

# A Historical Review of Cermet Fuel Development and the Engine Performance Implications

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**Abstract.** This paper reviews test data for cermet fuel samples developed in the 1960's to better quantify Nuclear Thermal Propulsion (NTP) cermet engine performance, and to better understand contemporary fuel testing results. Over 200 cermet (W-UO<sub>2</sub>) samples were tested by thermally cycling to 2500°C (2770 K) in hydrogen. The data indicates two issues at high temperatures: the vaporization rate of UO<sub>2</sub> and the chemical stability of UO<sub>2</sub>. The data show that cladding and chemical stabilizers each result in large, order of magnitude improvements in high temperature performance, while other approaches yield smaller, incremental improvements. Data is very limited above 2770 K, and this complicates predictions of engine performance at high I<sub>sp</sub>. The paper considers how this material performance data translates into engine performance. In particular, the location of maximum temperature within the fuel element and the effect of heat deposition rate are examined.

**Keywords:** Nuclear Thermal Propulsion, fuel development, cermet fuel,

## INTRODUCTION

The promise of nuclear thermal propulsion is that its high thrust and high specific impulse—twice that of the best chemical engines—significantly reduces rocket mass, size, and cost. NTP rocket engines use a nuclear reaction to heat propellant, in contrast to traditional chemical rockets, which heat propellant in a chemical reaction. High energy density nuclear fuel in high thrust engines operating at high specific impulse (I<sub>sp</sub> ≥ 900s) represents the next evolutionary step in liquid rocket engines.

To achieve high specific impulse, rockets often use materials and components near their performance limits; consequently, engine lifetimes can be short—from minutes to hours. For NTP rockets, the nuclear fuel and fuel elements operate near their structural and thermal limits. An understanding of high temperature behavior is important: What are the melting point, vaporization rate, and strength of fuel and cladding at high temperatures? Do chemical reactions appear in these extreme conditions? This paper examines historical test data for cermet fuel and attempts to clarify performance.

Nuclear thermal rockets were conceived in 1946 [1] at the beginning of the Atomic Age. The Rover and NERVA programs were initiated in 1955 and 1961, respectively, to develop NTP technology, and they were an integral part of the Space Race. NTP engines were a backup to intercontinental ballistic missile chemical propulsion, and envisioned uses included a lunar mission stage, Earth orbit-to-orbit transfer, and manned Mars missions. NTP's high I<sub>sp</sub> promised significant reductions in rocket mass and size. The NERVA program was an extensive program with a budget of ~\$8.9 billion (2014 \$) which designed, built, and tested the KIWI, NRX, PHOEBUS, PEWEE, and NF thermal spectrum reactors. This series of 20 rocket/reactors advanced graphite-based NTP fuels to the point where the NRX-XE rocket reactor performed 28 burns with more than 3.5 hours of operation. Yet, by 1970, the Space Race was won, chemical propulsion engines were well established, and priorities were reassessed: Apollo missions 18-20 were cancelled, and plans for manned Mars missions were curtailed. The Rover/NERVA program was cancelled in 1972. From 1987 to 1994, the Space Nuclear Thermal Propulsion (SNTP) program developed

graphite-based, particle bed reactors. Currently, NASA’s Nuclear Cryogenic Propellant Stage (NCPS) program is recapturing NTP fuel element fabrication techniques and design knowledge.

Although the NERVA program has become synonymous with graphite-based fuels, a second fuel type—ceramic metallic (cermet) fuel—was also investigated. Cermet fuel is used in fast and thermal spectrum reactors, and this composite material involves uranium dioxide (UO<sub>2</sub>) particles typically in a tungsten matrix. Early in the Rover/NERVA program, tungsten was a very promising material for high temperature nuclear reactors. It has the highest melting temperature of all metals (3680 K)—graphite sublimates at 3915 K, while the compounds hafnium carbide and tantalum carbide (early coating material for graphite-based fuels) both melt near 4150 K, (Table 1). Further, tungsten has the lowest vaporization rate of all materials (Table 1). Initially, tungsten appeared to be chemically compatible with both hydrogen and UO<sub>2</sub>. The performance of uranium dioxide fueled cermet fuels has been reviewed previously [2, 3].

