LA-UR-14-28633

A Computational Approach to the Dissolver Paradox

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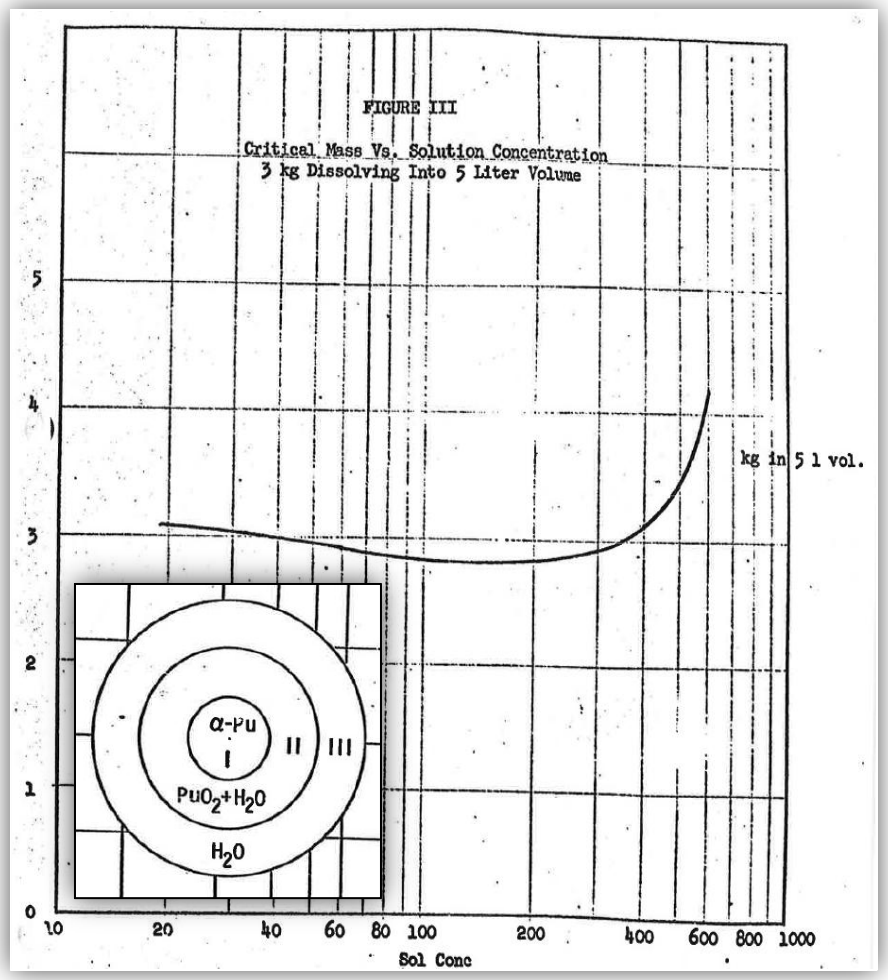
2014 ANS Winter Meeting

November 12, 2014

What is the dissolver paradox?

- **First reported in 1963 by Reardon and Czerniejewski**
- **Three-layer arrangement:**
  - Region 1 = Plutonium metal
  - Region 2 = Plutonium-water solution
  - Region 3 = Water reflector
- **Looked at 3000g dissolving into 5L**
- **Showed that some intermediate points in dissolving process were supercritical, even when both end points were subcritical**



THEORETICAL ANALYSIS

Previous Work and Basis

- **Lutz and Webb (1993) attempted to explore the dissolver paradox mathematically**
  - Based calculations on Avery (1958): “Theory of Coupled Reactors”

- **Calculated Avery’s coupling parameters k_{12} and k_{21} two different ways**
  - Based on fission source in each cell
  - Based on single-region k_{eff} values and surface area ratios
  - Results were inconsistent between the two methods

- **Did not extrapolate their calculations to a total predicted k_{eff} for the three-region system**

Avery's Theory of Coupled Reactors (1958)

- **For any (not necessarily critical) system:**

$$\begin{bmatrix} k_{11} & k_{12} \\ k_{21} & k_{22} \end{bmatrix} \begin{bmatrix} S_1 \\ S_2 \end{bmatrix} = k \begin{bmatrix} S_1 \\ S_2 \end{bmatrix}, \text{ where:}$$

  - K_{11} is the multiplication of 'reactor 1' (i.e. the metal) by itself
  - K_{22} is the multiplication of 'reactor 2' (i.e. the solution) by itself
  - K_{12} is the probability that a neutron born in reactor 2 will give rise to a neutron in reactor 1
  - K_{21} is the probability that a neutron born in reactor 1 will give rise to a neutron in reactor 2
  - S_1 and S_2 are the fission neutron source in reactors 1 and 2

