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COMPARISON OF REACTOR FUELS FOR HIGH TEMPERATURE APPLICATIONS

S. C. Weaver and J. L. Scott

Abstract

Continuing interest by the Atomic Energy Commission and the National Aeronautics and Space Administration in high energy, compact nuclear reactors for space application has precipitated a study of the relative advantages and disadvantages of three ceramic fuel materials: UN, UO_2, and UC. In addition, UO_2 as a dispersion in a refractory metal was considered. The fuels were compared at the design conditions for SNAP-50 [2 Mw(th)] and the Medium Power Reactor Experiment [1 Mw(th)] which have operating clad temperatures of 2200°F (1200°C) and 1500°F (850°C), respectively.

The results of the investigation show that UN exhibits much less radiation damage than does UC for SNAP-50 irradiation conditions. The use of UO_2 was precluded from the Pratt and Whitney SNAP-50 reactors because of its incompatibility with lithium. However, for an advanced refractory metal reactor cooled by potassium, UO_2 could be an attractive fuel candidate. For the less severe irradiation environment of the MPRE, no important differences could be found between UN and UO_2. The use of UC as a fuel in either reactor is limited because of potential excessive fuel swelling and fission-gas release.

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COMPARISON OF REACTOR FUELS FOR HIGH TEMPERATURE APPLICATIONS

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Introduction

Oak Ridge National Laboratory is presently working on problems associated with the development of two types of reactor designed for space applications, the Medium Power Reactor Experiment and SNAP-50. These are to be compact, light weight reactors with a high degree of reliability for one to five years of operation. This study was initiated to evaluate the thermal and irradiation properties of three fuel materials, UN, UO_2, and UC, believed to be good candidates for these two reactor designs. The design requirements are listed in Table 1.[1-3]

The MPRE design by Oak Ridge National Laboratory and four reactor reference designs prepared by Pratt and Whitney Aircraft for the SNAP-50 application are summarized in Table 1. The MPRE is a single loop system of boiling potassium which eliminates many accessory parts found in conventional, multi-loop, heat exchanger designs. The SNAP-50 design is a conventional liquid-metal, multi-loop system employing lithium as the primary coolant. The first two reference designs shown in the table were based on early preliminary estimates of the irradiation and thermal stability of UC under reactor conditions. As irradiation test data became available it became evident that UN is superior to UC and neither could withstand the conditions assumed initially; hence the reactor

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[1]Pratt and Whitney Aircraft—CANEL, "SNAP-50/SPUR Program Engineering Progress Report, April 1, 1964, to June 30, 1964," PWAC-642, p. 82, July 30, 1964. CONFIDENTIAL-RESTRICTED DATA.

[2]Pratt and Whitney Aircraft—CANEL, "SNAP-50/SPUR Program Engineering Progress Report, October 1, 1963, to December 31, 1963," PWAC-634, p. 68, February 28, 1964. CONFIDENTIAL-RESTRICTED DATA.

[3]S. J. Ditto et al., "MPRE Safety Analysis Report," USAEC Report ORNL 64-6-46, Oak Ridge National Laboratory, April 1965. CONFIDENTIAL-RESTRICTED DATA.

Table 1. Reactor Design Parameters for P and W SNAP-50 and MPRE[a]

	SNAP-50 Reference Design 1	SNAP-50 Reference Design 2	SNAP-50 Reference Design 3	SNAP-50 Reference Design 4	MPRE
Description					
Power (Mwt)	8	8	2	2	1
Fuel Material	UC	UC	UN	UN	UO_2
Blanket Materials	BeO	BeO	BeO	BeO	BeO
Cladding	Cb-1 Zr	Cb-1 Zr	Cb-1 Zr	Cb-1 Zr	316 SS
Coolant	Li	Li	Li	Li	K
Core					
Power Density kw/liter	550	326	128	87	79
Height (in.)	10.4	13.2	11.8	12.9	11.625
Diameter (in.)	10.4	12.0	10.2	11.8	9.17
Lifetime (hr)	10,000	10,000	10,000	10,000	10,000
Fuel Pin					
Diameter (in.)	0.44	0.25	0.25	0.25	0.500
Max Heat Rating (w/cm)	1280	218	71.6	50	180
Max Power Density (w/cm^3)	1630	1220	400	280	170
Max Burnup (fissions/$cm^3 \times 10^{-20}$)	19.5	14.8	4.95	3.42	1.85
Max Temperature (°C)	1870	1370	1260	1260	1480
Ratio of Plenum Volume to Fuel Volume	0.385	0.68	1.0	0.68	0.30
Cladding					
Max Temperature (°C)	1200	1200	1200	1200	860
Thickness (mils)	15	25	25	25	20
Design Limit					
Fission-Gas Release (%)	10	10	20	20	20[b]
Pin Diametral Increase (%)	1	1	2	2	1
Fuel Swelling (Volumetric Increase, %)	4	4	8	8	9[b]

[a]See References 1, 2, and 3 in text.

[b]Amounts expected are much lower in the neighborhood of 1%.

design power level and burnup limitations were progressively reduced. To compare the three fuels for SNAP-50 conditions, the reactor irradiation conditions for Reference Design No. 3 will be used. No irradiation data are available for the severe conditions described in Reference Designs 1 and 2. Since irradiation conditions are less severe in Reference Design No. 4 than in No. 3, the results accumulated in a comparison of fuels with the requirements of Reference Design No. 3 should also be applicable to Reference Design No. 4.

Table 2 shows a list of the fuels considered for high temperature applications. From this list the three bulk fuels, UO_2, UC, and UN, and three dispersion fuels, $BeO-UO_2$, $W-UO_2$, and $Mo-UO_2$ were chosen for comparison. Lack of irradiation and fabrication data on the other fuels prevents proper evaluation.

For a qualitative comparison of the thermal capability of the fuels, two limiting heat ratings were calculated. These are H_m, the heat rating to produce centerline melting, and H_f, the heat rating to produce radial cracking from thermal stress. H_m was calculated for rods with a fuel surface temperature, T_s, of 1000°C. This quantity is the product of the fuel cross-sectional area and the power density.[4]

$$H_m = \pi R^2 Q = 4\pi \int_{T_s}^{T_m} K dT$$

where,

R = radius of fuel rod,

Q = power density,

T_m = fuel melting temperature,

T_s = fuel surface temperature, and

K = thermal conductivity of fuel.

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[4]J. L. Scott, "High Temperature Fuel Element Development," Paper presented at the ARS Space-Nuclear Conference, Gatlinburg, Tennessee, May 1963.

Table 2. Properties of Candidate Fuels

Fuel	Matrix Crystal Structure	Matrix Density (g/cm^3)	Uranium Density (g/cm^3)	Liquidus Temp (°C)	Mean[a] Matrix Thermal Cond., K (w/cm/°C)	Coefficient of Expansion ($°C^{-1} \times 10^6$)	$(\sigma/E) \times 10^3$	Heat Rating[b] to produce Central Melting H_m(w/cm)	Heat Rating[c] to produce Radial Cracking H_f(w/cm)
UO_2	FCC, CaF_2	10.97	9.6	2800	0.03	10.52	1.15	680	65
UN	FCC, NaCl	14.32	13.6	2850	0.25	9.9	1.20	5,680	669
UC	FCC, NaCl	13.63	12.97	2400	0.17	11.0	1.20	2,990	645
UB_4	Tetragonal	9.3	8.58	2550	0.268	--	--	5,250	--
US	FCC	10.87	10.19	2460	0.12	--	--	2,200	--
UB_2	Hexagonal	12.87	12.35	2440	--	--	--	--	--
UO_2-BeO	Hexagonal	3.01	--	2150	0.188	5.5	0.38	1,580	130
UO_2-W	BCC	19.30	--	3750	1.46	4.3	2.00	23,000	8,300
UO_2-Mo	BCC	10.22	--	2610	1.42	4.9	2.10	21,100	7,900

[a]Values listed are approximate mean values between 1000°C and the melting point.

[b]Heat rating to produce centerline melting: $H_m = 4\pi \int_{1000°C}^{T_m} k\, dt$; where T_m = Liquidus temperature.

[c]Heat rating to produce radial cracking: $H_f = \frac{8\pi K(1-\mu)\sigma}{\alpha E}$; where μ = Poisson's ratio, σ and E are the values for rupture stress and Poisson's ratio, respectively.

The results of this calculation are shown in Table 2. The heat rating to produce radial cracking, H_f, is given by the equation[5]

$$H_f = \frac{8\pi\ K(1 - \mu)\sigma}{\alpha\ E}$$

where,

σ = rupture stress, psi,

E = elastic modulus, psi,

α = linear coefficient of thermal expansion, $°C^{-1}$, and

μ = Poisson's ratio.

Since the fuel will be clad, the heat rating is not necessarily limited by radial cracking, but H_f does provide a limit above which some effect on the fuel would be expected.

Other fuel performance factors to be compared are (1) the volumetric increase of the fuel during irradiation, (2) fission-gas release, (3) fuel pin diametral increase, and (4) the compatibility of fuel with clad and coolant materials. These fuel performance factors were examined as functions of temperature and irradiation exposure. A detailed examination of the advantages and disadvantages of each fuel is discussed with respect to these thermal and irradiation factors in the following section.

Uranium Dioxide

Some of the advantages of UO_2 as a reactor fuel are:

(1) Low fuel swelling and fission gas release below fuel centerline temperatures of 1600°C.

(2) Low fabrication costs.

(3) Good compatibility with potassium.

(4) Good compatibility with most cladding materials.

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[5]J. L. Scott, "High Temperature Fuel Element Development," Paper presented at the ARS Space-Nuclear Conference, Gatlinburg, Tennessee, May 1963.

Many extensive research and development programs have been carried out in the past few years on UO_2 and considerable data have been compiled. As a result, UO_2 has become a prominent fuel for many commercial reactors. Fabrication methods for UO_2 fuel elements are well established, and most production and handling problems have been eliminated. The principal advantage of UO_2 as a fuel for space reactors is the low fission-gas release and pellet swelling below 1600°C.

A disadvantage of UO_2 is its low thermal conductivity which limits the maximum power rating. At a given power level this causes a large temperature differential between the fuel surface and fuel center in comparison with UN or UC.

The low thermal conductivity of UO_2 (Fig. 1) also results in low values for H_f and H_m (Table 2). The value of H_f for UO_2, 65 w/cm, is well below the maximum MPRE design power rating of 180 w/cm. The SNAP-50 (Reference Design No. 3) linear heat rating of 72 w/cm (400 w/cc) also falls within this range. This indicates that UO_2 would be subject to thermal-stress cracking under the design irradiation conditions of either reactor. The cracking of UO_2 in clad fuel elements, however, is ordinarily not a limiting factor since many thousands of UO_2 fuel elements have been irradiated under conditions that induced thermal-stress cracking of the UO_2 without significant detrimental effects. Some reduction in thermal conductivity may occur because the cracks act as thermal barriers. The surface area generated by cracks may enhance the fission-gas release rate.

Fuel element dimensional changes resulting from radiation damage are of major importance in comparing one fuel with another. The tendency for fuel pins to increase in diameter may result from either or both of two irradiation phenomena: (1) swelling of the fuel matrix, and (2) high temperature creep of the clad from the internal pressure of released fission gases. The swelling of the fuel matrix is caused by accumulation of solid fission products (two atoms are formed from each atom fissioned), and the agglomeration of fission gas atoms into bubbles.

To determine the stability of a fuel in an irradiation environment, the fuel radiation damage is usually examined with respect to two parameters,

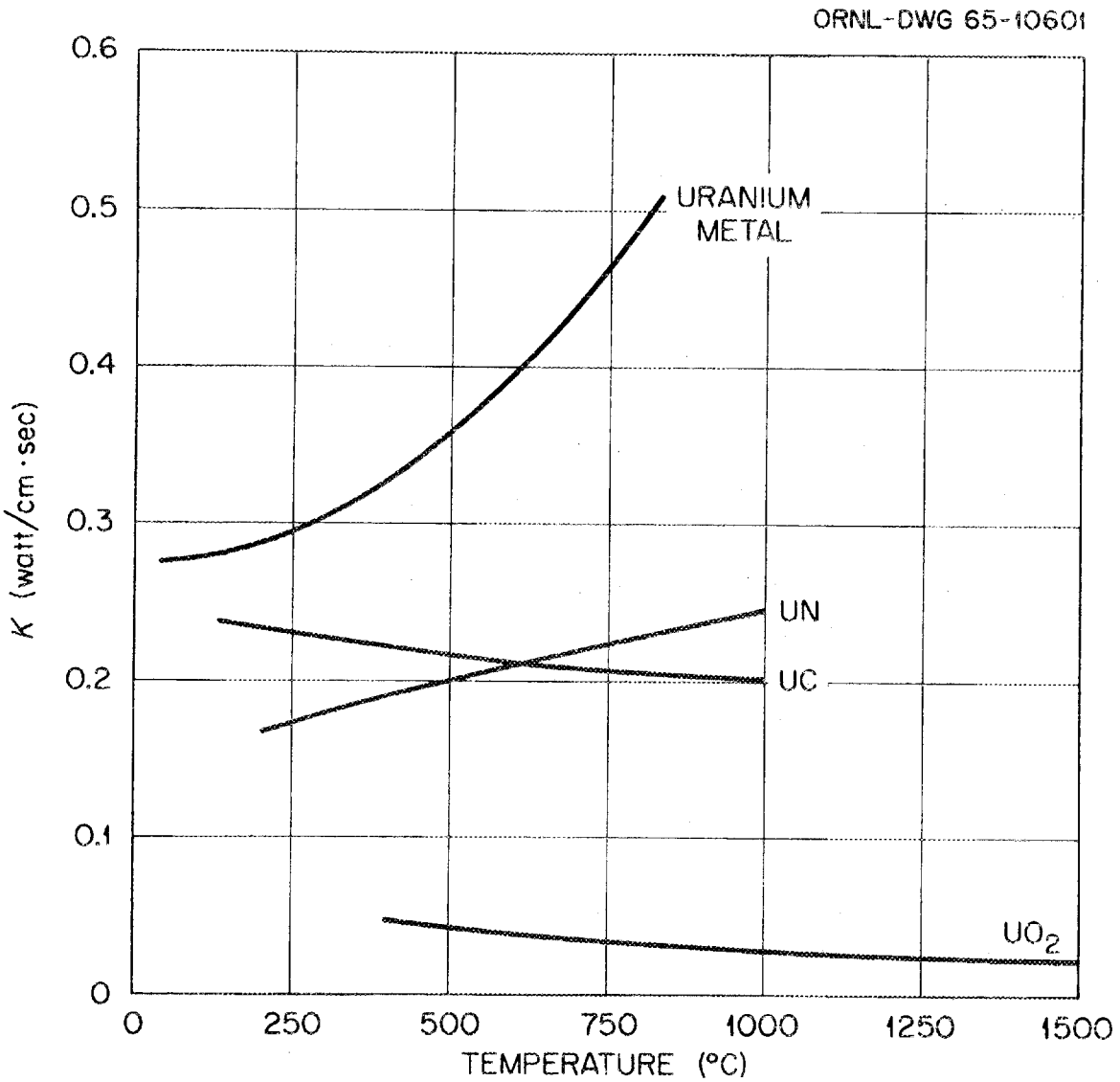

Fig. 1. Thermal Conductivity of Reactor Fuel Materials.

fuel temperature and burnup (fission density). Fuel centerline temperatures were used for comparison because clad temperatures are less representative of the fuel temperature. Clad temperatures and fuel temperatures may vary widely depending on the power density, the cladding used, and the gap conductance. It must not be overlooked, however, that diametral increases of the cladding depend in part on the creep strength of the cladding, and this is a function of the cladding temperature.

Fuel Volumetric Changes

Swelling of UO_2 at low temperatures is somewhat less than for UN and UC at comparable temperatures and burnups. The initial swelling in the low temperature irradiation of UO_2 is offset by the disappearance of porosity.[6] For specimens irradiated in the temperature range 150–375°C, no fuel volume increase was noted up to burnups of 3×10^{20} fissions/cm^3, the volume increases linearly with burnup. It should be noted, however, that these data were obtained from UO_2 dispersed in a stainless steel matrix. In bulk UO_2 the swelling will be greater. At higher temperatures (725–1760°C, Fig. 3)[7] volumetric increases begin during the initial stages of irradiation, although they are still somewhat less than for UC or UN at similar temperatures. The curve in Fig. 3 is a least squares fit of the high-temperature swelling data shown. The maximum swelling exhibited by the UO_2 fuel plates is higher than this curve by 2-4% of the plate volume.

Fission-Gas Release

Uranium dioxide fission-gas release results from the agglomeration into bubbles of gaseous atoms produced during fissioning.[8-9] These

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[6]M. L. Bleiberg, R. M. Berman, and B. Lustman, "Effects of High Burnup on Oxide Ceramic Fuels," WAPD-T-1455, March 1962.

[7]M. L. Bleiberg, R. M. Berman, and B. Lustman, "Effects of High Burnup on Oxide Ceramic Fuels," WAPD-T-1455, March 1962.