**TABLE 1.** Melting Points and Surface Vaporization Rates (Vacuum) for NTP Materials and Fuels. From [4, 3]

Material	Melting Point (K)	Surface Vaporization Rate at 2800 K (mil/hr)	Material	Melting Point (K)	Surface Vaporization Rate at 2800 K (mil/hr)
Tungsten, W	3680	< 0.01	Uranium Dioxide, UO <sub>2</sub>	3075	6×10 <sup>3</sup>
Graphite, C (sublimes)	3915	10	Uranium Carbide, UC <sub>2</sub>	2835	10
Rhenium, Re	3453	0.1	Uranium Nitride, UN	Unstable	-
Tantalum, Ta	3270	0.07	UC-40ZrC	3050	2.
Molybdenum, Mo	2890	>>10	Hafnium Carbide, HaC	4160	~1.
Zirconium Carbide, ZrC	3805	>>10	Tantalum Carbide, TaC	4150	0.1

Why were much less resources devoted to cermet, rather than graphite-based fuels? Rom [4] claims that, “The potential for tungsten reactors needed intensive experimental investigation for verification. Aside from its use as a light bulb filament, very little was known about the properties of tungsten. There was essentially no data base.” In contrast, graphite had been extensively used in high temperature industrial applications including crucibles and furnace electrodes. Graphite and related carbides have high melting temperatures, moderate vaporization rates, and good chemical compatibility at high temperatures (Table 1). Very high purity reactor grade graphite had been developed and could be fabricated with precision [4].

In the 1960’s, Argonne National Laboratory (ANL) designed two rocket reactors based on cermet fuel. The ANL200 [5] and ANL2000 were 200 MW<sub>t</sub> and 2000 MW<sub>t</sub> fast spectrum propulsion reactors. The performance goals of this rocket/reactor were a maximum fuel temperature of at least 2500°C (2770 K) (I<sub>sp</sub> = 821 to 832 s), ten hours of operation with at least 25 thermal cycles, and a fuel loss target of less than 1%. The program advanced to the point where many fuel samples and several fuel elements were tested in high temperature hydrogen. NASA’s Lewis Research Center (LeRC) performed extensive fuel and reactor development work and designed the thermal spectrum Tungsten Water Moderated Rocket (TWMR) [6, 7] to similar performance goals. Honeycomb and concentric cylinder fuel elements were developed and a critical assembly experimental program was completed.

In 1961, General Electric (GE) started the High-Temperature Materials Program (HTMP) [8] involving extensive reactor materials development for high temperature (>2200 K), fast spectrum, gas reactors. The program involved extensive thermal cycling of material samples of UO<sub>2</sub> with W, Re, and Mo matrices, with various cladding, plus reactor tests of fuel samples. The 710 Reactor Project [8], a reactor design sub-element of GE’s HTMP, involved developing brazing, sintering, and fabrication techniques for fuel elements, and culminated in reactor tests. Initially, it developed a design for the GE 710 reactor for NTP, but the program’s focus moved to closed-cycle, land and space power systems using inert gas working fluid at lower fuel temperatures. Consequently, GE 710 sample testing is less relevant to NTP, although GE HTMP continued research into high temperature cermet fuel in hydrogen.

Although cermet fuel elements were not tested as a rocket/reactor, extensive fuel sample development and testing took place at the Department of Energy’s (DOE) Argonne, Pacific Northwest, and Los Alamos National Laboratories (LANL), NASA Lewis Research Center, and General Electric. During the Nuclear Energy for the Propulsion of Aircraft (NEPA) project, W-UO<sub>2</sub> and Mo-UO<sub>2</sub> cermet materials were investigated [8]. LANL was

testing cermet fuel samples in 1957 [9], and by 1962 LeRC had a rocket reactor concept [10] and sample testing. By May 1962, when GE's 710 Reactor Project started, GE's HTMP was already performing in-pile tests of cylindrical, seven-channel, Ta- and Nb-clad W-60% UO<sub>2</sub> fuel system specimens. Tighter budgets terminated cermet fuel development about 1968, and final reports were published between 1966 and 1968.

The next section of this paper examines published cermet sample test results. The intent is to understand the significance to performance of surface cladding, fuel stabilizers, fuel particle coatings, and processing techniques. The final section attempts to place high temperature fuel performance in the engine context and understand the engine performance implications.