- **Avery reduced this equation to:**

$$(1-k_{11})(1-k_{22}) + (k-1)\big((1-k_{11})(1-k_{22})\big) + (k-1)^2 = k_{12}k_{21}$$

Further Reduction of Avery's Formula

- Solving the previous into a quadratic equation gives:

$$k^2 - (k_{11} + k_{22})k + (k_{11}k_{22} - k_{12}k_{21}) = 0$$

- Applying the quadratic formula gives:

$$k = \frac{(k_{11} + k_{22}) + \sqrt{(k_{11} + k_{22})^2 - 4(k_{11}k_{22} - k_{12}k_{21})}}{2}$$

Reducing the Formulae per Lutz & Webb (1993)

- **Lutz & Webb describe simple ways to calculate the parameters k_{21} and k_{12} for the particular situation in question (concentric spheres).**

- **All neutrons leaking from Cell 1 (metal center) enter Cell 2.**

$$k_{21} \approx \left(1 - \frac{k_{eff,1}}{k_{\infty,1}}\right) k_{eff,2}$$

- **The neutrons in Cell 2 will leak either back to Cell 1 or out to the reflector. Leakage to Cell 1 is derived from ratio of inner surface area to total.**

$$k_{12} \approx \left(1 - \frac{k_{eff,2}}{k_{\infty,2}}\right)\left(\frac{R_i}{R_o + R_i}\right)^2 k_{eff,1}$$

These can be applied back into the quadratic version of Avery's formulas.

Correction to Lutz & Webb's Formulas

- **The equation for k_{12} contains an error. It was stated as:**

$$k_{12} \approx \left(1 - \frac{k_{eff,2}}{k_{inf,2}}\right)\left(\frac{R_i}{R_o + R_i}\right)^2 k_{eff,1}$$

with $\left(\frac{R_i}{R_o+R_i}\right)^2$ representing the ratios of the surface areas at the metal-solution interface to the total surface area of Region 2 (solution).

- **The correctly derived ratio of surface areas is:**

$$k_{12} \approx \left(1 - \frac{k_{eff,2}}{k_{inf,2}}\right)\left(\frac{{R_i}^2}{{R_o}^2 + {R_i}^2}\right) k_{eff,1}$$

Do the formulas work?

- **Applied calculated parameters from single-unit calculations and surface area ratios to quadratic equation discussed earlier**

- **Compared resulting values with full-system MCNP values**

- **Then used cosine-binned current tallies in MCNP to directly calculate interaction parameters k_{12} and k_{21}, and applied those to formulas**

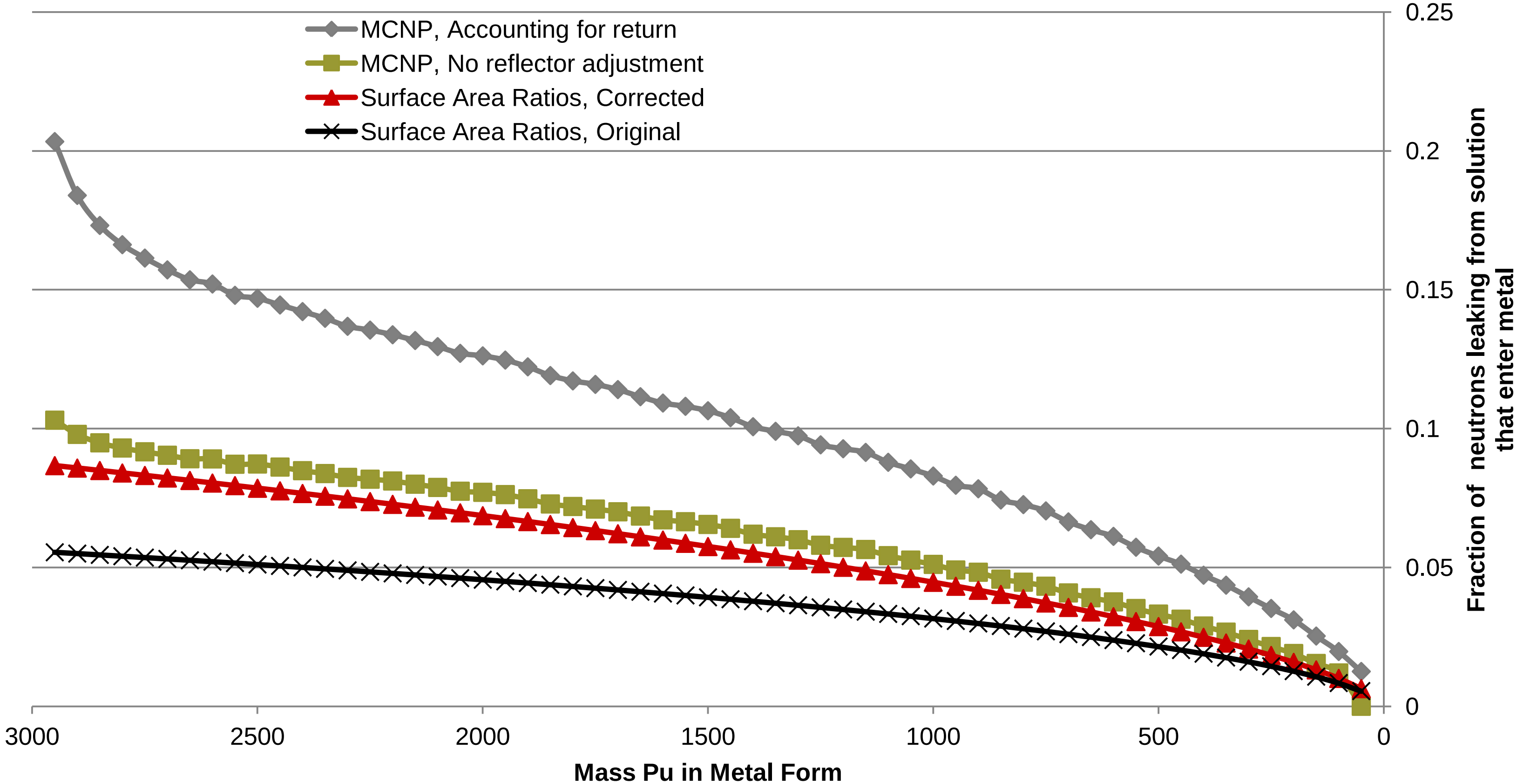
Do the formulas work?

