ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

STUDIES OF FAST REACTOR FUEL ELEMENT BEHAVIOR
UNDER TRANSIENT HEATING TO FAILURE

*III. In-pile Experiments on
Irradiated UO₂ Fuel Pins
in the Absence of Coolant*

by

R. R. Stewart, C. E. Dickerman,
L. E. Robinson

Reactor Physics Division

and

W. B. Doe

Reactor Operations Division

Part I is ANL-6334
Part II is ANL-6845

March 1969

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	7
1. INTRODUCTION	7
II. EQUIPMENT AND PROCEDURES	10
A. TREAT	10
B. Transparent Meltdown Apparatus and Instrumentation	11
C. Reactor Settings	12
III. EXPERIMENTAL RESULTS	12
A. TREAT Irradiation Conditions	12
B. Post-TREAT Transient Results	14
1. Transient 1018 (Pin 1)	14
2. Transient 1020 (Pin 11)	14
3. Transient 1022 (Pin 3)	16
4. Transient 1024 (Pin 9)	16
5. Transient 1072 (Pin 16)	18
IV. TEMPERATURE CALCULATIONS	23
V. DISCUSSION	26
ACKNOWLEDGMENTS	27
REFERENCES	28

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	TREAT Cutaway, Showing Viewing Slot Extending into Center of Core	10
2.	Transparent Meltdown Assembly	11
3.	Cladding Temperature Data from Transient 1017 (Pin 1)	14
4.	Cladding Temperature Data from Transient 1018 (Pin 1)	14
5.	Post-TREAT Condition of Pin 1	15
6.	Cladding Temperature Data from Transient 1020 (Pin 11). . . .	15
7.	Post-TREAT Condition of Pin 11	15
8.	Cladding Temperature Data from Transient 1022 (Pin 3)	16
9.	Post-TREAT Conditions of Pins 3 and 9	17
10.	Cladding Temperature Data from Transient 1024 (Pin 9)	18
11.	Cladding Temperature Data from Transient 1072 (Pin 16). . . .	18
12.	Still Frames from 2000-frame/sec Photographic Record of Transient 1072 (Pin 16)	19
13.	Post-TREAT Condition of Pin 16 before Slot Liner Was Removed from Core.	20
14.	Post-TREAT Condition of Pin 16 after Slot Liner Was Removed from Core.	21
15.	Closeup of Pin 16 Remains, Showing Two Views of Cylindrical 6 a/o Burnup Oxide with Surface Bubbles Typical of Oxide Fuel Remains after Transient 1072.	21
16.	Closeup of Pin 16 Remains, Showing Typical Pellet Fusion and Typical Pellet Cross Section after Transient 1072.	22
17.	Photomicrograph of 6 a/o Burnup UO_2 , Showing a Porous Bubble at the Fuel Surface	22
18.	Calculated Radial Temperature Distributions of Fuel during Steady-state Irradiation	23
19.	Calculated Temperature Curves of Cladding Surface for Transient 1017, with Fuel-cladding Gap Conductance as Parameters	24
20.	Experimental Values of Maximum Cladding Temperature as a Function of Estimated Sample Energy Input	24

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
I.	TREAT Irradiation Conditions	12
II.	Sample Temperature Data.	13
III.	Relative Radial Power Factors	24
IV.	Summary of Transient-temperature Calculations on "Final" Transients	25
V.	Effects of Oxide Conductivity on Calculated Temperatures . . .	25

STUDIES OF FAST REACTOR FUEL ELEMENT BEHAVIOR UNDER TRANSIENT HEATING TO FAILURE

III. In-pile Experiments on Irradiated UO_2 Fuel Pins in the Absence of Coolant

by

R. R. Stewart, C. E. Dickerman,
L. E. Robinson, and W. B. Doe

ABSTRACT

Studies of the behavior of stainless steel-clad uranium oxide fast-reactor type fuel pins under accident-simulating temperature excursions are under way in the TREAT reactor.

This report discusses experiments on five preirradiated gas-bonded UO_2 pins. The pins were tested in TREAT inside transparent capsules to permit recording of events by high-speed color photography. Fast-response thermocouples provided cladding surface-temperature data. These pins had been previously irradiated to a maximum of 6 a/o nominal burnup, at an initial power density of 2.5 kW/cc, producing initial fuel centerline and surface temperatures of 2400 and 1215°C respectively.

One pin was heated to a temperature just below that of cladding failure due to melting. Experiments on two other pins produced maximum fuel temperatures equal to the oxide melting point and resulted in severe cladding failure. Two pins were heated to temperatures in the intermediate range between these two extremes.

In theory, fission gases are capable of producing rapid fuel motion or even explosive fuel disassembly under transient heating. However, no fuel motions of these types were observed.

I. INTRODUCTION

The study of core-meltdown phenomena for fast reactors has occupied a prominent position in fast-reactor safety efforts.¹ This arises principally for three reasons: First, the core design of the typical fast power reactor is far from the most efficient nuclear arrangement, so that large

amounts of excess reactivity are available if the core were rearranged into a more compact mass by a meltdown. Second, for many designs of interest, the worth of sodium is negative (at least in some parts of the core), and expulsion of coolant during an accident could sustain or increase the severity of the incident. Third, the prompt-neutron lifetime is so short that shutdown following an accident that could lead to a super-prompt-critical excursion must be done by the physics of the core itself (Doppler feedback due to heating of the fertile atoms, increased neutron leakage due to thermal expansions, disassembly of fuel under internal pressures, etc.). Because of these reasons, the accident analyst must not only consider the "large" accidents leading directly to serious core damage, but also continue the analysis of smaller more credible accidents to their conclusions in order to ascertain if a large accident can be triggered.

