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FUEL SYSTEMS FOR COMPACT FAST SPACE REACTORS

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MASTER

FUEL SYSTEMS FOR COMPACT FAST SPACE REACTORS

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ABSTRACT

About 200 refractory metal clad ceramic fuel pins have been irradiated in thermal reactors under the 1200 K to 1550 K cladding temperature conditions of primary relevance to space reactors. This paper reviews performance with respect to fissile atom density, operating temperatures, fuel swelling, fission gas release, fuel-cladding compatibility, and consequences of failure. It was concluded that UO_2 and UN fuels show approximately equal performance potential and that UC fuel has lesser potential. W/Re alloys have performed quite well as cladding materials, and Ta, Nb, and Mo/Re alloys, in conjunction with W diffusion barriers, show good promise. Significant issues to be addressed in the future include high burnup swelling of UN, effects of UO_2-Li coolant reaction in the event of fuel pin failure, and development of an irradiation performance data base with prototypically configured fuel pins irradiated in a fast neutron flux.

INTRODUCTION

A substantial program was conducted in the 1960's to develop fuel elements for space nuclear reactors. Much of the program emphasized refractory metal clad UN, UC, and UO_2. This paper reviews the state of development of these fuel systems and discusses their technical feasibility issues as they relate to the current interest in space reactors. Previous reviews were provided by Weaver and Scott (1965) and by Gluyas and Watson (1975).

DESIGN CONSIDERATIONS

The fundamental requirements for a space nuclear reactor are relatively low weight and long lifetimes. For the reactor core, this has been translated to a compact liquid metal cooled fast reactor with fuel goal burnups of 3 to 5 at.% and fast neutron fluences of 2 to 3 x 10^{22} n/cm^2. Specific powers are modest by comparison with terrestrial reactors but coolant temperatures are much higher, up to 1500 K versus about 800 K. The high operating temperatures are required to increase thermal efficiency and minimize the size and weight of the heat rejection system. This has a significant effect on the design of the reactor: Li rather than the more conventional Na or NaK must be used as the liquid metal coolant, because of its lower vapor pressure; refractory metals must be used as fuel pin cladding, rather than more conventional ferrous alloys; and fuel melting temperatures must be high, forcing use of a ceramic fuel material. In selecting a fuel system there are several considerations that must be addressed including: fissile atom density, fuel operating temperature, fuel swelling, fission gas release, fuel-cladding compatibility, and the consequences of cladding failure.

IRRADIATION DATA

In reviewing these fuel selection issues, the relevance of data is largely controlled by the operating environment, namely the linear heat rate (or fuel temperature), fuel burnup, cladding temperature and fast neutron fluence. Irradiation data were compiled from multiple reports, primarily from NASA, ORNL, and BMI, Mayer et al. (1973) provide a good source of references. Earlier work on a Pratt and Whitney/ORNL Program was summarized by Weaver et al. (1969). The primary irradiation data base of interest, defined here as having a cladding temperature range of 1200 K to 1550 K, is shown in Figure 1. This figure plots the cladding temperature and burnup for each fuel pin. Fuel pin failures identified on this plot were determined primarily from visual examinations after irradiation. The data shown here represent 124 UN, 61 UC, and 9 UO_2 fuel pins. All these data suffer the nonprototypic feature of having been irradiated in a thermal rather than a fast reactor; thus, the effects of fast neutrons on cladding properties were not simulated, and the fission rate distribution across the fuel diameter had a marked depression in the center of the fuel. Additionally, in many of the early experiments the length-to-diameter (l/d) ratios of the fuel columns were quite low. This complicated the heat transfer analysis and also magnified the effects of power peaking at the ends of the fuel columns. To obtain a relatively reliable pedigreed set of data with respect to operating power and temperatures, we discarded data with an l/d less than 4, resulting in the data field shown in Figure 2. The irradiation data fields shown in Figures 1 and 2 supplemented by reviews of relevant laboratory and theoretical studies form the bases for the discussion of fuel selection issues which follows.

FISSILE ATOM DENSITY

The high fissile atom densities of theoretically dense UN and UC make these materials good candidates for space reactor fuels. UO_2, which has a 30% lower theoretical fissile atom density, is less attractive, and cermet fuels with their rather dilute fissile material density are probably unacceptable due to their large core volume requirements. Part of the difference between UO_2 and the higher fissile density fuels is usually offset by use of somewhat higher smear density for UO_2 fuel.

FUEL OPERATING TEMPERATURES

Figure 3 displays typical operating temperatures of the three fuels at a linear heat rate of 200 W/cm (6 kW/ft) and with 1500 K cladding temperature. UN, UC, and UO_2 under such conditions operate at 54%, 60% and 67% of their respective melting temperatures. It should be noted that these temperatures are relatively independent of fuel pin diameter.

FUEL SWELLING

Figure 4 compares estimates of fuel swelling as a function of temperature at a burnup of 1 at.% for UN, UO_2, and UC based on the work of Chubb, Storhok, and Keller (1973). The data, largely "unconstrained" show considerable scatter reflecting experimental difficulties and the effects of compositional variables. Fuel swelling, and the related gas release are dependent on stoichiometry of UC and UN and to a lesser degree, the O_2 content of UN. Similar experiments reported by Zimmermann (1978) have been conducted to higher burnups with UO_2 and show (Figure 5) that the swelling rate decreases with burnup. Comparable high burnup data were not found for UN and UC. However, limited work reported by Zimmermann and Dienst (1974) with high oxygen content UN indicates the swelling rate may increase with burnup. Considering these uncertainties UO_2 and UN, with low oxygen content are expected to exhibit comparable fuel swelling and UC, significantly greater swelling under space reactor conditions.

Although UO_2 has a much higher creep rate than UN and UC at temperatures below 1100 K, the creep rate of all three fuels increases very significantly and may be comparable above 1400 K. Thus, high unconstrained swelling at temperatures above, say, 1600 K is of little technical significance because it is apparently limited in fuel pins by cladding restraint or restraint of the colder fuel regions-Dienst et al. (1979).

In summary fuel swelling considerations favor UO_2 over UN because the UO_2 operates at higher temperatures and is thus more plastic, leading to lower cladding stresses. UC is expected to exhibit the greatest fuel swelling.

FISSION GAS RELEASE

Fission gases released from the fuel form a major loading on the fuel pin cladding. Figure 6 compares the fission gas releases at 4 at.% burnup in the operating temperature range of interest. This was derived from our compilation plus the work of Dollins and Nichols (1977), Weaver and Scott (1965), and Grando et al. (1971). UC and UO_2 are shown to have comparable fission gas release. UN demonstrates significantly less fission gas release; thus, unless fuel pins are vented, fission gas release considerations favor UN.

FUEL-CLADDING COMPATIBILITY

Figure 7 summarizes the understanding of fuel-cladding chemical compatibility. This came from review of the earlier summary by Weaver and Scott (1965), from the irradiation data base summarized for this paper, and from review of several laboratory experiments.