- **MCNP-derived values for k_{12} were calculated in two different ways – accounting for the reflector, and not.**

- **Not accounting for reflector:**
  - (Total leakage from R2) = (Leaving R2 for R1) + (Leaving R2 for R3)
  - (Leakage fraction 2→1) = (Leaving R2 for R1) / (Total leakage from R2)

- **Accounting for reflector:**
  - ("Lost" to reflector) = (Leaving R2 for R3) – (Leaving R3 for R2)
  - (Total leakage from R2) = (Leaving R2 for R1) + ("Lost" to reflector)
  - (Leakage fraction 2→1) = (Leaving R2 for R1) / (Total leakage from R2)

Do the formulas work?



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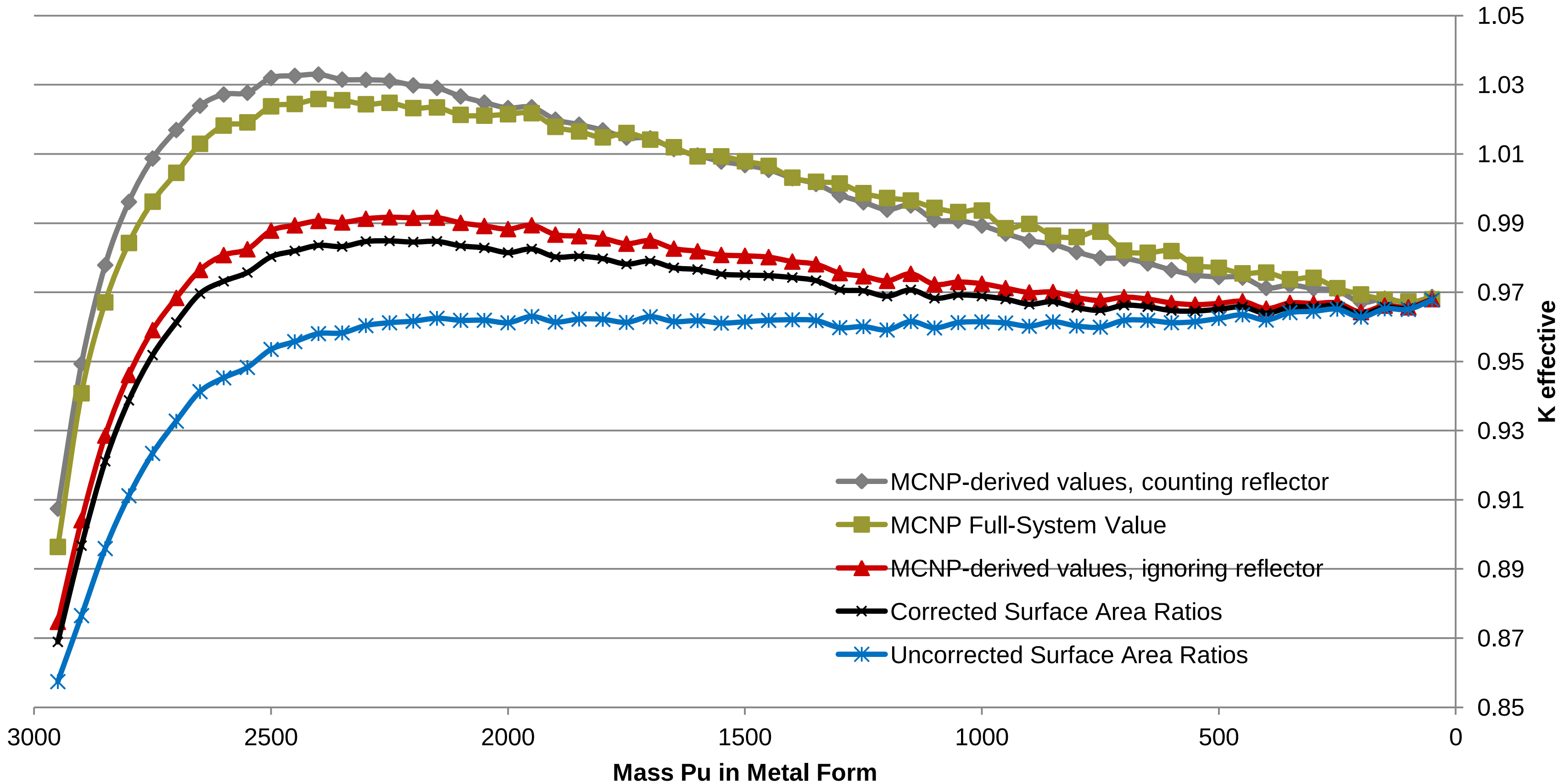