[8]R. S. Barnes and R. S. Nelson, "Theories of Swelling and Gas Retention in Reactor Materials," AERE-R 4952, June 1965.

[9]V. Scotti et al., "Advanced Materials Program for July and August, 1964," PWAC-1016, pp. 39-46, November 11, 1964. CONFIDENTIAL-RESTRICTED DATA.

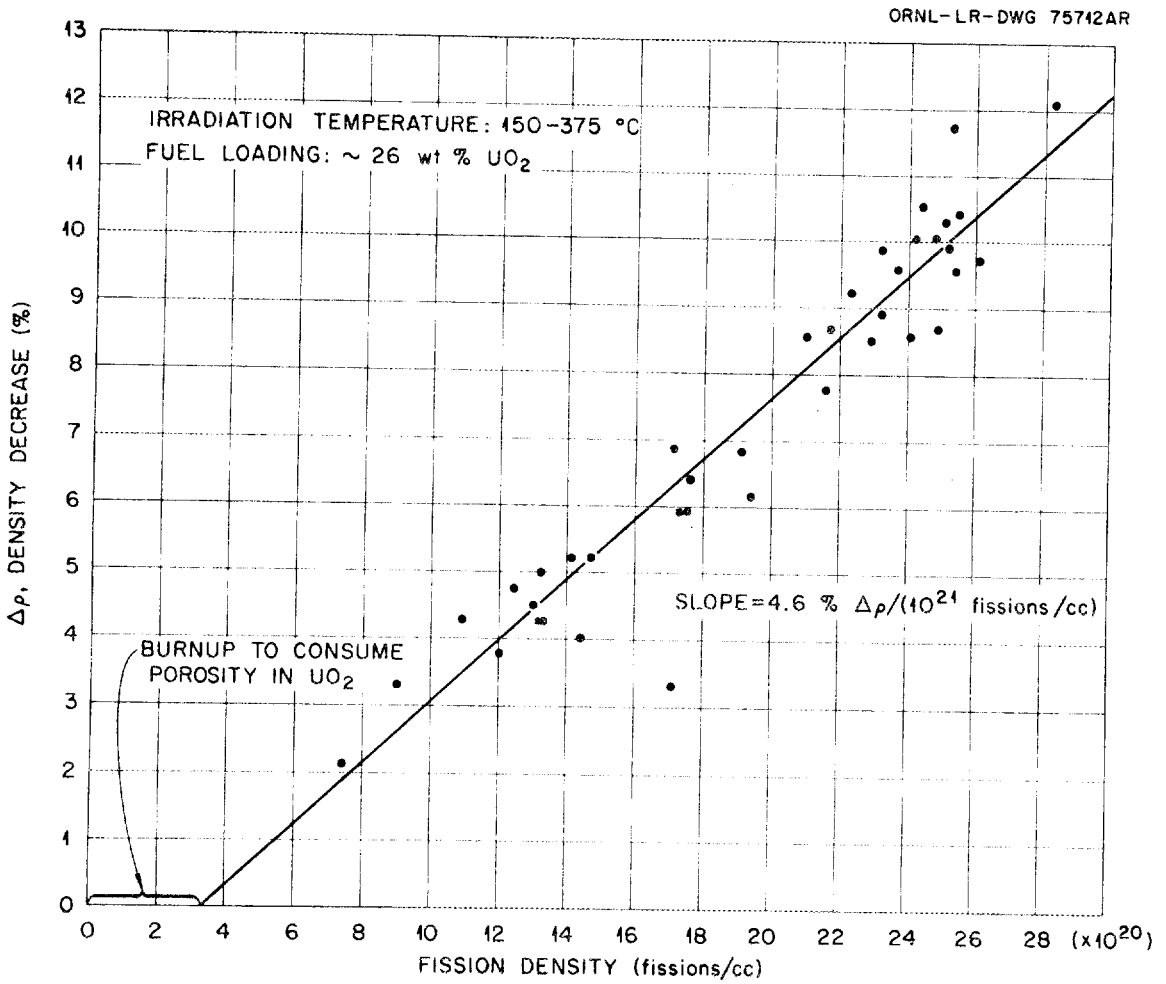

Fig. 2. Matrix Swelling of UO_2-Type 347 Stainless Steel Dispersion Fuels.

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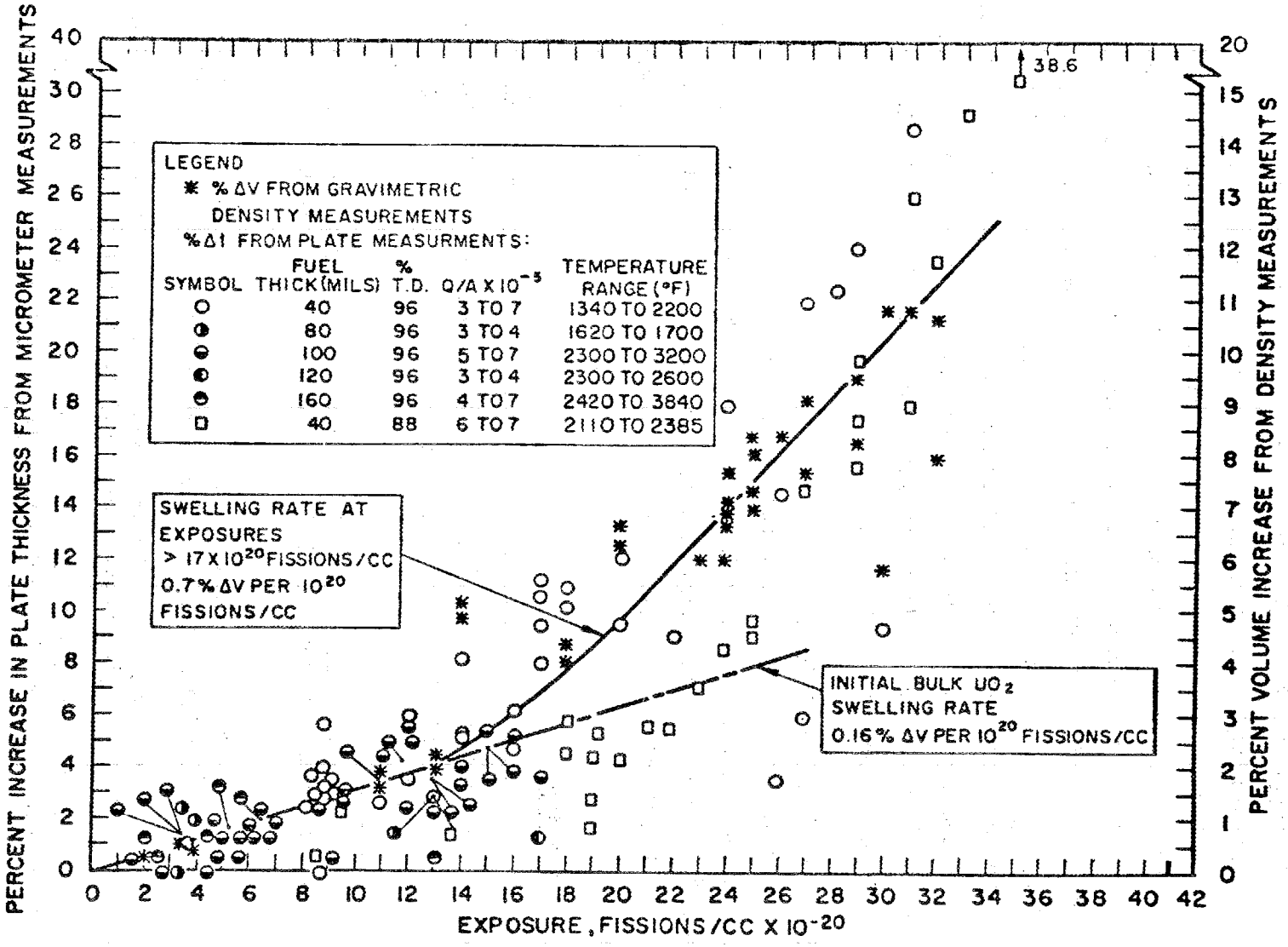

Fig. 3. Solid Volume Swelling of Bulk UO_2.

bubbles migrate up a temperature gradient and collect to form a central void at the center of the fuel pellet. Uranium dioxide fission-gas release is small (approximately 1%) for centerline temperatures below 1600°C,[10] but increases rapidly with increasing temperature above 1600°C. The curve in Fig. 4 shows very little change in gas release with centerline temperature in the temperature range below 1600°C.[11] Figure 5 shows the effect of fuel burnup in this temperature range.[12] At temperatures exceeding 1600°C, however, the bubble migration becomes more rapid causing an increase in gas release. At 1700°C growth of columnar grains is observed and attributed to the migration of large transverse voids which move up the temperature gradient by a sublimation or surface diffusion process.[13] Under this latter condition, essentially all fission gas formed is released from the UO_2.

Fuel Pin Diametral Change

Because pin diametral increases are a result of fuel swelling and fission-gas pressure, one would expect very little diametral change below 1600°C. For centerline temperatures above 1600°C, provision must be made for a high fission-gas release if diametral increases are to be kept small.

Compatibility

Other criteria to be considered are the compatibility of the fuel material with both the cladding and the coolant. Ideally the fuel should never come in contact with the coolant, however, if the fuel and coolant

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[10]B. Lustman, "Irradiation Effects in Uranium Dioxide, Properties and Nuclear Applications," J. Belle, ed., U.S.G.P.O., Washington 25, D. C., 1961.

[11]H. K. Hardy, L. M. Wyatt, and S. F. Pugh, "Metallurgy of Fuel and Cans," British Nuclear Engineering Society Journal, 2: 243 (1963).

[12]M. L. Bleiberg, R. M. Berman, and B. Lustman, "Effects of High Burnup on Oxide Ceramic Fuels," WAPD-T-1455, March 1962.

[13]J. R. MacEwan and U. B. Lawson, "Grain Growth in Sintered Uranium Dioxide: II. Columnar Grain Growth," J. Am. Ceram. Soc., 45: 42-46 (1962).

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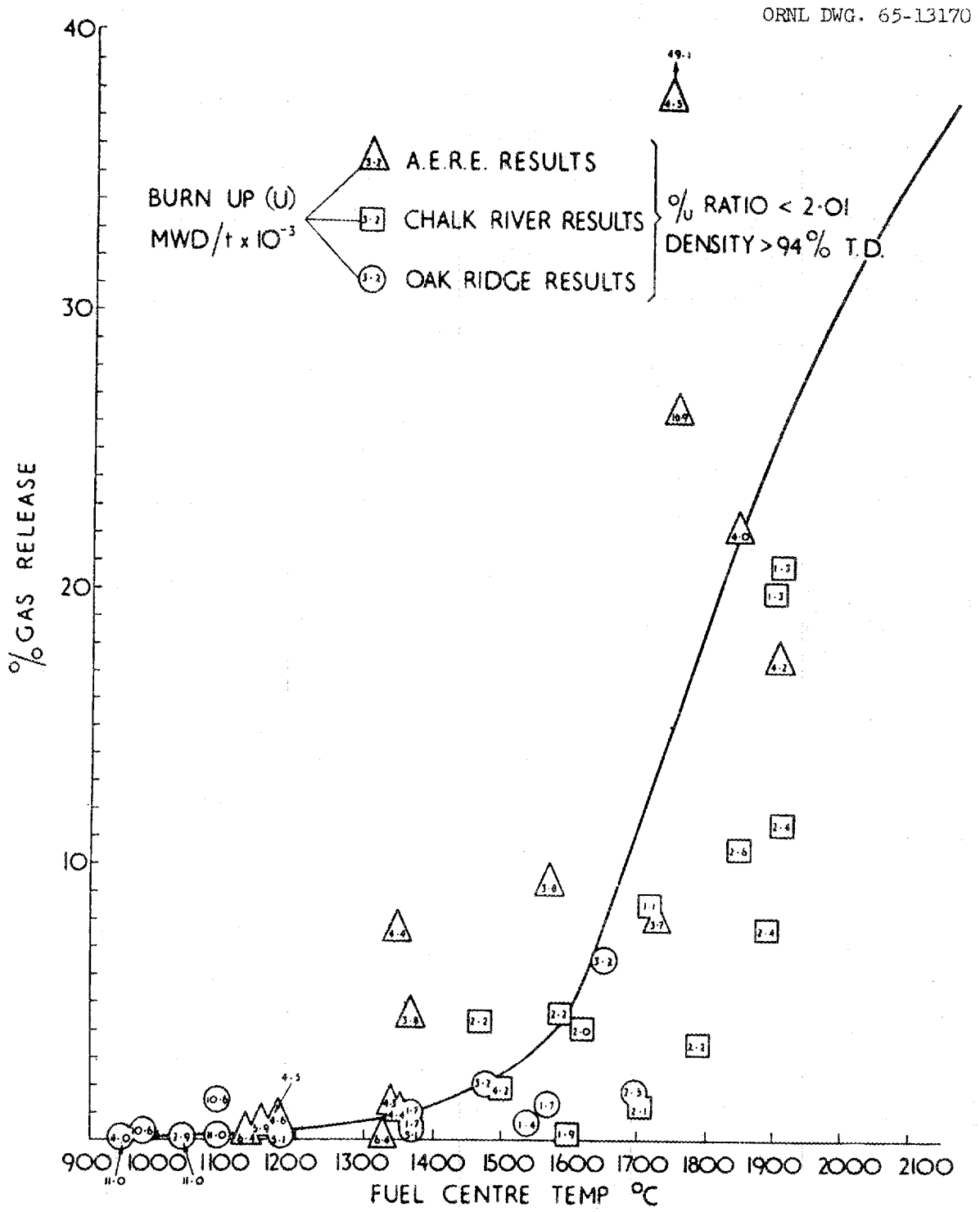

Fig. 4. Collected Fission Gas Release Data as a Function of UO_2 Centre Temperature.

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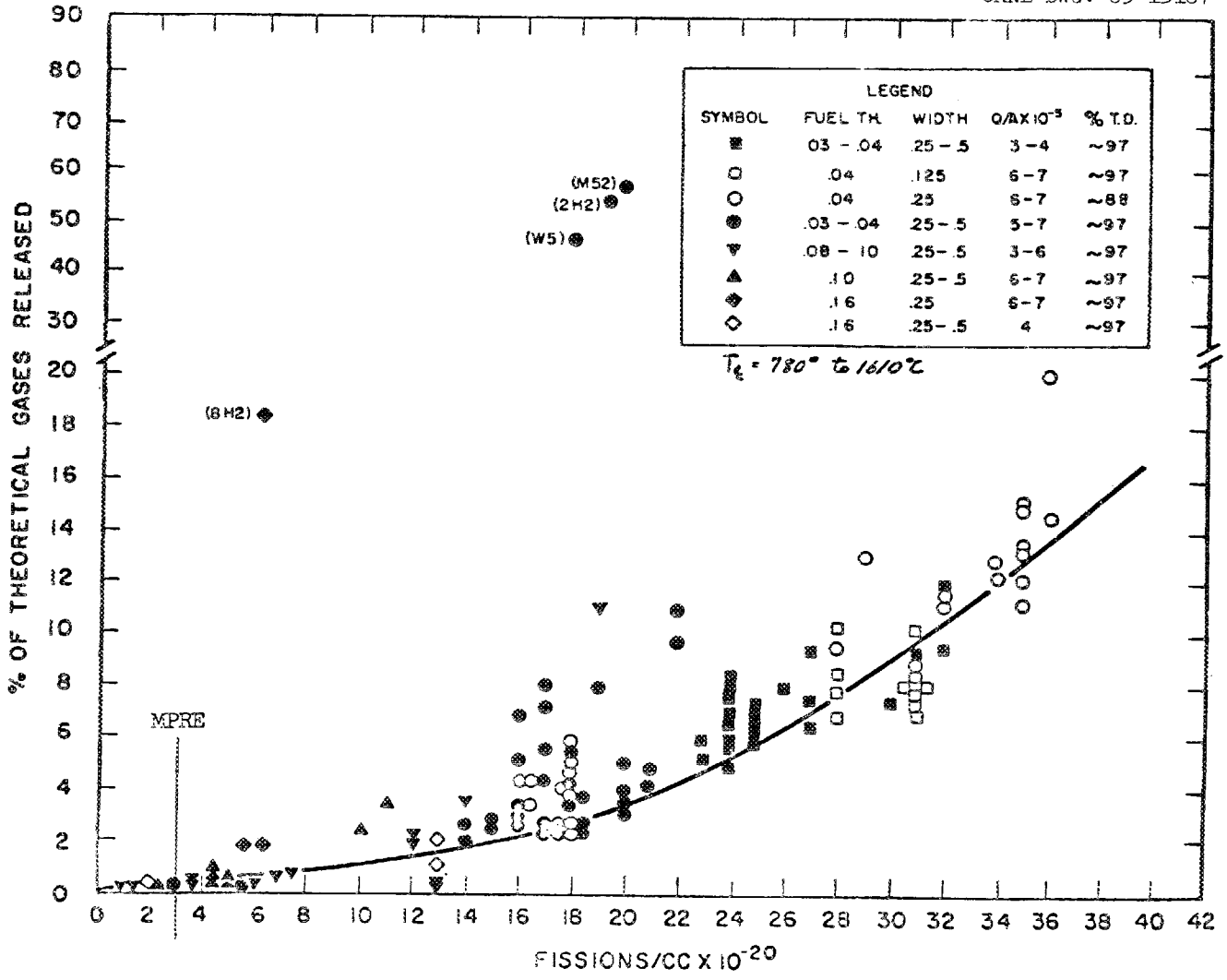

Fig. 5. Fission Gas Release From UO_2 Plates.

are compatible no serious effect will result if only one or a few cladding failures occur. On the other hand, in a reactor where the fuel and coolant are reactive, the failure of a single pin might be sufficient to initiate a reactor shutdown. For example, UO_2 and lithium react exothermically so that under reactor operating conditions, a failed fuel pin would result in the dispersal of large amounts of fuel and fission products throughout the primary coolant system. Thus, failure of a single pin could create a serious environmental hazard.