## RESULTS OF HISTORICAL CERMET MATERIAL TESTING

This section summarizes over 200 cermet samples from the 1960's that were tested by thermally cycling to high temperature in hydrogen (Table 2). The following sub-sections explain the underlying mechanisms and significant design improvements. The reports show two issues at high temperatures: the vaporization rate of UO<sub>2</sub> and the chemical stability of UO<sub>2</sub>. The data also show that cladding and chemical stabilizers each result in large improvements in high temperature performance, while other approaches yield smaller, incremental improvements.

The columns of Table 2 correspond to tests of different groups of samples. The design features are indicated across the top. Samples include unstable UO<sub>2</sub> samples (indicated in red), to stable fuel samples with greater than 5% fuel loss (indicated in yellow), to stable samples with lower, less than 5%, fuel loss (indicated in green). Unstable fuel cracks or turns to powder due to free uranium forming uranium hydride (UH<sub>3</sub>) (see next section); stable fuel does not. The results indicate the most effective design features are surface cladding and fuel stabilizers. The following two sections consider these two issues. Other design and processing techniques are valuable, and Baker et al [11] describes process improvement.

Sample Group	Unstable: Cracks or Forms Powder			Stable: Mass Loss > 5 %				Stable: Mass Loss < 5 %						
	1	2	3	4	5	6	7	8	9	10	11	12	13	14
UO <sub>2</sub> Only	✓						✓							
W-UO <sub>2</sub>		✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓
Partial Clad (Not Edges)								✓			✓			
Full Clad									✓	✓		✓	✓	✓
Coated Fuel Particles			✓							✓		✓	✓	✓
Stabilizers (Various)						✓	✓	✓	✓		✓	✓	✓	✓
Temperature														
(C)														
(K)														
2000														
2300														
2350														
2500														
2600														
2650														
2700														
2800														
Cycles Tested								25	>25				<30	<10
Fuel Samples Tested		29+14	46	19	2		25+	~30	~20	6	2	1	2	2
Reference	[17]	[11]	[11]	[11]	[18]	[18]	[17]	[13]	[13]	[9]	[18]	[18]	[18]	[18]

**TABLE 2.** Stability and Mass Loss of Tested Cermet Samples from Historical Reports. Each column corresponds to a different sample group, and the group's features are indicated with check marks near the top of each column. From left to right cladding and stabilizers are added. Moving from left to right in the table moves from unstable fuel (indicated in red), to stable fuel samples with > 5% fuel loss (indicated in yellow), to stable samples with low (< 5%) fuel loss (indicated in green). The number of tested samples is shown at the foot of the column with a reference to the data source.

Table 2 samples are a fraction of all the samples tested since many additional ones are not detailed or relevant. For example, Lenz compares cooling in helium and hydrogen to detect UH<sub>3</sub> formation from free uranium [9]; these samples cannot be easily included within the scope of this table. GE 710 sample tests were conducted in an inert gas, not hydrogen, as the program's focus moved to closed-cycle power generation.

### High Temperature Vaporization of $UO_2$ and the Need for Cladding

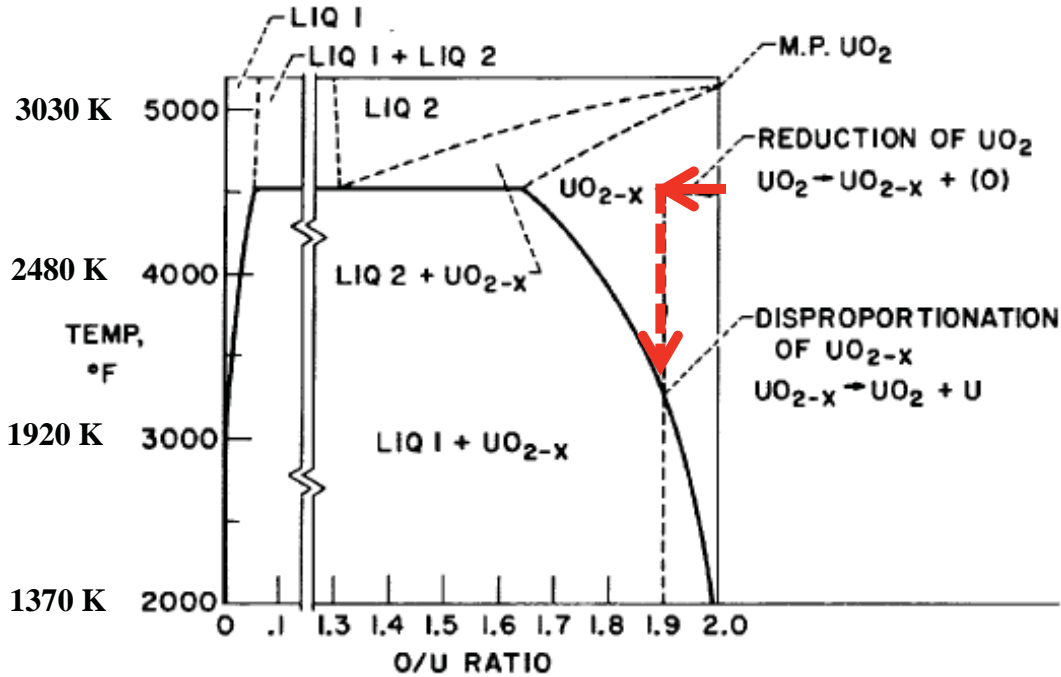
At temperatures above 2000 K, uranium dioxide vaporizes rapidly (Table 1), and cladding or coating is necessary. This need for cladding was quickly recognized as a successful method for reducing fuel loss at high temperatures. At the 1962 nuclear propulsion conference, Lenz and Mundinger [9] reported that thin tungsten coatings significantly reduced  $UO_2$  vaporization and loss (Group 10). At the same time, Saunders et al [10] and McDonald [12] reported that fuel vaporization was a major issue with unclad samples, and could be reduced by an order of magnitude with cladding.