The Four Interaction Parameters

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- **The formula for k_{eff} from the four parameters is:**

$$k = \frac{(k_{11} + k_{22}) + \sqrt{(k_{11} - k_{22})^2 + 4k_{12}k_{21}}}{2}$$

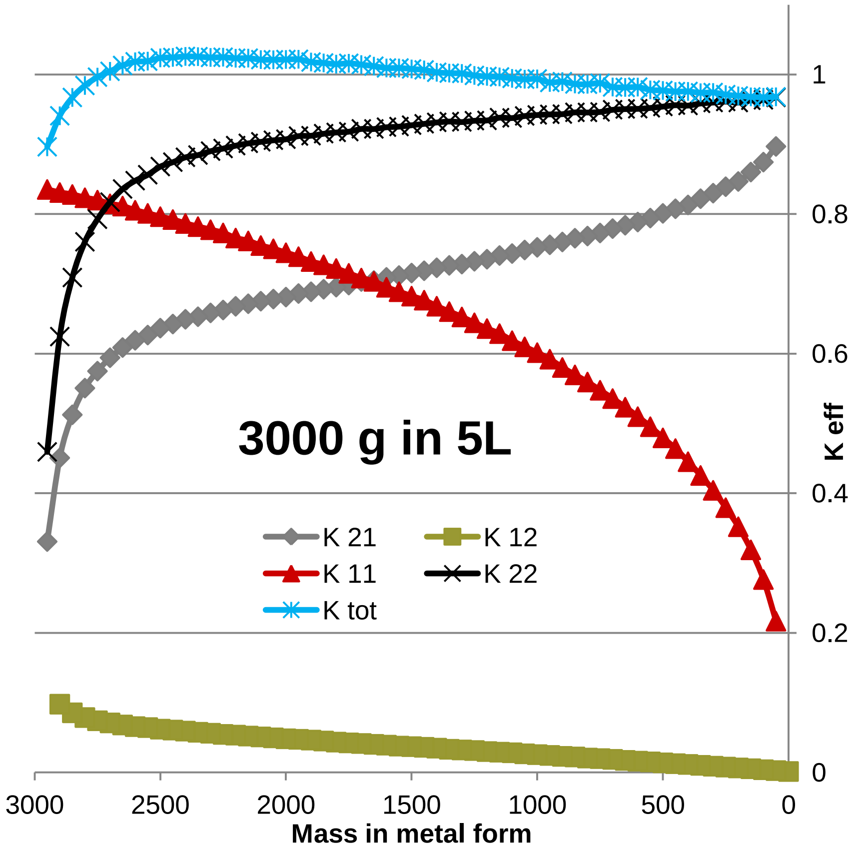
- **The values of k_{11} and k_{22} are most important in determining k_{eff}.**
  - The first portion of the formula is a simple average of the two.

