Uncertainty Quantification for Compositions in Benchmark Evaluations

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Nuclear Criticality Safety Program Technical Program Review
Santa Fe, NM
December 3–5, 2024

Compositions

- Fuel
  - Enrichment and other uranium isotopics
  - “Enrichment” and other plutonium isotopics
  - How much of the composition is the fuel element (uranium or plutonium vs. other)?
  - Impurities

- Material of interest—e.g., Teflon parts for fluorine studies, PVC/CPVC parts for chlorine studies
  - How much of the composition is the main element/compound?
  - Impurities?

- Structural materials
  - How much of the composition is the main element/compound?
  - Impurities?

First-Order Uncertainty Quantification

- The standard uncertainty u_{R,x_i} of response R due to the standard uncertainty u_{x_i} in input parameter x_i is *approximately*

$$u_{R,x_i}^2 = \sum_i \left(\frac{\partial R}{\partial x_i} \right)^2 u_{x_i}^2$$

$$= \sum_i \left(R_0 \right)^2 \left(\frac{x_0}{R_0} \frac{\partial R}{\partial x_i} \right)^2 \left(\frac{u_{x_i}}{x_0} \right)^2$$

or

$$\left(\frac{u_{R,x_i}}{R_0} \right)^2 = \sum_i \left(S_{R,x_i} \right)^2 \left(\frac{u_{x_i}}{x_0} \right)^2$$

where

$$S_{R,x_i} \equiv \frac{x_{i,0}}{R_0} \frac{\partial R}{\partial x_i} \bigg|_{x_i = x_{i,0}}$$

- The relative sensitivity S_{R,x_i} can be computed with the adjoint method or the direct perturbation method.

Treatment of Uranium Isotopics in Historic HEU-MET-FAST Evaluations

- • HEU-MET-FAST-005, Rev. 1 was 1999 but when was Rev. 0?

Table 3. Isotopic Composition of Uranium.

Uranium Isotope	Wt.%
^{234}U	0.9
^{235}U	90.0 ± 0.3
^{236}U	0.4
^{238}U	8.7 ± 0.3

The uncertainty in k_{eff} caused by inaccuracy of the uranium isotopic composition (see Table 3) was estimated for Case 1 and was found equal to 0.16%.

- HEU-MET-FAST-007, Rev. 1 was 1999 but when was Rev. 0?

APPENDIX E: EFFECTS OF VARIATIONS IN URANIUM COMPOSITION

E.1 Calculated Effects of Variations in Uranium Isotopic Composition

The effects of increasing ^{235}U, ^{234}U, and ^{236}U by 0.05 wt.% were calculated, one at a time for each case, with CSAS1X (SCALE4.4) using XSDRN, 238-group ENDF/B-V cross sections, and a MULTIREGION buckled-slab unit cell. Because the amount of ^{238}U was reported as being obtained "by difference," the 0.05 wt.% increase in each isotope was balanced by an equal decrease in ^{238}U. The effects are shown in Table E.1.

Because the effects of increasing ^{234}U and ^{236}U were smaller than increasing ^{235}U by the same amount (as expected), and because the effect of the standard uncertainty (1/5 of the calculated variation) was almost negligible for ^{235}U, the effects of uncertainty in ^{234}U and ^{236}U are neglected.

Treatment of Uranium Isotopics in Historic HEU-MET-FAST Evaluations (cont.)

- • HEU-MET-FAST-086 (Godiva-IV), 2012

2.2 HEU Enrichment and Isotopics

The average enrichment for the 12 HEU parts is 93.15 wt.%, with a standard deviation of ± 0.03 wt.% for the individual parts. Calculations were performed that increased the enrichment of all 12 parts by 0.05 wt.% and decreased the enrichment of all 12 parts by 0.05 wt.%. The difference in the values of k_{eff} from those two calculations is negligible. Furthermore, to produce a standard deviation that result would have to be reduced substantially to account for the fact that the enrichment was increased in all of the fuel parts. Consequently, it is clear that enrichment can be discarded as a source of uncertainty for the k_{eff} of the configuration.

A calculation was performed in which the ^{234}U in the fuel was replaced by ^{238}U while preserving the mass of the fuel. The result from that calculation indicated that the worth of the ^{234}U content of the fuel is 0.0043 ± 0.0004 Δk. However, the standard deviation determined above is substantially less than 1% of the ratio. Even if the effective standard deviation were increased to 1% to account for the fact that swipes were not taken from all the fuel parts, the resulting increment in k_{eff} would be only marginally significant. Furthermore, that result would have to be reduced substantially to account for the fact that the change was made to all of the fuel parts simultaneously. Consequently, it is clear that the ^{234}U content of the fuel can be discounted as a source of uncertainty in the k_{eff} of the configuration.