Good compatibility of UO_2 with potassium has been demonstrated in long endurance tests[14] in which the only reaction has been some grain boundary attack. Thus, failure of a fuel pin would probably not significantly affect reactor operation.

A compilation of data on the compatibility of UO_2 with possible cladding materials[15-16] is shown in Table 3. Uranium dioxide appears to be compatible with most of these materials except Ta or Zr base alloys. For example, ORNL has irradiated many stainless steel clad UO_2 fuel pins in the ORR with no appreciable fuel-clad reactions.

In summary, UO_2 exhibits many desirable features as a candidate fuel for the MPRE reactor. Among these features are:

(1) Good compatibility with potassium.

(2) Good compatibility with stainless steel.

(3) High resistance to radiation damage at the MPRE design temperatures with low fission-gas release and low fuel swelling.

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[14]W. H. Cook, "Medium Power Reactor Experiment Quarterly Progress Report for Period Ending March 31, 1965," USAEC Report ORNL-3818, pp. 118-23, June 1965. CONFIDENTIAL-RESTRICTED DATA.

[15]DeMastry and Griesenauer, "Investigation of High Temperature Refractory Metals and Alloys for Thermionic Converters," AFAPL-TR-65-29, Supplement 1, Battelle Memorial Institute, April 1965. CONFIDENTIAL-RESTRICTED DATA.

[16]Uranium Dioxide: Properties and Nuclear Applications, USAEC Report, Naval Reactors, Division of Reactor Development, pp. 344-71, 669, ed. by J. Belle.

Table 3. UO_2 Compatibility

Temp (°C)	Stainless Steel	Zr	Cb	W	W-0.9Cb	W-15Mo	W-10Re	W-25Re	TZM	Mo-50Re	Re	Ta-12W	T-111	B66(Cb-5Mo 5V-12Re)
850	Com													
870		Significant Reaction at 15 mils After 4 Days												
1000			Reacts											
1371				Com	Com	Com	Com	Com	Com	Com	Com	Com	Heavy Oxide ppt after 100 hr	General ppt throughout Matrix
1538				Com	Com	Com	Com	Com	Com	Com	Com	Slight Reaction	Incom	Incom
1705				Com	Com	Com	Com	Com	Com	Com	Com	Incom 100 hr	Incom	Incom
1871				Com	Com	Com	Com	Com	Com	Com	Com	Incom	Incom	Incom

Com — Compatible, no reaction after 1000 hr.

Incom — Incompatible, gross reaction.

Extensive testing under MPRE irradiation conditions with good success is also a strong recommendation for UO_2. In addition, most fabrication and handling problems of UO_2 have been worked out thereby yielding lower fabrication costs than for UN or UC.

Uranium Carbide

Some of the advantages of uranium carbide as a refractory ceramic fuel are:

(1) High thermal conductivity.

(2) High uranium density.

(3) High heat rating to produce radial cracking, H_f.

(4) High heat rating to produce centerline melting, H_m.

The higher thermal conductivity (Fig. 1) of UC makes it possible to obtain higher power ratings from UC than UO_2. For the power densities of this study, UC gives a relatively low temperature differential between the center and surface of the fuel compared to UO_2 at the same power level. The values of H_f and H_m, the heat ratings to produce radial cracking and melting, respectively, are high (Table 2) which insures integrity of the fuel bodies under reactor operating conditions.

To determine the advantages of UC in an irradiation environment, irradiation data were examined from four sources — BMI, AI, CANEL, and United Nuclear. Workers at Atomics International and Battelle Memorial Institute organized their data by assuming a linear relationship between burnup and the irradiation effects (fuel pin diametral increase, fission-gas release, and fuel pellet swelling). The irradiation effects for a given burnup were then plotted as a function of temperature. CANEL irradiation data, however, suggests that the linear relationship proposed by AI and BMI is not a good assumption. The quantity of data available is small in comparison with UO_2, although more data are available for UC than for UN.

Fuel Volumetric Change

The effect of irradiation and temperature on the fuel volumetric increase are demonstrated in Figs. 6 and 7 from data compiled by AI

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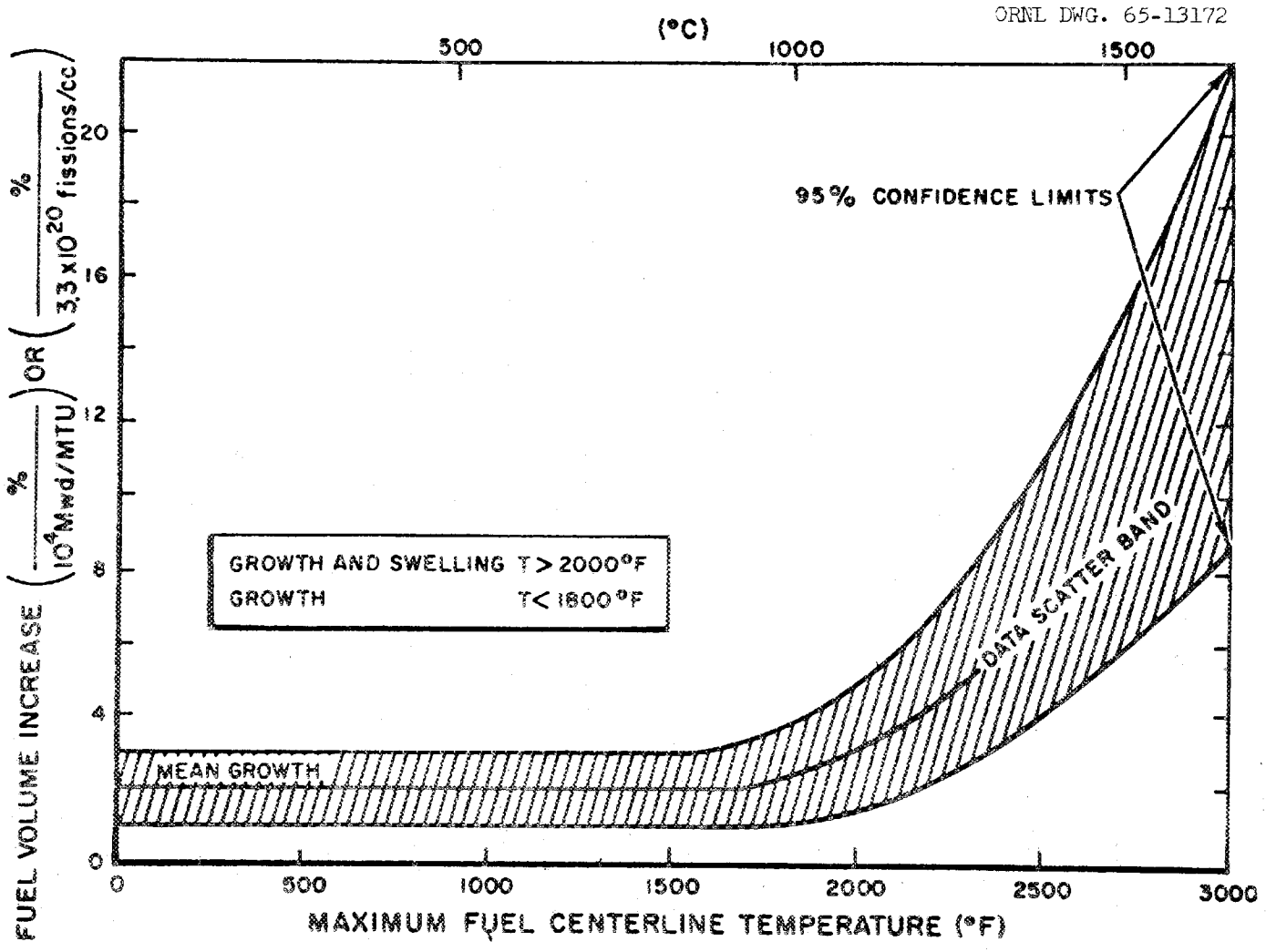

Fig. 6. Uranium Carbide Fuel Growth and Swelling Characteristics as a Function of Temperature.

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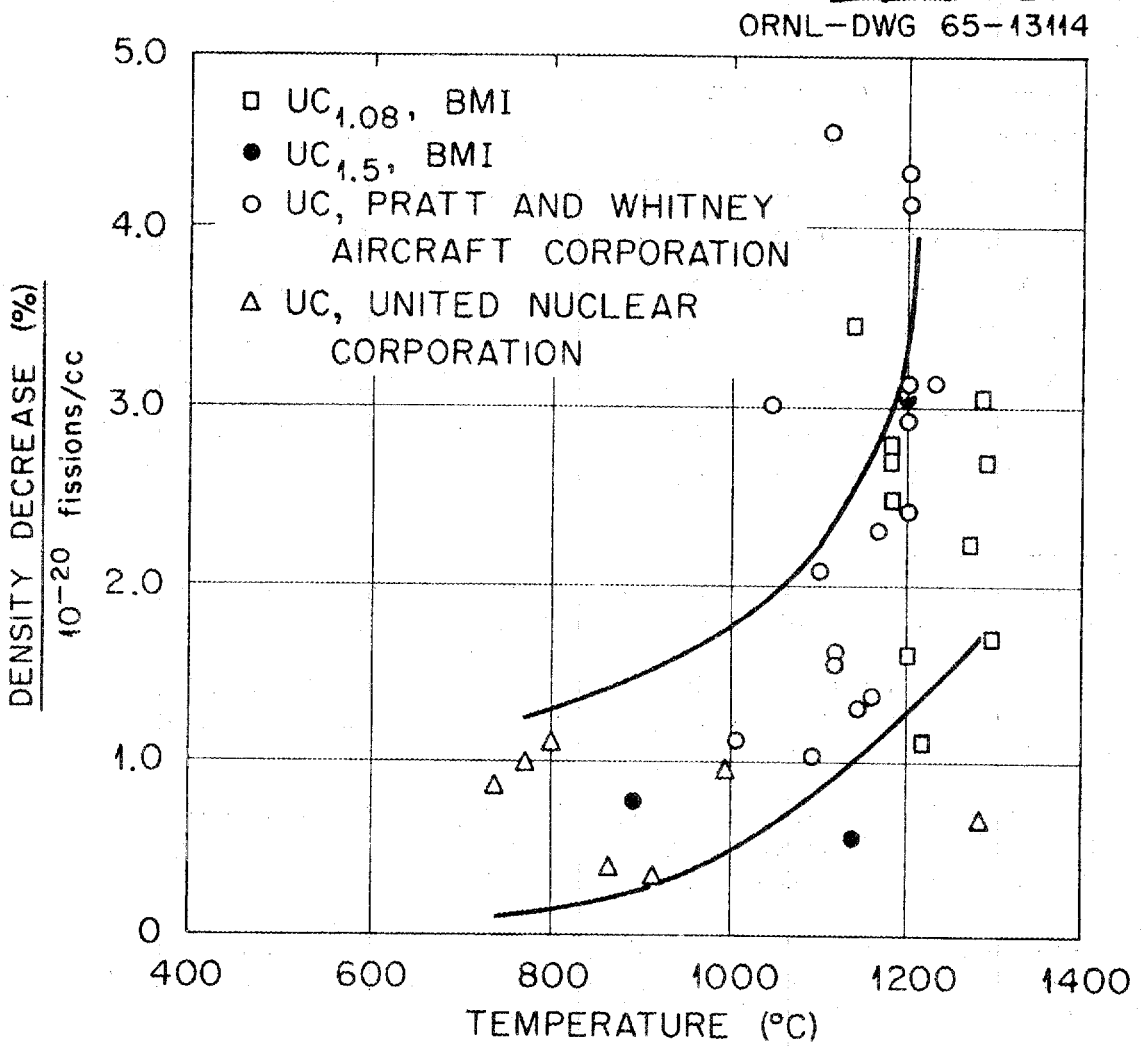

Fig. 7. UC Fuel Volumetric Change.

[redacted]

and BMI.[17-18] Both curves show a large change in slope at 1100-1200°C indicating that fuel swelling is greatly dependent on temperature. At a burnup of about 4.9×10^{20} fissions/cm^3, CANEL irradiation data[19-20] indicate that breakaway swelling occurs and the slope of the isothermal curve representing density decrease vs fuel burnup increases rapidly (Fig. 8). This suggests that fuel volumetric changes limit the use of UC to burnups [redacted] 4.9×10^{20} fissions/cm^3 at temperatures of 1500°C.

One method proposed to reduce UC fuel swelling is to increase the carbon content of the fuel. Results obtained at United Nuclear indicate that the volumetric change of UC caused by irradiation decreased with increasing carbon content of the fuel at 900°C (Fig. 9).[21] However, while the increase in carbon content may reduce the fuel swelling slightly, it may also have a detrimental effect since CANEL personnel observed carbon migration down a thermal gradient at centerline temperatures of 1500°C.[22] This carbon migration results in the formation and reformation of second phases in the structure during irradiation.[23]

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[17]"Annual Technical Progress Report, AEC Unclassified Programs," USAEC Report NAA-SR-9999, Atomics International, August 31, 1964.

[18]R. J. Bayuzick and J. E. Gates, "Development of Advanced High-Temperature Nuclear Materials During May through July 1965," BMI-1741, Battelle Memorial Institute. CONFIDENTIAL-RESTRICTED DATA.

[19]Pratt and Whitney Aircraft—CANEL, "SNAP-50/SPUR Program Engineering Progress Report April 1, 1964, to June 30, 1964," PWAC-642, p. 135, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

[20]Pratt and Whitney Aircraft—CANEL, "SNAP-50/SPUR Program Engineering Progress Report October 1, 1964, to December 31, 1964," PWAC-644, p. 151, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

[21]J. Crane and E. Gordon, "The Development of Uranium Carbide as a Nuclear Fuel, Final Report," USAEC Report UNC-5080, United Nuclear Corp., February 29, 1964.

[22]M. A. DeCrescente et al., "Advanced Materials Program for July and August, 1963," PWAC-1010, pp. 33-44, October 25, 1965. CONFIDENTIAL-RESTRICTED DATA.

[23]M. A. DeCrescente, "Advanced Materials Program for July and August, 1963," PWAC-1010, p. 38, October 25, 1963. CONFIDENTIAL-RESTRICTED DATA.

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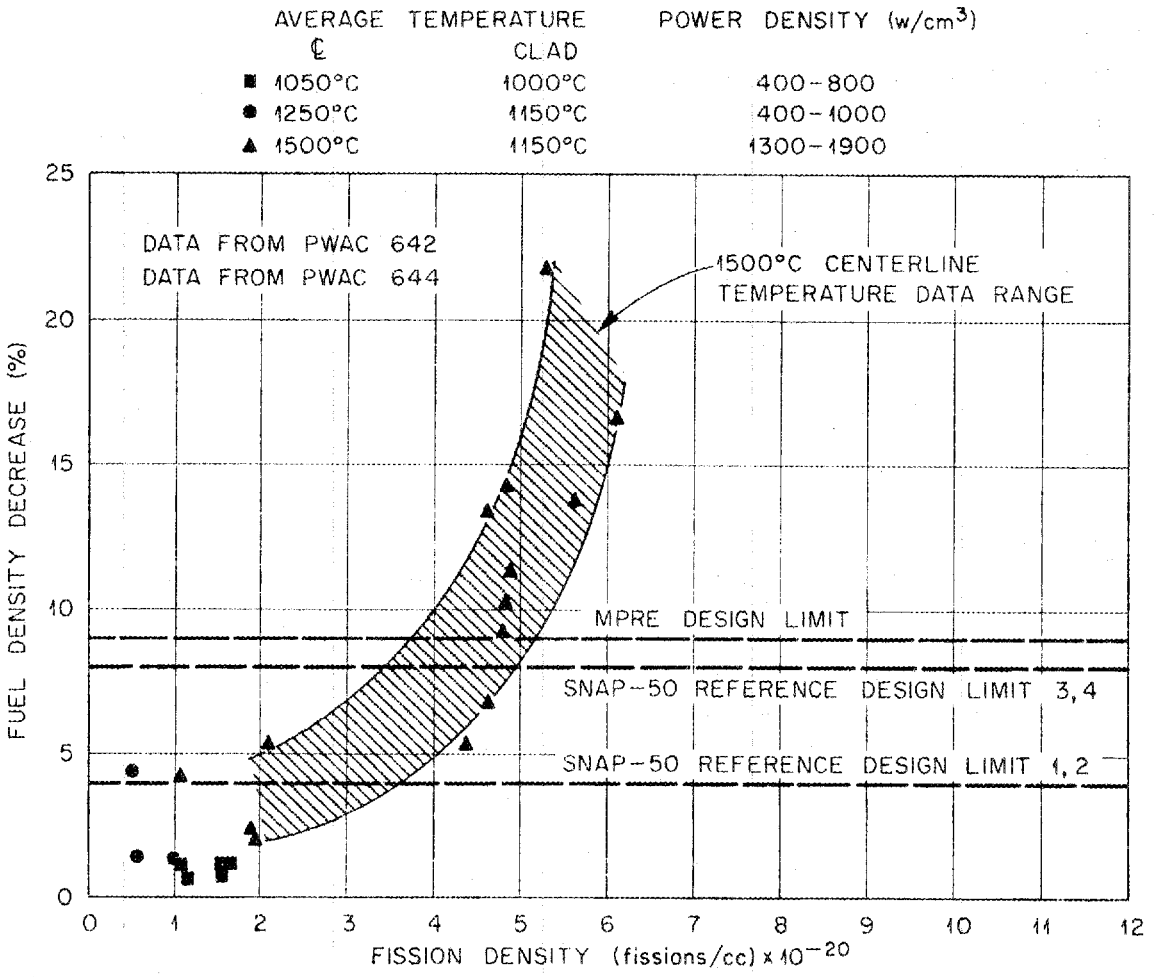

Fig. 8. UC Fuel Volume Change.