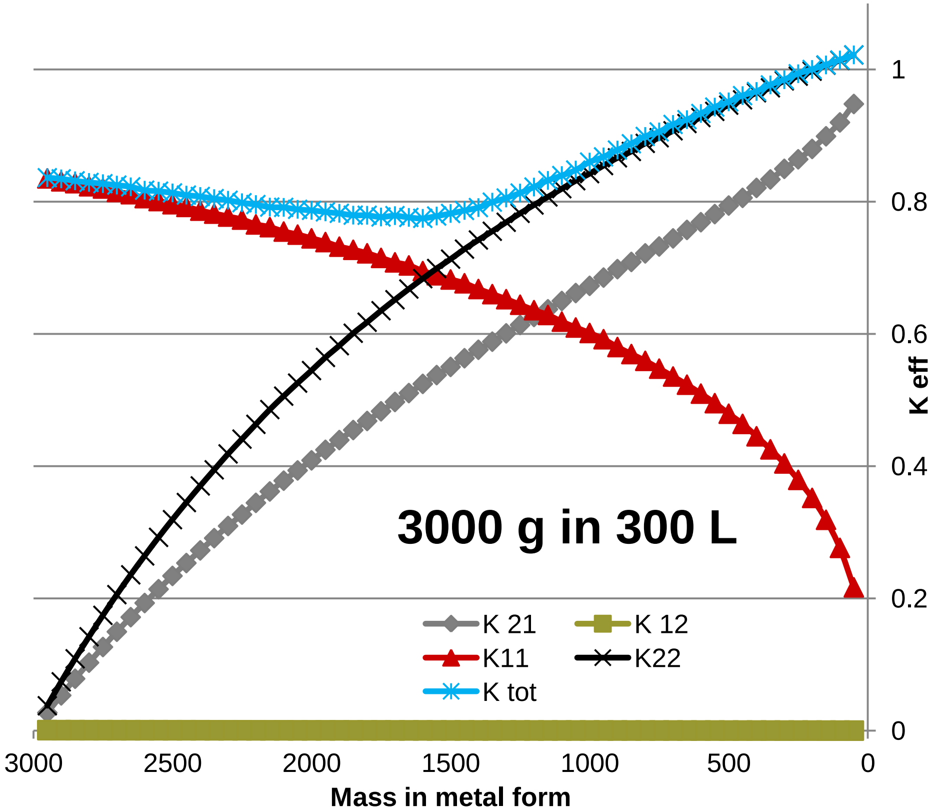
- **K_{12} [solution neutrons causing fissions in the metal] is very small, and becomes completely insignificant for larger volumes.**

- **For the same mass in different volumes, k_{11} always has the same curve.**

- **The initial slope of k_{22} (as compared to k_{11}) is the main driver for the increase.**

The Four Interaction Parameters





NUMERICAL ANALYSIS

Calculational Method

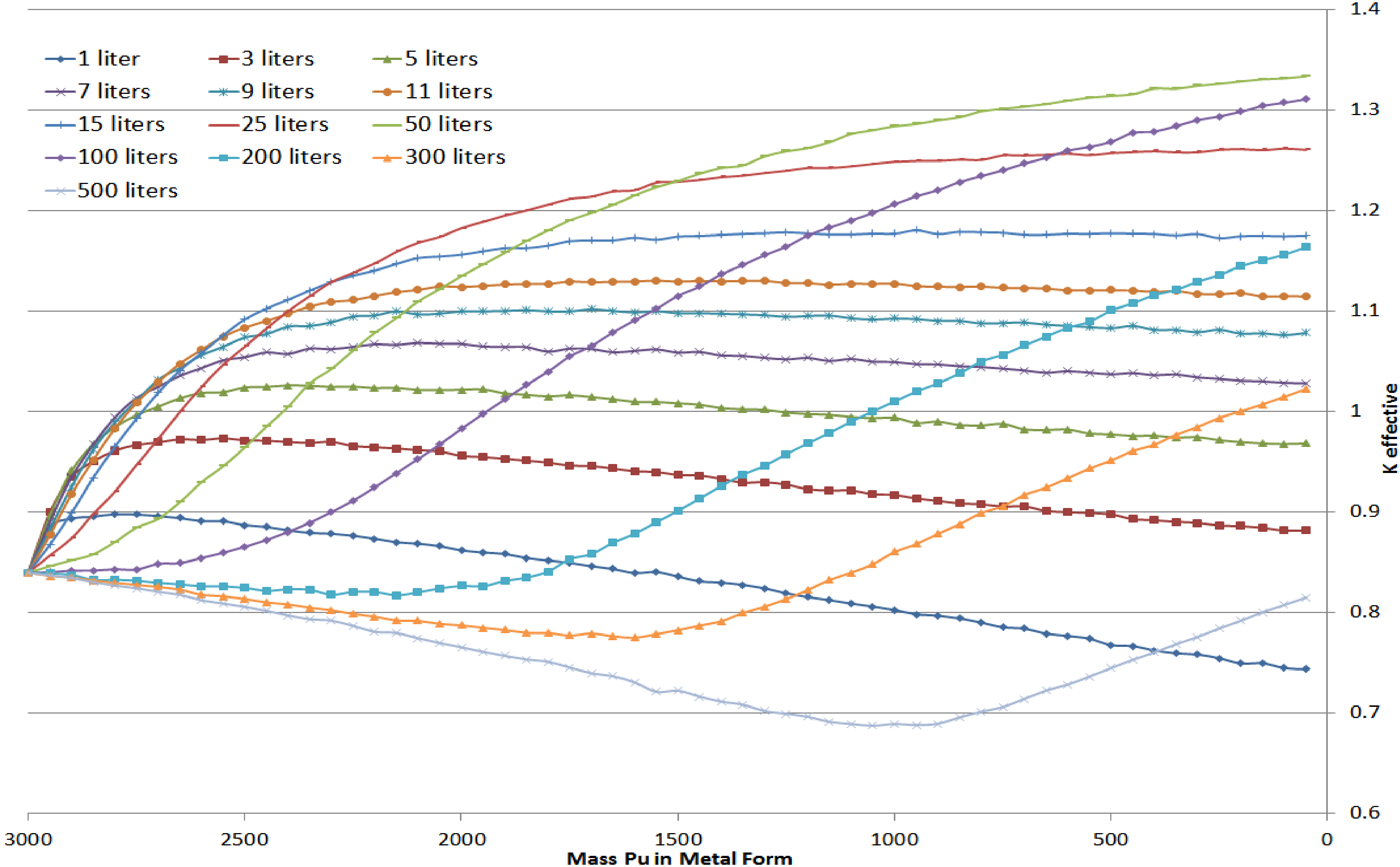
- **All calculations performed with MCNP5, using ENDF/B-VI cross-sections**