There are very limited relevant irradiation data for UO_2. These data in conjunction with laboratory experiments show UO_2 to be compatible with W/Re alloys for temperatures well in excess of 1500 K. Ta and Cb alloys were found to be incompatible with UO_2 for temperatures above 1250 K but have been used successfully with a W diffusion barrier for temperatures in excess of 1300 K. Mo/Re alloys have not been used under relevant reactor experiment conditions. Laboratory studies and in-reactor fuel pin irradiations using other Mo alloys indicate compatibility should be adequate at temperatures in excess of 1500 K; however, O_2 and Mo transport issues remain. W diffusion barriers may thus also be required with Mo alloys. Compatibility considerations for UO_2 thus indicate preference in decreasing order for W/Re, Mo/Re, and Cb or Ta alloys, with use of W diffusion barriers probably required for the latter three.

Greater, relevant irradiation data exist for UN (Figure 8). Again, W/Re alloys have had unqualified success at cladding temperatures above 1500 K. Ta and Cb alloys clearly require a W barrier above 1250 K. Compatibility of Mo/Re alloys is in doubt above 1300 K.

Relevant irradiation data for UC are limited to W/Re and Cb alloys (Figure 9). These plus laboratory studies have indicated that UC, in particular hyperstoichiometric UC, is incompatible with all refractory metals other than W alloys above 1300 K. W/Re alloys have been found to be compatible with uranium carbide to at least 1500 K.

Low ductilities have been observed in many refractory alloys, notably T-111 and to a lesser extent Cb-1Zr after fuel pin irradiation. This has been primarily attributed to hydrogen embrittlement from handling during post-irradiation examination or to poor fabrication practices but other sources have been postulated, such as (n, p) reaction, experimental anomalies, and carbon pickup.

CONSEQUENCES OF FAILURE

Three issues have been considered relative to the consequences of fuel pin failure. These are: fuel-Li reactions, the potential for fuel dissociation, and fuel-refractory metal reactions resulting from the formation of a lithium diffusion path.

Thermochemical data at temperatures ranging from 1000 K to 2000 K indicate that reactions between UN and Li are not thermodynamically feasible. Compatibility experiments have been conducted with UN pellets immersed in a pool of Li and a defected UN fuel pin in a Li loop-Weaver and Scott (1965) Gluyas (1975). Results from these experiments have shown good compatibility of UN with Li although some pellet spalling and washout were reported. Stoichiometric UC is also compatible with Li; however, excess carbon is readily soluble in liquid Li and will react with cladding and structural materials. A significant potential problem has been identified for failed UO_2 fuel pins. Laboratory experiments have shown UO_2 to react readily with Li at temperatures of 1400 K to 1500 K, Kangilaski et al. (1965). The reaction forms U_3O_8 and free U that may form low melting, ∿1300 K, eutectics with cladding and structural materials. Thus the use of UO_2 fuel pins in Li systems requires either a very reliable fuel pin design or accommodation of the effects of the UO_2-Li reaction.

Although all three fuels will disassociate at low pressures, UN has the highest vapor pressure. The primary concern is formation of free U which could react with cladding and structural materials and N_2 which also might react with cladding and structural materials. This does not appear to be a problem in the temperature range of interest since the equilibrium nitrogen pressure for the reaction UN(s) = U(l) + 1/2 N_2 is only $\sim 10^{-10}$ atmosphere-Kubaschewski, Evans, and Alcock (1967); Inouye and Leitnaker (1968); Oetting and Leitnaker (1972).

If Li enters a failed fuel pin, it will form a diffusion path between the fuel and cladding. When this happens, at some point the fuel will react with the cladding by diffusion of excess interstitial atoms through the Li. For example, nitriding has been reported of T-111 cladding in a defected UN pin in a Li loop Gluyas and Watson (1975).

SUMMARY

Irradiation of about 200 fuel pins at prototypic space reactor temperatures indicates that refractory metal clad ceramic fuel pins can perform adequately. However, there are few data with both prototypic temperatures and prototypic configurations. Furthermore, all relevant irradiations have been conducted in thermal reactors whereas the more likely application is a fast reactor. Thus the achievable operating temperature/lifetime envelope of such fuel pins cannot be defined.

UN and UO_2 show approximately equal performance potential. UC appears of less promise since it is indicated to have the highest fuel swelling, the least compatibility with refractory metal claddings, and relatively high release of fission gases.

Fuel swelling appears to favor UO_2 since the low burnup unconstrained swelling rates of UO_2 and UN are similar, but UO_2 is more plastic at its operating temperature, and the UO_2 swelling rate appears to decrease with burnup. High burnup swelling of UN is not well defined but the swelling rate may increase with burnup. Fission gas release considerations favor UN such that use of UO_2 might require stronger cladding, larger fission gas plena or venting of fission gases.

W/Re cladding appears chemically compatible with both UO_2 and UN to at least 1500 K. Mo/Re cladding has not been used in irradiation tests but is expected based on performance of other Mo alloys to be compatible with UO_2 to 1500 K; however, a W diffusion barrier may be required. Ta and Cb alloys when used with a W diffusion barrier appear usable with UO_2 and UN to greater than 1300 K.

There is a potential problem for failed UO_2 fuel pins, namely reaction with Li to form free U. This may result in low melting eutectics with refractory metals and thus requires either a very reliable fuel pin design or accommodation of the effects of this reaction.

Significant issues to be addressed in the future include high burnup swelling of UN, effects of UO_2-Li coolant reaction in the event of a fuel pin failure, and development of an irradiation performance data base with prototypically configured fuel pins irradiated in a fast neutron flux.