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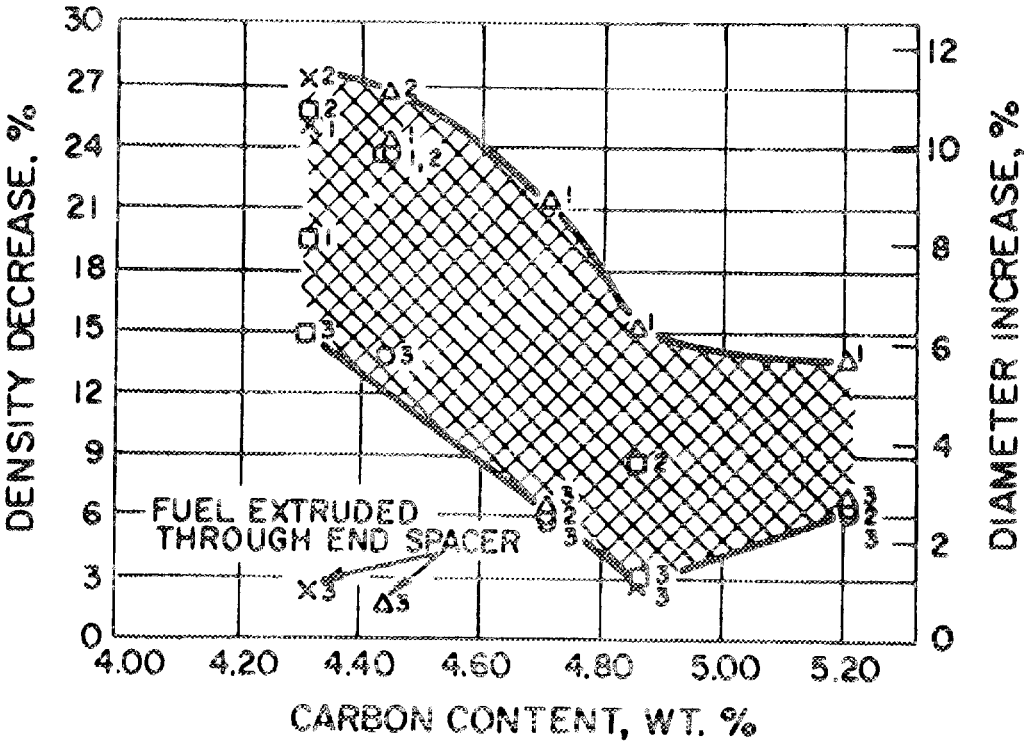

Fig. 9. *Radiation-induced density and diameter changes in uranium carbide as a function of carbon content.*[55] *(Capsule numbers are noted next to each point; burnups to 5.4 at.% at surface temperatures to 900°C.) Density change: ○, cast; □, sintered. Diameter change: △, cast; ×, sintered.*

[redacted]

These phase changes are thought to enhance bubble formation and fission-gas release. The end result may be a reduction in fuel volume with diametral pin swelling remaining the same or increasing due to creep of the clad from the hydrostatic pressure of fission gases.

This reported increase in fission-gas release with carbon content is in contrast (Fig. 10) with United Nuclear's low temperature data[24] (900°C) in which they observed a decrease in fission-gas release with increasing carbon content. This is probably due to a reduction in the kinetics of carbon migration at the lower temperature.

Fission-Gas Release

Three sets of data are also available to compare fission-gas release effects. Atomics International (Fig. 11) irradiation data[25] show a large temperature dependency of fission-gas release above 1100°C. This corresponds to the same temperature at which they observe breakaway fuel swelling. CANEL's high temperature data (1500°C)[26-27] suggests a burnup limit of approximately 4×10^{20} fissions/cm^3. At this burnup the slope of the curve showing gas release vs burnup increases rapidly (Fig. 12). Unfortunately, considerable spread can be observed in CANEL's irradiation data at 1500°C. High fission-gas release was observed at relatively low burnups compared to UO_2 and UN. Fission-gas release data from BMI are plotted as a function of fuel surface temperature (Fig. 13).[28]

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[24]J. Crane and E. Gordon, "The Development of Uranium Carbide as a Nuclear Fuel, Final Report," USAEC Report UNC-5080, United Nuclear Corp., February 29, 1964.

[25]"Annual Technical Progress Report, AEC Unclassified Programs," USAEC Report NAA-SR-9999, Atomics International, August 31, 1964.

[26]Pratt and Whitney Aircraft-CANEL, "SNAP-50/SPUR Program Engineering Progress Report April 1, 1964, to June 30, 1964," PWAC-642, p. 135, July 30, 1964. CONFIDENTIAL-RESTRICTED DATA.

[27]Pratt and Whitney Aircraft-CANEL, "SNAP-50/SPUR Program Engineering Progress Report October 1, 1964, to December 31, 1964," PWAC-644, p. 151, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

[28]Freas et al., "High Temperature Irradiation of Uranium Carbide," BMI-1622, p. 29, Battelle Memorial Institute, October 25, 1963. CONFIDENTIAL-RESTRICTED DATA.

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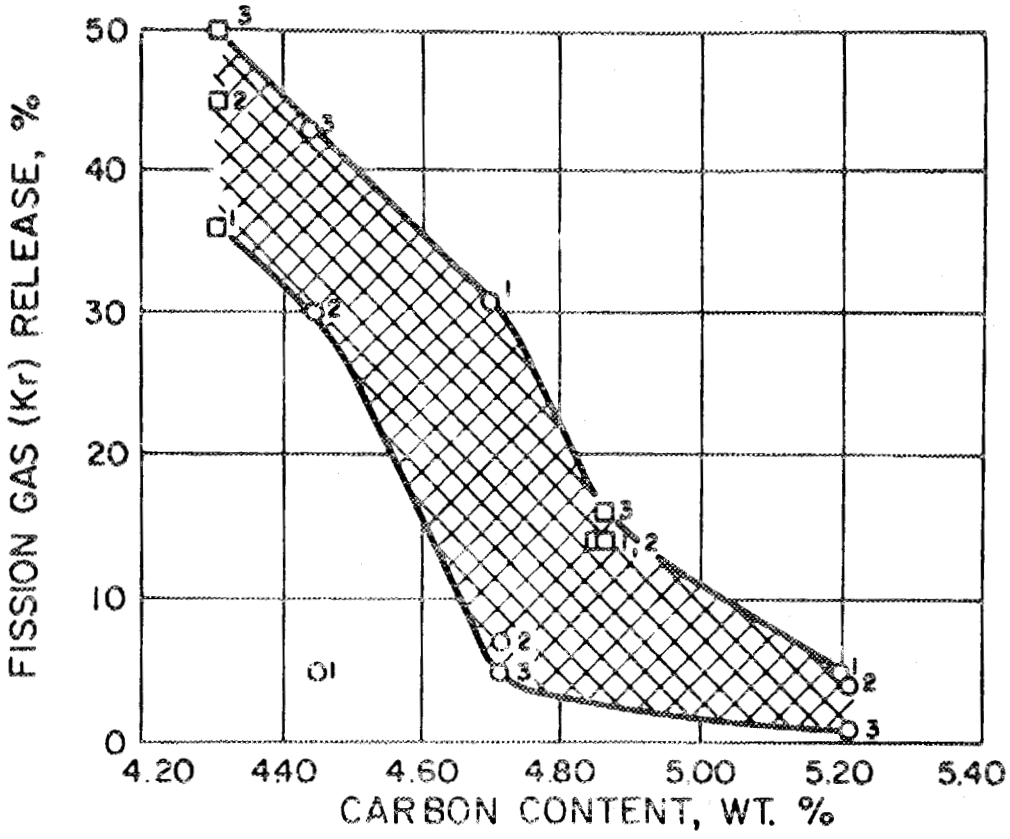

Fig. 10. *Percentage fission-gas release of the total formed in irradiated uranium carbide, as a function of carbon content.*[55] *(Capsule numbers are noted next to each point; burnups to 5.4 at.% at surface temperatures to 900°C.)* ○, *cast;* □, *sintered.*

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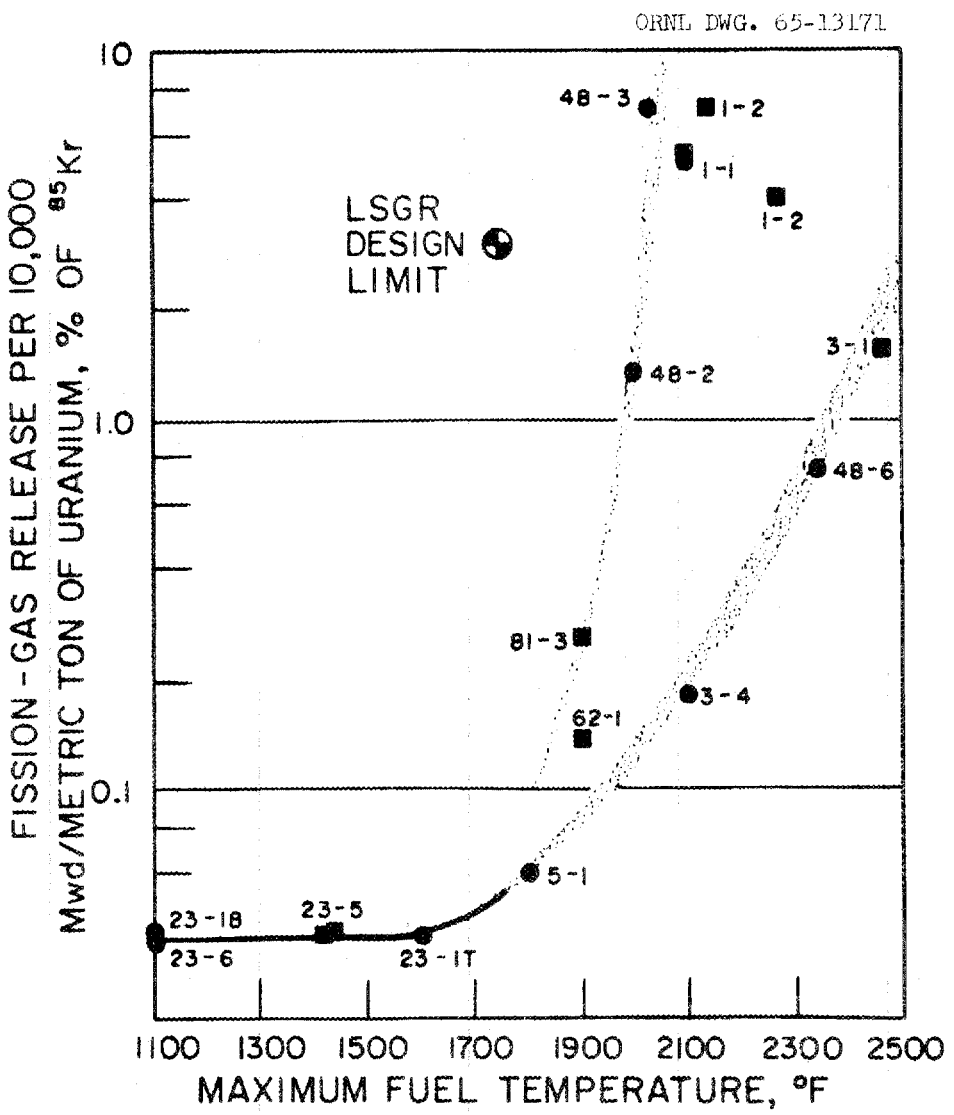

Fig. 11. *Fission-gas release from UC due to irradiation.*[58] ■, *hypostoichiometric UC (< 4.8 wt.% carbon);* ●, *hyperstoichiometric UC (> 4.8 wt.% carbon).*

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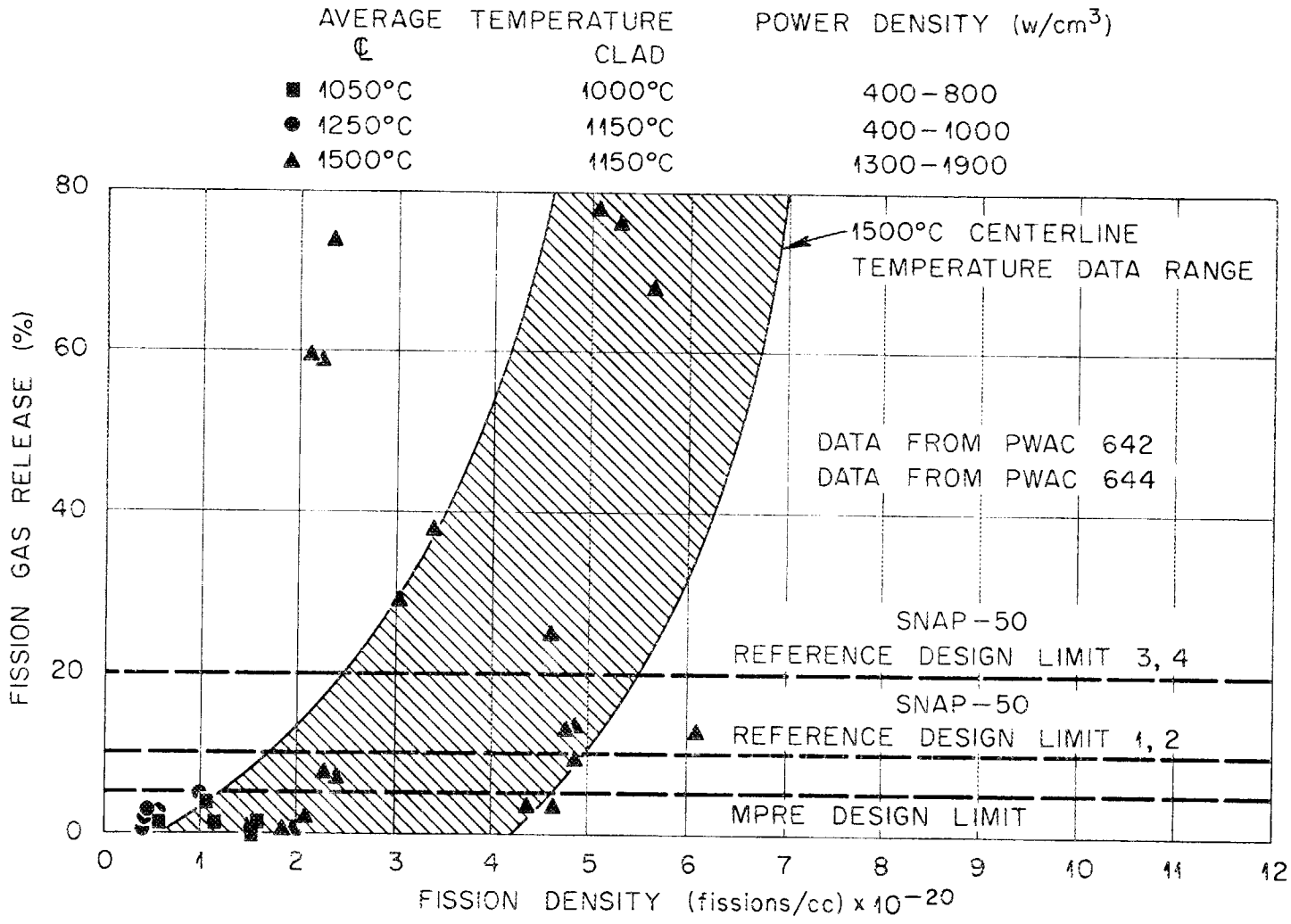

Fig. 12. UC Fission Gas Release.

