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A Review of the Effects of Burnup on the Thermal Conductivity of UO_2

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A REVIEW OF THE EFFECTS OF BURNUP ON THE THERMAL CONDUCTIVITY OF UO_2

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INTRODUCTION

In the temperature range below 500°C, irradiation damage strongly affects the thermal conductivity of UO_2.[1-7] Above 500°C, which is in the primary region of operation for commercial LWR fuels, much of the damage is quickly annealed out, and the resultant effects on thermal conductivity are generally considered to be minimal. However, in a recent report[8] on the behavior of ThO_2-2.3 w/o UO_2 fuel, the trend of decreasing thermal conductivity due to irradiation seemed to be more pronounced and conceivably could imply a basic oxide fuel characteristic that may have been underestimated in previous evaluations of UO_2 fuel work.

The purpose of this review was to re-evaluate the current licensing assumption that fuel pellet thermal conductivity does not change significantly with increasing burnup and to assess the applicability of the ThO_2 based data to LWR operation.

LITERATURE REVIEW

A review of the literature indicates work with UO_2 thermal conductivity in three major temperature regions: 1) below 500°C, 2) between 500°C and 1600°C, and 3) above 1600°C to melting (∿2850°C).

Low Temperature (<500°C)

Several investigators [1-6] have shown a significant decrease in UO_2 thermal conductivity below 500°C at relatively low burnups. For example, Ross[1] reported a 26% decrease in conductivity relative to the unirradiated value at 60°C after irradiation at 450°C to an exposure of 2×10^{16} fissions/cc, but no further decrease after 7×10^{18} fissions/cc.* Annealing at temperatures up to 1000°C removed some of the irradiation produced damage with the amount

* fission/cc = burnup (MWD/MTM) x density x 2.578×10^{16}

removed decreasing with increasing exposure. Daniel, et al[2] found a decrease of 50% of the unirradiated value after irradiation at <100°C out to a burnup of 11×10^{18} fissions/cc; however, no detectable minimum was observed. Other reports [3-6] also show decreasing thermal conductivity at temperatures below 500°C with the largest affect appearing between 10^{15} and 10^{16} fissions/cc. Above 10^{18} fissions/cc, the rate of decrease is usually reported to be small and apparently approaches a constant with higher exposures.

Thus, the general trend below 500°C is for the thermal conductivity to decrease with the degree of reduction a function of both burnup and irradiation temperature. These results have been interpreted[7] to mean that more than one damage mechanism is operating. Radiation initially produces isolated point defects with small clusters of displaced atoms and fission products which cause a reduction in the effective mean free path between phonon collisions. Nucleation and growth of larger clusters follow and these subsequently migrate to grain boundaries, thus, the effective mean free path approaches a steady state condition.

Mid-Range (500°C - 1600°C)

The thermal conductivity of stoichiometric UO_2 does not seem to show measurable reduction in this temperature region for exposures up to 4×10^{19} fissions/cc. [3,9,10] However, above 2.5×10^{20} fissions/cc, a decrease in thermal conductivity is found. [9,11,12] This appears to be true for mixed oxide fuel as well[11] where a 75% decrease was reported at 1000°C after 11×10^{20} fissions/cc, except at 12×10^{20} fissions/cc where the decrease was only 25%. The latter results on mixed oxide were attributed to the formation of a uranium/plutonium solid solution. In general, the degree of reduction in UO_2 seems to vary from 25-50% [9,12] and this variability is apparently due, at least in part, to differences in O/M ratios for the fuels studied. In most instances, the reduction in thermal conductivity approached a near constant value with increasing exposure.

The pattern of decreasing thermal conductivity for ThO_2-2.3 w/o UO_2 as reported by Berman[8] was only for one sample at 8.6×10^{20} fissions/cc but the trend was generally the same as for UO_2 except below 500°C where the decreases appear larger. In the mid-temperature range, reductions

from unirradiated values on the order of 20-30% were reported at a fission depletion of > 10^{19} fissions/cc which is in line with UO_2 results. Other contributing factors such as oxygen potential, microstructural features, transmutation products or basic differences in physical properties of the two materials make relative comparisons difficult and may be the reason for differences in the absolute values. Thus, the same general trend is seen for both types of fuel as a function of burnup and temperature.

High Temperature (>1600°C)

Little work has been done in this region because of the experimental difficulties associated with obtaining accurate data at high temperatures. Annealing of radiation damage affects are expected to be quite rapid and hence the effects on thermal conductivity minimal. For these reasons, this review did not address the effects of burnup on thermal conductivity above 1600°C under the premise that the overall affects could have minimal impact on normal fuel operating behavior.