Calculations to determine the worth of the ^{233}U and ^{236}U content of the fuel were performed by replacing those isotopes with ^{238}U while preserving the mass of the fuel. The worths of the ^{233}U and the ^{236}U were found to be 0.0006 ± 0.0004 Δk and 0.0012 ± 0.0004 Δk , respectively.

The three isotopes for which there are no measurements for the Godiva-IV fuel are ^{233}U, ^{236}U, and ^{238}U. The ^{233}U content is minuscule, and therefore an increase or decrease in the ^{238}U content would have to be accompanied by a decrease or increase, respectively, in the ^{236}U content. The uncertainty of ± 0.28 wt.% in the ^{236}U content implicitly included a change of ∓ 0.28 wt.% in the ^{238}U content. Scaling that change from 0.28 wt.% to 0.35 wt.% would not change the standard deviation of ± 0.0002 in k_{eff} associated with the uncertainty in the ^{236}U / ^{238}U content. Consequently, the impact on k_{eff} of the uncertainty in the ^{238}U concentration already has been addressed implicitly.

Treatment of Plutonium Isotopics in Historic PU-MET-FAST Evaluations

- PU-MET-FAST-006 (Flattop-Pu) (Rev. 1 was 1999 but when was Rev. 0?) is the first to mention Pu isotopic uncertainties

Isotopic uncertainties were not given in the references. Isotopic uncertainties in weight percent were typically, for weapons-grade plutonium using mass spectrometry, ±0.05 for ^{239}Pu, ±0.05 for ^{240}Pu, and ±0.01 for ^{241}Pu.

But these uncertainties are not propagated:

3.5 Experimental and Benchmark-Model k_{eff}

Since delayed critical was obtained, the experimental k_{eff} was 1.0000. Using the given experimental uncertainty of ±30 g in the Pu mass, an uncertainty in k_{eff} of ±0.0016 can be derived using ONEDANT as shown in Appendix B. The benchmark-model k_{eff} is 1.0000 ± 0.0030. The uncertainty in k_{eff} was based upon engineering judgment and personal communication with T. P. McLaughlin.

- PU-MET-FAST-044, 2007

The weight percent values used to estimate the ^{239}Pu wt.% uncertainty were taken from Table 2 (93.72 wt.%, 93.53 wt.%, and 93.87 wt.%). The uncertainty for the plutonium enrichment is considered a Type B uncertainty with the wt.% ^{239}Pu values having an equal probability over the entire range (93.53 wt.% - 93.87 wt.%). Equation 1 of Section 2.1.2 was used to estimate the 1σ uncertainty of ±0.098 wt.% for the ^{239}Pu isotopic content. The weight percent of each of the other plutonium isotopes, ^{240}Pu and ^{241}Pu, is varied inversely with the change in wt.% ^{239}Pu according to the relative nominal weight percent of ^{240}Pu and ^{241}Pu (see Table 14). The plutonium isotopic ranges listed in Table 14 correspond to the minimum ^{239}Pu wt.% and the maximum ^{239}Pu wt.% in the plutonium-gallium alloy.

Table 14. Plutonium Isotopic Uncertainty.

Isotope/ Nuclide	Nominal wt.%	wt.% (Reference 2)			σ, wt.%	Isotopic Range, wt.%	
		Section I	Section II	Section III		Minimum ^{239}Pu (-1σ)	Maximum ^{239}Pu (+1σ)
^{239}Pu	93.642	93.87	93.72	93.53	0.098	93.544	93.740
^{240}Pu	5.061					5.153	4.970
^{241}Pu	0.347					0.353	0.340
Ga	0.950					0.950	0.950
Total	100.000					100.000	100.000

In other words, CPA with ^{240}Pu + ^{241}Pu as the control parameter

Treatment of Plutonium Isotopics in Historic PU-MET-FAST Evaluations (cont.)

- PU-MET-FAST-046, 2008

Inaccuracies in other parameters of the assembly described in Section 1 lead to even smaller uncertainties in k_{eff}. The component uncertainties in k_{eff} for all cases are given in Table 14.

Table 14. Parameters Bringing to Uncertainties and Uncertainties in k_{eff} (1σ, %).

