and when the diameter of the sphere was varied indicates that most of the strong dependence of c_k on H/Pu (in Fig. 2) is due to changes produced in the neutron energy spectrum. Using the bounding curves, $c_k > 0.9$ for systems with EALF from ~2 eV – 25 keV (2.36×10^{-19} – 2.96×10^{-15} ft-lb); $c_k > 0.8$ for EALF from ~0.4 eV – 100 keV (4.73×10^{-20} – 1.18×10^{-14} ft-lb). This is qualitatively consistent with the results of Ref. 3, in which the neutron energy screening criteria require only that EALF be in the same energy range (i.e., thermal, intermediate, or fast). This is perhaps explained by the fact that even systems with very different EALF can have neutron spectra with considerable overlap over much of the range from thermal to fast neutron energies.

Similar investigations were also done with regard to other system parameters, including powder density, geometry, and plutonium isotopics (^{240}Pu assay). These studies showed that there was relatively little effect on c_k from any of these parameters, so that they may be regarded as of secondary importance. The theoretical density of powder in the PuO_2-water system was gradually decreased from 11.46 – 3 g/cm^3 (716.4 – 187.5 lb/ft^3), while maintaining H/Pu = 3.79 (thus increasing the effective void space). This had a very small

effect on the correlation coefficient, which dropped to ~0.97 for a system at full theoretical density compared to one at 3 g/cm^3 (187.5 lb/ft^3).

The effect of system geometry was similarly small. Because the geometric size and shape can affect the value of EALF, spheres, cylinders, and slabs of varying sizes were modeled and compared to the reference (spherical) case. When c_k was graphed as a function of EALF, the curves corresponding to spheres, cylinders, and slabs were almost coincident. This indicates that it is the material properties that are of primary importance to benchmark applicability, and the geometric form is much less important. Size appears to be significant only in that it affects the neutron energy spectrum.

The effect of ^{240}Pu assay was similarly found to be of secondary importance. The weight percent of ^{240}Pu in the plutonium models was varied from 2 – 30wt% (MOX fuel will be analyzed at 4wt% ^{240}Pu). A two-isotope ^{239}Pu-^{240}Pu mixture is assumed. Even at a ^{240}Pu content of 30wt%, the value of c_k when compared to a nominal 4wt% reference case was > 0.97. This was not unexpected, as the sensitivity of these systems to ^{240}Pu accounted for only a few percent of the total sensitivity to the fissile mixture. Direct perturbation of number densities was employed as a spot check on the sensitivity calculations.

One application concerned miscellaneous Pu compounds present during the aqueous polishing process in the proposed U.S. MFFF. Various compounds (including PuO_2, PuO_2F_2, $Pu(NO_3)_4$, and $Pu(C_2O_4)_2$) are to be modeled as PuO_2F_2. The optimum nature of PuO_2F_2 over the range of interest in H/Pu can be confirmed by a traditional sensitivity calculation. However, none of the benchmarks proposed in Refs. 5 or 6 contained PuO_2F_2 as the fissile medium. Curves provided in the validation report showed similar EALF values for subcritical (k_{eff} = 0.93), fully reflected cylinders containing PuO_2 to those containing PuO_2F_2, over a broad range in H/Pu of 0.3 – 30. However, the difference in EALF values gradually increased at low H/Pu. This raised a two-fold question: (1) whether a comparison of the EALF values is sufficient to demonstrate that two cases are closely related; and (2) if so, what difference in energy represents when two cases are not closely related. To test this methodology, the NRC staff ran several cases (at H/Pu = 0.3, 1, 3, 10, and 30) and, at each H/Pu, computed the c_k value for the PuO_2 system vs. the PuO_2F_2 system. The results showed that, across the range of H/Pu = 0.3 – 30, there was a very high degree of correlation ($c_k \geq 0.98$) between the PuO_2 and PuO_2F_2 systems. This can be explained by the low value of ^{16}O and ^{19}F sensitivity coefficients as compared to the total mixture sensitivity.

Another area in which TSUNAMI proved useful was the validation of calculations with strong neutron absorbers (such as borated steel, borated concrete, and Cd). Frequently, available benchmarks did not contain these materials, so decisions had to be made with regard to when design applications that had strong absorbers were applicable to benchmarks that did not. Traditionally, benchmarks were required to have the same materials as design applications. This is not always feasible. Using the current approach, however, limited use of absorbers may be appropriate even when benchmarks do not contain these materials, provided that the sensitivity of k_{eff} to these materials is low.