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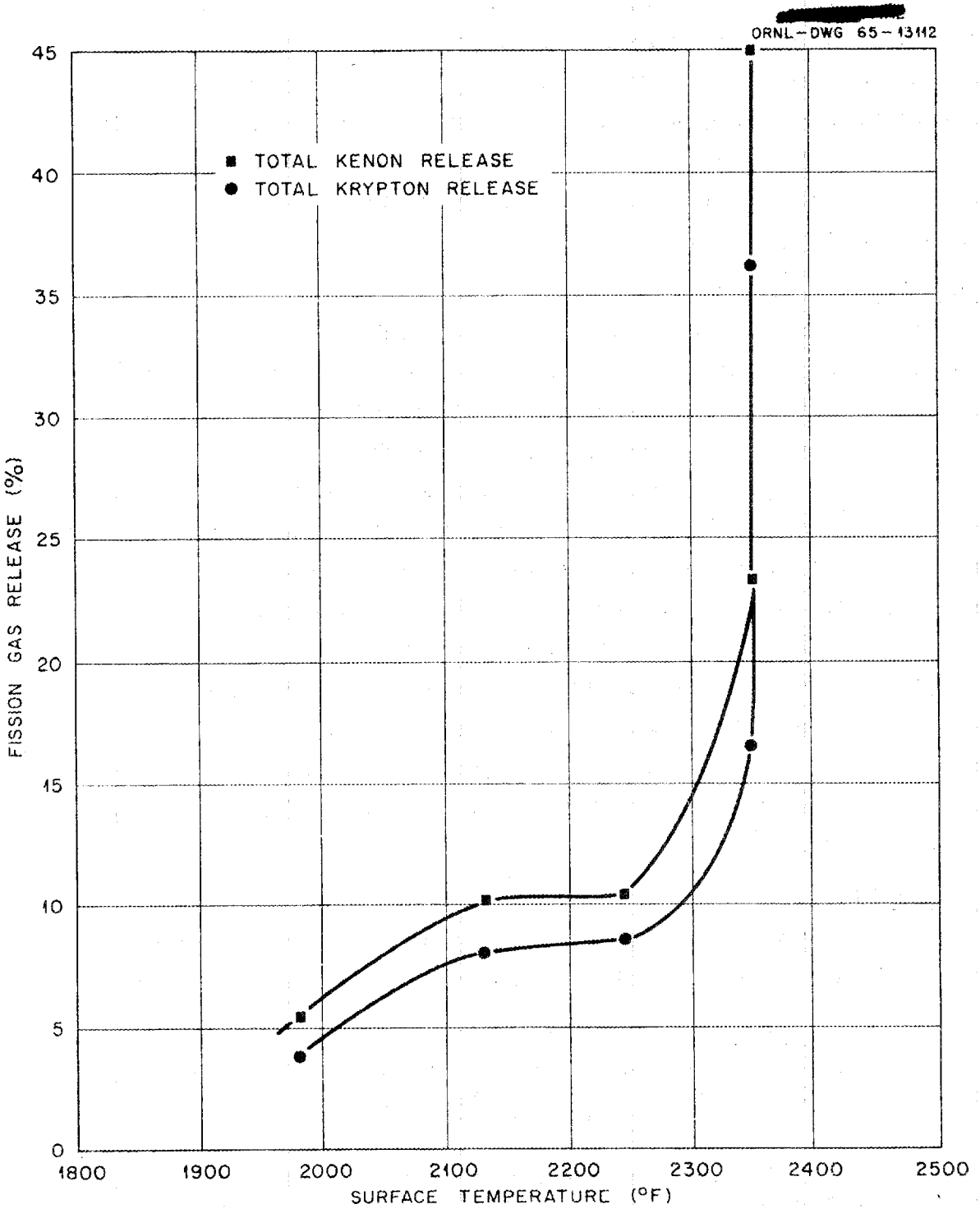

Fig. 13. UC Fission-Gas Release.

[redacted]

These data were taken from hyperstoichiometric UC ($UC_{1.45}$) irradiated to approximately 2 at. % burnup in which there was some fuel-clad reaction. These data indicate a strong temperature dependence at a surface temperature greater than 1230°C. A similar temperature dependence is shown by the xenon gas release which increases from 10% at 1230°C to 45% at 1290°C. For reactor designs based on a burnup of 2.0 at. % U and a fission-gas release of 20% or less, 1290°C appears to be a limiting temperature.

Fuel Pin Diametral Change

The fuel pin diametral increase is a result of the fuel swelling and hydrostatic pressure of released fission gases. At 1500°C the CANEL data indicate that the burnup limit for UC is about 4.9×10^{20} fissions/cm^3 (Fig. 14).[29-30] At this burnup the slope of the curve increases very rapidly. This change in slope corresponds to the same burnup at which UC exhibits a large fuel volume change. This suggests that the large pin diametral increase is a result of the excessive fuel swelling. The lack of data on fuel pin diameter changes for lower temperatures prevents a further analysis of CANEL data.

Atomics International irradiation data (Fig. 15)[31] suggests a temperature limit of about 1000°C. Below 1000°C the pin diametral increase is low and appears to be independent of temperature, while above 1000°C the diametral change increases more rapidly with increasing temperature.

Compatibility

Compatibility of UC with lithium has been studied extensively at CANEL and BMI. This is little reaction between lithium and stoichiometric

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[29]Pratt and Whitney Aircraft-CANEL, "SNAP-50/SPUR Program Engineering Progress Report April 1, 1964, to June 30, 1964," PWAC-642, p. 135, July 30, 1964. CONFIDENTIAL-RESTRICTED DATA.

[30]Pratt and Whitney Aircraft-CANEL, "SNAP-50/SPUR Program Engineering Progress Report October 1, 1964, to December 31, 1964," PWAC-644, p. 151, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

[31]"Annual Technical Progress Report, AEC Unclassified Programs," USAEC Report NAA-SR-9999, Atomics International, August 31, 1964.

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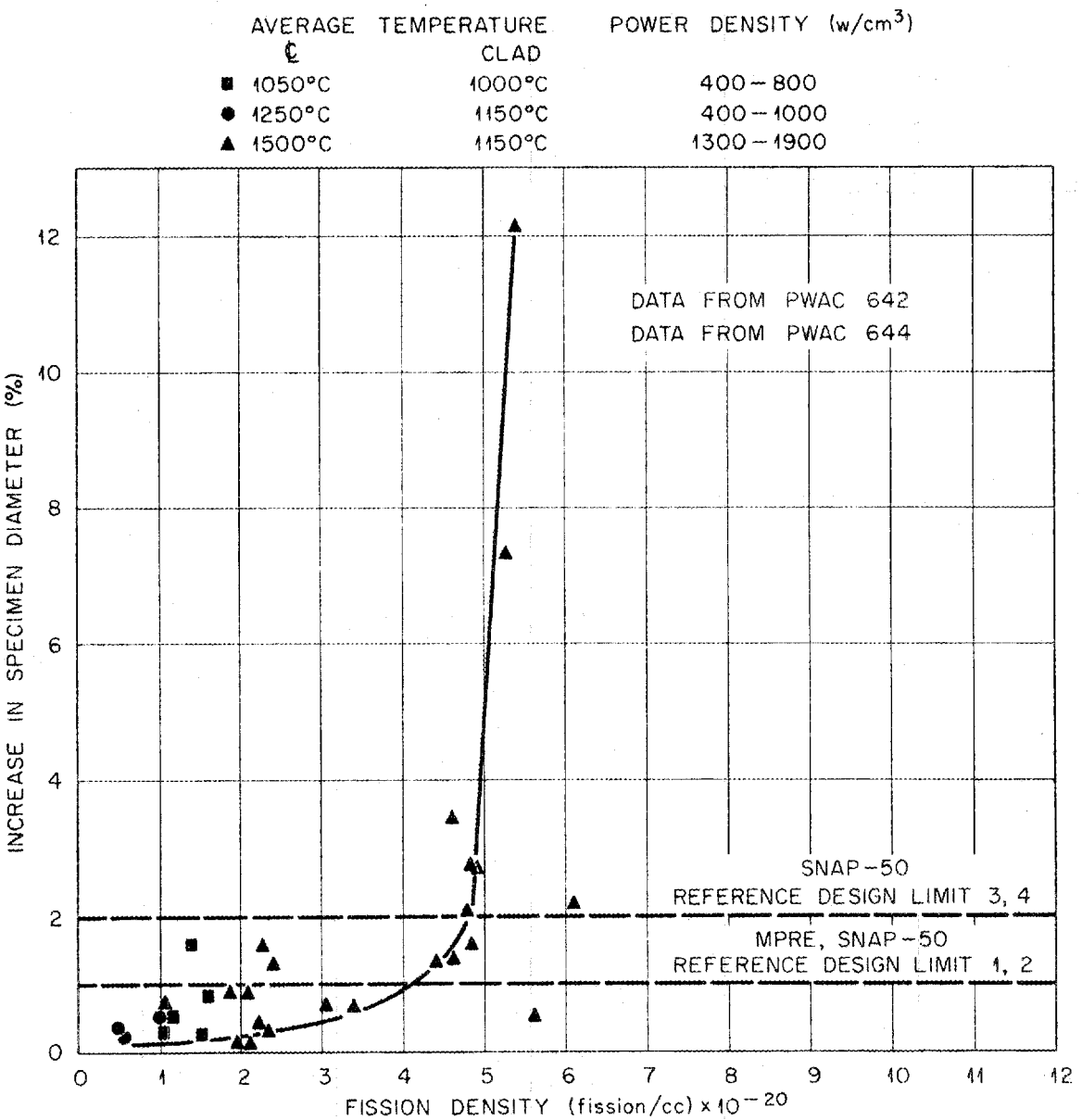

Fig. 14. UC Fuel Pin Diametral Change.

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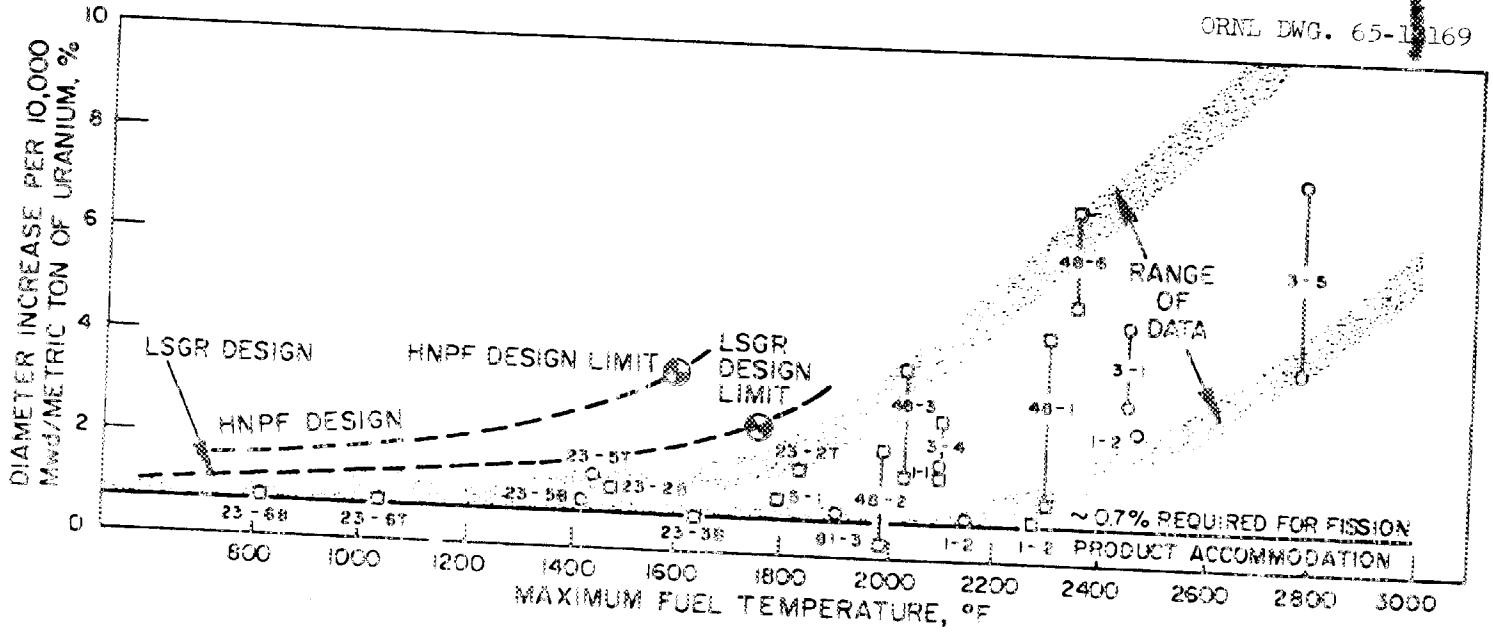

FIG. 15. *Diameter increase in UC due to irradiation.*[58] ○, *hypostoichiometric UC (< 4.8 wt.% carbon)*; □, *hyperstoichiometric UC (> 4.8 wt.% carbon).*

UC.[32] However, lithium has a high solubility for carbon and the carbon in hyperstoichiometric UC will migrate out of the fuel into the lithium. Under irradiation this carbon migration will be enhanced with a temperature gradient.

The compatibility of UC-cladding systems is shown in Table 4.[33] Compatibility does not appear to be a major problem though UC is less stable than UN. Hyperstoichiometric UC, however, is unstable in the presence of many cladding materials; for example, UC is stable in the presence of tungsten and molybdenum but forms UWC_2 and $UMoC_2$, respectively, in the presence of excess carbon.

For cladding materials that react with stoichiometric UC, the introduction of excess carbon into the UC may be beneficial. A reaction can then occur with the cladding material producing a reaction layer between the fuel and the clad. This reaction layer serves as a protective coating to prevent a detrimental fuel-clad reaction during irradiation which could result in the formation of free uranium.

Uranium Mononitride

Uranium mononitride has many advantages as a refractory ceramic reactor fuel. Among these advantages are:

(1) High thermal conductivity.

(2) High uranium density.

(3) Good compatibility with lithium.

(4) High heat rating to produce radial cracking, H_f.

(5) High heat rating to produce centerline melting, H_m.

(6) Good compatibility with most cladding materials.

The thermal conductivity (Fig. 1) of UN is the highest of the three primary fuels under investigation above approximately 700°C (Table 2). This makes possible higher power ratings from UN than from UC or UO_2.

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[32]Fackelmann et al., "Development of Advanced High-Temperature Nuclear Materials During November 1963 through January 1964," BMI-1666, p. 30, Battelle Memorial Institute, March 5, 1964. CONFIDENTIAL-RESTRICTED DATA.

[33]DeMastry and Griesenauer, "Investigation of High Temperature Refractory Metals and Alloys for Thermionic Converters," AFAPL-TR-65-29, Supplement 1, Battelle Memorial Institute, April 1965. CONFIDENTIAL-RESTRICTED DATA.

Table 4. UC Compatibility

Temp (°C)	Zr	Stainless Steel	Cb	Mo	W	W-0.9Cb	W-15Mo	W-10Re	TZM	Mo-50Re	Fe	Ta-12W	T-111	B-66	W-25Re
800	Reacts Rapidly														
870		No Reaction 6000 hr													
1100		Very Rapid Reaction	Slight Reaction												
1371					Com	Com	Com	Com	Com	Com	Com	Incom	Incom	Incom	Com
1500				No Reaction											
1538					Com	Com	Com	Com	Com	Com	Com	Incom	Incom	Incom	Com
1705					Com	Com	Com	Com	Incom	Incom	Forms UFe_2	Incom	Incom	Incom	Com
1871					Com	Com	Reacts Peritectically 1000 hr	Com	Incom	Incom	Incom	Incom	Incom	Incom	Reacts Peritectically 1000 hr

Com — Compatible, no reaction after 1000 hr.

Incom — Incompatible, gross reaction.

These data are true for stoichiometric UC only. Hyperstoichiometric UC will react with some materials in which stoichiometric UC is stable. Two examples of this, the reaction of tungsten and molybdenum with hypostoichiometric UC producing the phases: UWC_2 and $UMoC_2$.

Or, for a given power rating and surface temperature, UN will have a lower centerline temperature. The uranium density for UN, an important parameter for reducing reactor size and weight, is 13.6 g/cm^3, the highest of the refractory ceramic fuels (Table 2). The very high values for H_f, the heat rating to produce fracture, and H_m, the heat rating to produce centerline melting, insure good integrity of the fuel under extreme reactor operating conditions.

Fuel Volumetric Change

The resistance of UN to irradiation damage is a major factor in favoring its use as a reactor fuel. The volumetric increase of the fuel as denoted by the fuel density decrease is shown in Fig. 16.[34-35] The fuel densities used for these calculations were measured by standard immersion techniques. The curves denoting volume change were plotted to show the effect of temperature and fuel burnup. Examination of these curves shows that the effects of temperature are not well defined. The scatter in the 1500°C data is low for burnups above 2.5×10^{20} fissions/cm^3. At fuel burnup values less than 2.5×10^{20} fissions/cm^3 the volumetric change during irradiation drops off rapidly. No reason for this decrease is apparent. The scatter in the 1100°C curve is considerable for the range of data available (maximum 3.4×10^{20} fissions/cm^3). No experimental evidence that could account for this scatter is apparent from the data.