DISCUSSION

During the irradiation of UO_2 fuel, additional scattering centers are introduced into the fuel matrix which behave similarly to lattice impurities. The multiplication of these lattice defects, caused by irradiation, tends to saturate at very low burnups ($\sim 10^{19}$ fissions/cc) and low temperatures. The observation that the decreases become less severe as the temperature increases can be attributed to an annealing affect which reduces the number of lattice defects. The influence of annealing is more pronounced at low burnups because smaller numbers of lattice defects will be introduced into the UO_2 matrix.

Changes in thermal conductivity due to burnup can depend on factors other than irradiation induced lattice changes such as the resultant solid and gaseous fission products, pore size and shape and microstructural features. At temperatures between 1000°C and 1600°C, fuel densification may offset increases in pore size due to swelling caused by fission gases and thereby increase the burnup threshold. Additional thermal conductivity decreases for fission depletions greater than 2.5×10^{20} fissions/cc may

also, be influenced by microstructural changes which have sometimes been evidenced by the disruption of the original grain boundary structure into very fine subgrains, or by the complete absence of resolvable structure for fuel which has experienced very high exposure. Fission gas release rates are known to increase with burnup and will contribute to reduced conductivity because of the accumulation of significant quantities of foreign fission products in the UO_2 matrix and from the expected net increase in O/M ratio accompanying fission.

A few mathematical relationships have been suggested[4,9,13] to correlate UO_2 thermal conductivity with temperature and burnup. The most generally applicable of these was proposed by Daniel and Cohen[9]:

$$(1) \quad K_{irr} = \left[\frac{1}{K_o} + \frac{AF}{T}\right]^{-1}$$

where K_{irr} = thermal conductivity of irradiated UO_2 (W/cm°C)

K_o = thermal conductivity of unirradiated UO_2 (W/cm°C)

A = Constant (W/cm)

F = fission depletion (fissions/cc$\times 10^{-20}$)

T = temperature (°C)

The unirradiated thermal conductivity (K_o) can be found from the work of Lyons, et al.[15] for 95% TD UO_2.

$$(2) \quad K_o = \frac{B}{T} + C(T+273)^3$$

where B = 0.095

and C = 6.1256×10^{-13}

T = °C

and equation (2) can also be adjusted for density (D) to yield:

$$(3) \quad K_o = 1.08\left[\frac{D}{1+0.5(1-D)}\right]\frac{B}{T} + C(T+273)^3$$

To determine the reduction in thermal conductivity as a function of burnup, several regions of burnup and temperature were defined and a representative value for the constant (A) in equation (1) assigned:

Fission Depletion Range	A
1) fission depletion (FD) $< 1.0 \times 10^{15}$ fissions/cc,	0
2) $1.0 \times 10^{15} \leq$ FD $< 1.0 \times 10^{16}$ and temperature (T) $< 500°C$	20
3) $1.0 \times 10^{16} \leq$ FD $< 1.0 \times 10^{17}$ and $T < 500°C$,	200
4) $1.0 \times 10^{17} \leq$ FD $< 1.0 \times 10^{18}$ and $T < 500°C$,	75
5) $1.0 \times 10^{18} \leq$ FD $< 1.0 \times 10^{19}$ and $T < 500°C$,	20
6) $1.0 \times 10^{19} \leq$ FD $< 1.0 \times 10^{20}$ and $500 \leq T < 1600°C$,	45
7) $1.0 \times 10^{20} \leq$ FD $< 1.0 \times 10^{22}$ and $T > 500°C$.	450

Equations 1 and 3 were incorporated into a special subroutine and inserted into GAPCON-THERMAL-2[14]. Thermal conductivity was then calculated from inputed values of temperature, burnup, and density. A plot of the percent decrease in thermal conductivity as a function of burnup (expressed as fission depletion) for a range of temperatures and for a 94% density case is shown in Figure 1. Reductions of up to 50% are indicated at temperatures below 300°C and for fission depletions of $<10^{19}$ fission/cc. Above 500°C, the reductions are much less severe and diminish rapidly with increasing temperature owing to the annealing out of irradiation damage. A plot of thermal conductivity as a function of temperature for different burnups is presented in Figure 2, and shows that the differences in thermal conductivity at temperatures above 1000°C are relatively small.

The net calculated effect on fuel centerline temperature for a case in which the fuel would be operated at a linear heat rating 12 KW/ft is shown in Figure 3. The maximum difference in temperature occurs at about 24,000 MWd/MTM and indicates an approximate 20°C increase if burnup effects are taken into account. This difference would most likely have minimal impact on fuel restructuring and fission gas release.