ACKNOWLEDGMENTS

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FIGURE CAPTIONS

1. Fuel Pin Irradiation Data Base.

2. Irradiation Data Base with Fuel Column Length: Diameter >4.

3. Fuel Pin Temperatures at 200 W/cm.

4. Fuel Swelling at 1 at.% Burnup.

5. Burnup Dependence of Oxide Fuel Swelling at 1750 K.

6. Fission Gas Release at 4 at.% Burnup.

7. Fuel-Cladding Compatibility Summary.

8. Postirradiation Observations of Cladding Reactions with UN.

9. Postirradiation Observations of Cladding Reactions with UC.

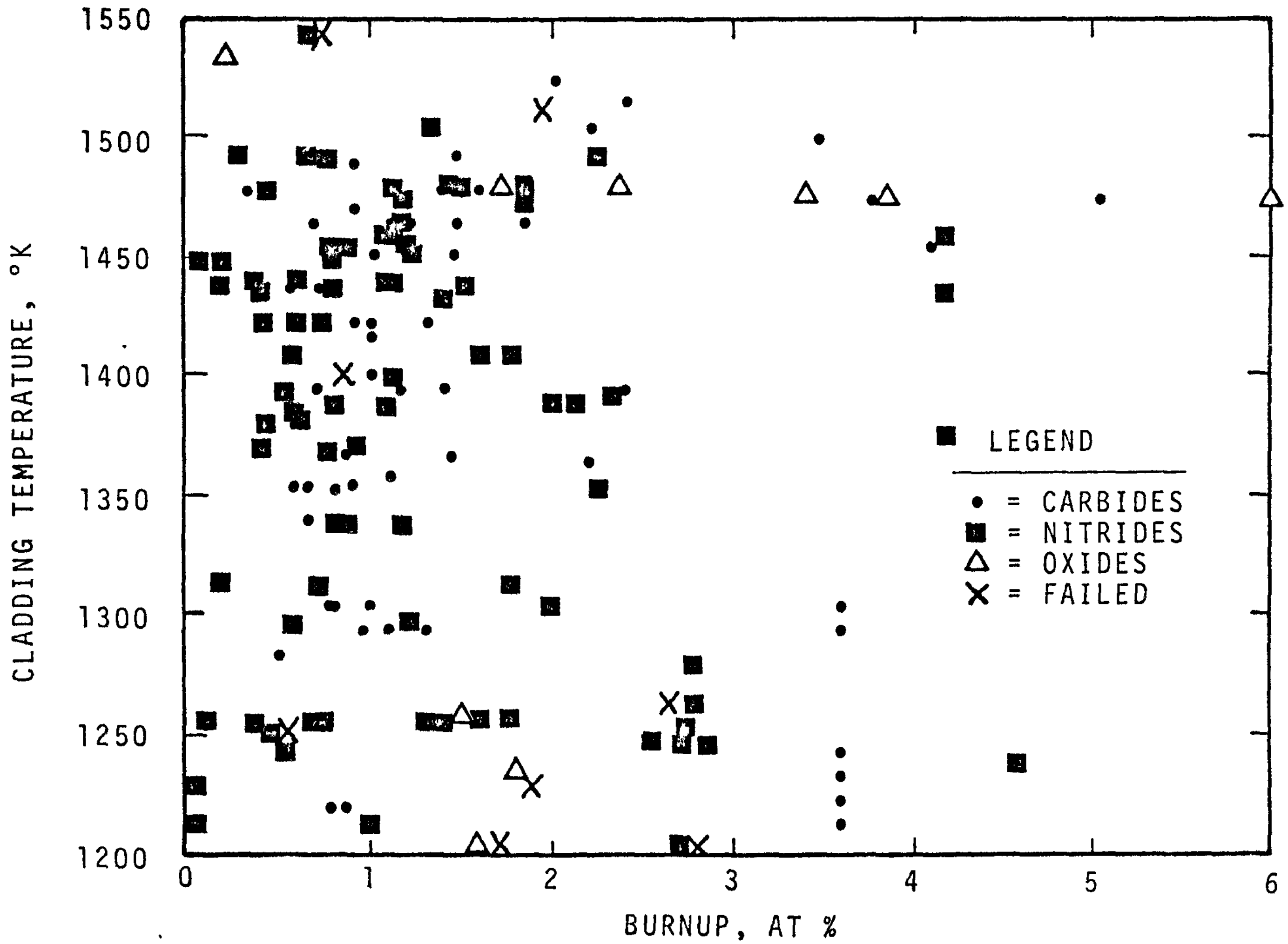
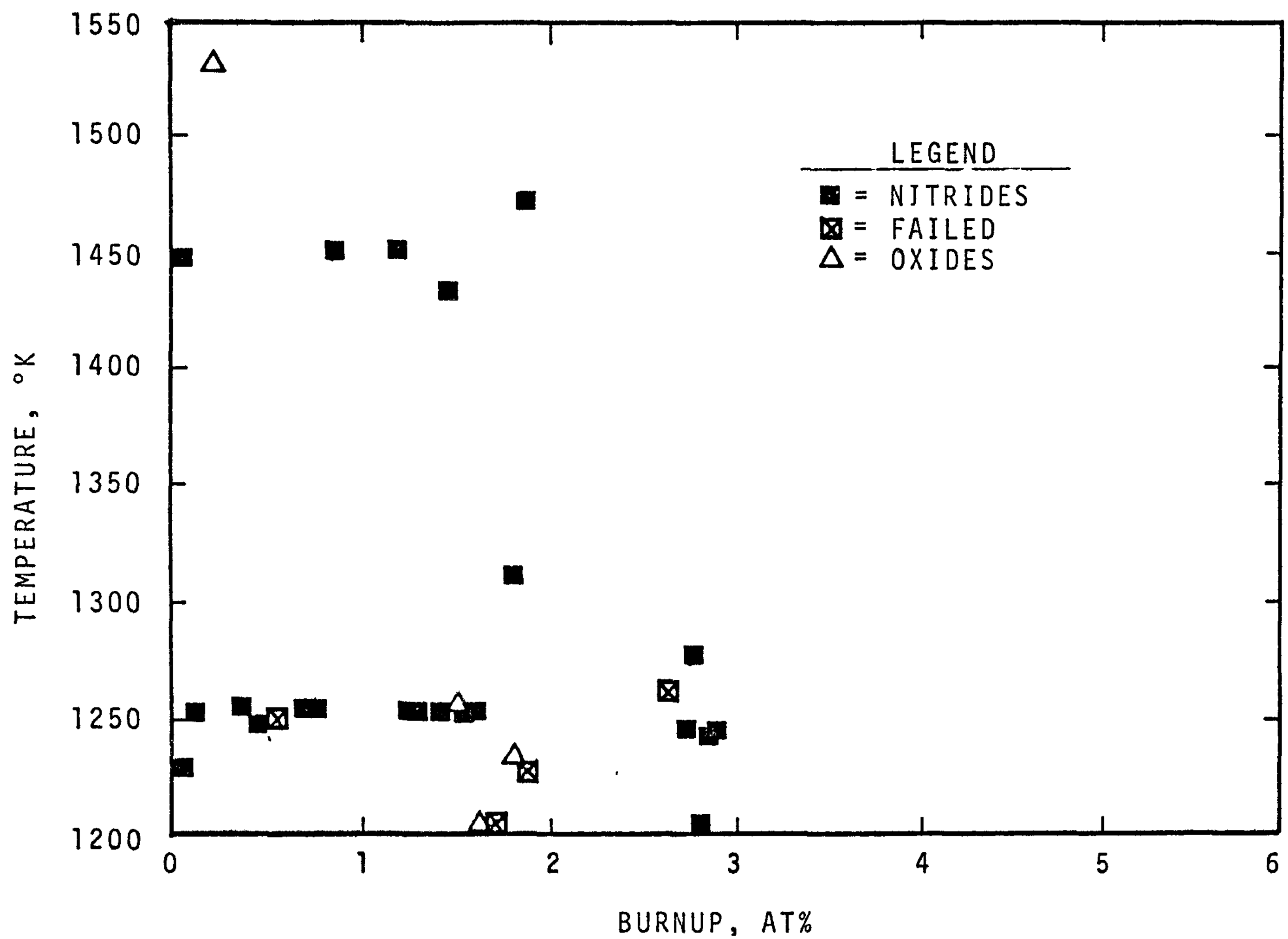


FIGURE 1. Fuel Pin Irradiation Data Base.