Phenomena that must be understood, at least in a semiquantitative manner, include: types of prefailure fuel movement, pressure generation inside fuel, thresholds of failure, modes of failure, changes in fuel during temperature excursions, fuel motion arising from failure, pressure generation associated with failure, coolant movements arising from failure, and a broad range of possible postfailure phenomena, which could propagate and cause a super-prompt-critical secondary accident. These phenomena are being examined in an experimental program of in-pile studies using the Transient Reactor Test Facility (TREAT).²

Metallic fuel of the Experimental Breeder Reactor II (EBR-II) Mark-I design³ has been the type most extensively investigated in TREAT thus far.² The behavior under transient heating in an inert gas was studied first;⁴ the fuel movements and failure modes were observed directly, using high-speed photography;⁵ kinetics of cladding dissolution were studied;⁶ effects of a stagnant coolant environment on failure were investigated;⁷ fuel-behavior characteristics arising from prior irradiation were surveyed;⁸ and the initial experiments were performed on samples in flowing-sodium loops.^{9,10}

The properties that influence the behavior of oxide fuel are different from those of the sodium-bonded, metallic, EBR-II pins. The oxide has a much higher melting point, a much lower thermal conductivity, and a low-conductance region between fuel and cladding, instead of a high-conductance thermal bond; it is a brittle material, which develops cracks during steady-state service; and it has a tendency to form a central void in operation.

Results have been reported for TREAT experiments on the transient behavior of unirradiated gas-bonded UO₂ pins, clad with half-length EBR-II Mark-I cladding tubes.¹¹ Those studies investigated failure mechanisms for steel-, tantalum-, and niobium-clad oxide pins, the physical changes brought about in the fuel material itself by rapid heating, the magnitude and variations in fuel-cladding gap conductance, and effects of trapped gas on fuel-pin

behavior. Transient experiments using TREAT are also being performed by Argonne National Laboratory on larger-diameter oxide fuel, as part of the USAEC safety program on water-cooled thermal reactors.¹²

Engineering tests are being conducted by General Electric in TREAT, on gas-bonded oxide fuel pins bonded by NaK to a heat sink. Large-diameter oxide pins of the SEFOR type were tested to establish transient operating limits.¹³ Operating limits and incipient failure were also studied for mixed oxide fuel of typical fast-reactor design dimensions, both unirradiated and irradiated.¹⁴⁻¹⁷

The experiments reported here were conducted as a continuation of the systematic investigation into accident behavior of oxide fuel, begun with unirradiated specimens.¹¹ Twenty steel-clad, gas-bonded pins of the same type were irradiated in the Materials Testing Reactor (MTR) to a range of burnups extending from a nominal 0.7 a/o to a nominal 6 a/o. Preliminary reports have been issued on the postirradiation condition of the lower-burnup (0.7 and 3.0 a/o nominal burnup levels) pins.¹⁸ Five of the irradiated pins were run in TREAT transient experiments; the results from the first four have been summarized.¹⁹

These experiments had two principal goals: first, characterization of the fuel behavior arising from transient heating, including the interrelated effects from fission products and the changes in physical structure of the oxide fuel; second, characterization of the gross behavior of the entire pins, including failure modes, checks on magnitude and degree of consistency of fuel-cladding thermal conductance, possible fuel-cladding interactions, fuel slumping or ratcheting inside cladding, and fuel motion upon failure of the cladding.

Each TREAT sample was exposed, in an inert-gas environment, in a standard TREAT transparent meltdown facility developed for experiments²⁰ with irradiated fuel pins. With this type of experimental arrangement, we could instrument the fuel pins with fast-response cladding thermocouples, follow the course of each experiment directly using high-speed color photography, and observe the sample remains afterward without the possible misleading results of liquid-metal, coolant-induced motions on pressure pulses. For experimental simplifications in handling, UO_2 was used for these experiments instead of the mixed oxide ($\text{PuO}_2\text{-UO}_2$) under development for fast breeder reactors. The results from these studies should prove adequate for providing insight into the behavior of irradiated mixed oxide under accident conditions. With these experiments as a basis, and the firming up of specific fuel designs, however, mixed oxide samples would seem to be required.

Of the 20 UO_2 pins irradiated, five were sacrificed as control specimens, and five were run in TREAT transparent capsules. The

remaining have been set aside for experimentation on the effects of stagnant or flowing-sodium environments with respect to failure and transient-pressure-generation phenomena.

The five irradiated UO_2 pins that were sacrificed as control specimens are the subject of a separate report,²¹ which discusses the uranium-dioxide fuel fabrication, the irradiation conditions, and the postirradiation condition. The postirradiation condition is described by an extensive fuel analysis and characterization study, which is documented by charts, tables, neutron and X-ray radiographs, photographs, photomicrographs, and beta-gamma and alpha radiographs, which may be used as a comparison for future studies involving uranium oxides and/or mixed plutonium-uranium oxides. A large portion of the extensive fuel analysis presented in Ref.21 was made possible through the use of an electron-probe microanalyzer.

II. EQUIPMENT AND PROCEDURES

A. TREAT

Figure 1 is a cutaway view of the TREAT reactor. TREAT is a graphite-moderated and -reflected, pulsed engineering test reactor designed and built to meet the needs of the Fast Reactor Safety Program for a versatile facility capable of safely generating a high, time-integrated, neutron flux

112-771

Fig. 1. TREAT Cutaway, Showing Viewing Slot Extending into Center of Core

over a comparatively large sample volume.²² The reactor engineering design has been reported.²³ Briefly, the reactor core is approximately 120 cm high, with top and bottom reflectors about 60 cm thick. Each fuel element is 10 cm square. Special elements with large slots, nominally 56 cm high and 7 cm wide, can be loaded into the reactor to provide a large core-viewing slot. One such slot is shown in Fig. 1, extending from the floodlight to the center of the core.