The cladding's permeability to oxygen is important. GE found that W-Re-W was  $10^3$  times less permeable to oxygen than tantalum [8, pp. 32, Vol 3], hence a much better cladding material.

Furthermore, full cladding appears to be significantly better than face cladding. Gluyas et al [13] thermally cycled face and full clad specimens with various stabilizers in hydrogen (groups 8 & 9 in Table 2). The 3.5 x 2.5 cm samples with 0.0046 cm thick face cladding had 0.045 cm thick unclad edges. The results demonstrate the need for full cladding.

### Decomposition of Uranium Dioxide, Hydride Formation, and the Need for Stabilizers

Early in the cermet fuel program, uranium dioxide's instability at high temperatures was recognized. Further, thermal cycling and flowing hydrogen all accelerate the instability. In 1960, Anderson et al [14] reported that uranium dioxide decomposes (or reduces) at high temperature and becomes hypostoichiometric in oxygen,  $UO_{2-x}$ . At the nuclear propulsion conference in 1962 [15], papers from LeRC [10] and LANL [9] noted the  $UO_{2-x}$  reduction issue, free uranium formation,  $UH_3$  formation from this free uranium, and sample cracking. A similar understanding by GE was also cited. Towards the end of the program, Baker et al [11] carefully explained this basic behavior.

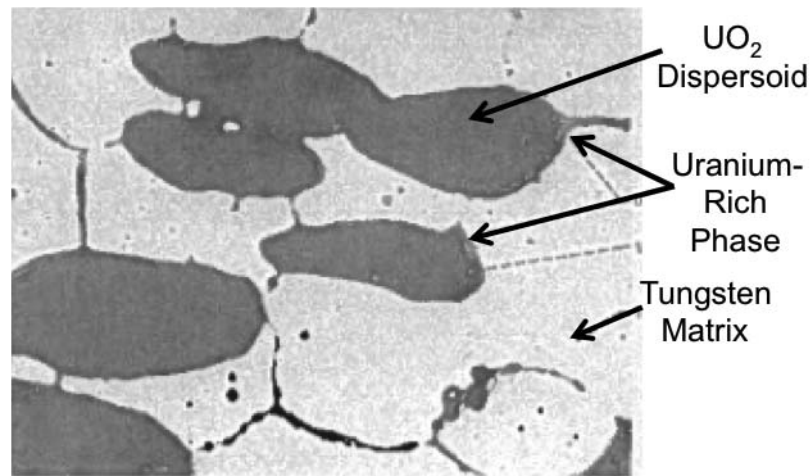


**FIGURE 1.** Oxygen Uranium Phase Diagram shows Reduction of Uranium Dioxide is Significant at Temperatures Above 2000 K. From [16].