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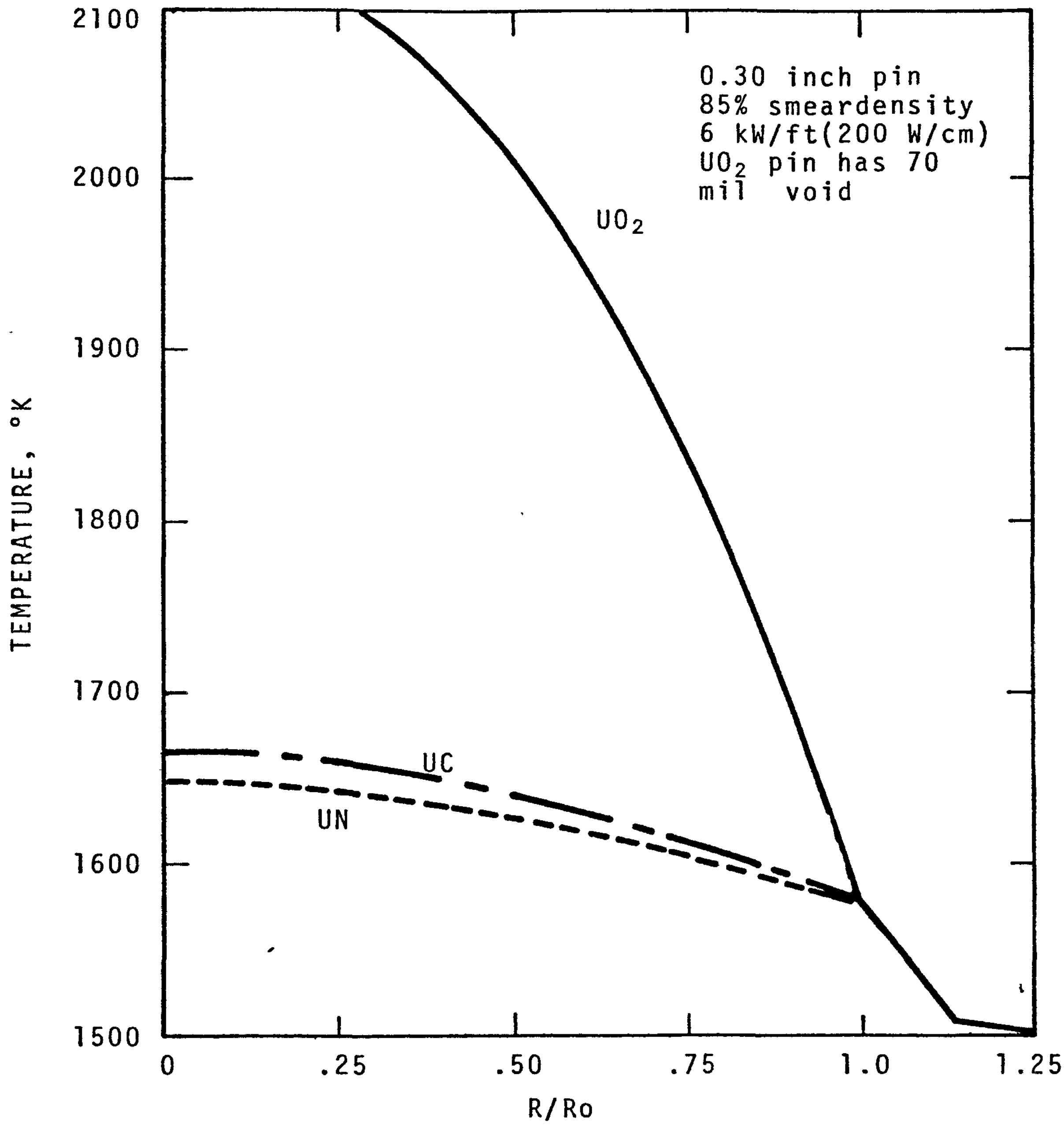


FIGURE 3. Fuel Pin Temperatures at 200 W/cm.