- **Modelled classic 3-zone problem**
  - Metal
  - Metal-water mixture (“solution”)
  - Water reflector

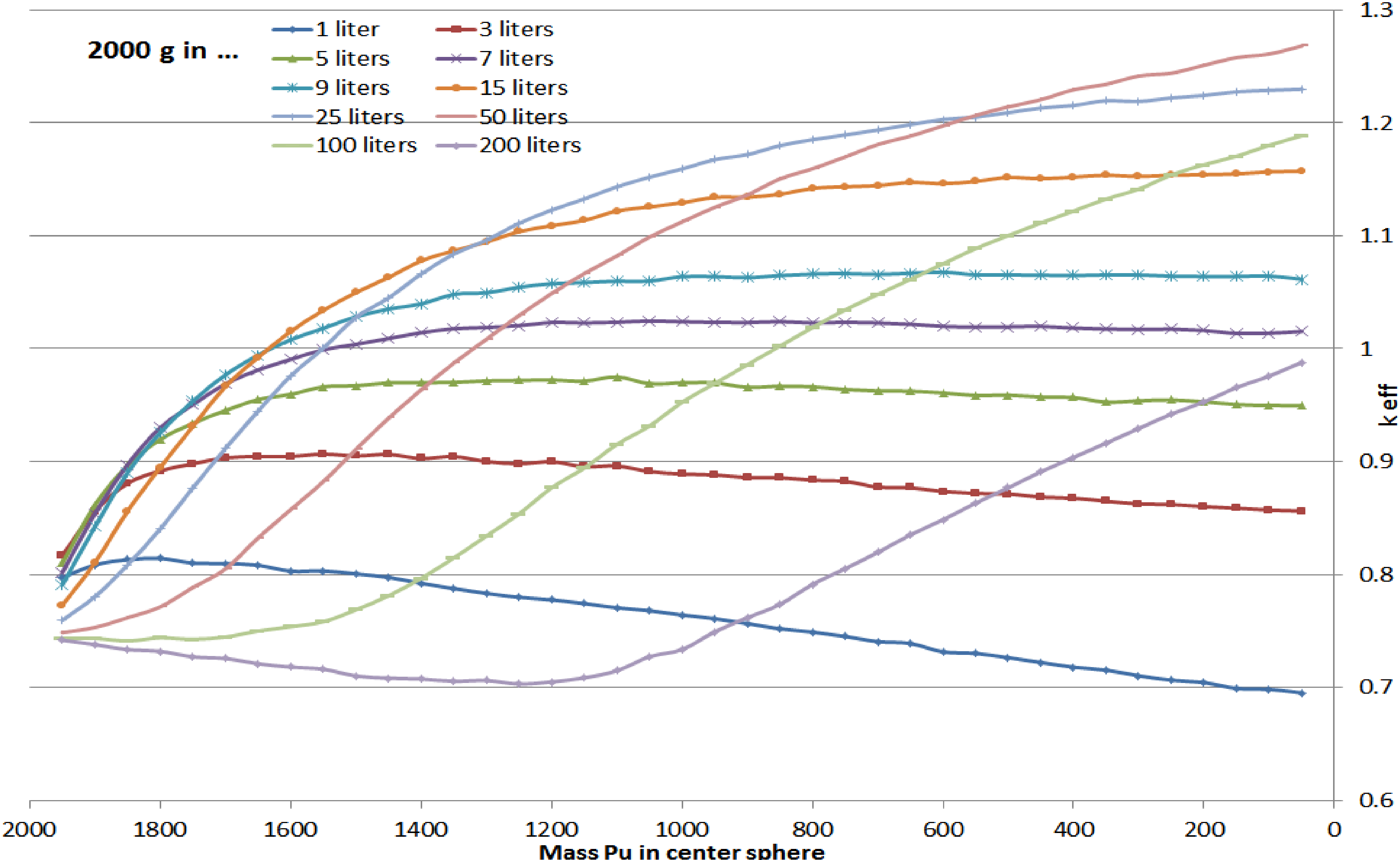
- **Traditional approach: vary solution radius or concentration, tabulate critical mass**