MOX Powder Systems

Low-moderated MOX powder systems can be expected to have many of the same properties as low-moderated PuO_2 systems, in terms of relative sensitivity to various system parameters. The actual permissible ranges of fundamental parameters may be different, however, because very pure ^{235}U or ^{239}Pu systems tend to have greater sensitivity to parameter changes than low ^{235}U or ^{239}Pu systems. The most significant physical difference between PuO_2 and MOX powders is the presence of large quantities of ^{238}U (88 and 93.7wt% ^{238}U for the master and final blend respectively). ^{238}U capture can contribute a significant fraction of the total fissile mixture sensitivity in the thermal to intermediate energy range, while ^{238}U fission contributes in the fast energy range. Typical sensitivity profiles for ^{239}Pu fission and capture are shown in Fig. 4, for a case with ~30wt% Pu content. Typical sensitivity profiles for ^{238}U fission and capture are shown in Fig. 5.

The total sensitivity coefficients for the dominant reactions for this case are shown below in Table 3, in decreasing order. The subscripts (f), (c) , and (s) refer to fission, capture, and scattering. The total sensitivity for this fissile mixture was 0.306.

Table 3. Dominant sensitivity coefficients for NSE55T5-07

$^{239}Pu(f)$	$^{239}Pu(c)$	$^{1}H(s)$	$^{238}U(c)$	$^{240}Pu(c)$	$^{238}U(f)$
0.424	-0.209	0.115	-0.085	-0.054	0.029

Note that ^{240}Pu was previously found to be of secondary importance, so anything with a coefficient less than that for ^{240}Pu capture is also a minor contributor.

What we see from Fig. 4 is that in the fast energy range, almost all of the sensitivity of k_{eff} to ^{239}Pu (the dominant nuclide) is due to ^{239}Pu fission. In such a regime, it would not be unreasonable to base acceptance largely on a comparison of the ^{239}Pu fission spectra. However, in the thermal and intermediate ranges, ^{239}Pu capture becomes increasingly important. Fig. 5 shows that ^{238}U fission is an important contributor at fast energies and ^{238}U capture becomes significant at intermediate energies—even rivaling the sensitivity of ^{239}Pu fission and capture in this regime.

Figure 4. ^{239}Pu total, fission, and capture sensitivity profile for NSE55T5-07

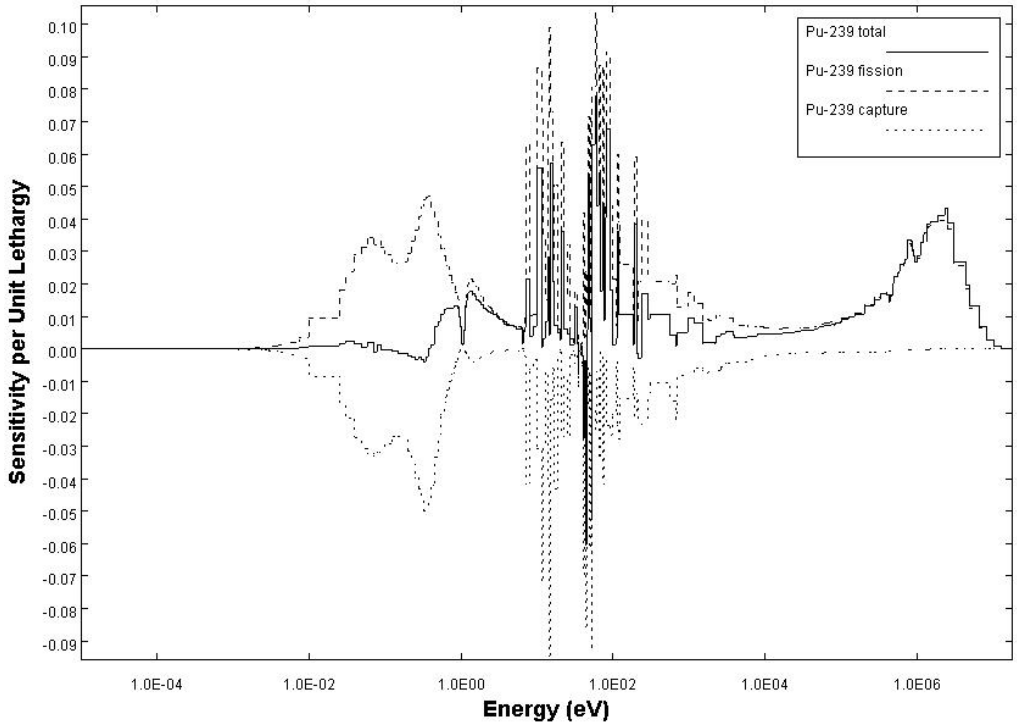
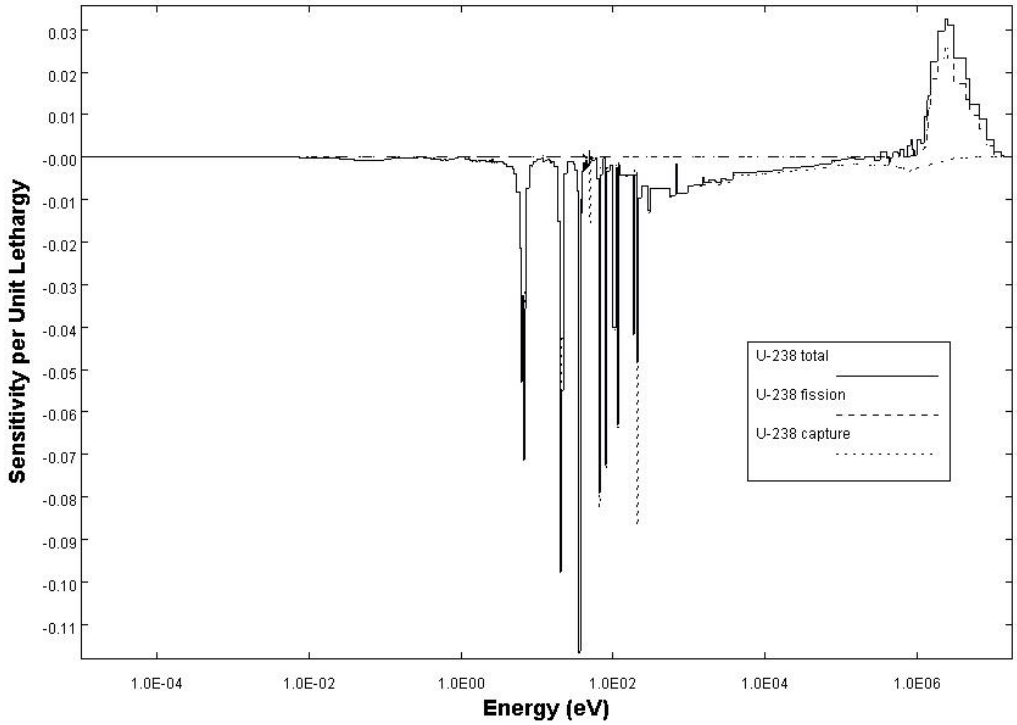