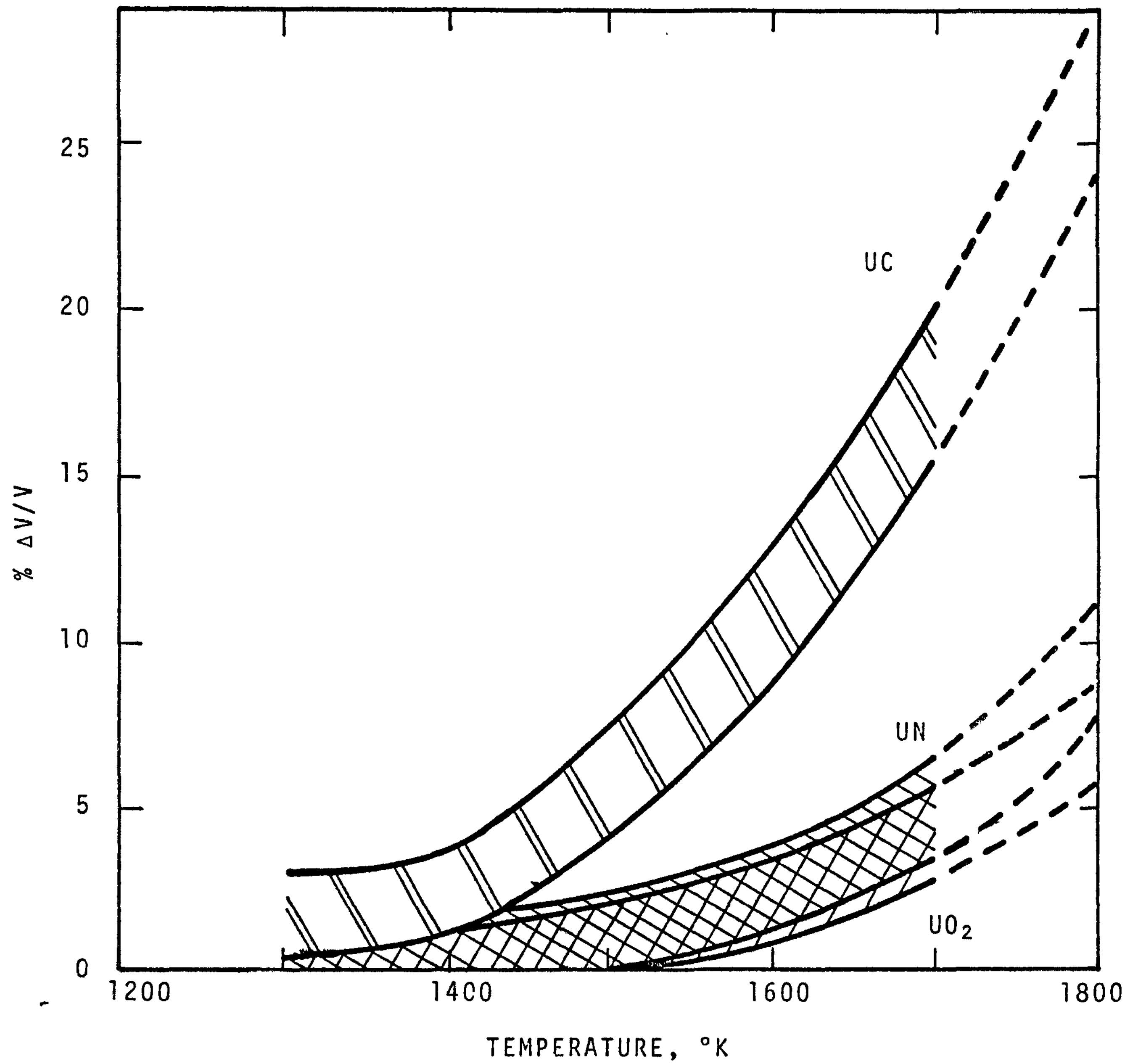
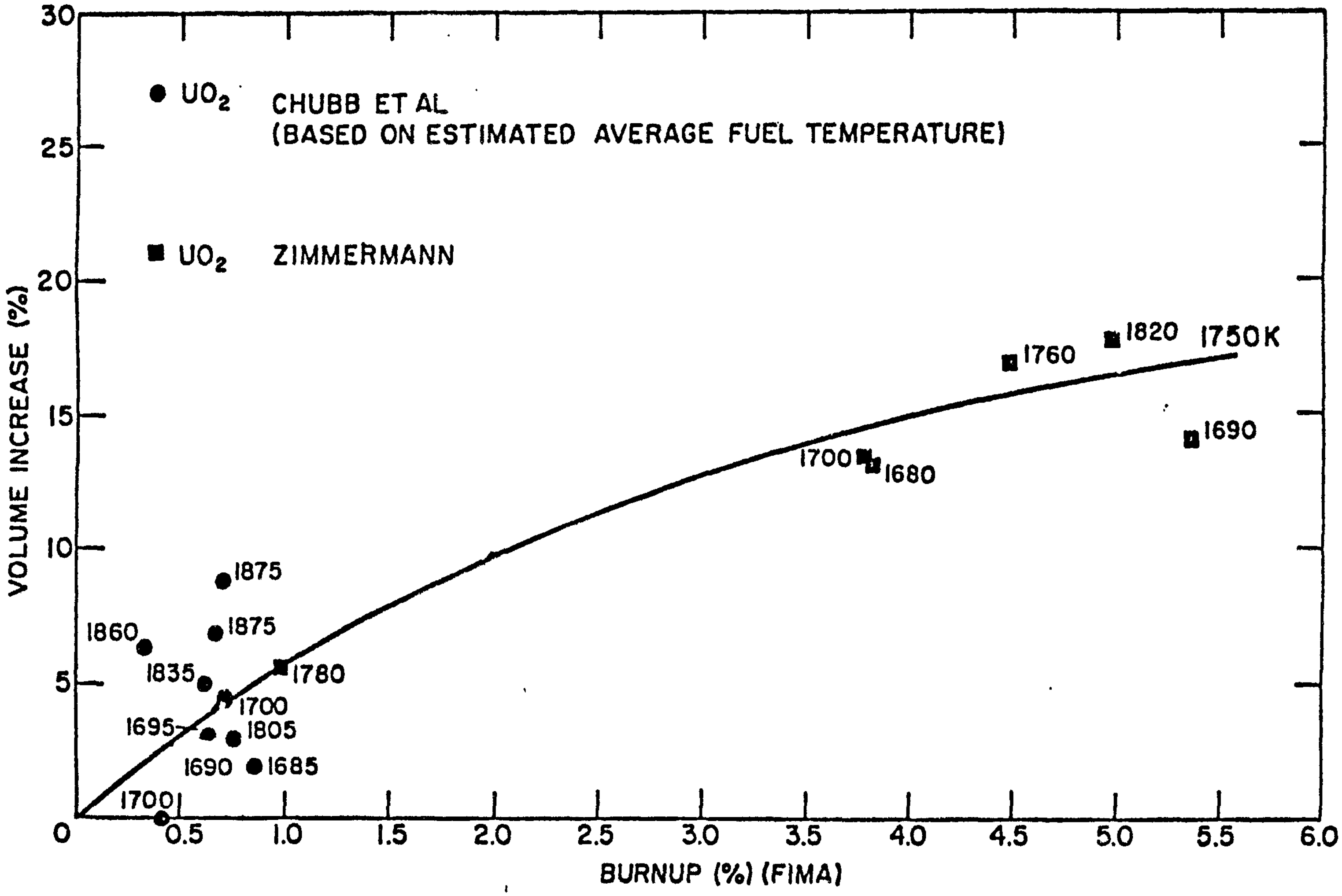


FIGURE 4. Fuel Swelling at 1 Atom % burnup.

BURNUP DEPENDENCE OF OXIDE FUEL SWELLING AT 1750°K



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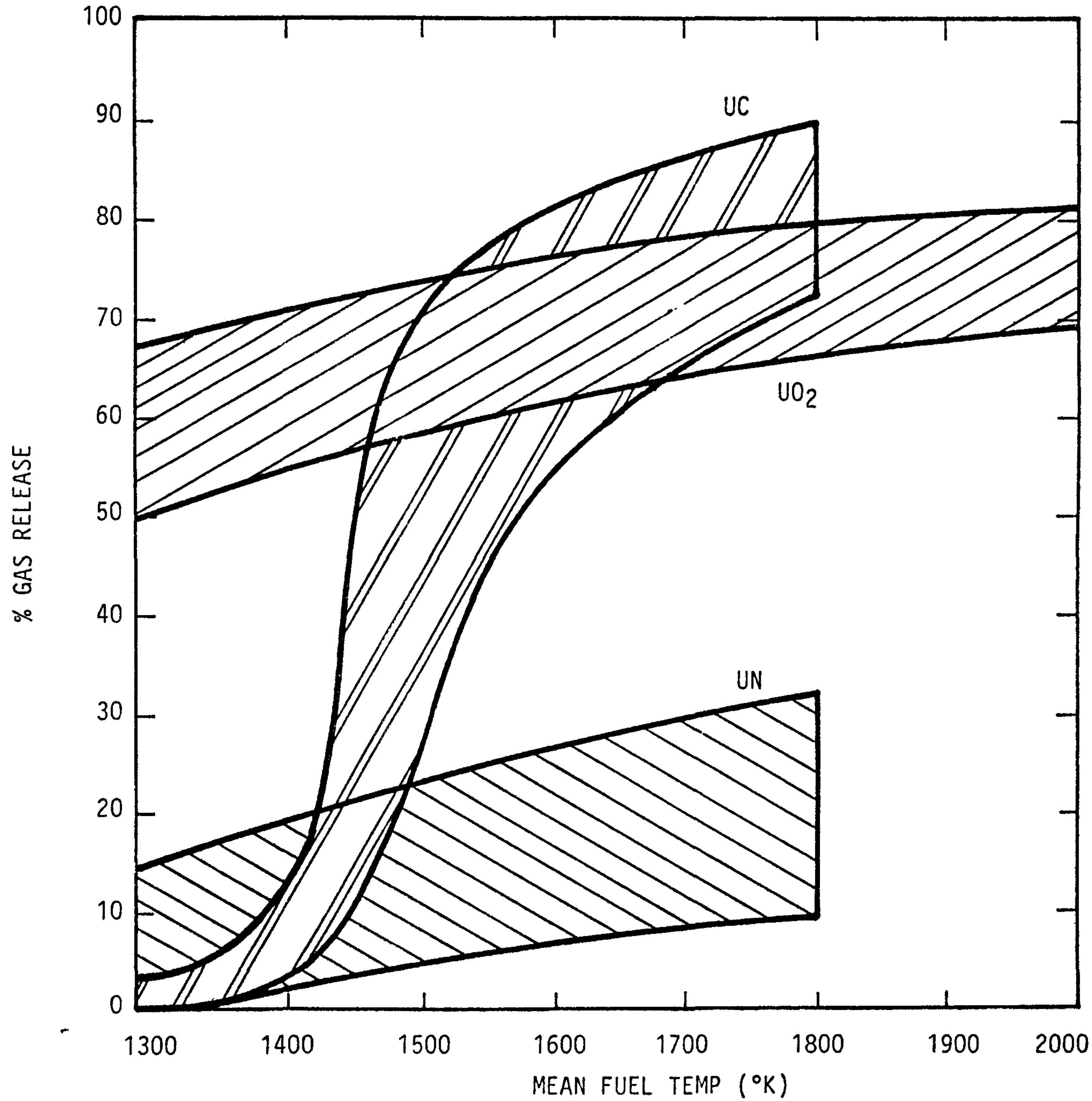