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[34]Pratt and Whitney Aircraft, "SNAP-50/SPUR Program Engineering Progress Report, January 1, 1965, to March 31, 1965," PWAC-651, p. 115, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

[35]Pratt and Whitney Aircraft, "SNAP-50/SPUR Program Engineering Progress Report, April 1, 1964, to June 30, 1964," PWAC-642, July 30, 1965. CONFIDENTIAL-RESTRICTED DATA.

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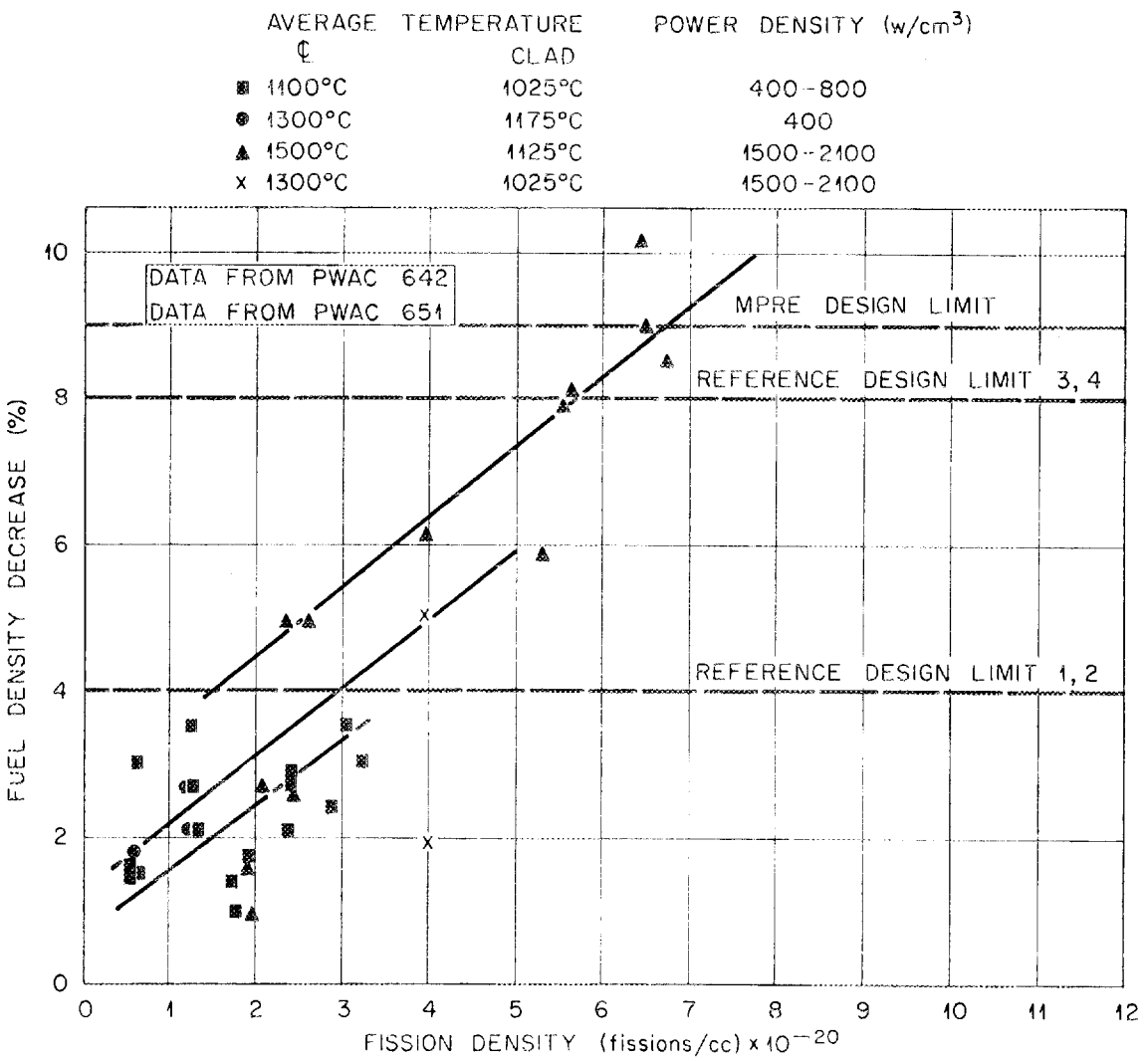

Fig. 16. UN Fuel Volume Change.

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Fission-Gas Release

For UN, fission-gas release increases with increasing temperature as shown in CANEL's irradiation data (Fig. 17).[36-37] For the limited data collected (maximum burnup 3.4×10^{20} fissions/cm^3) at centerline temperatures of 1100°C, the fission gas released is very small (approximately 0.1%). At 1500°C, however, fission-gas release is appreciable and strongly dependent on fission density. Although 1300°C data are limited, the preliminary data indicate a significant fission-gas release but less burnup dependence than the 1500°C curve of Fig. 17.

A relationship can be shown between fuel swelling and fission-gas release. The fission gas atoms agglomerate into bubbles, migrate through the fuel, and are eventually trapped at grain boundaries. The grain boundary porosity arising from these bubbles eventually becomes interconnected providing a path for gas-release to the fuel surface. Thus, as the fuel swells due to the formation of fission gas bubbles, one would expect an associated increase in fission-gas release. This relationship is demonstrated in Fig. 18.[38]

Fuel Pin Diametral Change

An increase in the diameter of fuel pins containing UN results from swelling and fission-gas release. Figure 19 shows curves from CANEL's irradiation data that illustrate the effect of burnup on changes

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[36]Pratt and Whitney Aircraft, "SNAP-50/SPUR Program Engineering Progress Report, January 1, 1965, to March 31, 1965," PWAC-651, p. 115, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

[37]Pratt and Whitney Aircraft, "SNAP-50/SPUR Program Engineering Progress Report, April 1, 1964, to June 30, 1964," PWAC-642, July 30, 1965. CONFIDENTIAL-RESTRICTED DATA.

[38]F. J. Huegel and T. Sturiale, "Advanced Materials Program for March and April, 1965," PWAC-1014, p. 49, June 15, 1964. CONFIDENTIAL-RESTRICTED DATA.

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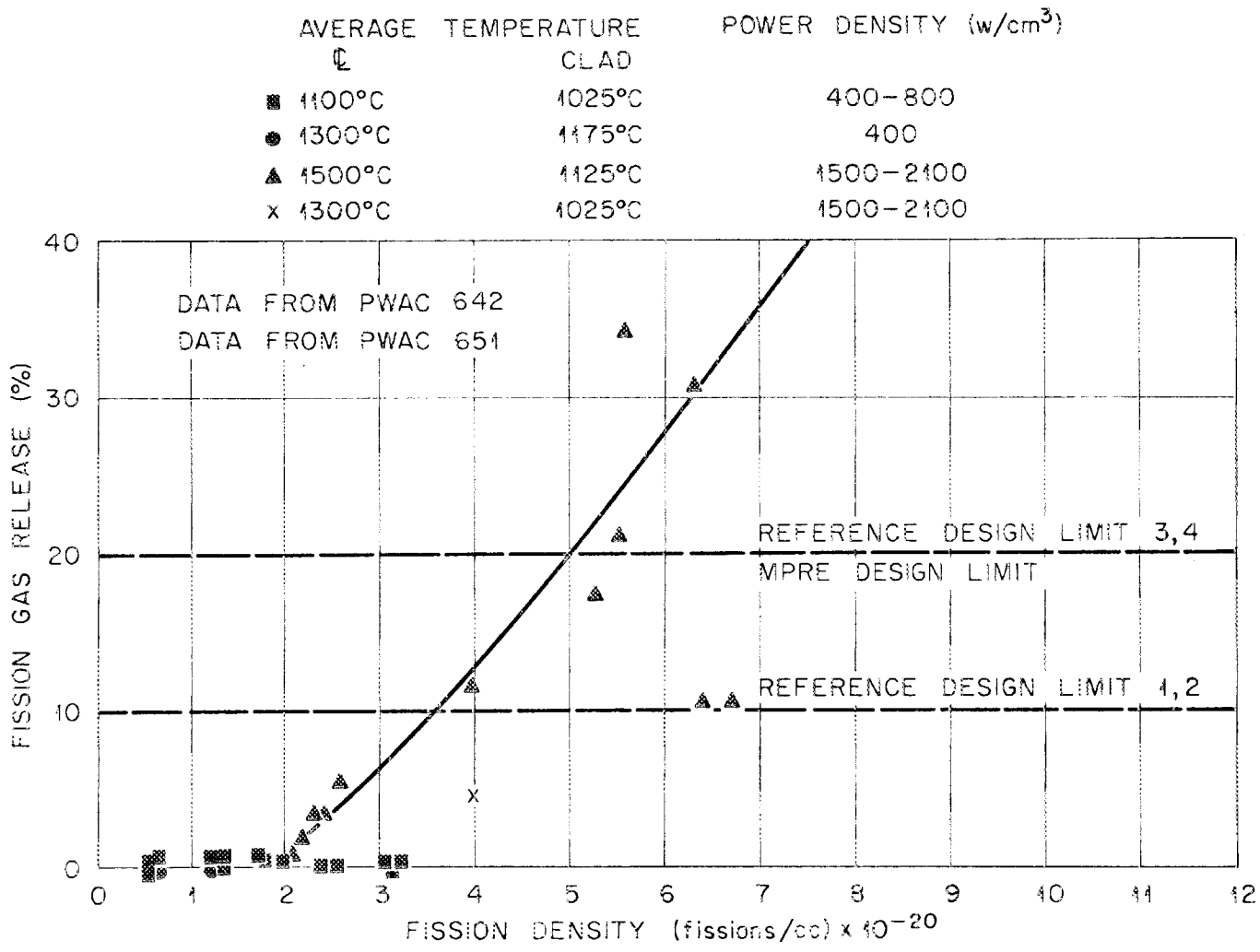

Fig. 17. UN Fission Gas Release.

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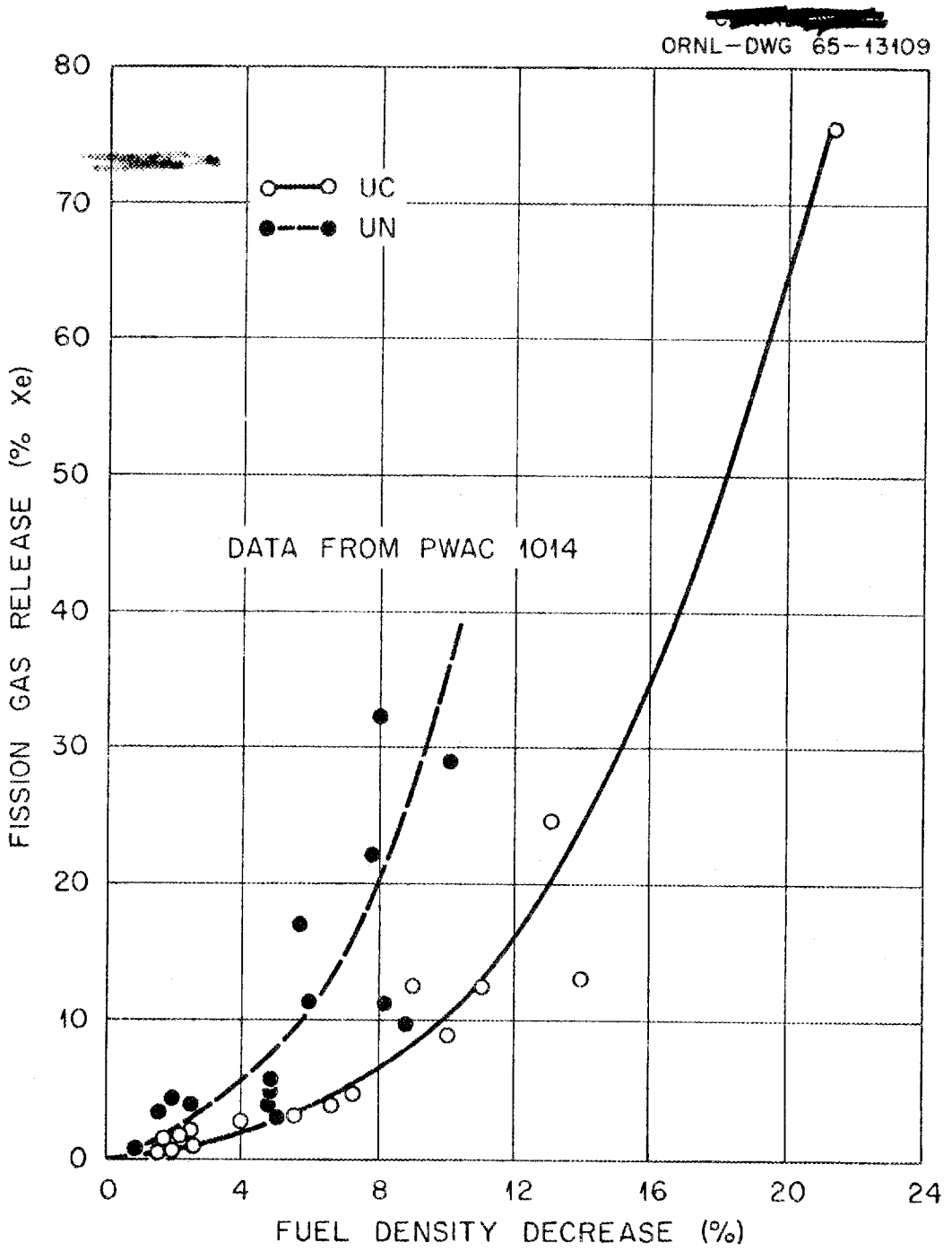

Fig. 18. Comparison of Fission Gas Release vs Fuel Density Decrease for UN and UC.

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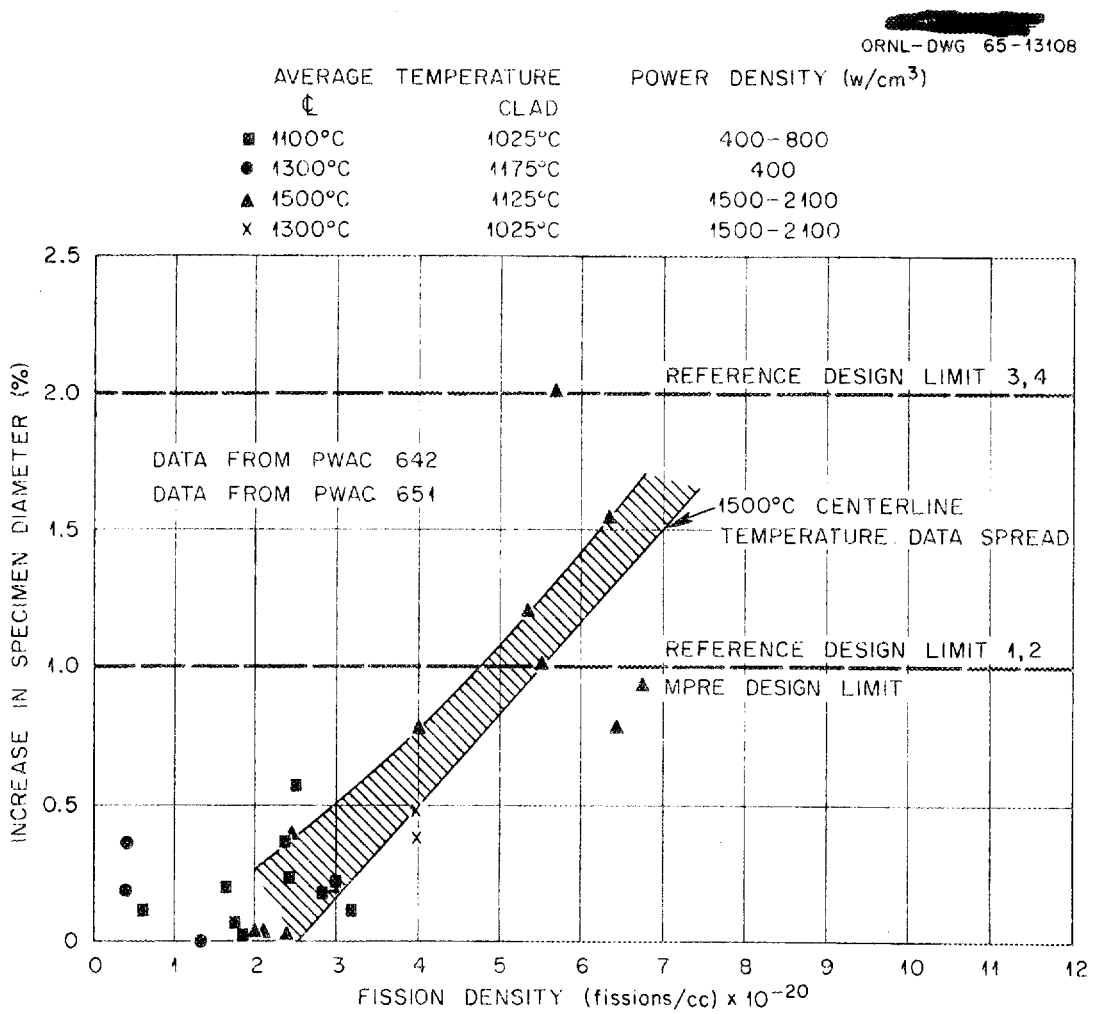

Fig. 19. UN Fuel Pin Diametral Change.