CONCLUSIONS

The general trends which relate changes in thermal conductivity of UO_2 fuel as a function of temperature and burnup can be summarized as follows:

1) At temperatures below 500°C, reductions in UO_2 thermal conductivity relative to the unirradiated values can be expected up to a saturation level of approximately 10^{19} fissions/cc.

2) At temperatures above 500°C, the thermal conductivity will undergo little change at low burnups, ($<10^{19}$ fissions/cc) but at higher exposures some decrease can be expected which should, in turn, diminish with increasing temperature.

3) A review of the data reported by Berman[8] on the ThO_2-UO_2 fuel indicates that the basic behavior is the same as for UO_2 in the temperature range of major interest. The applicability of this data to LWR UO_2 fuel is somewhat questionable because of basic physical property differences, and limited data on irradiation effects, and would not seem to support concerns that the effects of burnup on thermal conductivity for LWR fuel may be of more significance than currently believed.

4) A mathematical expression of the type proposed by Daniel and Cohen[9] seems to provide a reasonable approximation for the behavioral trends reported in the literature which relate changes in thermal conductivity to increasing burnup in certain temperature regimes. Calculations indicate that only small incremental increases in the fuel centerline temperature might be expected if burnup effects are taken into account.

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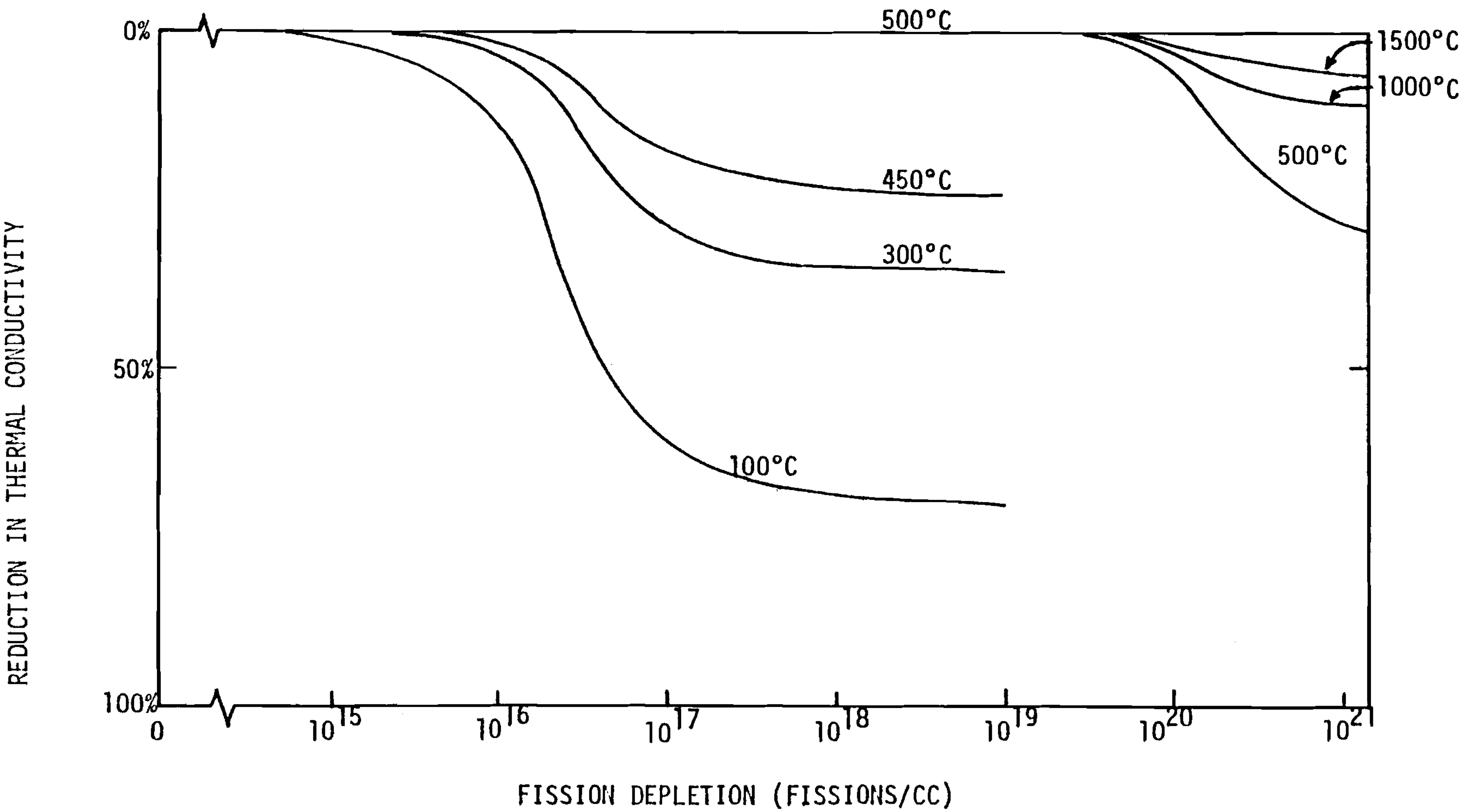

FIGURE 1. Percent Decrease in Thermal Conductivity of 100% TD UO_2 vs. Fission Depletion for Various Temperatures.

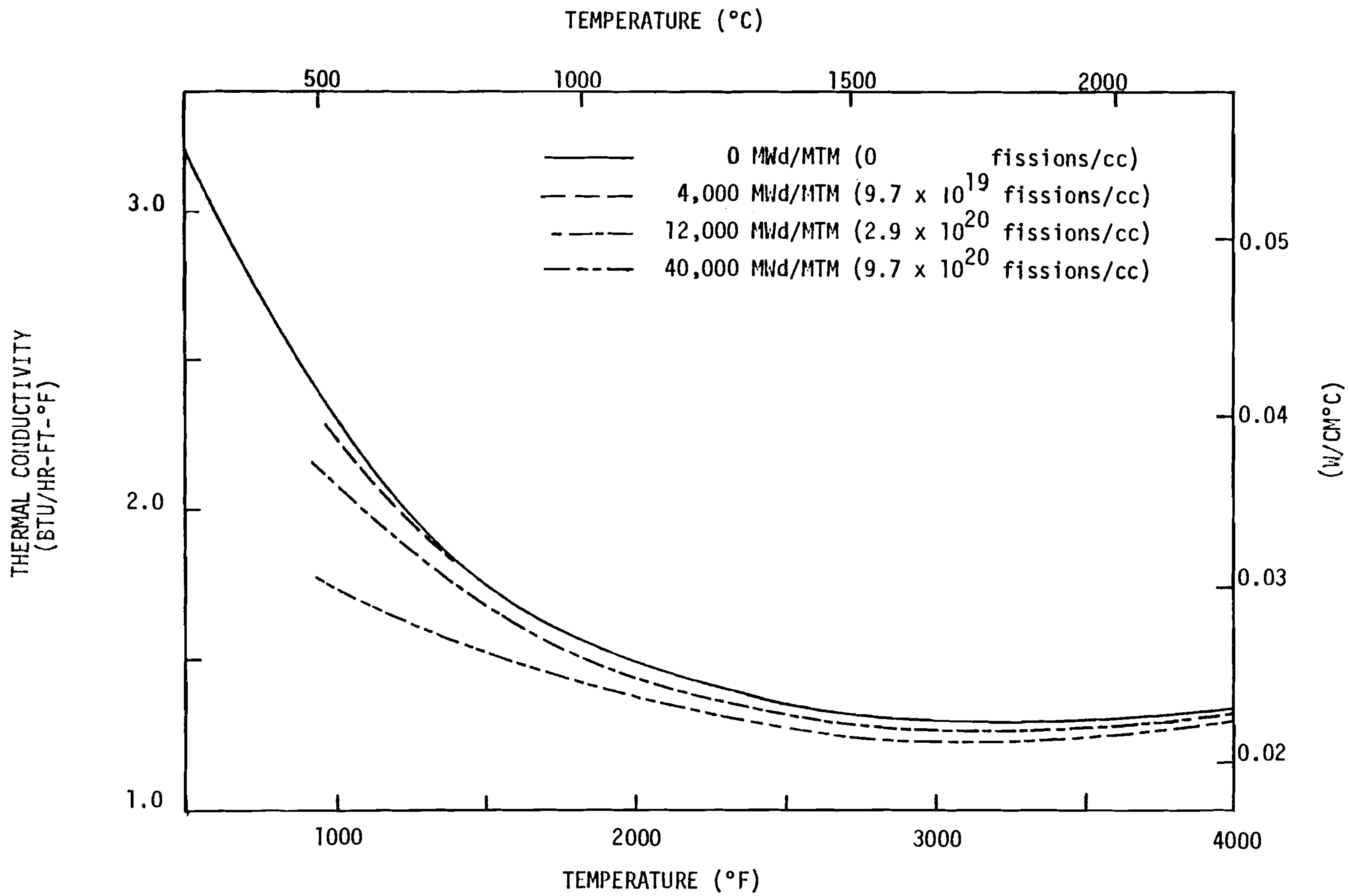

FIGURE 2. Thermal Conductivity of 94% TD UO_2 vs. Temperature For Various Burnup Values.

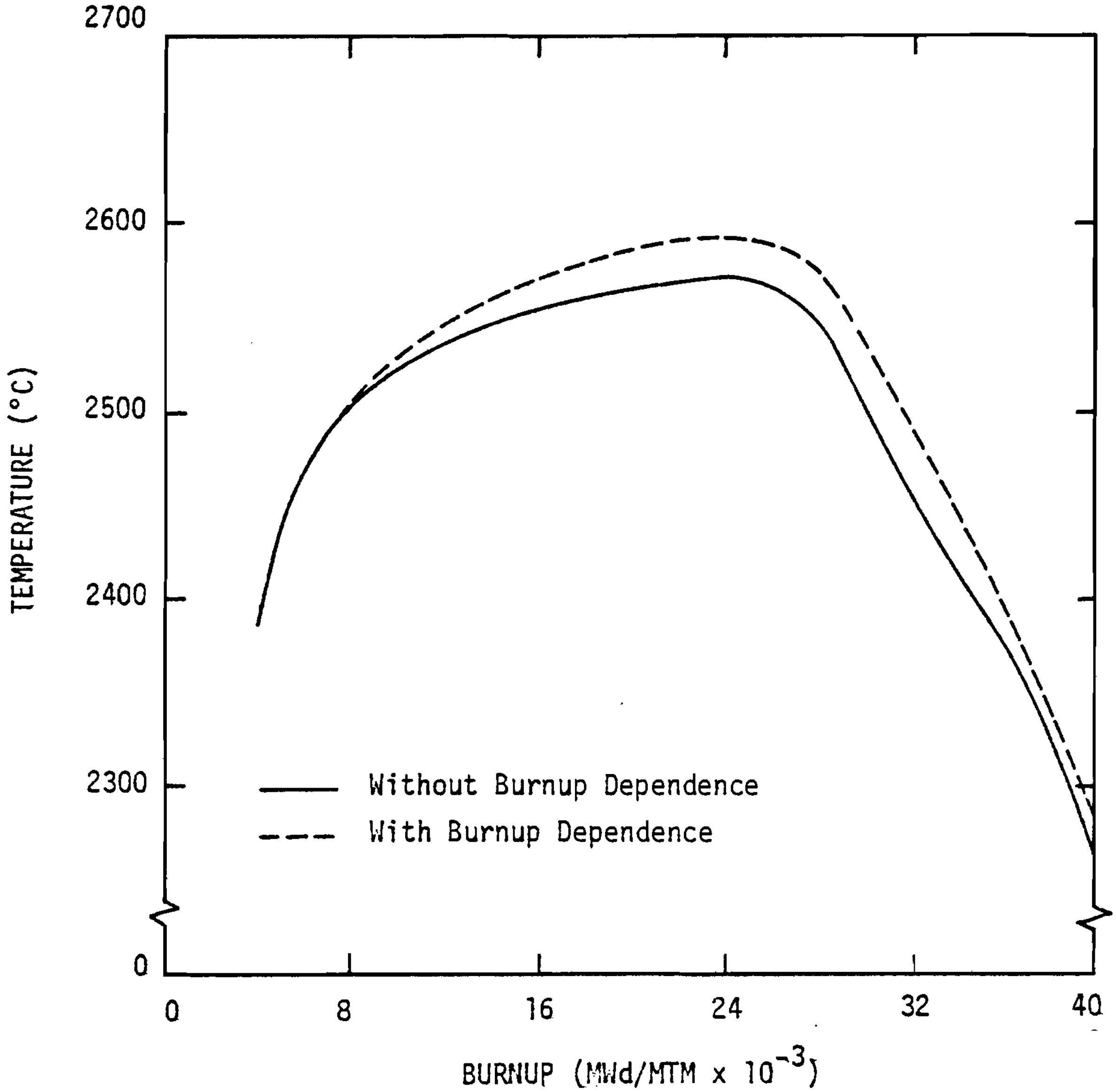

FIGURE 3. Centerline Temperature of 94% TD UO_2 with 12 KW/ft linear Heat Rating. (Predicted with GAPCON-THERMAL-2).

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