FIGURE 6. Fission Gas Release at 4 At.% Burnup.

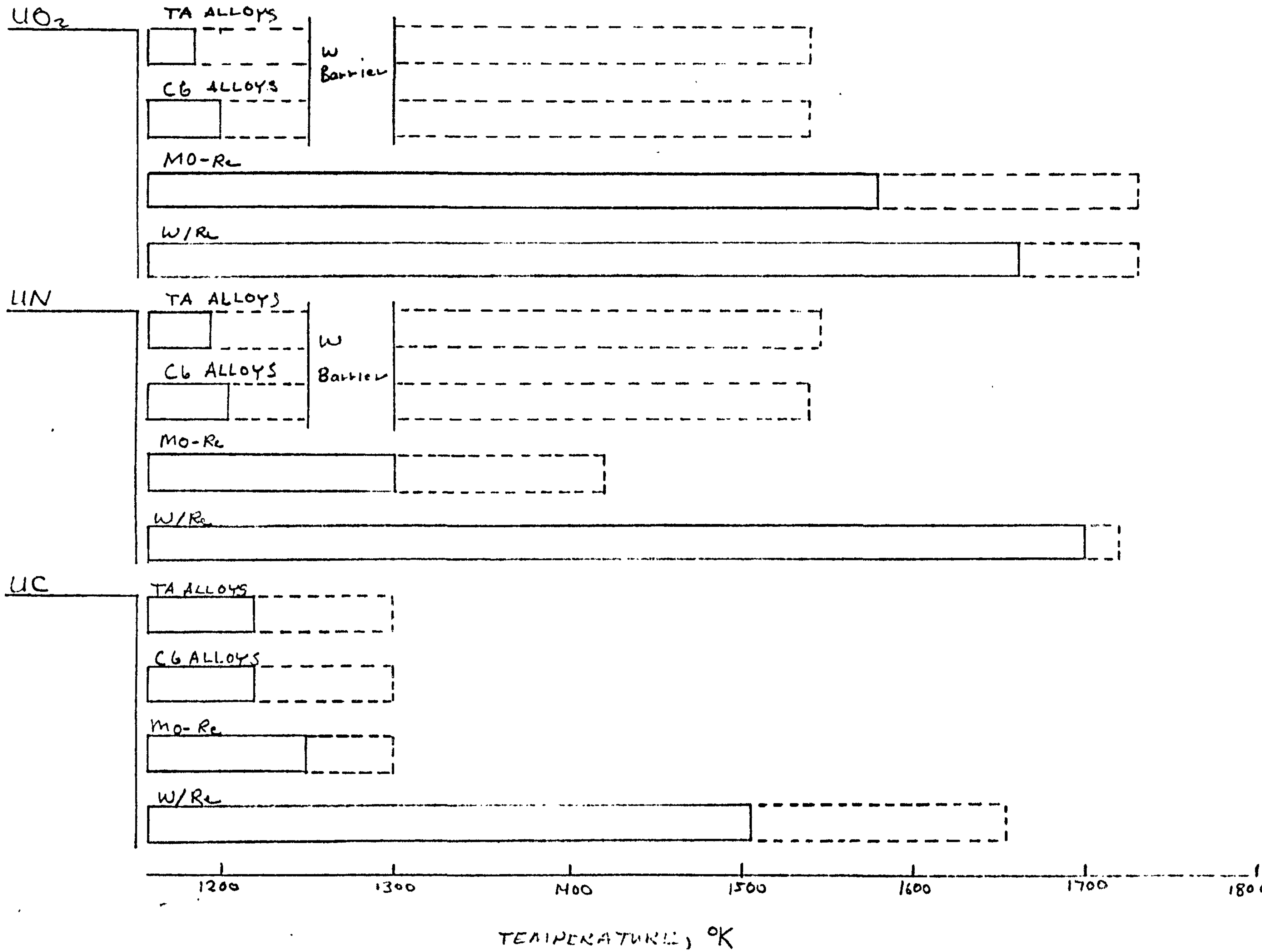

FIGURE 7. Fuel-Cladding Compatibility Summary.

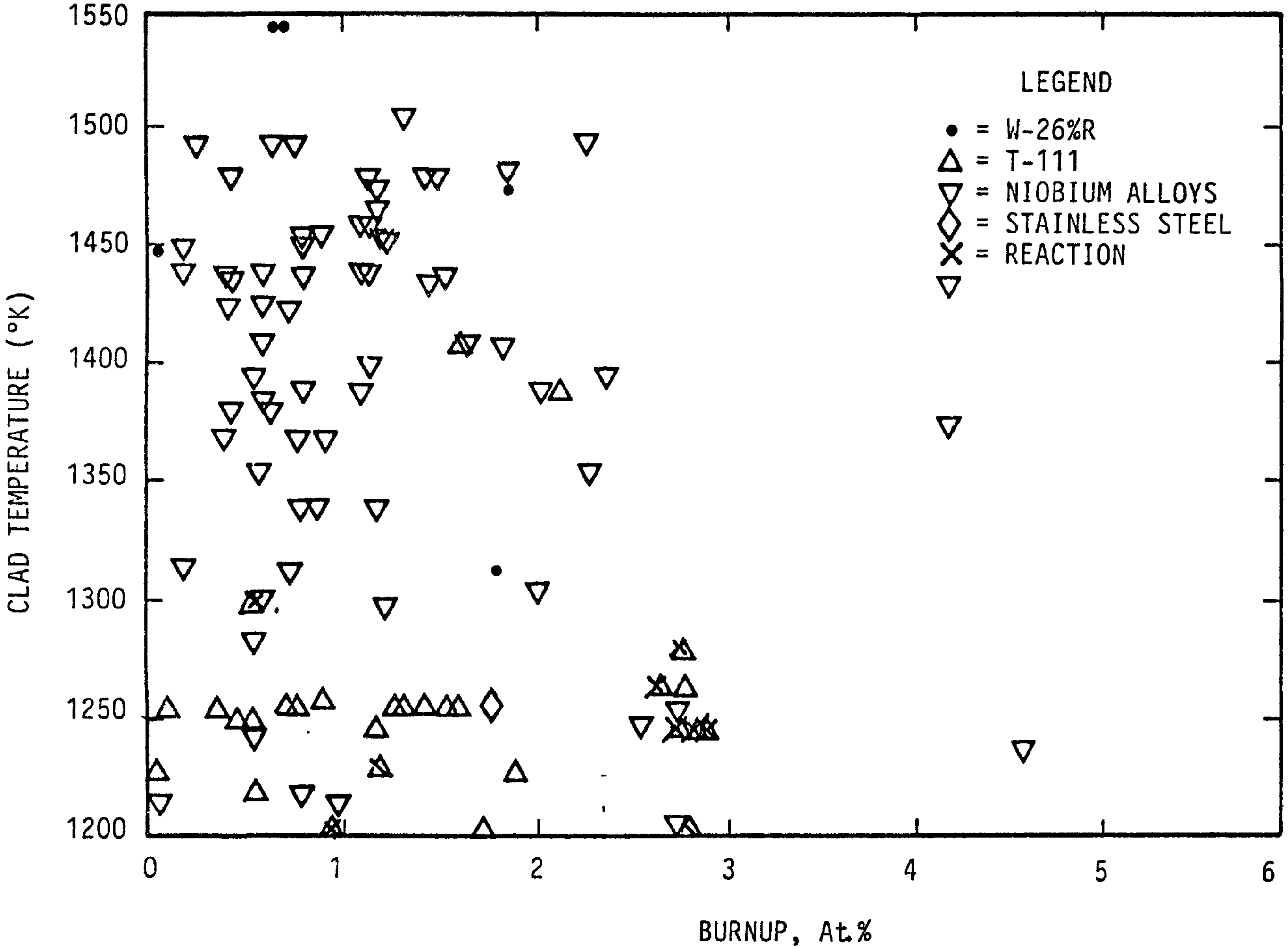

FIGURE 8. Postirradiation Observations of Cladding Reactions with UN.

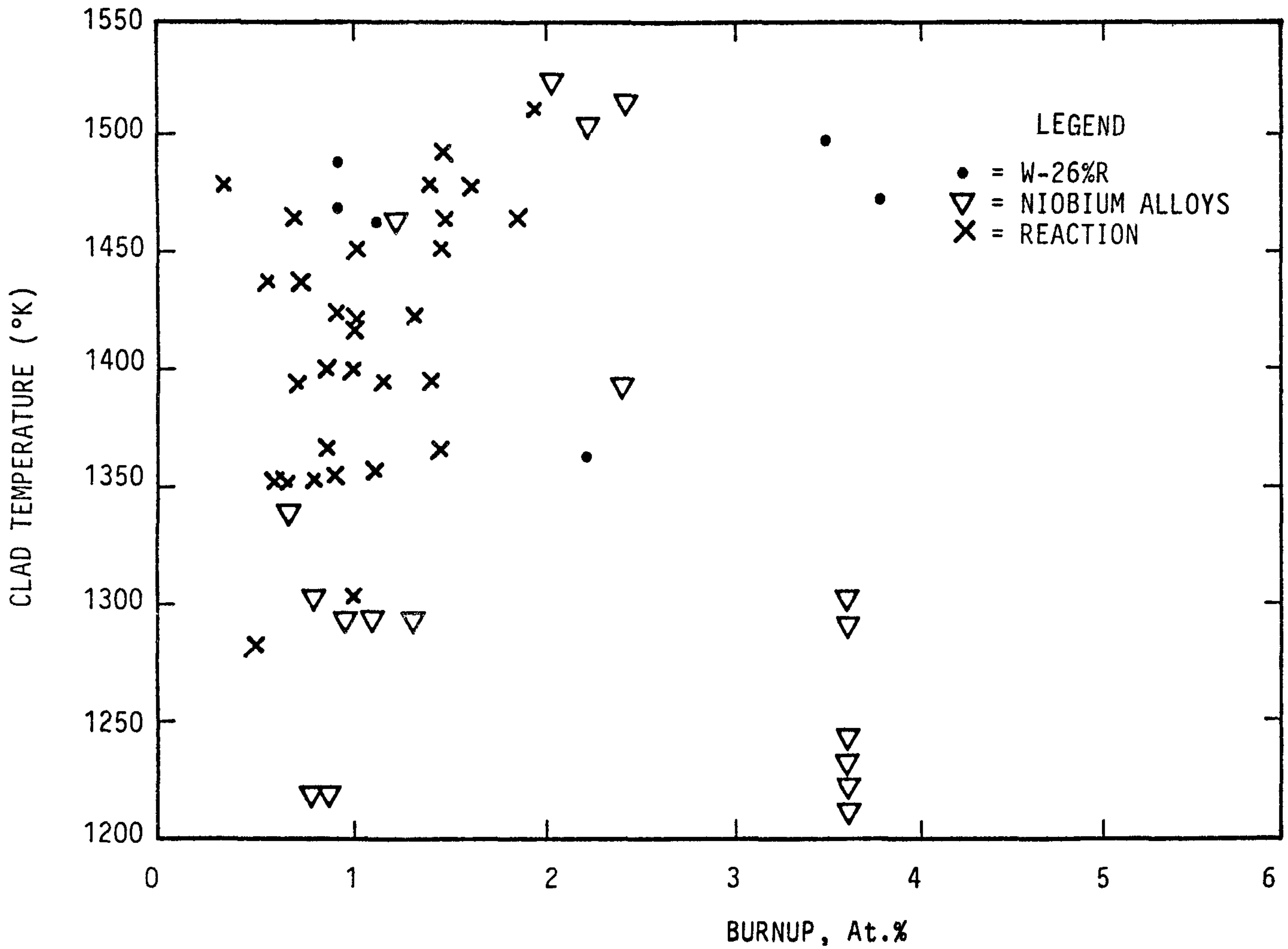

FIGURE 9. Postirradiation Observations of Cladding Reactions with UC.

SUMMARY

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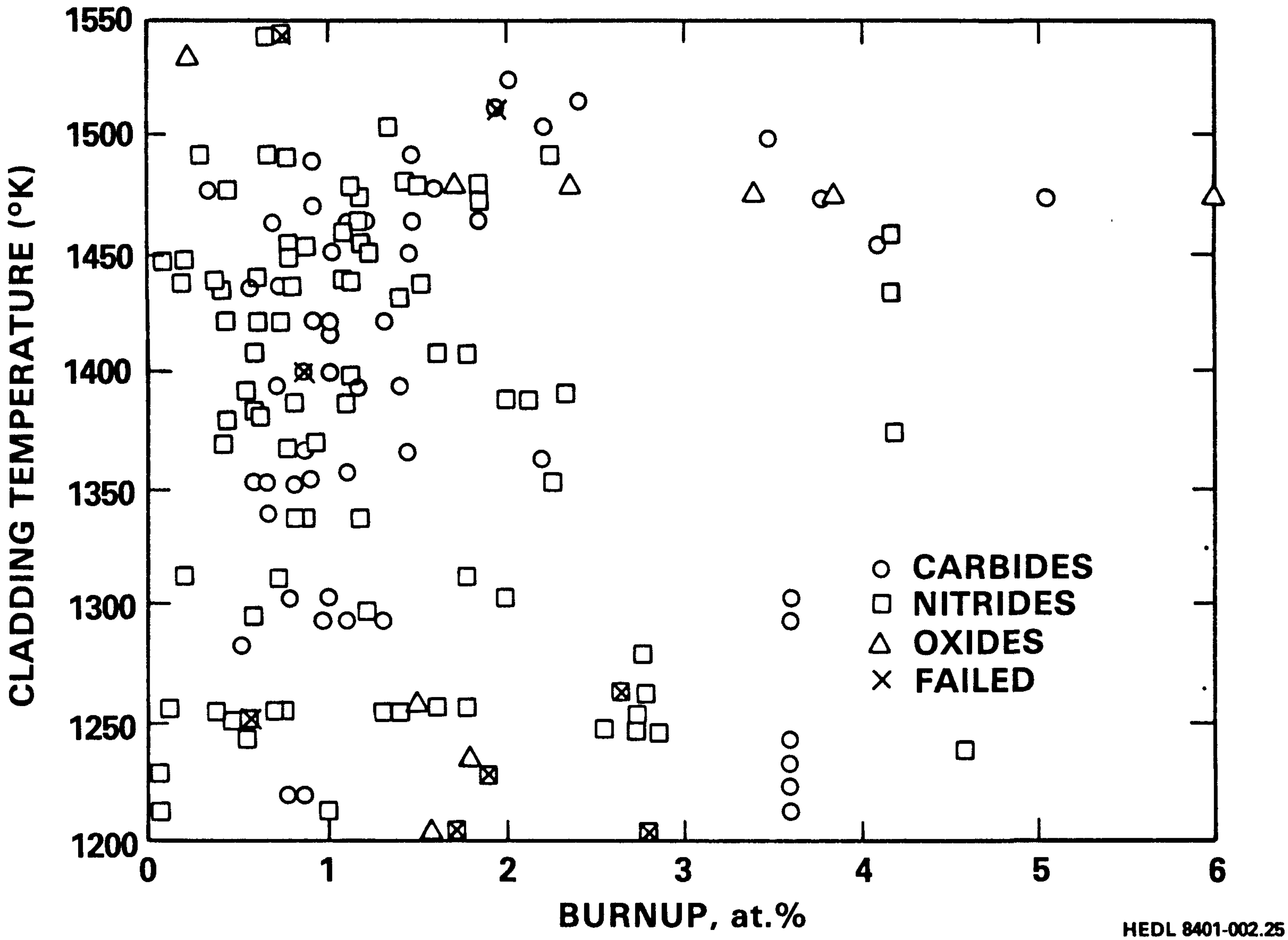

FIGURE 1. FUEL PIN IRRADIATION DATA BASE.

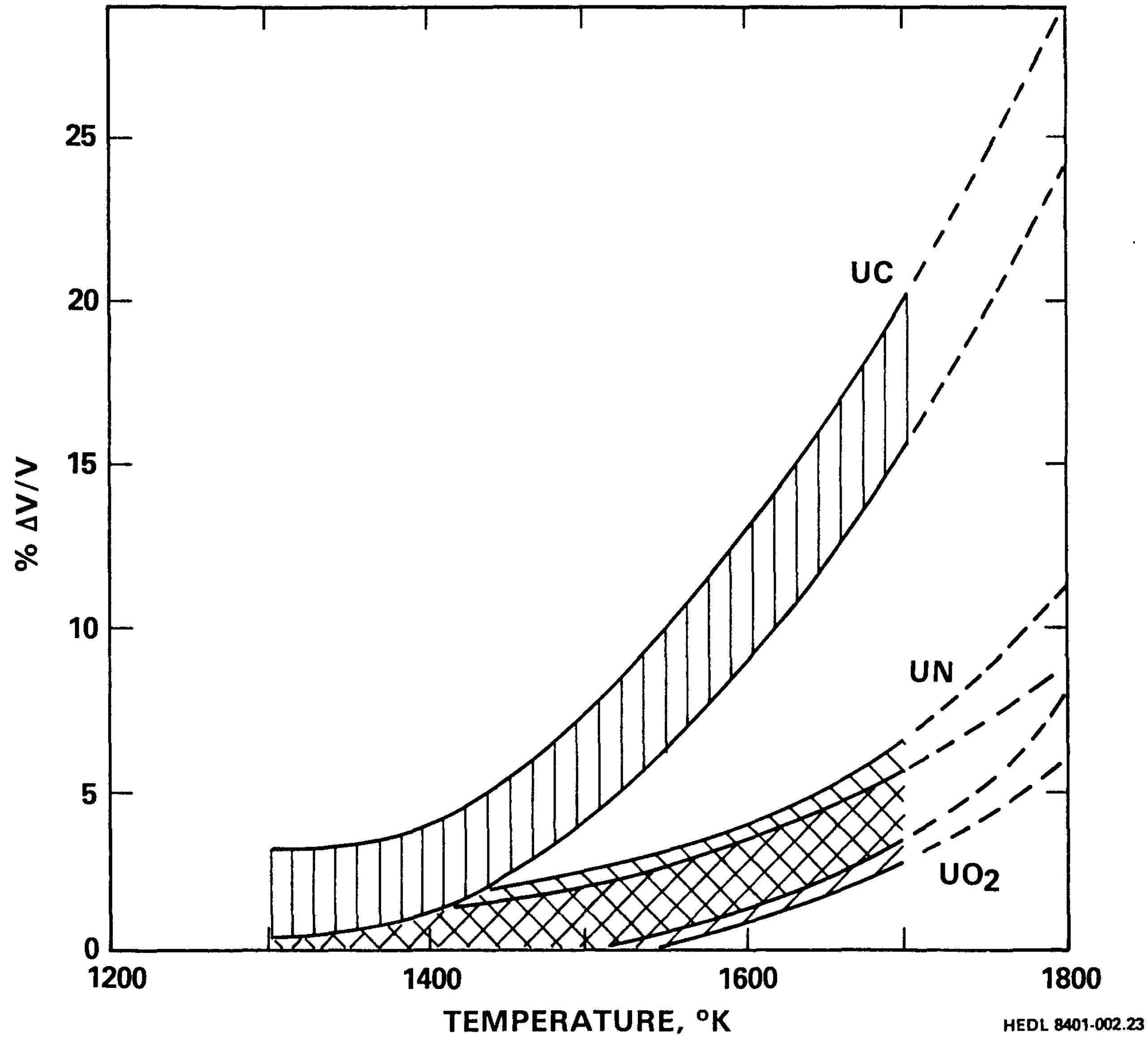

FIGURE 2. FUEL SWELLING AT 1 ATOM % BURNUP.