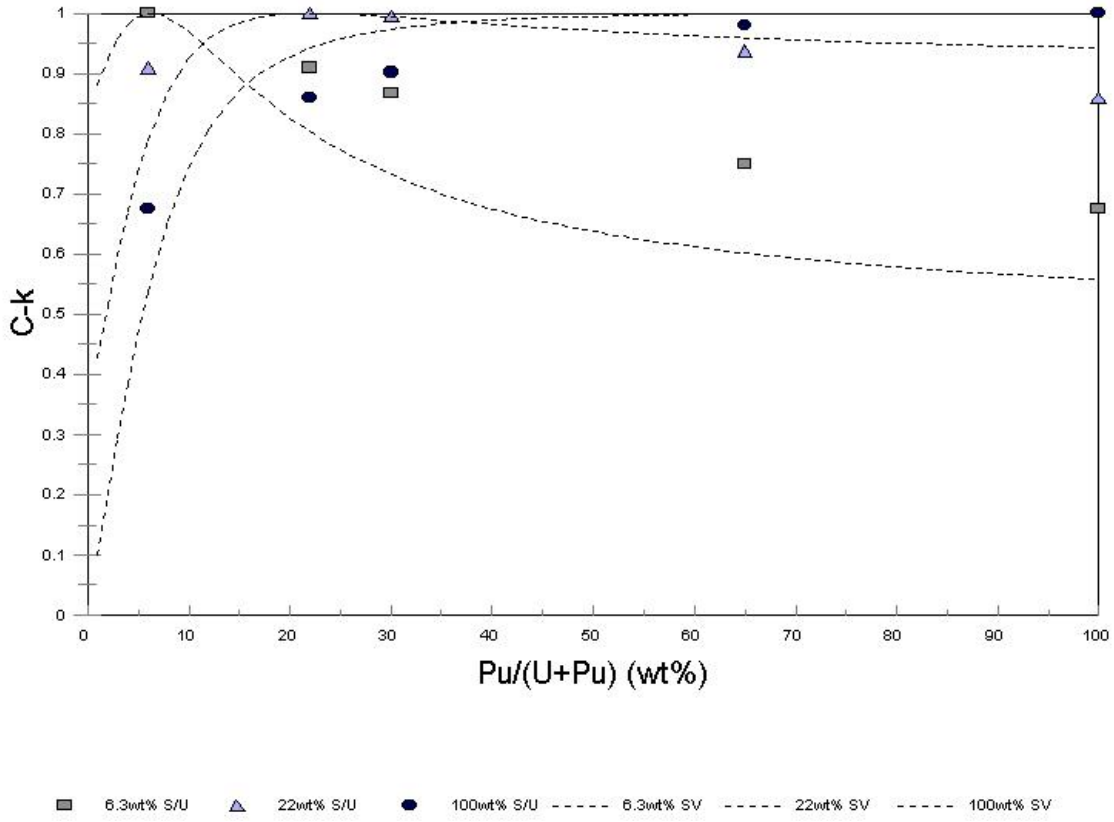
Figure 5. ^{238}U total, fission, and capture sensitivity profiles for NSE55T5-07