Experimental samples may be exposed in containers designed to fit into the hole left by removal of one or more TREAT fuel elements, or in containers that fit in a core slot. Despite the large perturbations in core neutron-flux distribution caused by the slots and experimental samples, the kinetics analysis²⁴ that fits the transient behavior of the reactor loaded with a simple solid core²⁵ was found to fit (with a 10% reduction in the temperature coefficient of reactivity) data for a typical slotted-core loading with a sample.²⁶ TREAT has an inherent negative coefficient of reactivity produced by the change in effective thermal-neutron temperature as the core heats up during a transient. Because of the finely divided fuel-moderator mixture, heat transfer to the moderator is much faster (time constant of the order of 1 msec) than the shortest reactor period (about 40 msec at the time these experiments were performed).

B. Transparent Meltdown Apparatus and Instrumentation

The irradiated UO_2 pins were exposed in TREAT, using the standard transparent meltdown apparatus developed for use with highly active, pre-irradiated sample pins.²⁰ Figure 2 shows the basic layout of the transparent

112-1223

Fig. 2. Transparent Meltdown Assembly

meltdown assembly. (This general layout is the same for both unirradiated and preirradiated specimens, although the design details of the two types of apparatus are different.) The sample is held inside a Zircaloy capsule, which provides primary (but not sealed) containment. The capsule is held within a graphite- and Zircaloy-lined, steel slot liner, which provides secondary containment and a sealed system for an inert-gas cover during the experiment. The slot liner assembly was loaded at the ANL Illinois Hot

Laboratory and shipped to TREAT in special shipping casks.²⁰ At TREAT, each liner assembly was transferred to a shielded handling coffin, and loaded into the TREAT viewing slot depicted in Fig. 1.

Each sample was instrumented with two bare-wire Chromel-Alumel thermocouples spot-welded to the cladding surface at about ± 2.5 cm from the axial center of the fuel.

Camera speed for the experiments was set at 2000 frames/sec.

C. Reactor Settings

TREAT settings were provisionally based on the relationship between reactor power and sample power used in the earlier experiments on unirradiated UO_2 pins,¹¹ corrected for the 30% change in sample enrichment and the ^{235}U depletion produced by steady-state irradiation. Each reactor power burst was of about a half-second duration at half maximum power. Sample fuel transient-energy input ranged from about 700 J/g to about 1150 J/g. Sample fuel heating rates were about 2000°C per sec. Each pin was given a low-energy preliminary burst of about 450 J/g to produce a cladding temperature rise of 800°C to provide checks on the calculated dependence of sample temperature on reactor energy release (this relationship is dependent on sample burnup) and the magnitude and consistency of heat-transfer rates from fuel to cladding.

III. EXPERIMENTAL RESULTS

A. TREAT Irradiation Conditions

Table I summarizes the TREAT irradiation conditions. The values of estimated sample burnup are based on radiochemical results from the monitor wires²⁷ (see Table I, normalized* to agree with direct measurements of burnup performed by mass spectrometer²⁸ on four control samples. Energy

TABLE I. TREAT Irradiation Conditions^a

Sample	Pin Reference No.	Estimated Burnup a/o	Energy Input, J/g	Calculated Max Fuel Temp, °C	Calculated Fuel Surface Temp at Time of Max Fuel Temp, °C
1	1	0.56	757	2131	1582
2	11	3.10	1141	2750 ^b	2011-2220 ^b
3	3	0.73	869	2464	1809
4	9	3.66	728	2045	1508
5	16	5.85	1151	2750 ^c	2060-2175 ^c

^aLow-power preliminary transients are omitted for clarity.

^bFuel at melting point for about 0.4 sec.

^cFuel at melting point for about 0.5 sec.

*The mass spectrometer gave results about 29.5% below the monitor wire data for low burnup, and results about 6% below the monitor wire values for high burnup. (See Table VI in Ref. 21 for comparison with values given in Table I.)

inputs (joules/gram of UO_2) in the table are based on the ratio of sample power to reactor power used previously,¹¹ corrected for sample burnup, with an additional correction of -9% applied to obtain agreement between calculated and measured cladding surface temperatures for the preliminary transients. (This is considered to be good agreement. It is well within the change in power ratio that resulted from the change in core loading from that used previously¹¹ to the loading with full viewing slot used in these experiments. This core change was made to permit simultaneous tests on operation of the fast-neutron hodoscope.)²⁹ Temperatures are those calculated using the ARGUS code³⁰ in which a fuel-cladding interface conductance of $0.25 \text{ W/cm}^2\text{-}^\circ\text{C}$ was used. The latter was deduced from analyses of the curves of cladding surface temperature versus time from the preliminary transients.

The first sample, a 0.7 a/o burnup pin, survived a transient producing a maximum cladding temperature of 1275°C without cladding failure. One 0.7 a/o and one 3 a/o burnup sample was given a transient calculated to produce temperatures in excess of those attained in steady-state irradiation, but without melting the fuel; cladding on both pins failed. The remaining 3 a/o burnup pin was given a high-energy transient (programmed to attain the fuel melting point) and experienced cladding failure. To complete this study, a 6 a/o burnup pin was given an energy input nominally identical with the most severe of the previous exposures. This 6 a/o burnup pin also underwent cladding failure.

Temperature data are presented in Table II. For reference the reactor energy-release values are listed along with the sample energy inputs. The preliminary transients are included in the table.

