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DOES CRITICAL MASS DECREASE AS TEMPERATURE INCREASES:

A REVIEW OF FIVE BENCHMARK EXPERIMENTS THAT SPAN A RANGE OF ELEVATED TEMPERATURES & CRITICAL CONFIGURATIONS

DISCLAIMER

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LIST OF ACRONYMS & SYMBOLS

ACRYNOM	DEFINITION
k_{eff}	*effective neutron multiplication factor*
SRNS	*Savannah River Nuclear Solutions, LLC*
SRS	*Savannah River Site*
DOE	*Department of Energy*
α_T	*temperature reactivity coefficient*
g	*Gram*
l	*liter*

REVISION LISTING

Rev. 1: Corrigenda -

page 4 of 10: Summary, 2nd para., "In four of the five benchmarks ... " has been corrected to, "In three of the five benchmarks ... ."

page 6 of 10: top para., last sentence, "While the results of Table 1-2 are interesting, it does alter ..." has been corrected to, "While the results of Table 1-2 are interesting, it does not alter ... "

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SUMMARY

Five sets of benchmark experiments are reviewed herein that cover a diverse set of fissile system configurations. The review specifically focused on the change in critical mass of these systems at elevated temperatures and the temperature reactivity coefficient (α_T) on the system. Because plutonium-based critical benchmark experiments at varying temperatures were not found at the time this review was prepared, only uranium-based systems are included, as follows [1-5][1]:

1. **HEU-SOL-THERM-010:** *UO_2F_2 solutions with high U^{235} enrichment,*
2. **HEU-COMP-THERM-016:** *uranium-graphite blocks with low U concentration,*
3. **LEU-COMP-THERM-032:** *water moderated lattices of UO_2 with stainless steel cladding, and intermediate U^{235} enrichment,*
4. **IEU-COMP-THERM-002:** *water moderated lattices of annular UO_2 with/without absorbers, and intermediate U^{235} enrichment,*
5. **LEU-COMP-THERM-026:** *water moderated lattices of UO_2 at different pitches, and low U^{235} enrichment.*

In three of the five benchmarks (1, 3, and 5), modeling of the critical system at room temperature is conservative compared to modeling the system at elevated temperatures, i.e., a greater fissile mass is required at elevated temperature. In one benchmark (4), there was no difference in the fissile mass between the room temperature system and the system at the examined elevated temperature. In benchmark (2), the system clearly had a negative temperature reactivity coefficient.

Some of the high temperature benchmark experiments were treated with appropriate (and comprehensive) adjustments to the cross section sets and thermal expansion coefficients, while other experiments were treated with partial adjustments. Regardless of the temperature treatment, modeling the systems at room temperature was found to be conservative for the examined systems, i.e., a smaller critical mass was obtained.

While the five benchmarks presented herein demonstrate that, for the conditions examined, modeling of the systems at room temperature is conservative as compared to modeling the systems at elevated temperatures, it is possible to design a system in which the critical mass at room temperature is non-conservative compared to a system at elevated temperatures. As the temperature of the systems evaluated in this review was increased, the system's overall α_T was negative at elevated temperatures.

Furthermore, the review demonstrates that to accurately assess the effect of increased temperature on a system's k_{eff}, changes in fissile, moderator, cladding, and, in some cases, structural material cross sections must be combined with other factors that influence reactivity, such as volumetric thermal expansion of fissile, moderating, reflector, and other interacting media. Altering the microscopic cross sections of fissile and moderating regions for temperature changes, without adjusting the corresponding densities at elevated temperatures, can lead to an incorrect assessment of the impact of elevated temperature on a fissile system.

[1] *Numbers appearing inside brackets correspond to Reference numbers listed on page 10.*

DISCUSSION

It should be noted that in assessing the five benchmark critical experiments presented herein, references to α_T are made and a clarification on its use is warranted. When discussing the fissile material cross section, α_T is used to denote changes in the microscopic reactions of the nuclide or fissile material due to temperature changes. However, when the system response is being discussed, i.e., such as referring to the system's α_T, then it is used to denote the over-all effect on k_{eff} due to the temperature changes. An increase or decrease in a nuclide's microscopic cross section due to temperature changes is not the only mechanism that dictates the behavior of the system's reactivity. These temperature changes also affect the microscopic cross sections, density, and volume of all other interacting media (moderator, reflector, absorbers), as well as the volume and density of the fissile material, and each of these cited changes influence k_{eff}.