Therefore, in systems in which there is a large amount of ^{238}U, the effect on this nuclide on the overall system k_{eff} cannot be discounted. ^{239}Pu fission dominates in the fast energy range but for systems with sizeable spectral contributions in the intermediate and thermal ranges, the relative sensitivity of k_{eff} to other nuclide-reaction pairs must be considered.

The results of a parametric analysis in which MOX-water spheres were modeled with varying Pu-content are presented below in Fig. 6. In addition to changing the number densities of ^{239}Pu and ^{238}U, increasing the Pu-content of the mixture results in a hardening of the fission spectrum, which may be responsible for part of the drop-off in c_k with increasing Pu-content.

Figure 6. Correlation of MOX-water spheres to 6.3, 22, and 100wt% cases



This graph shows that there is a considerable drop-off in c_k as the difference in Pu-content between the cases compared grows. The points represent c_k values calculated using the TSUNAMI code; the MOX-water spheres were modeled using the XSDRNPM discrete ordinates code. This shows that 100wt% Pu benchmarks have a high degree of applicability to applications with much lower Pu-content, but that there is a sudden drop-off below ~30wt% Pu. The breadth of this range is thought to be due to the dominance of ^{239}Pu fission, which is responsible for almost all of the system reactivity. Removing the ^{238}U results in a net change in k_{eff} of at most a few percent over the range of 30-100wt% Pu.

The dashed curves are the result of another type of analysis. Ref. 7 derives an analytical approach for the correlation between a pair of two-isotope systems. This was originally derived to investigate the behavior of a partially enriched system, but can be applied to a ^{239}Pu-^{238}U system as well. The dashed curves are the tabulated values of the following function for the correlation parameter:

$$\rho = \frac{\{\sigma_1\sigma_2\varepsilon_1\varepsilon_2\delta_P^2 + \sigma_1\sigma_2(1-\varepsilon_1)(1-\varepsilon_2)\delta_U^2\}}{\sqrt{\{\sigma_1^2\varepsilon_1^2\delta_P^2 + \sigma_1^2(1-\varepsilon_1)^2\delta_U^2\}}\sqrt{\{\sigma_2^2\varepsilon_2^2\delta_P^2 + \sigma_2^2(1-\varepsilon_2)^2\delta_U^2\}}}$$

In this application, σ_P and σ_U are the total ^{239}Pu and ^{235}U cross sections, δ_P and δ_U their uncertainties, and ε_1 and ε_2 the Pu-content of the two cases. The total macroscopic cross sections of the two mixtures can be expressed as $\sigma_1 = \sigma_P\varepsilon_1 + \sigma_U(1-\varepsilon_1)$ and $\sigma_2 = \sigma_P\varepsilon_2 + \sigma_U(1-\varepsilon_2)$. This simple expression does not take into account energy- and reaction-dependent differences in the cross sections, but only makes a gross comparison that depends on the ratios of the total cross sections and their uncertainties. In the intermediate-to-fast energy range, the ratio of the ^{239}Pu to the ^{238}U total cross section is ~1. In the thermal energy range, however, the ratio ranges from ~10 to ~100 due to the prevalence of thermal ^{239}Pu fission. The curves above were derived using the assumption that the uncertainties are in the same ratio as the cross sections, with σ_P / σ_U (and thus δ_P / δ_U) = 100. Although a much less sophisticated treatment, this gives results qualitatively similar to that using the full TSUNAMI approach.

Based on the TSUNAMI analysis, the following range in Pu-content is derived, based on a c_k cutoff of 0.90. For 100wt% Pu applications, the screening range is 30 – 100wt% Pu. For 22wt% Pu, the screening range is 4 – 82wt% Pu. And for 6.3wt% Pu, the screening range is ≤ 23wt% Pu.

Application to Existing Benchmarks

Based on the foregoing parametric analysis for low-moderated PuO_2- and MOX-water systems, the parameters of greatest significance to benchmark applicability are the H/X ratio and total Pu-content. Other parameters, including geometric form, physical and chemical form, density, and plutonium isotopics (^{240}Pu content) appear to play only a minor role.

The following set of screening criteria are based on the application ranges in Table 2, the results of TSUNAMI calculations, and the screening criteria in Table 2.3 of Ref. 3. The parametric ranges covered by the experiments analyzed in Refs. 5 and 6 are also included for comparison.

Table 4. Screening Ranges and Benchmarks for AOA(3)

Parameter	Ref. X	TSUNAMI	Applications	Benchmarks
H/X	0 – 19.8	0 – 25	0 – 16.5	0 – 15.1
Pu-content	90 – 100wt%	30 – 100wt%	100wt%	29.3 – 100wt%
^{240}Pu	0 – 8wt%	0 – 30wt%	4wt%	2.2 – 18.4wt%
EALF	1 – 2x10^{7} eV (1.18x10^{-19} – 2.36x10^{-12} ft-lb)		3 – 2.66x10^{5} eV (3.55x10^{-19} – 3.14x10^{-14} ft-lb)	4 – 1.25x10^{6} eV (4.73x10^{-19} – 1.48x10^{-13} ft-lb)

Table 5. Screening Ranges and Benchmarks for AOA(4)

Parameter	Ref. X	TSUNAMI	Applications	Benchmarks
H/X	0.88 – 1.92		1.1 – 1.6	2.77 – 15.1
Pu-content	3.8 – 37	≤ 82wt%	6.3, 22wt%	8.1 – 100wt%
^{240}Pu	0 – 8wt%		4wt%	2.2 – 11.5wt%
EALF	0 – 10^{4} eV (0 – 1.18x10^{-15} ft-lb)		0.28 – 850 eV (3.31x10^{-20} – 1.00x10$^{=16}$ ft-lb)	0.6 – 92.6 eV (7.09x10^{-20} – 1.09x10^{-17} ft-lb)

Note that TSUNAMI analyses were not run for every combination of parameters. For MOX powders, only the effect of Pu-content was investigated; the effect of the other parameters is presumed roughly similar to that for PuO_2 powders.

Based on this table, it appears that the primary limitation of existing benchmarks is their ability to cover the lowest energy powder systems, as well as low-Pu content systems. The application of TSUNAMI results in general in less restrictive screening ranges than those derived using Table 2.3 of Ref. 3 (especially in regard to H/X, Pu-content, and ^{240}Pu-content). The use of a different cutoff value for c_k would have a corresponding effect on these values.

There are two issues that are of particular interest—(1) the applicability of Pu-metal benchmarks to validate MOX-power systems, and (2) the applicability of PuO_2 systems to validate low-Pu MOX systems. The dependence of c_k on Pu-content has already been explored, and this reveals the need for additional low-Pu MOX benchmarks. With regard to Pu-metals, replacing low-density PuO_2 powder with full-density Pu-metal is expected to have a very slight effect on system c_k. This is expected based on the demonstrated low dependence on powder density, and on the low sensitivity to the unique element present in powder (i.e., the ^{16}O cross sections). The primary issue with regard to use of Pu-metal benchmarks, then, would seem to be the fact that these benchmarks have H/Pu = 0.

Conclusions

The primary area of concern with regard to subcritical margin appears to be the low-energy (thermal to intermediate) range of PuO_2 and especially MOX powders design

applications. This was based on a consideration of relative risks, sensitivity of k_{eff} to traditional trending parameters, and a survey of existing benchmarks. In the fast energy range ^{239}Pu fission plays a dominant role, but other absorbing and moderating materials such as ^{238}U come to play a significant role in the lower-energy regime. The intermediate energy range is of particular interest for MOX systems, where the ^{239}Pu and ^{238}U cross sections are in competition. This is expected to occur in the powder blending process, which is also an area of key criticality control.

The parameters of primary importance appear to be the H/X ratio and Pu-content, based on parametric curves derived in this study and on the observation that ^{239}Pu and ^{1}H account for most of the system k_{eff} sensitivity in low-moderated PuO_2 and MOX systems. Density, geometry, and neutron energy appear to have a much less significant effect. This study shows that use of the TSUNAMI codes, with a conservative c_k cutoff value can, together with traditional trending approaches and other analytical methods, provide valuable insights into which parameters and behavior are important for validation of process in an MFFF.

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