TABLE II. Sample Temperature Data

Sample No.	Pin No.	TREAT Transient	TREAT Energy, MW-sec	Sample Energy, J/g	Max Temp of Cladding Thermocouples, $^\circ\text{C}$	
					Lower	Upper
1 ^a	1	1017	48	448	815	750
1	1	1018	81	757	1220	1145
2 ^a	11	1019	54	391	b	815
2	11	1020	155	1141	b	1370
3 ^a	3	1021	47	437	750	750
3	3	1022	94	869	1320 ^c	1340 ^c
4 ^a	9	1023	53	351	830	815
4	9	1024	114	728	1300	1330
5 ^a	16	1071	74	419	889	850
5	16	1072	207	1151	1310 ^c	1300 ^c

^aPreliminary transient experiment, run as a check on calculations.

^bThermocouple opened before transient.

^cCladding melting occurred.

Figure 3 shows the cladding thermocouple data from the first preliminary transient (1017). Although the two curves are similar, a clearly discernible change in heat transfer to the two thermocouples occurs just before peak power.

113-1311

Fig. 3. Cladding Temperature Data from Transient 1017 (Pin 1)

experiment. Figure 5 shows the post-TREAT condition of pin 1 with the thermocouples still attached to the cladding. No change in the cladding diameter or distortion of cross section was detected. Two sets of micrometer measurements of cladding diameters were taken at 1.3-cm intervals along the length of the pin, one set at 90° with respect to the other. (Estimated uncertainty in these remote micrometer measurements is ± 0.001 cm.) Evidence for localized overheating was found in the presence of two areas of darkened cladding surface, one extending from the fuel center up approximately 3.5 cm, and one extending between 3.0 and 4.5 cm below the fuel center. No actual damage to the cladding was found in those areas. Microscopic optical examination of the fuel showed it to resemble very closely the condition of control pins 2 and 6.¹⁹

2. Transient 1020 (Pin 11)

Figure 6 is the cladding temperature data from transient 1020 (pin 11). Only one thermocouple was operative; it broke away from the fuel as the cladding melted away. Figure 7 shows the post-TREAT condition of the pin. The remains of the cladding are shown in the capsule load frame, at the left of the figure. Note that one segment of fuel pellet remains at the bottom. The fuel fragments and the cladding remains are shown at the right in the figure. Although

although the two curves are similar, a clearly discernible change in heat transfer to the two thermocouples occurs just before peak power. Cladding temperature traces from transients 1019, 1021, 1023, and 1071 were similar to those in Fig. 3.

B. Post-TREAT Transient Results

1. Transient 1018 (Pin 1)

Transient 1018 carried pin 1 cladding close to the melting point, but did not produce cladding failure. Figure 4 shows the smooth cladding temperature curves for this

113-1310

Fig. 4. Cladding Temperature Data from Transient 1018 (Pin 1)

112-7443 Rev. 1

Fig. 5

Post-TREAT
Condition of
Pin 1

112-7444

113-1517

Fig. 6. Cladding Temperature Data
from Transient 1020 (Pin 11)

UO₂ Fuel

3.0 a/o Burnup Max Calc Fuel Temp 2750°C

Fig. 7. Post-TREAT Condition of Pin 11

extensive cladding melting occurred, the steel melted and fell away from the oxide; there is no microscopic evidence for explosive disintegration. No such disintegration was noted in the high-speed photographs, which provided a record of the sample behavior on a macroscopic level, although the photographs showed extensive darkening and localized production of dark vapor due to overheating of the capsule paint.*

3. Transient 1022 (Pin 3)

Figure 8 shows the transient cladding temperature data from transient 1022 (pin 3). Prior to thermocouple failure at 3 sec, there are two anomalies (included on Fig. 8) in the upper thermocouple trace, indicating a shift in the oxide near the thermocouple. As for pin 11, there was no photographic evidence of explosive failure. The cladding melted away, and fuel fell to the floor of the capsule under the influence of gravity. The post-TREAT condition of pin 3 is shown in the left and center of Fig. 9. Local overheating of the paint on the capsule load frame is clearly apparent. About 70% of the fuel was freed from the pin as a result of cladding failure. The freed fuel remains are shown in the center of Fig. 9. The twisting is attributed to unwinding of the sample around the spiral spacer wire. The wire was cooler than the cladding, thus making it stronger than the cladding, and also producing a bow, because the wire did not expand as much as the cladding tube. The cladding failed in two areas. The major failure ranged from 3.0 cm below the fuel center up to the top of the fuel stack, an area of about 5.0 cm², which is about 75% of the surface covering the fuel originally. The other failure occurred radially 180° from the first, leaving an opening of about 0.8 cm². As for pin 1, the optical microscopic examination of pin 3 showed it to resemble closely the condition of control pins 2 and 6.

113-1518

Fig. 8. Cladding Temperature Data
from Transient 1022 (Pin 3)

4. Transient 1024 (Pin 9)

Figure 10 shows the temperature curves from transient 1024. Both thermocouples show a break within about 5×10^{-3} msec of each other, the lower thermocouple signal disappearing and the upper thermocouple signal becoming erratic. Motion-picture photography of the experiment showed only mild failure. As shown at the right in Fig. 9, the paint on the load frame was subjected to localized overheating due to rapid release of hot gases. The posttransient condition

*A high-reflectance, vitreous silicone-base paint rated for operation up to 1000°C.

L. 0.7 a/o BURNUP (PIN 3)
C. CLOSEUP (PIN 3)
R. 3.0 a/o BURNUP (PIN 9)

112-7437

Fig. 9. Post-TREAT Conditions of Pins 3 and 9

113-1308

Fig. 10. Cladding Temperature Data
from Transient 1024 (Pin 9)

load frame. Spectrochemical analysis of the globule showed a strong concentration of iron (from cladding) and a very strong concentration of titanium (from the paint mixture), but no detectable uranium.