The configurations of the five benchmarks experiments are described in the following sections. Also included in these sections are results and discussions pertinent to the effects of temperature increases on the reactivity of these critical configurations.

1. **HEU-SOL-THERM-010:** Water Reflected Spheres of UO_2F_2 at Four Temperatures [1]

Four water-reflected sphere measurements were performed at the Oak Ridge National Laboratory (ORNL) in the 1950's with highly enriched uranium (93.13 wt% U^{235}) as uranium oxyfluoride (UO_2F_2) solutions. The examined temperature ranged from 27.5 to 85.5 °C. However, as the temperature increased, the required uranium mass and concentration to maintain the system critical also increased. For example, the critical uranium mass increased from 918 g to 1,010 g, and the uranium concentration increased from 102.06 to 111.52 g U/l, as the temperature increased from 27.5 to 85.5 °C.

Benchmark calculations were performed with MCNP and ONEDANT. For this set of benchmark calculations, the solution density was adjusted as the temperature varied. The aluminum core thickness was not adjusted for thermal expansion. However, the critical volume was measured at each temperature investigated. Table 1-1 presents the critical conditions for the four experiments.

Table 1-1: Critical Conditions of Water Reflected UO_2F_2 Spheres

Exp. No.	Volume (liters)	U^{235} Mass (g)	U^{235} Concentration (g/l)	Solution Density (g/cm^3)	Temperature (°C)
1	9.661	918.3	95.05	1.1159	27.5
2	9.675	935.3	96.67	1.1136	39.5
3	9.713	989.8	101.90	1.1015	74.0
4	9.726	1,010.1	103.86	1.0960	85.5

An analysis was done to investigate the effect of using room temperature cross sections rather than cross sections at elevated temperature using MCNP with ENDF-V continuous energy cross section data [1]. The cross sections were prepared at the experiment temperatures with NJOY. Each case was run with a hydrogen scattering kernel at 300 K and 400 K. The results were linearly interpolated to find the k_{eff} value for a scattering kernel at the experiment temperature. Table 1-2 compares the calculated k_{eff} using cross sections for room temperature and experimental temperatures. However, note that Doppler broadening at the three elevated temperatures is not included in the MCNP treatment of temperature.

Table 1-2: Effects of Only Employing Higher Temperature Cross-Sections

Exp. No.	Temp. (°C)	MCNP k_{eff} (Room Temp.)	MCNP k_{eff} (Exp. Temp.)	δk_{eff}, (Exp. - Room) Temp.
1	27.5	1.0040	1.0040	0
2	39.5	1.0055	1.0057	0.0002
3	74.0	1.0040	1.0046	0.0006
4	85.5	1.0015	1.0033	0.0018

The results of Table 1-2 indicate that the use of MCNP with room temperature cross sections, while including appropriate adjustments for solution density, tends to slightly under-predict k_{eff} compared to the use of cross sections with a scattering kernel at the actual elevated temperature. However, since Doppler broadening is not included in the treatment of cross sections at elevated temperatures, the difference between the use of room temperature cross sections and cross sections that include Doppler broadening may be closer than suggested in Table 1-2. While the results of Table 1-2 are interesting, it does not alter the results of Table 1-1; namely, that modeling the room temperature system is conservative (i.e., smaller critical mass) compared to modeling any of the systems at elevated temperature.

2. <u>HEU-COMP-THERM-016</u>: IGR Reactor - Uranium-Graphite Blocks Reflected by Graphite [2]

The reactor core is a stack of graphite blocks with a very low content of U^{235} (≈8,180 graphite nuclei per uranium nucleus). There were 147 uranium-impregnated columnar blocks and 21 un-impregnated graphite columnar blocks in the core. The overall core was a cube approximately 140 cm on a side. The core reflector was graphite. The six experiments produced critical configurations of the core at various stack temperatures and with different positions of the controls rods. Uranium enrichment was 90 wt % U^{235}.

Several types of control rods were used: startup rods (SR), manual rods (MR), automatic rods (AR), compensating rods (CR) and shim rods (TR). The shim rods served to compensate for the negative temperature effects of reactivity and to set the pattern of power variations during a burst.

The most significant approximations were the following:

- ✓ *use of the average core temperature instead of the actual temperature profile across the core, because it was difficult to determine, and,*
- ✓ *use of average fission product concentration across the core instead of the actual variable fission product distribution across the core.*

All 6 experiments were carried out with steady-state power of the reactor with k_{eff} set equal to 1.0. The uncertainty of the k_{eff} measurement was ±0.01. Critical configurations of the reactor with the core temperature variations in the range of <20 to 800 °C are found in Table 2-1. The rod positions are also presented in Table 2-1. The rod position value of 1,400 mm corresponds to complete rod insertion into the core and that of 0 mm corresponds to its full withdrawal to the top of the core. The movable part of the stack was in its normal position (fully inserted) for all configurations except the first, where it was withdrawn (106.1 cm lower). Only the first two configurations included an empty N-606 unit inside the core. α_T for this reactor system was -2.1×10^{-4} $\delta k/K$, indicating that as the temperature increases, reactivity addition was required to maintain the core critical [6].

The effect of the negative α_T for this system can be seen in Table 2-1 by examining critical configurations 4 (27 °C), 5 (476 °C), and 6 (794 °C). In configuration 4 (27 °C), the shim rods, whose purpose is to compensate for the negative α_T, are completely inserted into the core. In configuration 5 (476 °C), the shim rods are withdrawn about half way. In configuration 6 (794 °C), the shim rods are completely withdrawn from the core. As indicated above, it has been demonstrated that this system's $\alpha_T < 0$.

Table 2-1: Critical Configurations of the IGR Reactor

Critical Configuration	**1**	**2**	**3**	**4**	**5**	**6**
Movable Stack	out	in	in			
Empty N-606 Unit	yes		no			
Core Temperature, °C	**18**	**13**	**50**	**27**	**476**	**794**
Rod	**Rod Position, mm**					
SR1	1,400	785	400	0	0	0
SR2	1,400	0	461	0	0	0
SR3	309	718	1,400	796	800	1,400
TR1	0	1,240	1,240	1,400	715	0
TR2	0	1,240	1,240	1,400	715	0
TR3	0	1,240	1,240	1,400	715	0
TR4	0	1,240	1,240	1,400	715	0
TR5	0	1,240	1,240	1,400	715	0
TR6	0	1,240	1,240	1,400	715	0
TR7	0	1,240	1,240	1,400	715	0
TR8	0	1,240	1,240	1,400	715	0
AR	0	0	1,400	0	0	970
MR	0	0	1,400	0	945	1,400
CR1	0	1,400	0	1,400	1,400	1,400
CR2	0	1,400	1,400	1,400	1,400	1,400
CR3	0	1,400	0	1,400	1,400	1,400

3. LEU-COMP-THERM-032: Uniform Water Moderated Lattices of Rods with U (10%) O_2 Fuel in Range from 20 °C to 274 °C [3]

Three sets of critical experiments for uniform, fully flooded rods at lattice pitch values of 0.7, 1.4, and 1.852 cm were performed. For each rod pitch lattice configuration, the number of fuel rods needed to maintain the system critical was determined at three different temperatures were performed. The high temperature experiments were carried out in a pressure vessel. The rods were circular and were 856 mm long. Cladding was stainless steel. Rod diameter (including cladding) was 5.1 mm. The UO_2 sintered pellets had a 4.16 mm external diameter. Pertinent critical array configuration data for each of the nine experiments is presented in Table 3-1. The last column in Table 3-1 represents the temperature reactivity coefficient as determined by another set of measurements performed at the facility [7].

The following summarizes Table 3-1 data and pertinent conclusions for this set of critical experiments:

- ✓ *More rods were required to maintain a critical system as the temperature was increased, except between the temperatures of 20 to 193 °C for the rod pitch configuration of 1.852 cm. The number of rods required to maintain the system critical in this temperature range was the same.*
- ✓ *As the rod lattice pitch was increased, fewer rods were required to maintain a critical configuration as compared to the 0.7 cm lattice pitch system. This indicates that the system was under moderated at all examined temperatures at the rod pitch of 0.7 cm.*

Table 3-1: Critical Configuration of Water Moderated $U(10\%)O_2$ Lattice Rods

Array	Pitch (cm)	Temperature, °C	No. of Rods	System α_T, δk / °C
1	0.7	20	2,002	-0.2×10^{-04}
2		166	2,323	-3.0×10^{-04}
3		263	3,058	-5.2×10^{-04}
4	1.4	20	421	$+0.2 \times 10^{-04}$
5		206	481	-3.0×10^{-04}
6		274	565	-5.7×10^{-04}
7	1.852	20	523	$+0.7 \times 10^{-04}$
8		193	523	-1.0×10^{-04}
9		263	559	-2.0×10^{-04}

- ✓ *At the rod lattice pitch of 0.7 cm, α_T is negative for all examined temperatures. However, as the rod pitch was increased, α_T is initially positive at the lower temperature of 20 °C and becomes negative as temperature increases. The behavior of each of the three systems depends on the fissile mass to moderator ratio, the effect of Doppler broadening as temperature increases, self shielding of the fissile material, and the volumetric expansion of the fuel, moderator, and other interacting regions. These experiments also demonstrate that α_T is not constant and both its magnitude and worth decreases at higher temperatures for these types of systems.*

In conclusion, modeling this system at room temperature is conservative.

4. <u>IEU-COMP-THERM-002</u>: Water Moderated U (17) O_2 Annular Rods without Absorber and With Gadolinium or Cadmium in 6.8 cm Pitch Hexagonal Lattices at Different Temperatures [4]

Each lattice had one of three forms of fuel rods: rods without absorber, rods with a Gd absorber element, or rods with a Cd absorber element in the center of each fuel rod. Experiments involving the $U(17)O_2$ annular rods were as follows:

- ✓ **<u>Without Absorbers</u>**: *Examined temperature ranges were case 1 at 22.7 °C and case 2 at 218.4 °C. In both of these cases, 34 rods were required for to maintain a critical system.*
- ✓ **<u>With Gd Absorbers</u>**: *Examined temperature ranges were case 3 at 16.4 °C and case 4 at 151.0 °C. In both of these cases, 74 rods were required for to maintain a critical system.*
- ✓ **<u>With Cd Absorbers</u>**: *Examined temperature ranges were case 5 at 14.5 °C and case 6 at 150.6 °C. In both of these cases, 68 rods were required for to maintain a critical system.*

Reference 8 indicates that at room temperature, these systems exhibited a small, but positive α_T. However, as the temperature increased above room temperature, these system's α_T became negative and continued to decrease as the temperature further increased. The change in the system's α_T (from slight positive to negative and decreasing) highlights the fact that fissile system's feedback mechanisms are not restricted to a single parameter. Pertinent critical array configuration data for each of the three sets of experiments is presented in Table 4-1.

Table 4-1: Critical Configuration of the Water Moderated $U(17)O_2$ Annular Rods Lattice Rods

Rod Configuration	Case	No. of Rods	Temperature, °C	k_{eff}
Without Absorbers	1	34	22.7	1.0004
	2		218.4	1.0005
With Gadolinium	3	74	16.4	1.0007
			18.2	1.0011
			20.3	1.0013
			21.8	1.0014
	4	74	151.0	1.0004
			184.0	0.9940
With Cadmium	5	68	14.5	1.0002
	6		150.6	1.0000
			165.6	0.9980

For these specific systems, the number of fuel rods (i.e., critical mass) is the same at room temperature and at the elevated temperatures investigated. However, modeling the system at room temperature is much simpler. There is no need to include consideration of thermal expansion of the fuel, Doppler broadening of the resonances, self shielding of the fissile material, selection of scattering kernel, water density at elevated temperature and pressure, etc. The overall effect of elevated temperature is that the number of fuel rods (i.e., critical mass) does not change for each of the three cases.

5. LEU-COMP-THERM-026: Water moderated $U(4.92)O_2$ Fuel Rods in 1.29, 1.09, and 1.01 cm Pitch Hexagonal Lattices at Different Temperatures [5]

Six experiments were performed. Room temperature experiments were approximately 20 °C (cases 1, 3, and 5). Hot experiments were at 231.4 °C (case 2), 206 °C (case 4), and 212.1 °C (case 6). No absorber elements were present. The fuel rods were clad with zirconium alloy. Critical configurations are provided in Table 5-1. Table 5-1 demonstrates that in each of the three pitch configurations, the number of rods (hence, fissile mass) required to maintain a critical configuration increases as the temperature increases. Therefore, modeling these systems at room temperature is conservative.

Table 5-1: Critical Conditions of the Water Moderated $U(4.92)O_2$ Rods in a Hexagonal Lattice

Assembly	Lattice Pitch (cm)	Number of Fuel Rods	Temperature (°C)
1	1.29	621	20.1
2	1.29	889	231.4
3	1.09	1,951	19.3
4	1.09	2,791	206.0
5	1.01/1.29	325/680	20.8
6	1.01/1.29	325/912	212.1

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