	Parameter	Case 1	Case 2	Case 3
1	Reactivity excess	0.01	-	0.05
2	Position of the control elements	0.17	0.17	0.17
3	Mass of plutonium	0.04	0.04	0.04
4	Plutonium isotopic composition	0.03	0.03	0.03
5	Density of the uranium reflector	0.17	0.16	0.16
6	Plug height	0.07	0.07	0.07
7	Density of research rods	-	-	0.10
8	Exact layout	0.08	0.08	-
	Total uncertainty	0.27	0.26	0.27

- PU-MET-FAST-042, 2010

2.1.2 Isotopic Variation - Plutonium isotopic uncertainties were not provided. The isotopic uncertainties were estimated and are given in Table 8. To check the effect, the isotopes were first varied, one at a time, for one case. Then they were varied simultaneously according to expected effect on reactivity for the other cases, i.e., subtraction of ^{238}Pu, ^{240}Pu, and ^{242}Pu and addition of ^{241}Pu for expected positive effect and the opposite for expected negative effect, with ^{239}Pu adjusted to maintain plutonium mass. Adjustment of the isotopic abundance in such a fashion should, respectively, increase and decrease the effective multiplication factor. The results are given in Table 9. Only the most significant effect of the various perturbations for each case are reported and are included in the analysis. The isotopic uncertainties provided in PU-MET-FAST-034 that are used in this analysis are larger than an approach based upon the significant figures of the reported isotopic abundances, as performed in PU-MET-FAST-016. The latter approach would include uncertainties of 0.005% for Rocky Flats plutonium metal.

Table 8. Estimated Plutonium Isotopic Uncertainty.[a]

Isotope	(Weight Percent) ± 1σ Uncertainty
^{238}Pu	(0.01) ± 0.01
^{239}Pu	(93.58)
^{240}Pu	(5.90) ± 0.06
^{241}Pu	(0.49) ± 0.02
^{242}Pu	(0.02) ± 0.01

(a) PU-MET-FAST-034.

Calculating the Sensitivity of k_{eff} to U and Pu Isotopic Weight Fractions

- For uranium:
  - + Perturb isotope weight fractions and preserve total U mass by balancing the ^{238}U mass
  - + What to do about ^{238}U?
    - Balance with ^{235}U?
    - Balance with all other U?
    - Renormalize all isotopes including ^{238}U?
    - Renormalize all isotopes except ^{238}U?

- For plutonium:
  - + Perturb isotope weight fractions and preserve total Pu mass by balancing the ^{239}Pu mass?
  - + What to do about ^{239}Pu?
    - Balance with ^{240}Pu?
    - Balance with all other Pu?
    - Renormalize all isotopes including ^{239}Pu
    - Renormalize all isotopes except ^{239}Pu?

Calculating the Sensitivity of k_{eff} to U and Pu Isotopic Weight Fractions

- For uranium:
  - + Perturb isotope weight fractions and preserve total U mass by balancing with the ^{238}U mass
  - + What to do about ^{238}U?
    - Balance with ^{235}U?
    - Balance with all other U?
    - Renormalize all isotopes including ^{238}U?
    - Renormalize all isotopes except ^{238}U?

CPA with ^{235}U as balance

CPA with other U as balance

Full Normalization

Partial Normalization

Control Parameter Adjustment with ^{238}U as the control parameter

- For plutonium:
  - + Perturb isotope weight fractions and preserve total Pu mass by balancing the ^{239}Pu mass?
  - + What to do about ^{239}Pu?
    - Balance with ^{240}Pu?
    - Balance with all other Pu?
    - Renormalize all isotopes including ^{239}Pu?
    - Renormalize all isotopes except ^{239}Pu?

CPA with ^{240}Pu as balance

CPA with other Pu as balance

Full Normalization

Partial Normalization

Control Parameter Adjustment with ^{240}Pu as the control parameter

Calculating the Sensitivity of k_{eff} to Weight Fractions in Structural Alloys (e.g. Steel or Aluminum)

- For minor constituents and impurities, use CPA with the main element as the control parameter.
  - E.g., in steel, perturb chromium weight fraction and preserve the mass by balancing with the iron weight fraction.
  - When using adjoint methods, “perturb” is only notional!
  - We frequently refer to minor constituents as impurities when they are actually important components of the alloy!

- What to do for the major element?
  - Full normalization?
  - Partial normalization?
  - CPA with all other constituents as the balance?

Useful References

- …on how to do the calculations, but not which strategy to use or what the constituent uncertainties should be.