5. Transient 1072 (Pin 16)

Thermocouple data from transient 1072 (pin 16) are shown in Fig. 11. Although the curves for the two thermocouples are similar, there is a clear difference in heat transfer to the two sensors. The continued readings from the upper thermocouple indicate that it remained intact and attached to a cladding fragment, which cooled relatively slowly after pin failure. This interpretation was supported by the photographic record. Figure 12 shows eight still frames taken from the motion pictures of transient 1072. Frame a is the pre-transient condition of the sample: The pin is at the right, and the image of the back portion of the pin in the capsule mirror is at the left. (The bar across the image is a stiffener for the capsule.) In frame b, the sample has begun its temperature excursion, and bowing of the cladding is clearly shown in the mirror. Continued bowing, in frame c, has produced a hot spot at the top of the fuel stack, where the bowing has produced a region of better contact between cladding and fuel. This hot spot has increased in size, in frame d, which also shows a line of high thermal contact in the lower

112-8518

Fig. 11. Cladding Temperature Data
from Transient 1072 (Pin 16)

of the fuel fitted well in sequence of severity with the condition of pins 1, 11, and 3, when ranked by energy input (see Tables I and II). The cladding of pin 9 failed in two main areas, the major failure area ranging from 2.5 cm below the fuel center to 3.6 cm above it (and exposing 2.0 cm^2 of fuel, about 30% of the total surface). The other failure area was in approximately the same region as the minor failure of pin 3, and exposed about 0.3 cm^2 of fuel surface. Unlike the other (four) samples, there was a small globule of meltdown residue in the darkened area of the

Fig. 12. Still Frames from 2000-frame/sec Photo-graphic Record of Transient 1072 (Pin 16)

half of the sample. Note the relatively high degree of fuel plasticity shown in frame d. Failure, at the hot spot, occurs in frame e, with the emission of incandescent gas and fragments. Some dark vapor from the capsule paint partially obscures the postfailure movement of glowing fragments, shown in frame f. Because of the temperature, the sizes of the fragments cannot be accurately determined from the frames. Frame g shows the last stages of fragment motion, and frame h is the postexperiment condition of the pin, taken after the reactor power pulse was clipped, but while the sample was still glowing brightly. The specimen shown in frame h is the fuel stack, with what appears to be some sections of cladding that show

up as cooler (darker) regions. The fuel has maintained its general cylindrical shape, but its high-temperature plasticity is shown clearly. Figure 13 is a view of the sample, taken after transient 1072, directly through the reactor slot-viewing system before the slot liner was moved from the core. Fragments of cladding are shown, attached to the spiral wire at the top, and leaning against the back of the capsule. The bow-shaped curve of the UO_2 is readily apparent. (Note that the position of the mirror is shown reversed from that in Fig. 12. This is because the number of mirrors in the light path used for the still photograph of Fig. 13 was not equal to the number used for the high-speed camera.)

Fig. 13

Post-TREAT Condition of Pin 16, before Slot Liner Was Removed From Core

112-8520

Figure 14 shows the condition of pin 16, as examined in the ANL Illinois Hot Laboratory. The general cylindrical shape was maintained. Note the central fuel void appearing in the fuel section placed on end in Fig. 14b, and the "split" section of oxide at the top of the figure. Based on Figs. 12h and 13 and the postirradiation examinations (which are accurately reflected in Fig. 14b, we concluded that the bright fragments shown in the later frames of Fig. 12 were predominantly from cladding and did not result from extensive fragmentation of the oxide. Figure 15 shows details of the fuel surface. The surface bubbles shown in Fig. 15 were more pronounced than those of the control pins, or of the other four pins run in TREAT, and were typical of the entire surface of pin 16 oxide. Figure 16 shows two other closeup views of oxide from pin 16.

Figure 16a shows the interface where two oxide pellets had fused together. Figure 16b is an end view of a segment of pin 16 oxide, showing the well-defined central hole and bubble-like structures outside the columnar-grain growth region. Photomicrographs of the oxide from pin 16 showed that the bubbles appeared to be (high-enrichment) porous regions that were at the fuel surface. Figure 17 is a photomicrograph showing a high-enrichment porous bubble at the fuel surface. This bubbling phenomenon typified the entire surface of the fuel. Incipient bubbles were inside the peripheral regions, but disappeared in the columnar-grain growth region.¹⁹

112-8521

Fig. 14. Post-TREAT Condition of Pin 16 after Slot Liner Was Removed from Core; a, Remains of 6 a/o Burnup Oxide Pin in Its Load Frame after Transient 1072; b, Closeup of Remains of 6 a/o Oxide Fuel after Transient 1072

112-8522

Fig. 15. Closeup of Pin 16 Remains, Showing Two Views of Cylindrical 6 a/o Burnup Oxide with Surface Bubbles Typical of Oxide Fuel Remains after Transient 1072; a, View Showing Surface Bubbles; b, View Showing Surface Cracks and a Hole Left by a Bubble That Had Apparently Burst

112-8523

Fig. 16. Closeup of Pin 16 Remains, Showing Typical Pellet Fusion and Typical Pellet Cross Section after Transient 1072

40 μ

Fig. 17
Photomicrograph of 6 a/o Burnup UO_2 ,
Showing a Porous Bubble at the Fuel
Surface

112-8894

IV. TEMPERATURE CALCULATIONS

Figure 18 shows results of reference temperature calculations for steady-state irradiations. In addition, transient temperature calculations were made using the ARGUS code³⁰ to reconstruct the sample temperature conditions during the TREAT transients.