- **‘Transferred’ fissile mass from zone 1 to zone 2 to model dissolution process**
  - Constant volume; Tabulated k_{eff} values

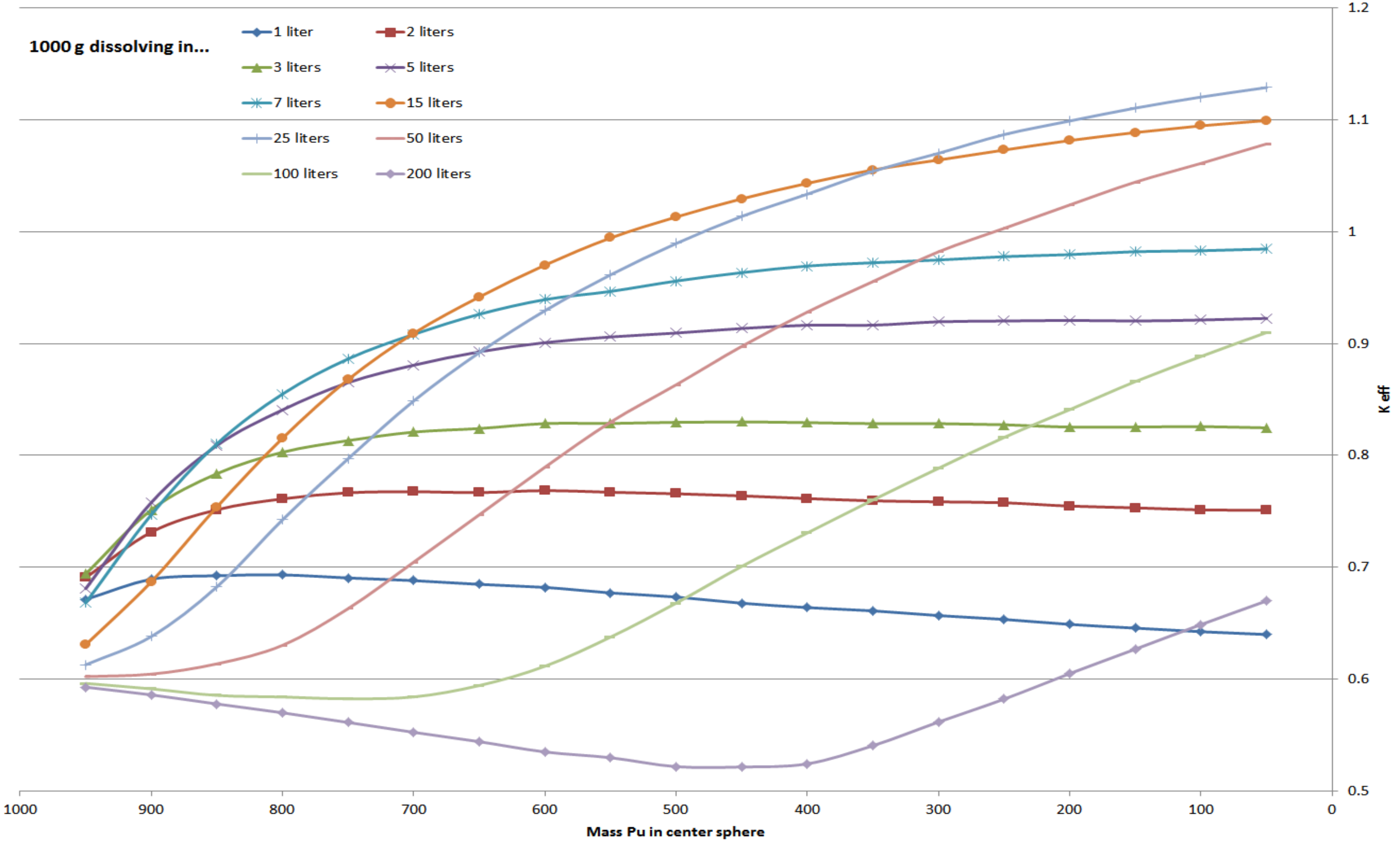
The Overall Curves – 3000 g dissolving in various volumes