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NUCLEAR SCIENCE AND ENGINEERING · VOLUME 185 · 384–405 · MARCH 2017

DOI: http://dx.doi.org/10.1080/00295639.2016.1272990

Adjoint-Based Sensitivity and Uncertainty Analysis for Density and Composition: A User's Guide

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Received August 31, 2016
Accepted for Publication October 8, 2016

Sensitivity of a Response to Stoichiometry

Sensitivity of a Response to Stoichiometry

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doi.org/10.13182/T125-36606

Using the Adjoint Method Can Lead to Large Uncertainties in the Constrained Sensitivities

- A perturbation of the weight fraction w_j of nuclide j must be balanced by perturbing other weight fractions in order to preserve the normalization ($\sum w_j = 1$).

- Constrained relative sensitivities to nuclide weight fractions are computed using the unconstrained sensitivities S_{k,N_i} output by the codes.

- If the balance is achieved by renormalizing all weight fractions, even w_j, then the constraint is called "full normalization" and the constrained sensitivity to w_j is

$$S^{FN}_{k,w_j} = S_{k,w_j} - \frac{w_{j,0}}{w_I} S_{k,\rho},$$

where subscript 0 indicates the initial, unperturbed weight fraction; w_I is the constrained total weight fraction of some relevant subset I of nuclides (if the "subset" is actually all the nuclides, then $w_I = 1$); and $S_{k,\rho}$ is the relative sensitivity to the material density.

- S_{k,w_j} and $S_{k,\rho}$ have statistical Monte Carlo uncertainties that must be propagated to S^{FN}_{k,w_j}.

- Jeffrey A. Favorite, "Correlated Sampling to Reduce the Uncertainty of Combinations of k_{eff} Sensitivities," *Transactions of the American Nuclear Society*, **131**, accepted (2024).

Example from PU-MET-THERM-005, Chlorine Worth Study

Table 343. u_k and Associated Uncertainty for the Composition of PVC Absorbers, CPVC Absorbers, and HDPE Moderators.

Component	Parameter	Case	Relative Uncertainty (u_T/wt%) (%)	$S_{k,x_i} \pm 1\sigma_{MC}$ (%/%)	Final u_k
HDPE Moderators	C_2H_4	1	± 1	1.50538E-04 ± 659 %	Negligible
		2	± 1	3.56196E-05 ± 2131 %	Negligible
		3	± 1	3.88722E-05 ± 2182 %	Negligible
	Cl	1	± 20/√3	-1.09214E-04 ± 1.62 %	± 0.00001
		2	± 20/√3	-2.07892E-06 ± 6.50 %	Negligible
		3	± 20/√3	-4.37300E-06 ± 6.41 %	Negligible
	H	1	± 0.1[(a)]	1.29652E-01 ± 0.44 %	± 0.00013
		2	± 0.1[(a)]	1.21694E-01 ± 0.36 %	± 0.00012
		3	± 0.1[(a)]	9.40401E-02 ± 0.52 %	± 0.00009

Table 353. u_k and Associated Uncertainty for the Composition of HDPE Reflectors.