What is this instability? Figure 1 shows the oxygen-uranium phase diagram, and the red path indicates the chemical changes at high temperature (solid line) and with cooling (dashed line). At temperatures above 2000 K, uranium dioxide undergoes reduction and becomes hypostoichiometric in oxygen,  $UO_{2-x}$ . The fluorite structure of  $UO_2$  develops vacant oxygen sites compensated with reduced  $U^{4+}$  ions. Stabilizers ( $Gd_2O_3$ ,  $Y_2O_3$ ) interfere with this reduction, and it may be the stabilizer provides free oxygen [17]. With cooling (dashed line in Figure 1), this reduction reaction reverses, and the uranium and oxygen would recombine into uranium dioxide—but some oxygen is no longer present.

Oxygen freed during reduction diffuses out of the fuel at high temperatures. With cooling, free uranium forms if oxygen has been removed. Figure 2 shows the formation of free uranium at grain boundaries adjacent to  $\text{UO}_2$  fuel particles. Cladding provides a barrier to hydrogen, oxygen, and water, and can slow the process.

With cooling below 770 K, uranium hydride forms from this free uranium. This large molecule forces apart grain boundaries, stresses the material, and can cause surface cracking [9] or reduce it to powder—sometimes in an explosive manner [17]. This is a specific example of hydrogen embrittlement. Some sources indicate that instead of forcing expansion,  $\text{UH}_3$  forms a brittle hydride on grain boundaries. Unstable fuel, where structural integrity is lost, is shown in red in Table 2.



**FIGURE 2.** Micrograph of Thermally Cycled W-20vol%  $\text{UO}_2$  Cermet Showing Free Uranium at Grain Boundaries. From [6] via [2].

Thermal cycling (corresponding to engine re-starts) amplifies this destructive process and eventually disrupts fuel integrity and allows rapid fuel vaporization. High pressure hydrogen (engine pressure  $\sim 1\text{MPa}$  (10 atm)) and flowing hydrogen accelerate fuel loss [13]. Higher temperatures should also accelerate the reaction. Cladding, stabilizers, and fuel particle coatings all slow this process, delay fuel failure (Table 2), and evidence exists of low fuel loss after many thermal cycles.

#### *High Temperature Cermet Fuel Performance*

Most of the samples in Table 2 were tested near  $2500^\circ\text{C}$  (2770 K)—for an  $I_{sp}$  of  $\sim 825$  s—because this was the design goal for the ANL and LeRC rocket/reactors. Yet, higher temperature data would help us understand higher performance cermet engines, which demand higher fuel temperatures to achieve a higher  $I_{sp}$ . Stoichiometric instability (Figure 1), and hydrogen and oxygen permeability in tungsten all increase with increasing temperatures, hence accelerated fuel loss is expected.

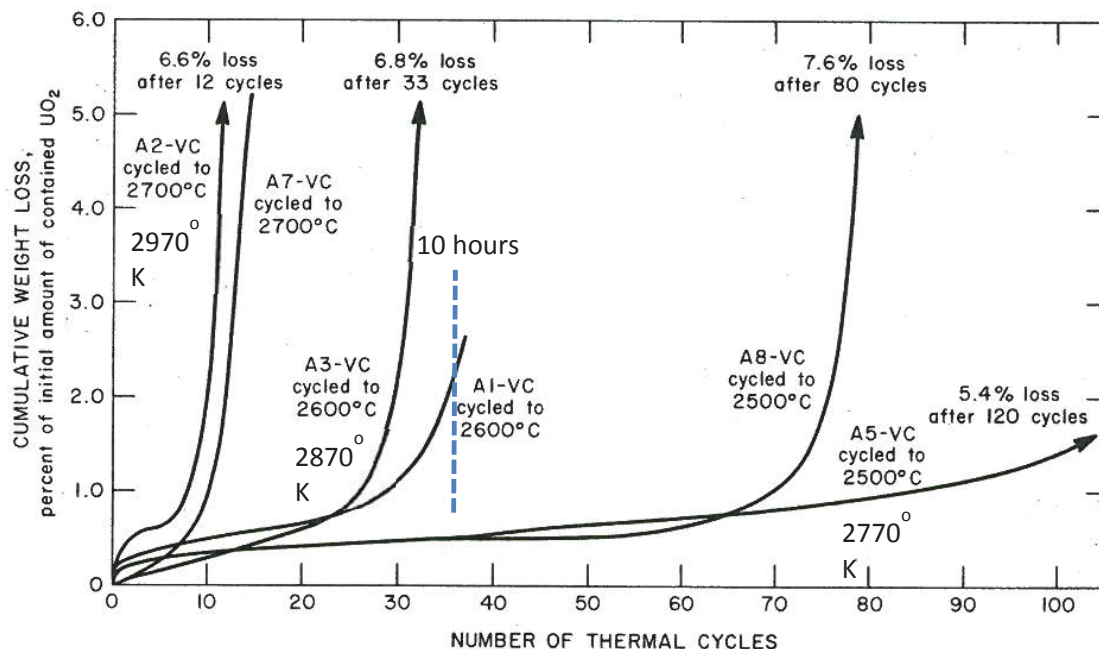
Limited data is available at higher temperatures. One group of data [17] tested  $\text{UO}_2$  with various chemical stabilizers and is not directly relevant to engine performance since it was only  $\text{UO}_2$ —matrix and cladding were not included. However, another group of 4 samples [18] (groups 13, 14) provides insight into high temperature behavior (Figure 3), and indicates drops in fuel endurance with each 100 K increase in fuel sample temperature. Baker et al [11] tested some samples to  $2500^\circ\text{C}$  and  $2600^\circ\text{C}$  in high pressure, flowing hydrogen (group 3). Again, the 100 K temperature increase shows higher fuel loss. The data [11] also show that flowing hydrogen significantly increases fuel loss, and hydrogen pressure can increase fuel loss.