The Overall Curves – 2000 g in various volumes



The Overall Curves – 1000 g in various volumes

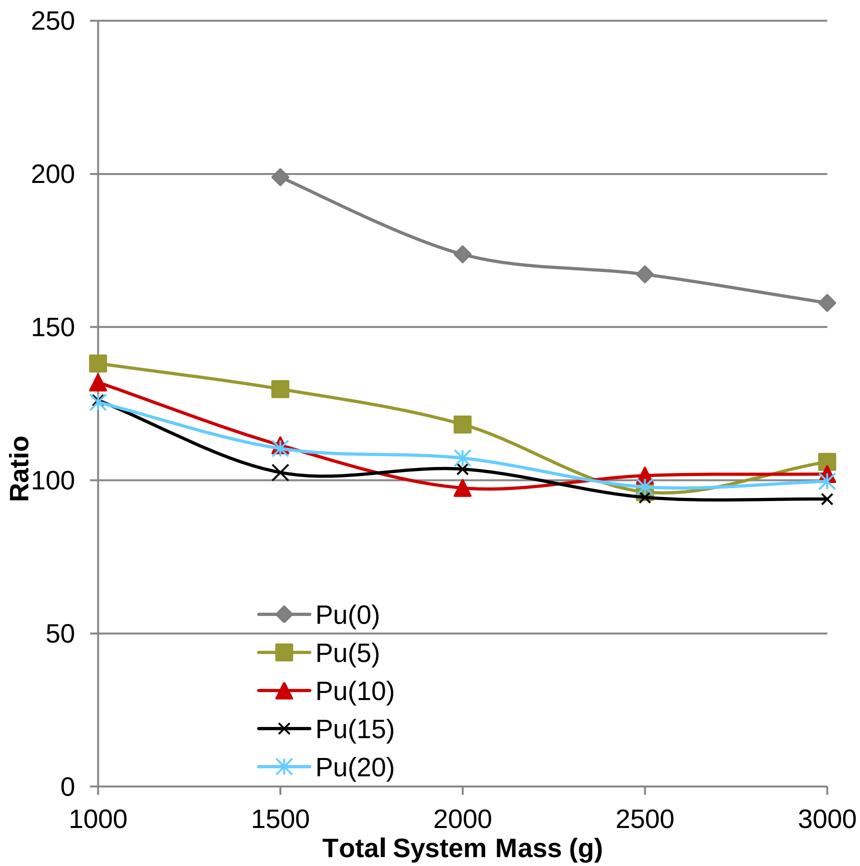


An Inverse Dissolver Paradox?

- **The curves with a minimum point were all high-volume (200L + for 3000g) systems.**
- **The turning point for all occurs when the solution is around 4-6 g/L**
  - This is the Limiting Critical Concentration for plutonium-water systems
- **Initial decline caused by the loss of mass in the central sphere**
  - Solution is too dilute to contribute much to k_{eff} - highly overmoderated
- **Once solution passes LCC, it can sustain its own chain reactions.**
  - k_{eff} increases as it approaches optimum moderation
- **A real system, particularly one that large, would not be nearly so evenly distributed as it dissolves**
  - System was modeled with mass in solution homogenously mixed with all water
  - An artifact of the calculation assumptions

The "Cutoff" Volume Ratio

- **Maximum volume where increase is seen, for given mass**
  - Increases linearly with total fissile mass of system
- **Plotted system fissile mass versus ratio of mass (g) over volume (L)**
- **Found a "cutoff" ratio of 100**
  - Consistent for Pu(5) through Pu(20)
  - Bounding of Pu(0)
- **Pu systems with mass-volume ratios greater than 100 should be explicitly modelled.**



Questions?

References

- **Avery, R. (1958). "Theory of Coupled Reactors".** ***Second United Nations International Conference on the Peaceful Uses of Atomic Energy*****.**

- **Reardon, W. A. and F. R. Czerniejewski (1963). "Idealized Plutonium Dissolvers and the 'Always Safe' Condition". Report HW-SA-2999.**

- **Lutz, H. F. and P. S. Webb (1993). "The Dissolver Paradox as a Coupled Fast-Thermal Reactor". LLNL Report UCRL-JC-113528.**