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in diameter.[39-40] The existing data are plotted for temperatures of 1100, 1300, and 1500°C. There appears to be no significant temperature dependence of the swelling. The lack of apparent temperature dependency probably results from insufficient data.

Compatibility

It has been found that UN exhibits very good compatibility with lithium under both isothermal and irradiation conditions.[41-42] The only appreciable effect detected in UN has been grain boundary attack and dissolution of UO_2 particles. By keeping the oxygen content low the effect of lithium attack should be small.

No data on compatibility of UN and potassium could be found. However, basic thermodynamic data indicate that corrosion by lithium should be much more severe than by potassium; hence UN should also be compatible with the MPRE coolant. However, before UN can be seriously considered as a fuel for the MPRE, some compatibility tests will be necessary.

An extensive study of the compatibility of UN with various cladding materials has been made by ORNL and BMI. The results of this investigation

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[39]Pratt and Whitney Aircraft, "SNAP-50/SPUR Program Engineering Progress Report, January 1, 1965, to March 31, 1965," PWAC-651, p. 115, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

[40]Pratt and Whitney Aircraft, "SNAP-50/SPUR Program Engineering Progress Report, April 1, 1964, to June 30, 1964," PWAC-642, July 30, 1965. CONFIDENTIAL-RESTRICTED DATA.

[41]W. H. Cook, "High-Temperature Materials Program Quarterly Progress Report for Period Ending October 31, 1964," USAEC Report ORNL-TM-980, pp. 111-15, Oak Ridge National Laboratory, December 1964. CONFIDENTIAL-RESTRICTED DATA.

[42]Pratt and Whitney Aircraft, "SNAP-50/SPUR Program Engineering Progress Report, January 1, 1965, to March 31, 1965," PWAC-651, p. 119, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

Table 5. UN Compatibility

Temp (°C)	W	W-0.9Cb	W-15Mo	W-10Re	W-25Re	TZM	Mo-50Re	Re	Ta-12W	T-111	B-66	Mo	Ta
1000												Very Slow Reaction	
1200												Com	125 μ Gross Intergranular Attack
1300												Com	
1371	Com	Com	Com	Com	Com	Com	Com	Com	Com	Heavy Matrix ppt, 100 hr	Grain Boundary ppt-100 G.B. Reaction Zones 1000 hr	2000 hr ppt, reaction ~200 μ	
1500													248 hr 1000 μ Gross Intergranular Attack
1538	Com	Com	Com	Com	Com	Com	Com	Com	Slight Reaction	Incom	Incom		
1705	Com	Com	Com	Com	Com	Com	Com	Com	Incom	Incom	Incom		
1871	Com	Com	Com	Com	Com	Com	Com	Com	Incom	Incom	Incom		

Com – Compatible, no reaction after 1000 hr.

Incom – Incompatible, gross reaction.

Table 5 (Continued)

Temp (°C)	D-43	Cb-1 Zr	Cb	304 Stainless Steel	Cr	Fe	Ti	U	Zr
615				No Reaction 6 Mo					
1000				No Reaction 24 hr	Reaction	No Reaction 500 hr	Forms TiN and Intermetallic Phase	Reaction	Reaction to ZrN
1200		200 μ Gross Intergranular Attack 435 hr	125 μ	Slight Reaction 1600 hr					
1300	25-30 μ Reaction 152 hr		20 μ 152 hr	Slight Reaction 1600 hr					
1500	180 μ Gross Intergranular Attack	500 μ Gross Intergranular Attack 248 hr	430 μ 2000 hr						

are shown in Table 5.[43-44] Most tungsten and Re base alloys show good compatibility with UN. The interfacial reaction between UN and other clad materials that are incompatible may be considerably reduced by a diffusion barrier between the fuel and the clad. Cb-1 Zr with a 5-mil tungsten barrier has been used very extensively in CANEL's UN irradiation capsules to date. The only notable effect has been some migration of the uranium into the Cb-1 Zr forming a U-Cb intermetallic.[45] This reaction is reduced to a reaction zone of about 25 μ as a result of the tungsten barrier.

A potential disadvantage of UN as a fuel material is its high vapor pressure[46] relative to UO_2[47] and UC[48] (Fig. 20) which allows the UN to decompose by giving off nitrogen and leaving free uranium in the UN structure. However, the equilibrium nitrogen pressure is small for the UN temperatures encountered in reactor designs of interest here (SNAP-50: 5×10^{-11} atm N over UN; MPRE: 2×10^{-13} atm N over UN). Thus, the vapor pressures for these temperatures are negligible. It is only for much higher temperature reactors that the vapor pressure will begin to effect the cladding restraint. Even at 2670°C the partial pressure of nitrogen over UN will be only 0.1 atmosphere.

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[43]DeMastry and Griesenauer, "Investigation of High Temperature Refractory Metals and Alloys for Thermionic Converters," Battelle Memorial Institute, AFAPL-TR-65-29, Suppl. 1, April 1965. CONFIDENTIAL-RESTRICTED DATA.

[44]J. F. Murdock (to be reported in), "High Temperature Program Quarterly Progress Report for Period Ending October 31, 1965," ORNL-1350, Oak Ridge National Laboratory. CONFIDENTIAL-RESTRICTED DATA.

[45]Pratt and Whitney Aircraft, "SNAP-50/SPUR Program Engineering Progress Report January 1, 1965, to March 31, 1965," PWAC-651, pp. 116-117, May 14, 1965. CONFIDENTIAL-RESTRICTED DATA.

[46]P. A. Vozella, "Advanced Materials Program for March and April, 1964," PWAC-1014, p. 57, June 1964. CONFIDENTIAL-RESTRICTED DATA.

[47]A. T. Chapman and R. E. Meadows, "The Volatility of $UO_{2\pm x}$ and Phase Relations in the Uranium-Oxygen System," USAEC Report ORNL-3587, p. 17, Oak Ridge National Laboratory, April 1964.

[48]M. A. DeCrescente and A. D. Miller, "High Temperature Properties of Uranium Carbide," Carbides in Nuclear Energy, UKAEC, 1964.

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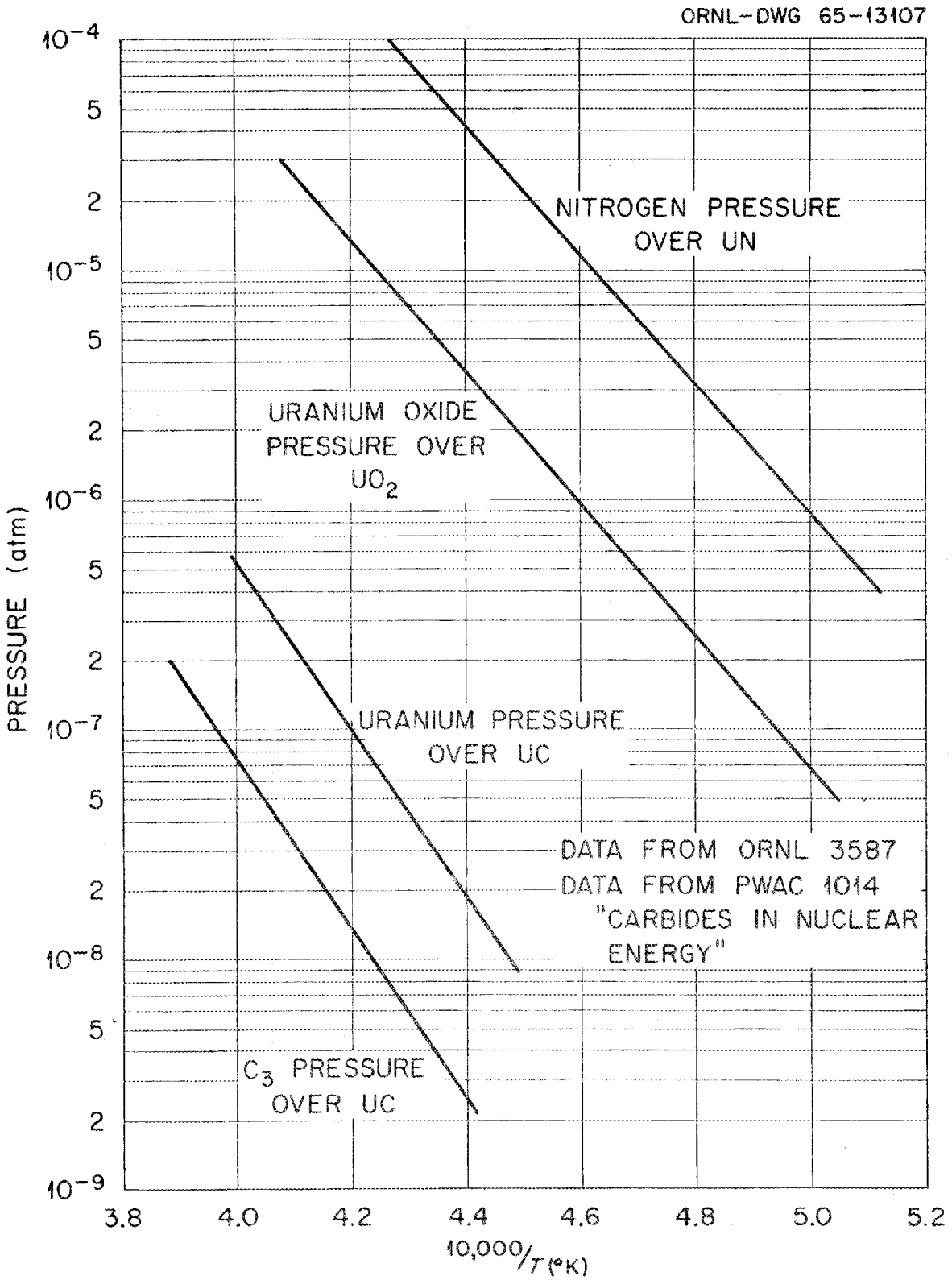

Fig. 20. Vapor Pressure of Fuels.

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Dispersion Fuels

The advantages of using dispersion fuels are:

(1) Increased thermal conductivity since most of the heat is conducted through the metal matrix material.

(2) Low fission-gas release.

(3) Constraint of fuel swelling.

A disadvantage of high temperature dispersion fuels is the difficulty in fabricating matrix materials which are stable at high temperatures. To circumvent this problem, the initial irradiation testing was done on UO_2 dispersed in stainless steel at low temperatures. As technology develops, the data obtained from irradiated UO_2-SS dispersions can be applied toward a dispersion matrix which can be used at higher temperatures. The dimensional stability of irradiated UO_2-stainless steel dispersions is shown as a function of fission density and surface temperature in Fig. 21.[49] A second disadvantage of dispersion fuels is the decrease in uranium loading which limits the total power output of the fuel elements.

BeO-UO_2

Considerable research was done on the system UO_2-BeO at Pratt-Whitney-CANEL.[50] Irradiation experiments showed UO_2-BeO pellets to be undesirable for such high-temperature irradiation environments as SNAP-50 (Table 1) since fission damage to the BeO in this high temperature range appears to cause extensive swelling and changes in properties of the material. Also, the low value of H_f, the heat rating to produce radial cracking, suggests that cracking and enhanced fission-gas release will occur at relatively low power ratings. In addition, a fast flux reaction occurs producing helium gas and tritium at the rate of approximately 3×10^{12} atoms/sec/cc. CANEL evaluation of the

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[49]W. C. Thurber and F. R. McQuilkin, "Metals and Ceramics Division Annual Progress Report for Period Ending May 31, 1963," USAEC Report ORNL-3740, p. 230, Oak Ridge National Laboratory, November 4, 1963.

[50]M. S. Freed, M. A. DeCrescente, and R. M. Kuhns, "Development of UO_2-BeO Fuels," PWAC-436, July 14, 1965.

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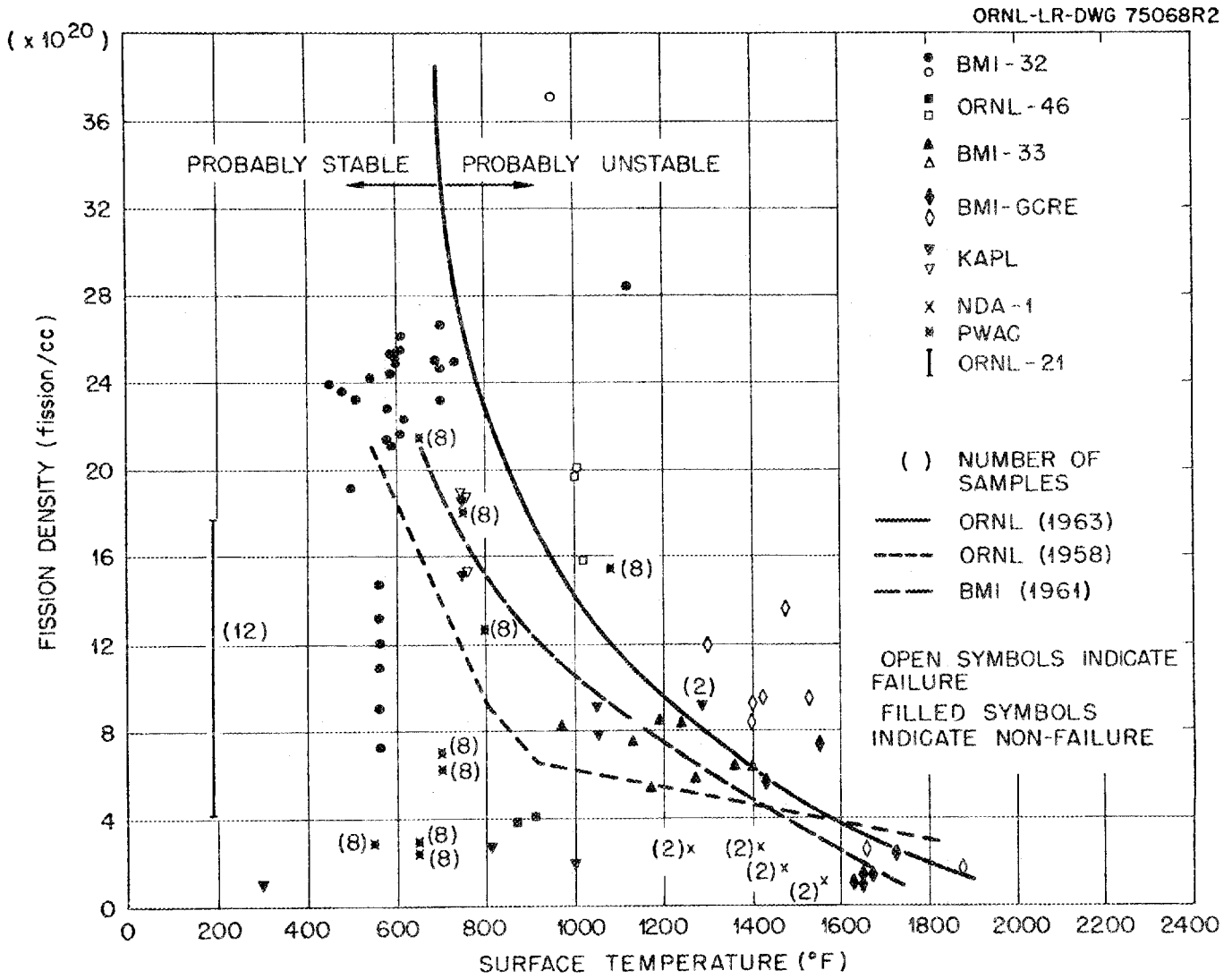

Fig. 21. Irradiation Stability of UO_2 -Stainless Steel Dispersions.

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irradiation data set the following limits on the use of UO_2-BeO (30 to 60 vol % UO_2) from the standpoint of diametral increase (2% maximum) and fission-gas release (10% maximum):

Burnup (fissions/cm^3 fuel)	Clad Temperature
11×10^{20}	1000°C
4.2×10^{20}	1010-1090°C
2.63×10^{20}	1100°C

Another factor to be considered in evaluating BeO-UO_2 is its cladding and materials compatibility. UO_2 and BeO exhibit good compatibility with most of the refractory metals. The temperature at which reactions are observed between BeO, UO_2, and these metals is shown in Table 6.

Table 6. Compatibility of BeO and UO_2 with Refractory Metals[51]

Metal	Approximate Reaction Temperature (°C)			
	BeO		UO_2	
	First	Fast	First	Fast
Niobium	1480	2135	2065	--
Tungsten	2140	2310	1785	2600
Tantalum	2245	2355	2295	2420
Molybdenum	No reaction to 2130°C		No reaction to 2155°C	

[51]J. M. Kerr, previously of Oak Ridge National Laboratory, personal communication to J. L. Scott, Oak Ridge National Laboratory.

UO_2-BeO systems cannot be considered for lithium-cooled reactors because of the exothermic UO_2–lithium reaction. Potassium being less reactive than lithium, however, does not pose any compatibility problems with UO_2–BeO.

W–UO_2 and Mo–UO_2

Two other dispersion fuels being examined for high temperature applications are W–UO_2 and Mo–UO_2. An advantage of W and Mo as a dispersion fuel matrix is the high thermal conductivity compared to UO_2 as a bulk fuel. The values of H_f and H_m for the UO_2 in W or Mo matrices are greater than bulk UO_2 by a factor of about 30. One major problem in producing a satisfactory fuel with Mo or W as a matrix is the difficulty in fabricating these materials. Both materials are brittle at low temperatures so high fabrication temperatures are required. Good atmospheric control is also important since they react rapidly with air at the high temperatures required for fabrication. An extensive research and development program will be essential prior to serious consideration of these materials for use in a reactor.

Irradiation data on W and Mo as dispersion fuel matrices is very limited. The Hanford Atomic Products Operation, General Electric[52] reports a successful short time irradiation experiment of three W–50 wt % UO_2 cermets to check the chemical and dimensional stability. The specimens were irradiated for 2-4 hr at temperatures greater than 2100°C. A literature search revealed no further irradiation data.

Conclusion

To determine the relative advantages and disadvantages of the fuels, important irradiation and thermal effects were evaluated. The results of this evaluation show that some temperature and burnup limitations can be placed on the fuels at points where the fuels show excessive swelling or fission-gas release.

UO_2

(1) In the temperature range 725 to 1610°C the slope of the fuel swelling curve increases by a factor of 4 at 15×10^{20} fissions/cm^3.

(2) Fission-gas release places a centerline temperature limitation of 1600°C on UO_2. Below 1600°C the fission-gas release rate is small. Above this temperature the gas release rate increases rapidly with temperature.

UC

(1) CANEL data indicate a burnup limitation of 4.9×10^{20} fissions/cm^3 at a fuel centerline temperature of 1500°C.

(2) BMI and AI data suggest a centerline temperature limitation of 1200°C at 2.0 at. % burnup.

UN

The available data for UN are not sufficient to define temperature and burnup limitations similar to those established for UO_2 and UC.

In comparing UN and UC, the curves for the maximum diametral increase (Fig. 22), the fission-gas release (Fig. 23), and fuel swelling (Fig. 24) with a 1500°C centerline temperature were drawn for both fuels on the same coordinates. At this temperature, UN is obviously superior to UC.

Differences in fuel properties and resistance to radiation damage suggest that some fuels are superior to others for specific reactor designs. To compare the relative advantages of these fuels for SNAP-50 and MPRE designs, the maximum radiation damage to each fuel corresponding to the design temperatures and burnups were taken from the curves in this report. These data are tabulated in Table 7. For the SNAP-50 design parameters UN is clearly superior to UC. UO_2 cannot be considered for lithium-cooled reactors because of its incompatibility with lithium. However, UO_2 should not be excluded from consideration of SNAP-50 designs since UO_2 should exhibit little radiation damage for the given design conditions. If potassium is used as the primary coolant in place of lithium, there should not be a compatibility problem. The

[redacted]

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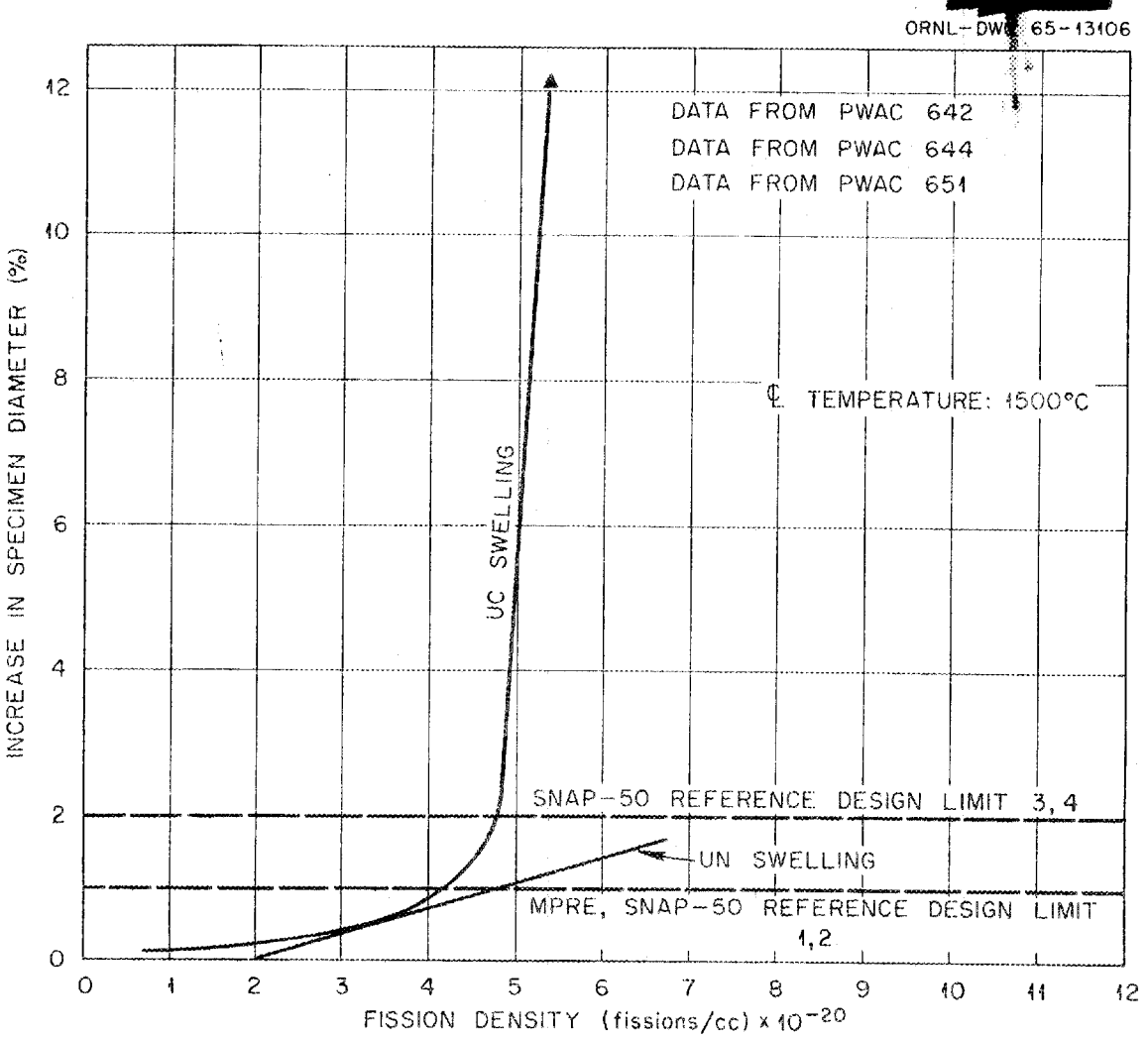

Fig. 22. UN - UC Diametral Pin Swelling.

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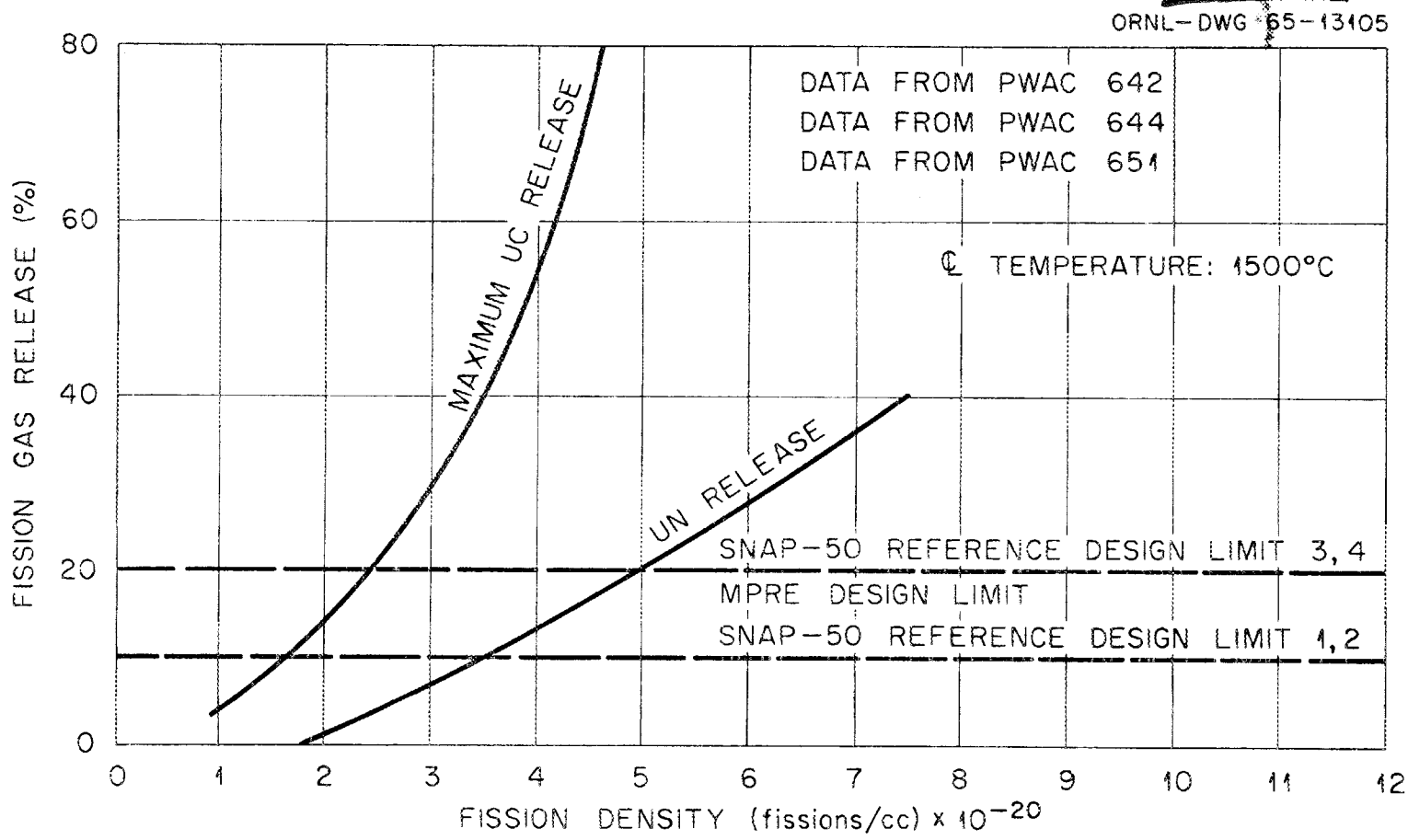

Fig. 23. UN – UC Fission Gas Release.

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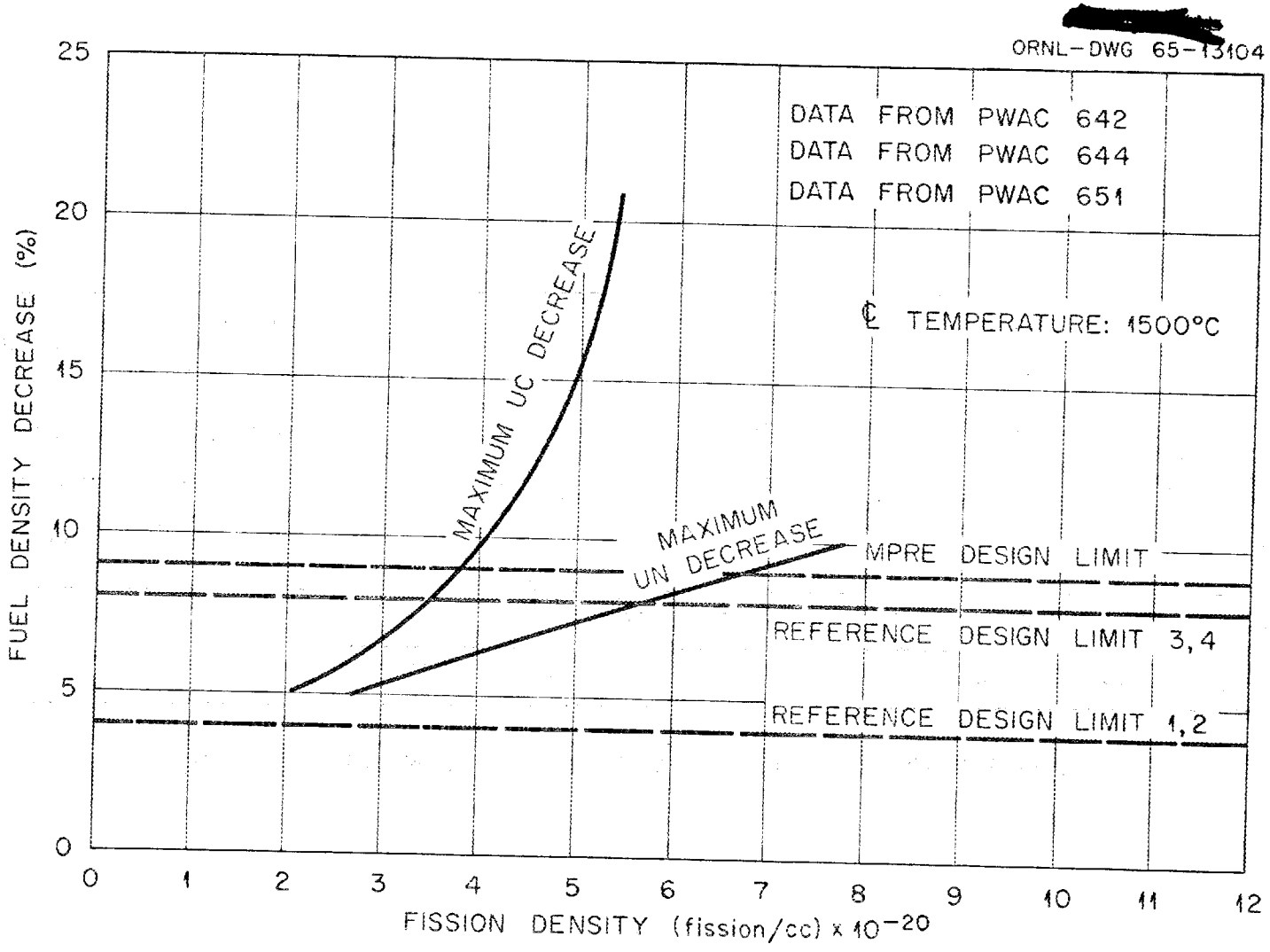

Fig. 24. UN - UC Fuel Volume Change.

[redacted]

Table 7. Postulated Irradiation Effects on Fuel for MPRE, SNAP-50

	MPRE				SNAP-50 (Reference 3)			
	UO_2	UN	UC	Design	UO_2	UN	UC	Design
Fission Gas Release, %	1	Neg	2–7	5%	1	8	100	20
Fuel Density Decrease, %	3	1	3–4	3%	3	4.6	11	8
Diametral Pin Swelling, %	Neg	Neg	2	1%	1	1%	5–9	2
Centerline Temperature (Assume: Gap Conductance of 0.25 w/cm^2)	1480°C	1100°C	1100°C		1450°C	1300°C	1300°C	

dispersion fuel UO_2–BeO must also be rejected as a SNAP-50 candidate fuel since SNAP-50 irradiation conditions would cause excessive swelling and fission-gas release. In addition, lithium is not compatible with the UO_2–BeO fuel system.

For the MPRE reactor design there appears to be no significant difference in fuel performance between UN and UO_2. UC exhibits more radiation damage than UN and UO_2 but might be considered as a borderline candidate fuel since the irradiation effects do not greatly exceed the design limits. The dispersion fuel UO_2–BeO show fuel swelling and fission-gas release which exceed the MPRE design specifications. Also, the maximum fuel power rating should exceed the calculated heat rating to produce fracture. From an economic standpoint UO_2 is superior to the other fuels since most fabrication and handling problems have been eliminated. Another advantage of considering UO_2 for MPRE designs is that the Oak Ridge National Laboratory has extensively tested UO_2 fuel rods at MPRE design conditions. These tests indicate that UO_2 is a good fuel for this reactor design.

Another important factor which should be considered when comparing different fuel materials for a given application is the gap conductance between the fuel and the cladding. If a poor gap conductance and a high heat flux exist, the surface temperature of the fuel may approach or exceed the performance limit set by fuel swelling or fission-gas release. Good gap conductance is particularly important for high temperature applications such as SNAP-50. For lower temperatures such as the 850°C maximum clad temperature for the MPRE, a poor gap conductance is less significant. For example, UO_2 has a temperature limit of 1600°C which is well above the maximum clad surface temperature of 850°C. Experimental work is needed in this area.

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