For the samples in Figure 3, swelling rates of 2-9% were reported [18]. Although mass loss is typically reported and swelling/shrinkage is not, it is not clear if this swelling was exceptional.

#### *Historical Material Property Data for Fuel Element Simulation*

Material property data for cermet fuel elements and reactors includes thermal conductivity, thermal expansion, Young's modulus, yield and ultimate strength all up to maximum temperature. This information is important for simulations that predict fuel element performance. Cermet material property data is limited, but component material

(W, UO<sub>2</sub>) is more readily available, and estimates can be made for the composite material. Stewart [19] reviews material property data for NTP fuel elements.



**FIGURE 3:** Fuel Loss Behaviors of 6 Samples of Tungsten Clad W-66v/o (10m/o GdO<sub>1.5</sub>-stabilized UO<sub>2</sub>) Cermet Thermally Cycled to 2770 K, 2870 K, and 2970 K in Low Pressure Hydrogen. The report suggests the hydrogen is static, or at a very low flow rate. From [18, p. 105]. Testing with flowing hydrogen at engine pressures would reduce performance.

## IMPLICATIONS FOR CERMET ENGINE PERFORMANCE

What does a NTP fuel element designer do with this high temperature material performance data? The designer must achieve the highest possible propellant outflow temperature and the minimum peak fuel temperature to maintain acceptable fuel integrity, fuel loss, nuclear and system performance. Here we will consider how fuel geometry and nuclear heat deposition rates influence existing fuel element/reactor designs for the fuel temperatures discussed above.

### *Size and Location of Peak Fuel Temperature in a Cermet Fuel Element*

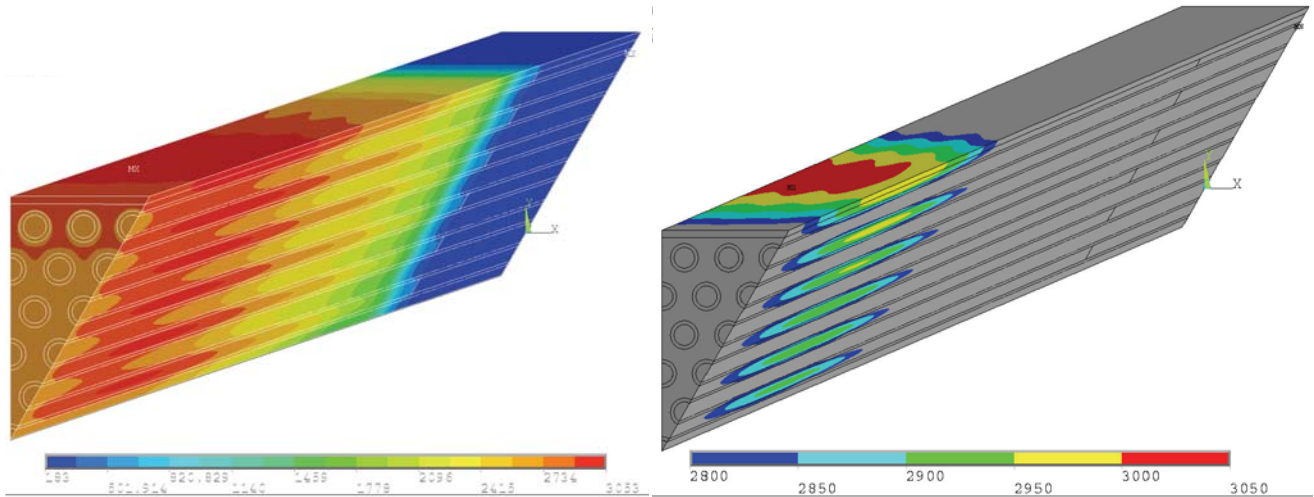
In NTP engines, the peak fuel temperature occurs within the solid fuel, and the gaseous propellant has a lower peak temperature and mean exit temperature. The nuclear reaction deposits heat in the solid fuel, and this heat diffuses to coolant channels carrying propellant. In contrast, in chemical rocket engines and jet engines, a chemical reaction releases heat in the gaseous propellant—away from the solid walls which are cooled.

The size of the high temperature fuel region within the fuel element is important. Figure 4 shows the predicted temperature distribution and the regions of highest fuel temperature from a GE 711 fuel element simulation. The high temperature fuel performance issues mentioned in the previous section would only be expected in the regions indicated. This high temperature region is a minority of the fuel element. Simulations of other cermet [20] and graphite-based [21] fuel element designs have similar temperature distributions.

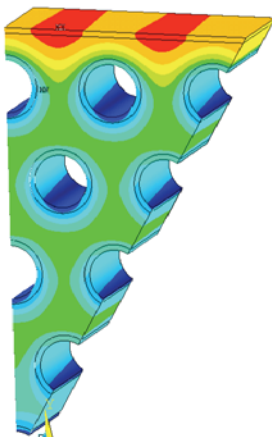
### *The Effect of Cermet Fuel Element Geometry and Heat Deposition Rate on Peak Fuel Temperature*

Fuel element geometry and heat deposition rate can have a significant effect on peak fuel temperature and propellant mean outflow temperature. In particular, some fuel element geometries are better than others.

To demonstrate this effect, we perform a thermal analysis of a short length of fuel element as shown in Figure 5. A short length can be used because the temperature gradients to the coolant channels are much greater (30X) than the gradients along the fuel element's length. The fluid flow and heat transfer effects are excluded by fixing the coolant channel surface temperature, and this greatly simplifies and clarifies the problem. Fluid flow and heat transfer effects must eventually be considered. The heat deposition rate given is the uniform rate within the fuel matrix. The heat deposition rate in the cladding is scaled based on MCNP simulation results [20].



**FIGURE 4.** Predicted Temperature Distribution Through a GE 711 Cermet Fuel Element (left), and Detail of the Hottest 250 K Region of the Fuel Element (right). The NCPs baseline cermet fuel element geometry is a modification of the GE 710 geometry, designated GE 711. Using symmetric sectors reduces computational requirements.



**FIGURE 5.** Temperature Distribution in a Fuel Element Length. Coolant Channels are Kept at a Fixed Temperature. Adiabatic External Surface.

To minimize peak fuel temperature while maximizing propellant mean outflow temperature, we are interested in two temperature differences which are plotted in Figure 6. The first, larger, temperature difference is between the fuel element edge (exterior surface) (red in Figure 5) and the coolant channel surface. These temperature differences form the solid lines in Figure 6. The second, smaller temperature difference is between the fuel matrix centerline (green) between coolant channels and the coolant channel surface. These differences form the dashed lines in Figure 6. These temperature differences are plotted against heat deposition rate (into the fuel matrix) since designers may choose to increase this rate to make a more compact reactor with heat deposited in fewer fuel elements. The engine's thrust to weight ratio increases. However, the propellant must remove the additional heat, and in smaller reactors, the nuclear reaction can be more difficult to control. As a footnote, the heat equation's mathematics confirms the linear variation of the results shown in Figure 6.