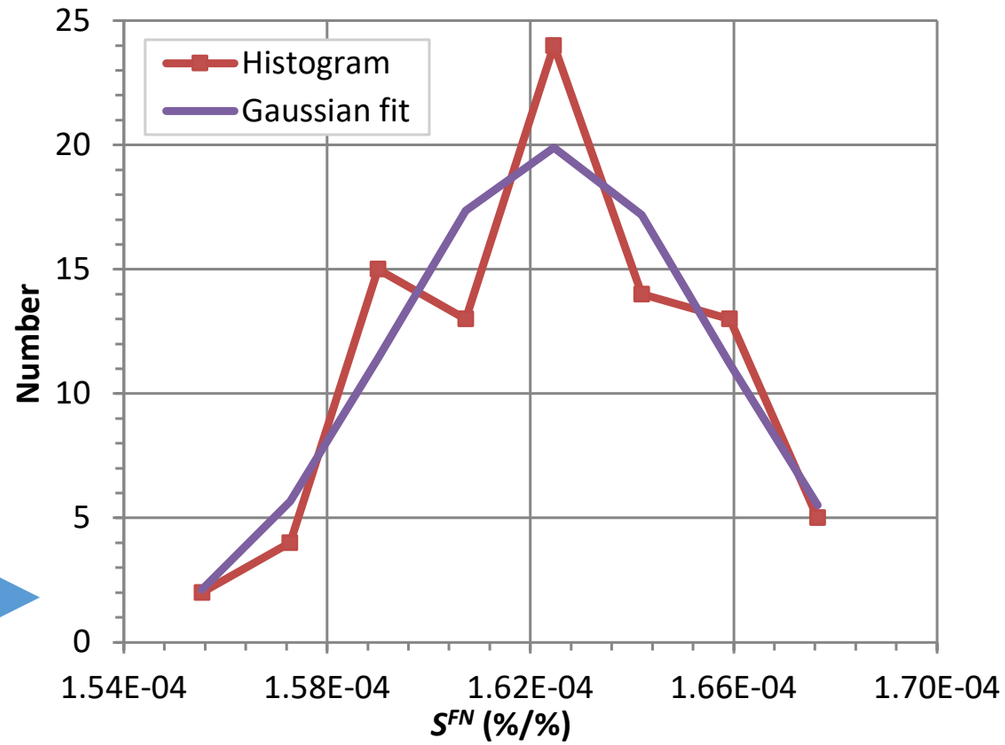
Component	Parameter	Case	Relative Uncertainty (u_T/wt%) (%)	$S_{k,x_i} \pm 1\sigma_{MC}$ (%/%)	Final u_k
HDPE Reflectors	C_2H_4	1	± 1	8.94343E-06 ± 6730 %	Negligible
		2	± 1	1.22097E-05 ± 5627 %	Negligible
		3	± 1	9.63690E-06 ± 7080 %	Negligible
	H	1	± 0.1	-3.74487E-03 ± 9.42 %	Negligible
		2	± 0.1	-5.08524E-03 ± 7.89 %	± 0.00001
		3	± 0.1	-1.00847E-02 ± 3.96 %	± 0.00001

Correlated Sampling for the Constrained Sensitivity

- Recall $S^{FN}_{k,w_j} = S_{k,w_j} - \frac{w_{j,0}}{w_I} S_{k,\rho}$.

- Propagating uncertainties without accounting for correlations, $\frac{\sigma_{S^{FN}_{k,w_j}}}{S^{FN}_{k,w_j}}$ was
  - 622 % for the HDPE moderators,
  - 5513 % for the HDPE reflectors.

- Using correlated sampling with 100 batches, $\frac{\sigma_{S^{FN}_{k,w_j}}}{S^{FN}_{k,w_j}}$ was
  - 1.829 % for the HDPE moderators,
  - 11.386 % for the HDPE reflectors.
  - Verified using 90 random samples.



$$\sigma^2_{S^C} = \frac{1}{N(N-1)} \sum_{n=1}^{N} \left[w_1^2 (g_n - S_1)^2 + w_2^2 (h_n - S_2)^2 \right] - \frac{2 w_1 w_2}{N(N-1)} \sum_{n=1}^{N} (g_n - S_1)(h_n - S_2)$$

On Fuel Impurities

- No impurities were reported in the Godiva-IV fuel. From Rev. 0:

> Information available for the fuel parts does not include details about impurities in those parts. Realistically, however, in a spectrum as hard as that produced by the Godiva-IV assembly impurities would have to be present at physically unrealistic levels to produce a statistically significant change in k_{eff}.

- But, from the Uncertainty Guide, estimate the effect of unknown impurities by
  1. subtracting fuel mass equal to the best estimate of the total mass of impurities, and
  2. adding best estimates of the impurities. Often, the first effect is the larger one.

- I used average impurities from the Jemima plates as an estimate of Godiva impurities (as Stolte did for the Flattop revision).

- Subtracting fuel mass $\Delta k_{eff} = -75$ pcm
- Adding impurities back in $\Delta k_{eff} = +120$ pcm

→ Replacing fuel with impurities (preserving mass density) $\Delta k_{eff} = +45$ pcm

Summary and Conclusions

- Our community has agreed on very few "rules" for composition uncertainties.
  - + For uranium isotopics, use CPA with ^{238}U as the control parameter.
  - + For minor constituents and impurities in structural alloys, use CPA with the main element as the control parameter.

- There are many more unsettled questions!
  - + Plutonium isotopics
  - + The CPA control parameter

- Very large uncertainties in adjoint-based constrained sensitivities can be reduced by properly accounting for correlations, and in MCNP6 we need sensitivities in batches.

- Canonical rules or "common knowledge" might not apply to your situation.

- I welcome your comments and suggestions: favorite@lanl.gov

- Acknowledgment:

This work was supported by the DOE Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy.