MODELLING OF PHENOMENA ASSOCIATED WITH HIGH BURNUP FUEL BEHAVIOUR DURING OVERPOWER TRANSIENTS

H.E. SILLS, V.J. LANGMAN AND F.C. IGLESIAS

Ontario Hydro, 700 University Avenue,
Toronto, Ontario, Canada M5G 1X6

ABSTRACT

Phenomena of importance to the behaviour of high burnup fuel subjected to conditions of rapid overpower (*i.e.*, LWR RIAs) include the change in cladding material properties due to irradiation, pellet-clad interaction (PCI) and "rim" effects associated with the periphery of high burnup fuel. "Rim" effects are postulated to be caused by changes in fuel morphology at high burnup.

Typical discharge burnups for CANDU fuel are low compared to LWRs. Maximum linear ratings for CANDU fuel are higher than those for LWRs. However, under normal operating conditions, the Zircaloy-4 clad of the CANDU fuel is collapsed onto the fuel stack. Thus, the CANDU fuel performance codes model the transient behaviour of the fuel-to-clad interface and are capable of assessing the potential for pellet-clad mechanical interaction (PCMI) failures for a wide range of overpower conditions. This report provides a discussion of the modelling of the phenomena of importance to high burnup fuel behaviour during rapid overpower transients.

1. INTRODUCTION

Computer models have been in place in Canada for many years to evaluate the behaviour of CANDU fuel during overpower transients associated with large break loss-of-coolant accidents (LOCAs). The computer code ELOCA (1) is capable of providing a temperature, stress and fuel morphology based mechanistic assessment of the transient thermal and mechanical behaviour of Zircaloy-4 clad, pelletized UO_2 fuel.

As part of the code's validation process, ELOCA has been successfully used to model the transient thermal and mechanical behaviour of LWR fuel in selected RIA fuel experiments from the Japanese Nuclear Safety Research Reactor (NSRR). The results of these assessments, along with a discussion of the modelling of the phenomena of importance to high burnup fuel behaviour during rapid overpower transients, will be discussed.

Commercial irradiations employ a once-through fuel cycle based on refuelling at power. Studies have not identified any life limiting phenomena which would preclude the use of the current fuel design to burnups of at least 600 MWh/kgU (25 GWd/t) (2).

A substantial database has been established for advanced fuel cycles using MOX [$(U,Pu)O_2$, $(Th,U)O_2$ and $(Th,Pu)O_2$] and slightly enriched uranium (SEU) for normal operation, including operational transients, to burnups in excess of 1000 MWh/kgU (40 GWd/t).

Figure 1 illustrates a typical overpower transient for a CANDU core for a critical break large LOCA. The overpower transient has a pulse width of approximately 1 second. Licensing considerations preclude centreline melting of the fuel. This limits the sum of the initial stored energy in a maximum powered fuel element and the energy in the overpower during the first 5 seconds to less than approximately 960 kJ/kg (230 cal/g).

2. EXPECTED OVERPOWER FUEL BEHAVIOUR

The transient thermal and mechanical aspects of fuel behaviour during overpower transients are described as a function of the rate of energy addition to the fuel. The expected behaviour of CANDU fuel during large break LOCA-induced overpower transients is shown to be predominantly affected by transient thermal processes and to be far less severe than the in-reactor fuel response to overpower transients representative of reactivity initiated accidents in light water reactors.

2.1 Fission Heat

The fission process is an energetic nuclear phenomenon. The rate of transformation of fission energy to thermal energy is very fast (*i.e.*, less than 10^{-10} seconds) (3). Each fission event adds 3.1×10^{-14} kJ to the lattice.

In the Reactivity Initiated Accident (RIA) tests conducted in the Power Burst Facility (PBF), the overpower transients typically have a duration of 0.050 seconds, with a maximum specific fission rate of approximately 5×10^{20} fissions/s/kg (4). This is slightly lower than the 1.5×10^{21} specific fission rate obtained in the NSRR RIA test series with a 0.012 second pulse duration (5). Since a fission path within the fuel is about 10 microns long by 150 Å in diameter, then approximately 40 per cent of the fuel volume is affected by fission events during a typical RIA overpower pulse.

The fission fragments produce local shock waves (0.1-1 GPa) and local thermal stresses (0.1-1 GPa) in the UO_2 lattice (3). These stresses dissipate at the speed of sound in UO_2. At fission rates in excess of 2×10^{21} fissions/s/kg, insufficient time would be available to dissipate these local stresses before the next fission event occurred. At higher fission rates, stresses in the lattice could provide a driving force for mechanical processes (*i.e.*, energetic fuel dispersal). At lower fission rates, stresses due to fission events have sufficient time to dissipate and energy transfer processes are thermally controlled.

2.2 Effect of Fission Rate on Fuel Behaviour during Overpower Transients

The fission rate determines the rate of energy deposition in the fuel. The fission rate increases from the fuel centreline to the pellet surface due to flux depression within the fuel pellet (6). The fission rate can be further enhanced at the fuel pellet surface by the distribution of fissile atoms (*i.e.*, Pu^{239}) associated with neutron capture by fertile atoms, such as U^{238} (Figure 2).

Figure 3 illustrates the specific fission densities associated with known "overpower" transients ranging from power ramps to pulsed reactor tests. The overpower transients predicted in the large break LOCA analyses for a CANDU large break LOCA, the OPTRAN test results from the PBF and the CANDU-PBF test have a similar energy deposition rate (7 and 8). The NSRR, SPERT-CDC and PBF-RIA tests form another group with energy deposition rates approximately two orders of magnitude more severe (4, 5, 9 and 10).

Figure 4 illustrates the thermal response of a fresh fuel element subjected to an NSRR type of overpower transient (*i.e.*, 10 ms pulse width) as predicted by the ELOCA code. The initial fuel radial temperature distribution is uniform at 300 K. As shown in Figure 4, the maximum fuel temperatures initially occur near the pellet surface. These predictions are consistent with observed fuel behaviour in the NSRR tests and are considerably different from the behaviour expected for CANDU fuel during large break LOCA-induced overpower transients.

2.3 Fuel Thermal Response Important to Overpower Transients

Heat transfer from the fuel during overpower transients is governed by the thermal diffusivity, the ratio of thermal conductivity to heat capacity, of the UO_2. The following sections examine the effect of irradiation, porosity and fission product content on these parameters.

2.3.1 Thermal Conductivity of UO_2

The thermal conductivity of UO_2 is needed for the assessment of fuel temperatures during normal operating and accident conditions. The thermal conductivity of UO_2 is influenced by the fuel stoichiometry, the presence of plutonium and the extent of irradiation (*i.e.*, pellet cracking, fission product generation and fuel porosity).

It is generally accepted that the UO_2 thermal conductivity can be expressed by the addition of two terms. These terms reflect the theoretical premise that heat is mainly conducted in UO_2 by mechanisms based on phonons and small polarons (11). At lower temperatures (*i.e.*, less than approximately 1500°C), the phonon-based mechanism is dominant. At higher temperatures (*i.e.*, greater than approximately 1800°C), the polaron-based mechanism is dominant.

Several authors (11 to 15) have proposed values for all the constants associated with determining the thermal conductivity of UO_2. Figure 5 shows UO_2 thermal conductivity versus temperature as predicted via the information in References 11 to 15, inclusive.

Neutron irradiation generally decreases the thermal conductivity of UO_2 through fission product generation and porosity changes.

2.3.1.1 Fission Product Generation

The generation of fission products during irradiation has a two-fold effect on fuel thermal conductivity. First, the production of fission products with high partial pressures at power contributes to the formation of bubbles which modify the existing porosity and can consequently affect the thermal conductivity of the fuel. The second effect is the production of fission products or compounds with low partial pressures that cause modifications to the phonon-impurity scattering contribution to fuel thermal conductivity.

Philipponneau (16) proposed that the effect of solid fission products on conductivity can be modelled by enhancing the value of the phonon term. Figure 6 shows the effect of this correction on the thermal conductivity for CANDU fuel with a burnup of 0, 225 and 700 MWh/kgU. It is apparent from the figure that the corrected thermal conductivity, for the solid fission product effects, is very small for the range of fuel burnups of interest to CANDU reactors. Recent measurements on irradiated fuel confirm that the effect of irradiation on thermal conductivity is negligible for burnups less than approximately 800 MWh/kgU (17).

2.3.1.2 Porosity

The fuel porosity is comprised of unsintered fabrication porosity and the bubbles formed by the fission release processes. Theoretically, it is expected that the porosity coefficient will decrease as fuel temperature increases since pores will become better conductors of heat (*i.e.*, gaseous and radiation conduction). From published results, reasonable bounds for this parameter, which include a wide range of pore shape and orientation, are $1 \leq$ coefficient ≤ 3 (14, 18, 19 and 20).

2.3.1.3 Effect of Plutonium Content

Olander concludes that the parameter representing the effect of phonon-impurity scattering is almost independent of the plutonium content (13 and 21). Conversely, he concludes that the results from experiments, in which the oxygen-to-metal ratio is held constant and the fraction of plutonium is varied, can be fitted by varying the parameter that stands for the phonon-phonon (Umklapp) scattering process.

2.3.2 Specific Heat of UO_2

The specific heat capacity of UO_2 is needed for fuel behaviour calculations during normal operating and accident conditions (22). As the heat capacity is an extensive material property, the addition of small amounts of fission products and/or other materials that do not modify the structure of the bulk fuel matrix for CANDU fuel will have little effect on the heat capacity.

Heat transfer from the fuel during overpower transients is governed by the thermal diffusivity, the ratio of thermal conductivity to heat capacity, of the UO_2. The diffusivity decreases with increasing burnup (23) due to the decrease in thermal conductivity. The effect of burnup on fuel specific heat appears negligible but a major effort worldwide is underway to confirm this observation.

2.4 Fuel Mechanical Response to an Overpower Transient

2.4.1 Plastic Core Formation and Pellet Cracking

At temperatures in excess of approximately 1000°C, UO_2 can behave as a plastic material. At temperatures near the melting point, UO_2 becomes a "viscous" solid (24). However, the temperature at which the UO_2 exhibits plasticity depends upon the heating rate. The results of fuel irradiations in the NRU reactor, indicate a temperature of plasticity close to 2000°C for fast ramps (*i.e.*, 10 second durations) to power (25).

The peripheral region of a fuel pellet, operating at temperatures less than the plasticity limit, is subject to brittle fracture in an attempt to reduce tensile stresses. The non-linear temperature profile produces tensile tangential stresses and compressive radial stresses in the fuel pellet. Fracturing of the peripheral fuel region (*i.e.*, outside the plastic region), is predominately by radial cracks. These radial cracks penetrate to the plastic inner zone. The deeper the crack penetration, the more the pellet can expand thermally and the greater the potential for PCMI.

For the very short overpower transients typical of NSRR type tests, the initial peak fuel temperature occurs near the rim of the fuel (Figure 4). Portions of the fuel pellet inboard and outboard of this high temperature region are put into tension which further assists fuel cracking and expansion.

2.4.2 Grain Boundary Gas Bubble Behaviour and Microcracking

The behaviour of grain boundary gas bubbles during overpower transients and the impact of this behaviour on the potential for extensive separation of the fuel grain boundaries (*i.e.*, referred to as fuel microcracking) are discussed. Fuel microcracking reduces the thermal conductivity of the fuel but appears to be negligible when the sheath tightly constrains the fuel (26).

Due to surface tension effects, grain boundary bubbles tend to be lenticular in shape with the long axis of the bubble oriented along the grain boundary. The dynamics of these gas filled bubbles is dependent on temperature and heating rate (27 to 29). Grain boundary bubbles would be expected to grow by vacancy diffusion in regions where the UO_2 is sufficiently plastic. In the colder regions of the pellet where the fuel is brittle, volume swelling of the grain boundary bubbles by overpressurization can be caused by i) crack propagation from the sharp ends of the lenticular bubble (27 and 29) or ii) early interconnection of bubbles by rapid grain boundary vacancy diffusion (27). At very high temperatures (*i.e.*, greater than 2300°C), grain boundary bubbles can take the form of large spheroid gas pools (29).

The overpressurization occurs at heating rates where gas atoms arrive at the grain boundary faster than the bubbles can grow to accommodate the arriving gas atoms. At very high heating rates (*i.e.*, greater than 5000°C/s), energetic microcracking (*i.e.*, "explosive fragmentation") has been postulated (28).

If low temperature, low power fuel is suddenly subjected to an extreme heating rate such as in the RIA tests in NSRR (5), then an essentially flat temperature distribution with a sharp temperature peak near the pellet perimeter exists for a short time at the peak of the power pulse (*i.e.*, approximately 10 ms elapsed time) (Figure 4). This temperature distribution causes the entire fuel pellet within the temperature peak to be in radial tension and the outboard rim of the fuel to be in tension in the tangential direction. As the fuel-to-sheath contact pressure is increasing rapidly at the same time, this tensile stress region will only exist for a short time. However, this tensile radial stress (the circumferential stress is also tensile) can assist in separating grain boundaries and releasing volatile grain boundary fission products.

Some fracturing of grain boundaries can also occur under fast cooling (*i.e.*, rewet) due to the thermal stresses generated. Gas filled boundaries would be prone to fracturing.

For the specific fission densities corresponding to a CANDU large break LOCA scenario, extensive UO_2 morphological changes are not expected. The OPTRAN and CANDU-PBF test results, with specific fission densities similar to the CANDU large break LOCA overpower transients, support this prediction since no unusual UO_2 morphologies are found.

2.4.3 Sheath Strain and Failure

One of the rim effects of concern at high burnup is the reduction of clad ductility and impact strength (6). Figure 7 illustrates the change in Zircaloy-4 yield stress for the high stress conditions typical of PCMI during overpower transients. Irradiation hardening increases the yield stress of Zircaloy-4 with this effect saturating at high dose levels (6). However, plastic strain increments as small as 0.3% can lead to "work softening" as irradiation damage is cleared from the lattice by "swathing" of dislocations (30 to 32). At high stress levels, Zircaloy-4 exhibits a high stress sensitivity leading to localized strain and failure at stress risers on the clad. The swathing of irradiation damage increases this sensitivity. At elevated temperatures (*i.e.*, >500°C), annealing of the irradiation damage would improve the clad ductility at high stress.

A related alloy, Zircaloy-2, used in the fuel channel structure of CANDU reactors, reaches very high fast fluences (>1 MeV). The change in both the ultimate tensile strength (UTS) and the failure strain saturates, in an exponential manner, by a fluence of 8×10^{25} n/m^2. Test results at 170°C and a strain rate of 10^{-3} s^{-1}, show the UTS increasing from 290 MPa to a final value of 525 MPa. For the same test conditions, the failure strain decreases from 40% total elongation to just under 10%.

3. CONCLUSIONS

A number of contributing factors to fuel rod failure during overpower at high burnups (>60 GWd/t) have been identified (6 and 33). These phenomena include:

i) increased energy deposition in the fuel rim;
ii) mechanical damage to the fuel from high pressure gas bubbles;
iii) reduced cladding ductility and impact strength;
iv) pellet clad mechanical interaction; and
v) corrosion (oxidation and/or hydriding).

These phenomena are modelled by Canadian fuel performance codes. Analysis and experiment have not identified any life limiting behaviour which would preclude the use of current the CANDU fuel design to burnups of at least 600 MWh/kgU (25 GWd/t) (2). Studies in support of advanced fuel cycles would extend our confidence to burnups in excess of 1000 MWh/kgU (40 GWd/t).

Current studies are extending this burnup range by tests on related alloys (*e.g.*, Zircaloy-2), related fuels (*e.g.*, SIMFUEL (23)), and re-examination of our high burnup fuels database.

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Figure 1
SMOKIN Predicted Overpower Transient

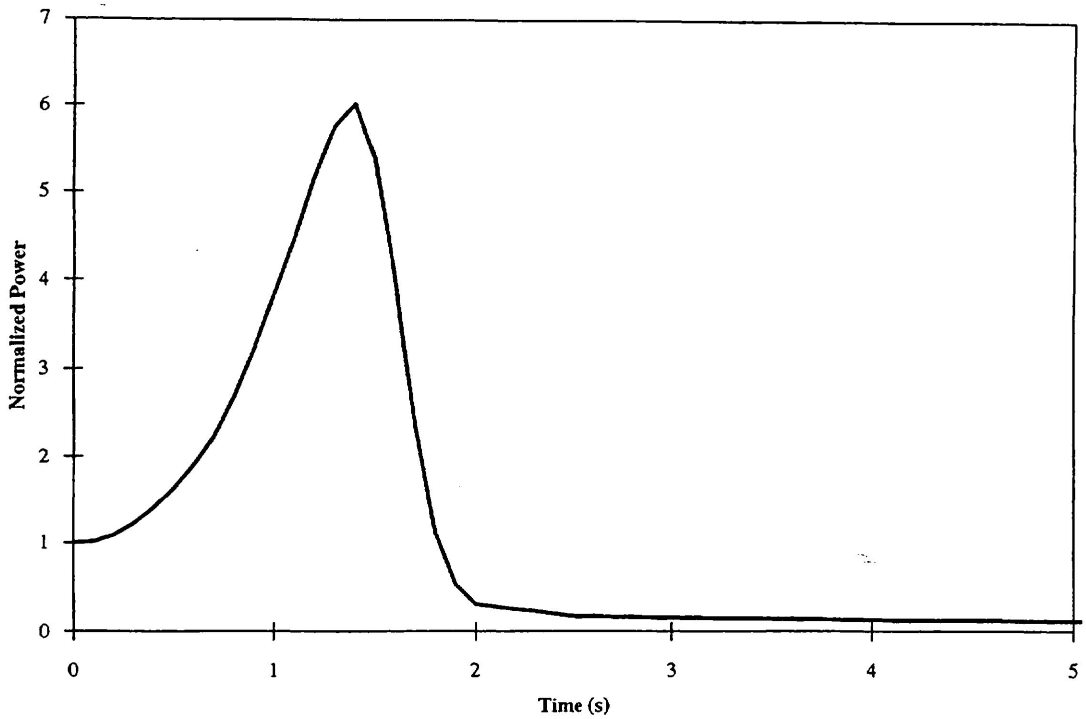


Figure 2
Variation of Radial Power Density with Pellet Radius and Burnup

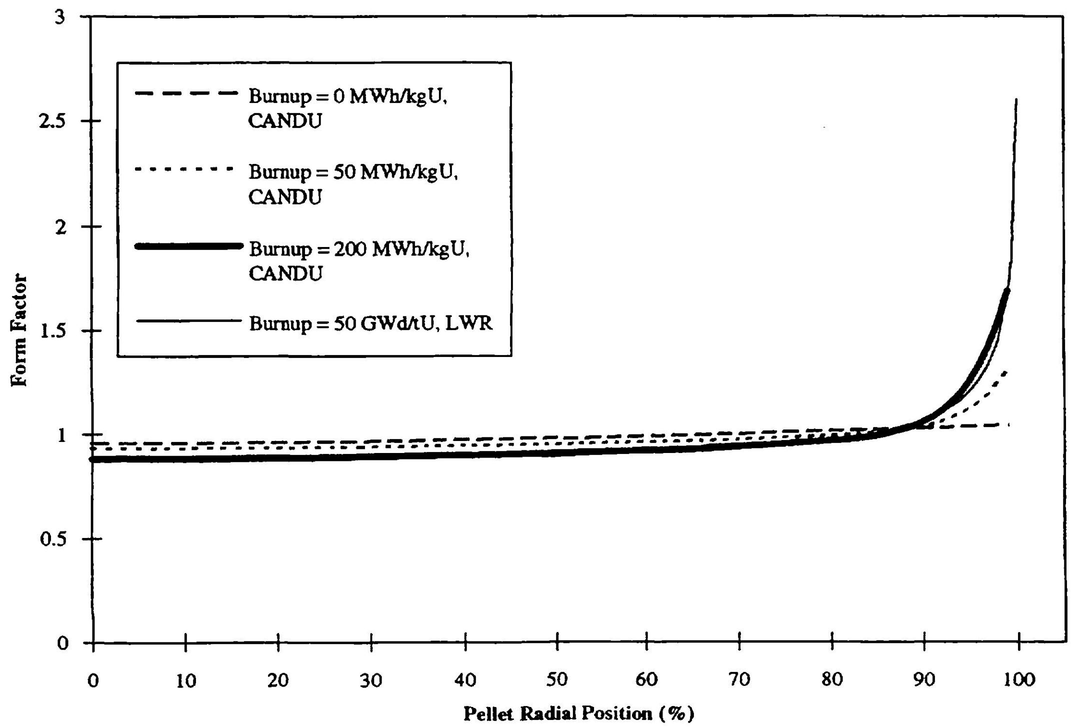


FIG_3_4.XLS Chart 2/9/29/95/12:55 PM

Figure 3
Fuel Self-Heating Rate as a Function of Specific Fission Density

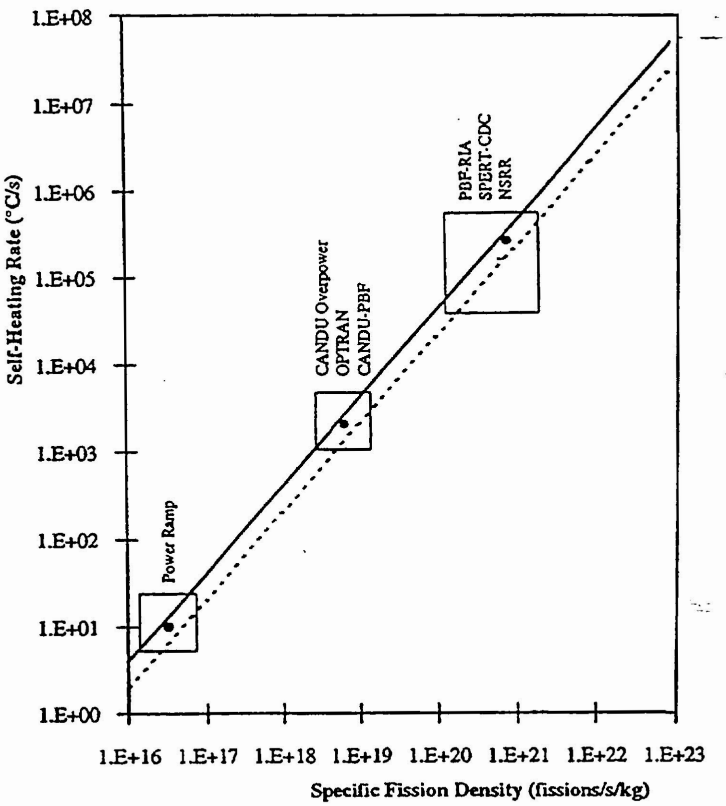


Figure 4
Transient Fuel Radial Temperature Profile
Generated during the Overpower Pulse of a Typical NSRR Test

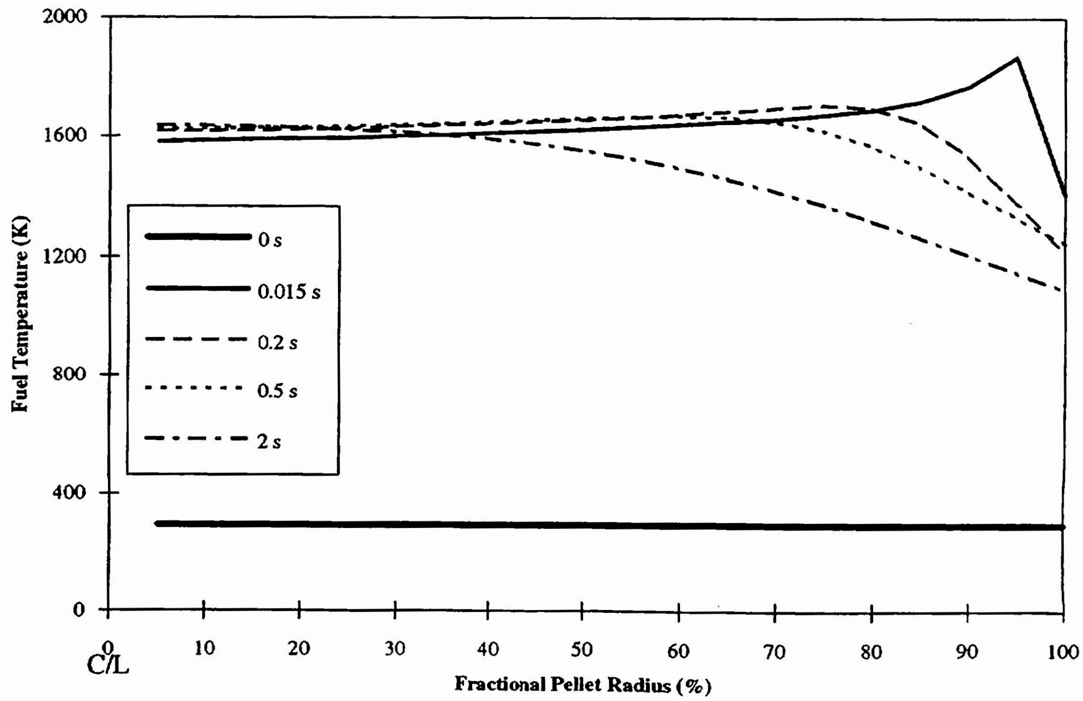
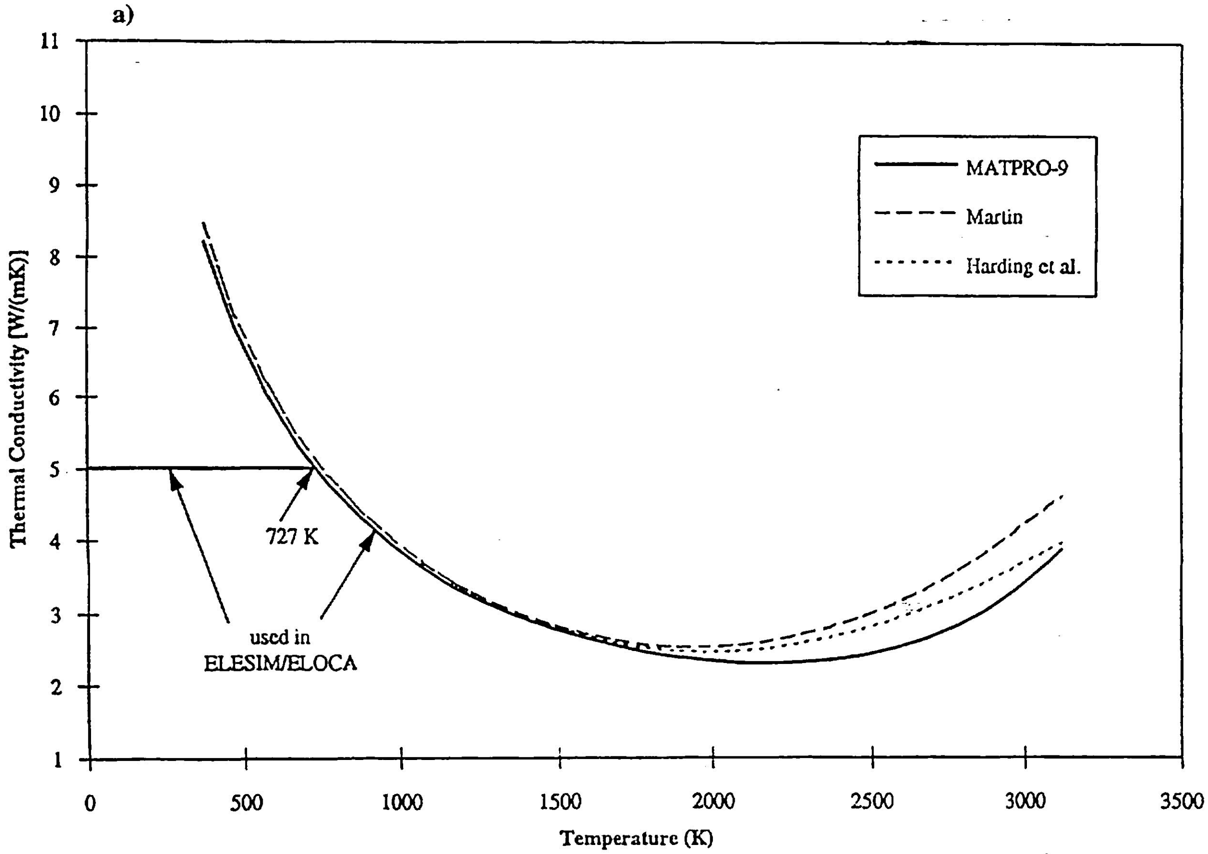


FIG_3-4.XLS Chart 3/9/29/95/1:03 PM

Figure 5
UO_2 Thermal Conductivity as a Function of Fuel Temperature



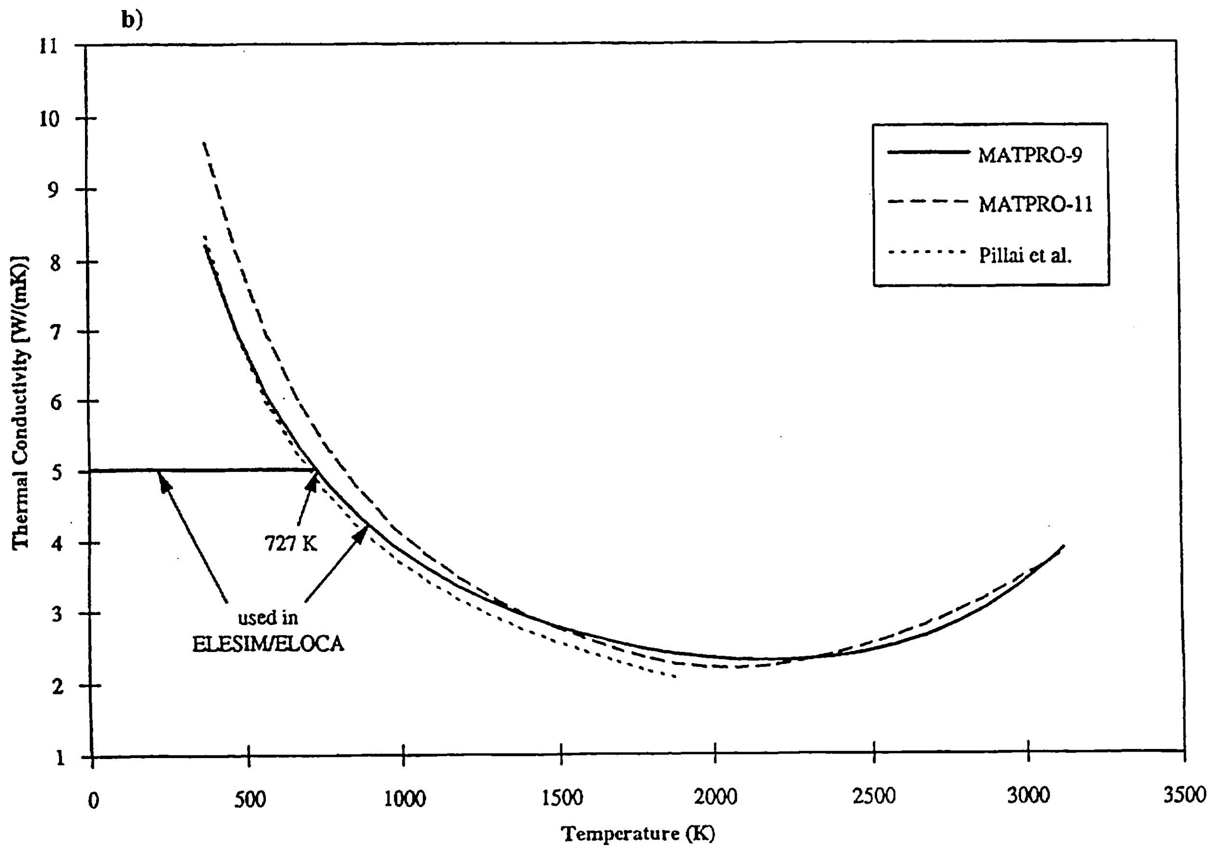


Figure 6
Effect of Burnup on Thermal Conductivity
(from Y. Philipponneau)

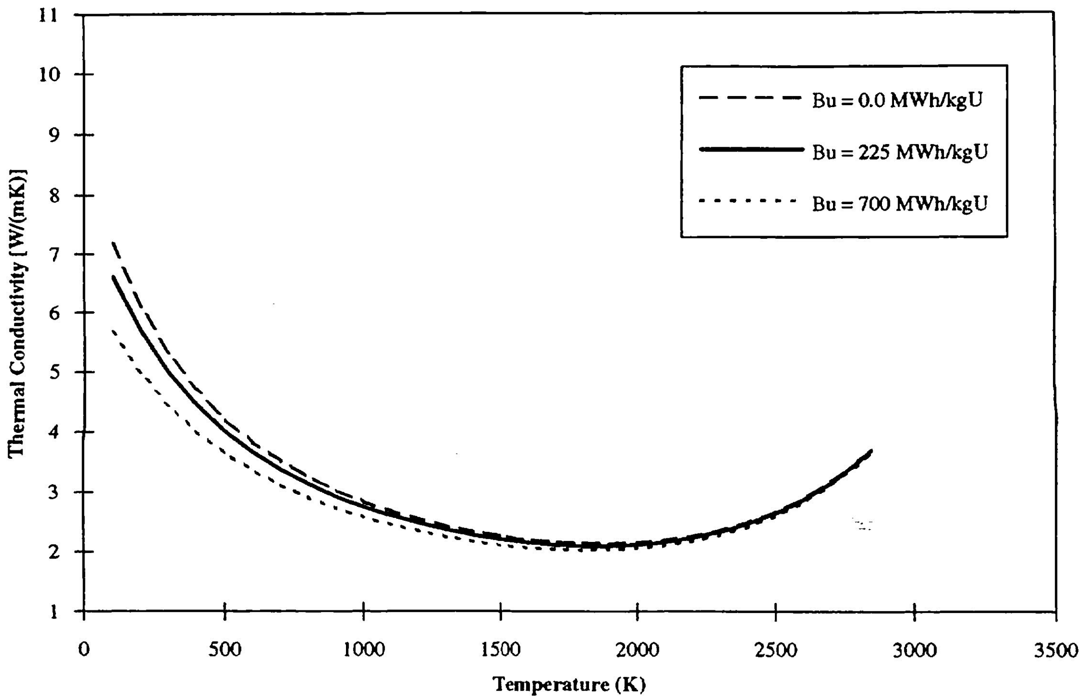


Figure 7
True Stress/True Strain for Zircaloy-4 Fuel Sheathing at Large Stresses