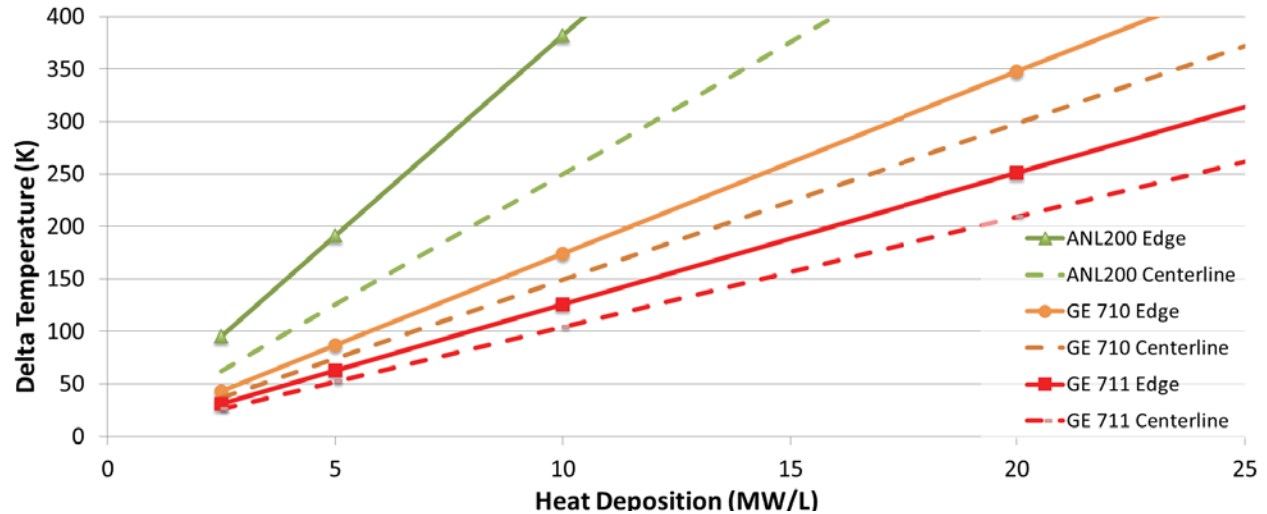
The results also show that different fuel element geometries perform differently. The ANL200 fuel element geometry has significantly larger temperature differences, than the GE 710 geometry, and, in turn the GE 711 geometry. The ANL nuclear rocket program's materials testing goal was a peak fuel temperature of 2500°C (2770 K) [18, p. 85]. Yet, the ANL200 design [5, p. 109] called for a peak centerline temperature of 3000 K at an average heat deposition of 3.27 MW/L (max. 5.16 MW/L) for an  $I_{sp}$  of 821s. Another fuel element geometry might have decreased the temperature differences and peak fuel temperature in the ANL200 design. For this

and other reasons, the NCPs baseline cermet fuel element geometry is a modification of the GE 710 geometry, designated GE 711.

Stress analysis for NTP fuel elements is not a typical linear stress analysis with a margin of safety. At high temperatures, the materials will creep and plastically deform. These deformations are driven by thermal stress, thermal expansion, and differences in thermal expansion at material interfaces. Thermal cycling results in large

deformations between the hot and cold shapes of each cycle. When hot, the material’s ductility can accommodate deformations, but when cold or cooling, the material is less able to avoid residual stress, deformation, or fracture.

Stress analysis for the fuel elements in Figure 6, indicate that stresses are dominated by mismatch of thermal expansion at material interfaces; thermal stresses are smaller.



**FIGURE 6.** Temperature Difference, Fuel Peak at Edge to Coolant Channel Wall (Solid Line) and Fuel Centerline to Coolant Channel Wall (Dashed Line), for Several Cermet Fuel Geometries at a Range of Heat Deposition Rates.

## CONCLUSION

In order to understand cermet fuel performance in NTP engines, this paper examined historical cermet material development reports. Fuel element and engine simulations were used to interpret the data. The reports indicate two issues at high temperatures: the high vaporization rate of  $UO_2$  and the chemical stability (high temperature reduction) of  $UO_2$ . The data show that cladding and chemical stabilizers each result in large, order of magnitude, improvements in high temperature performance. Contemporary fuel samples are unlikely to achieve the best historical performance without these features. Some coated, stabilized, cermet samples were tested above 2770 K, but they are a small sample to fully assess high  $I_{sp}$  (>900s) potential. Contemporary testing of fuel samples at high temperatures is justified. Fuel element development may be complicated by the stability of  $UO_2$  at high temperatures.

## NOMENCLATURE

$I_{sp}$  = Specific Impulse, s

$T$  = Temperature, K

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