113-1519

Fig. 18. Calculated Radial Temperature Distributions of Fuel during Steady-state Irradiation

For the transient calculations, the fuel was assumed to have a central void of 0.050-cm radius, and density 100% of theoretical. The outer radius of the fuel was 0.195 cm, the inner radius of the steel cladding was 0.195 cm, and the outer radius of the cladding was 0.223 cm. The value of ρC_p (specific heat per unit volume) used was 3.64 J/cc-°C, consistent with that input in the calculations for the earlier TREAT experiments on unirradiated oxide¹¹ (see Ref. 31). A constant value of oxide conductivity was used for each calculation, but the value was varied according to the fuel temperatures reached. The conductivity used for each case will be given along with results. (See Table IV.) For reference, the conductivities were calculated using the equation

$$k = \frac{35}{T + 300} \frac{W}{cm-^{\circ}C},$$

where T is in degrees centigrade. This equation is based on data reported by Kingery *et al.*,³² Hedge and Fieldhouse,³³ and Reiswig.³⁴ It does not include the increase in conductivity with temperature at higher temperatures, but because of the use of a constant k in the fuel in each calculation, this equation should have been adequate for the purpose. For future calculations using variable k in the fuel and the large temperature gradients typical of a sodium environment, greater detail like that of Belle *et al.*³⁵ would be appropriate. The oxide heat of fusion (an experimental value is given in Ref. 31) was not a significant quantity for these calculations since fuel energy input in the most severe transient was just sufficient to reach the melting point. Steel conductivity and specific heat were 0.25 W/cm-°C, and 5 J/cc-°C, respectively. A single set of relative radial power factors* was used for all calculations. It corresponds to the TREAT thermal flux depression for a nominal 10% enrichment, or conversely, a nominal 3.5 a/o burnup. Table III gives these factors, normalized to yield unity when integrated over the fuel annulus used in the transient calculations.

*Calculated using S_n transport theory.

TABLE III. Relative Radial Power Factors

Radius, cm	Radial Power Factor	Radius, cm	Radial Power Factor
0.059	0.913	0.159	1.023
0.087	0.957	0.186	1.056
0.123	0.968		

Reference transient temperatures were calculated, for a given sample energy input with U (the fuel-to-cladding thermal conductance per unit area) as a parameter, to compare with the experimental cladding temperature curves from the five preliminary transients. The power curve used for these calculations was taken from TREAT transient 1017, which had a width at half-maximum power of 0.33 sec. Fuel energy input for these calculations was set at 432 J/g (4740 J/cc). Fuel conductivity was 0.023 W/cm 2 - $^{\circ}$ C. Figure 19 shows results obtained for U of 0.1, 0.25, 0.5, and 1.0 W/cm 2 - $^{\circ}$ C and indicates the time of reactor peak power. To first approximation, the maximum cladding temperature is independent of U . In reality, because of heat loss to the surroundings, different values of U will result in different maximum cladding temperatures. As shown in Fig. 19, the difference in maximum-cladding-temperature rise produced by an order-of-magnitude change in U of 0.1 to 1.0 is only about 50 $^{\circ}$ C out of 800 $^{\circ}$ C, or 6%. The difference in temperature-curve shape is more pronounced: Inspection of the experimental curves for transients 1017, 1019, 1021, and 1023 indicated that the effective U was intermediate between 0.1 and 0.5 W/cm 2 - $^{\circ}$ C. On this basis, a nominal value of 0.25 W/cm 2 - $^{\circ}$ C was adopted.

113-1312

Fig. 20. Experimental Values of Maximum Cladding Temperature as a Function of Estimated Sample Energy Input

Rates of sample heat loss to the surroundings were calculated using the data reported previously for pins of the same size in the same capsule configuration.¹¹

113-1309 Rev. 1

Fig. 19. Calculated Temperature Curves of Cladding Surface for Transient 1017, with Fuel-cladding Gap Conductance as Parameters

Figure 20 shows maximum cladding temperatures from the five preliminary transients as a function of estimated sample fuel energy input. Data points are labeled by TREAT transient number. Also

shown is the spread of thermocouple readings. (Only one thermocouple was operational for transient 1019; two thermocouples were operational during all other tests.) Also shown in Fig. 20 is the reference calculation point for $U = 0.25 \text{ W/cm}^2\text{-}^\circ\text{C}$ and 472 J/g of fuel. (See Fig. 19.)

Sample energy inputs were obtained using as reference the ratio between sample energy and reactor energy established previously for the similar samples of 10% enrichment.¹¹ The reference ratio was corrected for the original enrichment of the irradiated sample (13.5% by mass spectrometric assay²⁸) burnup of ^{235}U , the small buildup of ^{239}Pu during irradiation in MTR, and the change in TREAT core loading between the single-viewing-slot loading used for the earlier photographic experiments on unirradiated pins,¹¹ and the double slot loading that was used for the irradiated sample transients. The reference ratio for the 13.5% enriched pins in the new core loading, with no burnup, is 9.92 J/g-MW-sec . Corrections were appreciable, with the pin of transient 1071 having a power ratio calculated to be only 5.64 J/g-MW-sec .

Transient temperatures were also calculated for each of the "final," or energetic, excursions on the pins. In each case the actual reactor power curve was used as input. Although sample pins were run past the failure threshold, no attempts were made to incorporate fuel motions in the transient-temperature calculations. No evidence was found in the post-experiment inspections for fuel movement down the central voids, or for significant fuel motion at temperatures below the melting point. Once the cladding failed and flowed, heat transfer conditions were not well-defined. Hence, the transient temperature calculations were based on intact geometry. Table IV summarizes the results of these calculations.

TABLE IV. Summary of Transient-temperature Calculations on "Final" Transients

Pin No.	TREAT Transient	Sample Energy Input, J/g	Max Fuel Temp, $^\circ\text{C}$	Fuel Surface Temp at Time of Max, $^\circ\text{C}$	Outer Cladding Surface Temp at Time of Max, $^\circ\text{C}$	Max Outer Cladding Surface Temp, $^\circ\text{C}$	Fuel k in Calculation, $\text{W/cm-}^\circ\text{C}$
1	1018	757	2131	1582	789	1321	0.016
3	1022	869	2464	1809	882	1476	0.015
9	1024	728	2045	1508	854	1290	0.017
11	1020	1141	2750 ^a	2011-2220 ^b	1153-1565 ^b	(1790) melted	0.015
16	1072	1151	2750 ^c	2060-2175 ^d	985-1570 ^d	(1774) melted	0.015

^aThe maximum extent of oxide at the melting point was a radius of 0.10 cm.

^bFuel was at the melting point for 0.4 sec.

^cThe maximum extent of oxide at the melting point was a radius of 0.14 cm.

^dFuel was at the melting point for 0.5 sec.

Because of the low oxide conductivity, little heat was lost from the pin during the power transient, and the results were not highly sensitive to the actual value of conductivity input to the calculations. Table V compares some maximum temperature calculated using a conductivity appropriate to 1200°C , $k = 0.023 \text{ W/cm-}^\circ\text{C}$, with numbers from Table IV.

TABLE V. Effects of Oxide Conductivity on Calculated Temperatures

Pin No.	TREAT Transient	Sample Energy Input, J/g	Maximum Fuel Temp, $^\circ\text{C}$		Maximum Cladding Temp, $^\circ\text{C}$	
			$k = 0.023 \text{ W/cm-}^\circ\text{C}$	Table IV	$k = 0.023 \text{ W/cm-}^\circ\text{C}$	Table IV
3	1022	869	2426	2464	1512	1476
9	1024	728	1995	2045	1309	1290

For slower transients than those of this study (for which the power pulse widths at half maximum power were about a half-second), fuel conductivity and interface conductance could have a large effect on maximum calculated fuel temperatures. It would be expected that variable oxide conductivity should be used for calculations on transient oxide pin experiments with power pulse durations of the order of seconds, particularly with a sodium environment.

V. DISCUSSION

In theory, fission gases resulting from irradiation of the UO_2 fuel cause pressure buildup within fuel pins. The fission gases are trapped inside the fuel, and the released gas is contained in the central void, the pellet gaps, and the plenum above the oxide. The energy stored in this pressurized gas is capable of producing rapid fuel motion, or even explosive fuel disassembly upon failure of the cladding. However, neither rapid fuel motion nor explosive fuel disassembly was observed during these experiments. No evidence was found for motion down the central voids during the transients. No pattern of fuel cracking peculiar to the transients was discovered. In contrast with the earlier experiments on unirradiated samples,¹¹ no central zone of loosely-bonded oxide was found for maximum fuel temperatures above 2200°C .^{*} Nonhomogeneity of structure did not produce gross effects on transient-induced behavior.

Thus, to first approximation, it would appear that accident models using the pretransient condition and geometry of the fuel, physical properties characteristic of the reactor exposure up to the time of the transient, and intact geometry should give a reasonable description of the transient fuel behavior up to the melting point. Significant swelling or change in dimensions of the fuel due to fission gases begins only at the fuel melting point. For analysis of accidents in which large radial temperature gradients of the fuel would occur, the results of these experiments should apply in a differential sense to the annulus of cooler oxide surrounding the fuel at (or above) the melting point. The behavior of such annuli should be amenable to direct analysis, assuming proper attention is paid to transient thermal stresses arising from thermal gradients, and to any stresses arising from expansion of interior plastic or molten regions that cannot be fully accommodated by the central void.

The degrees of swelling upon melting, kinetics of swelling, heat of fusion of the oxide, and viscosity of melted oxide are all of importance to extending the accident models into the range of fuel melting. Study of these phenomena and their degree of dependence upon burnup is necessary to provide a bridge to more detailed study of accident phenomena such as sodium-coolant flow starvation.

^{*}If that zone did result from transient stresses, the presence of a central void may have provided relief for the irradiated oxide material.

These studies were performed with a single type of UO_2 fuel pin. Extension of these results to other oxides should be done carefully. The detailed photographs in Section II of Ref. 21 can be used as comparison standards for other types of samples given transient exposures.

ACKNOWLEDGMENTS

We gratefully acknowledge the valuable assistance given by W. D. Jackson, B. J. Kestel, and A. Schmidt of the Reactor Operations Division and D. Matias of the Accelerator Division for their Hot Laboratory assistance.

Assistance is also gratefully acknowledged of N. R. Stalica, C. E. Crouthamel, and C. A. Seils, Jr., of the Chemical Engineering Division, and J. H. Handwerk and D. E. White of the Metallurgy Division.

The effective and efficient operation of TREAT by its staff, under the direction of TREAT Manager J. F. Boland, was essential to these studies.

We also wish to thank A. B. Rothman for his critical comments on the initial draft of this report.

REFERENCES

1. W. J. McCarthy, C. E. Dickerman, A. E. Klickman, and R. B. Nicholson, "Fast Reactor Safety Research," *Proc. Third U.N. International Conference on Peaceful Uses of Atomic Energy, Geneva, 1964*, Vol. 13, p. 26, U.N., New York (1965).
2. C. E. Dickerman, *Experiments and Analyses in Fast Reactor Safety*, Nuclear Safety 8(3), 220 (1967).
3. L. J. Koch, H. O. Monson, D. Okrent, M. Levenson, W. R. Simmons, J. R. Humphreys, J. Haugsnes, V. C. Jankus, and W. B. Loewenstein, *Hazards Summary Report: Experimental Breeder Reactor II (EBR-II)*, ANL-5719 (May 1957).
4. C. E. Dickerman, E. S. Sowa, D. Okrent, J. Monaweck, and L. B. Miller, *Studies of Fast Reactor Fuel Element Behavior under Transient Heating to Failure. I. Initial Experiments on Metallic Samples in the Absence of Coolant*, ANL-6334 (Aug 1961).
5. C. E. Dickerman, G. H. Golden, and L. E. Robinson, *In-pile Photographic Studies of EBR-II Mark I and Fermi Core A Sample Meltdown*, Nucl. Sci. Eng. 14, 30 (1962).
6. C. M. Walter and C. E. Dickerman, *TREAT Study of the Penetration of Molten Uranium and U-5 wt % Fissium Alloy through Type 304 Stainless Steel*, Nucl. Sci. Eng. 18, 319 (1964).
7. C. E. Dickerman, E. S. Sowa, J. H. Monaweck, and A. Barsell, *In-pile Experiments on Meltdown of EBR-II Mark I Fuel Elements in Stagnant Sodium*, Nucl. Sci. Eng. 18, 319 (1964).
8. C. E. Dickerman, L. E. Robinson, E. S. Sowa, J. H. Monaweck, and C. August, *TREAT Meltdown of Irradiated Metallic Fast Reactor Fuel in an Inert Atmosphere*, Nucl. Appl. 3, 461 (1967).
9. C. E. Dickerman, R. Purviance, L. E. Robinson, W. Stephany, and F. L. Willis, *Summary and Analysis of TREAT Sodium Loop Experiments on Behavior of Single EBR-II Mark I Pins under Accident Conditions*, Nucl. Eng. and Design 7, 442 (1968).
10. L. E. Robinson, C. E. Dickerman, and F. L. Willis, *Initial (7-Pin) EBR-II Mark I Scale-up Cluster Fast Reactor Safety Integral Loop Experiments in TREAT*, Trans. Am. Nucl. Soc. 10(2), 697 (1967).
11. C. E. Dickerman, L. E. Robinson, E. S. Sowa, and S. B. Skladzien, *Studies of Fast Reactor Fuel Element Behavior under Transient Heating to Failure. II. In-pile Experiments on UO_2 Samples in the Absence of Coolant*, ANL-6845 (Jan 1965).
12. L. Baker, Jr., *Metal-Water Reactions*, Nucl. Safety 8, No. 1, p. 20 (1966).
13. J. H. Field, *Experimental Studies of Transient Effects in Fast Reactor Fuels, Series I. UO_2 Irradiations*, GEAP-4130 (1962).

14. J. E. Hanson, J. H. Field, and S. A. Rabin, *Experimental Studies of Transient Effects in Fast Reactor Fuels, Series II. Mixed Oxide (PuO₂-UO₂ Irradiations)*, GEAP-4804 (1965).
15. J. E. Hanson, *Response of Low-burnup-irradiated Mixed-oxide (PuO₂-UO₂) Fuel to Transient Overpower*, Trans. Am. Nucl. Soc. 8, No. 2, p. 560 (1965).
16. J. H. Field and J. E. Hanson, *Response of High-burnup Irradiated Plutonia-Urania Fuel to Transient Overpower*, Trans. Am. Nucl. Soc. 9, No. 2, p. 396 (1966).
17. G. R. Thomas and J. H. Field, *Response of Axially Restrained Pellet Fuel to Transient Overpower*, Trans. Am. Nucl. Soc. 10, No. 2, p. 707 (1967).
18. C. E. Dickerman, L. E. Robinson, W. Doe, and R. Stewart, "Properties of Irradiated UO₂ Pins prior to Transient Reactor Test (TREAT) Facility Transients," in *Reactor Physics Division Annual Report: July 1, 1965 to June 30, 1966*, ANL-7210 (Dec 1966), Paper IV-4, pp. 227-240.
19. C. E. Dickerman, L. E. Robinson, and R. Stewart, *Photographic TREAT Fast Reactor Safety Experiments on Irradiated Oxide Pins*, Trans. Am. Nucl. Soc. 9, No. 2, p. 396 (1966).
20. L. E. Robinson and C. August, *Transparent Facility for TREAT Meltdown Experiments on Preirradiated Samples*, ANL-7572 (in preparation).
21. R. R. Stewart, C. E. Dickerman, N. R. Stalica, and W. B. Doe, *A Study of the Physical Properties of Nominal 0.1 a/o, 3 a/o and 6 a/o Burnup Oxide Fast Reactor Fuel Pins*, ANL-7571 (May 1969).
22. G. A. Freund, H. P. Iskenderian, and D. Okrent, "TREAT, A Pulsed Graphite-moderated Reactor for Kinetics Experiments," *Proc. Second U.N. Conference on Peaceful Uses of Atomic Energy*, Vol. 10, p. 461, U.N. Geneva (1958).
23. G. A. Freund, P. Elias, J. D. Geier, and J. F. Boland, *Design Summary Report on the Transient Reactor Test Facility (TREAT)*, ANL-6034 (June 1960).
24. D. Okrent, C. E. Dickerman, J. Gasidlo, D. M. O'Shea, and D. F. Schoeberle, *The Reactor Kinetics of the Transient Reactor Test Facility (TREAT)*, ANL-6174 (Sept 1960).
25. F. Kirn, J. F. Boland, H. Lawroski, and R. D. Cook, *Reactor Physics Measurements in TREAT*, ANL-6173 (Oct 1960).
26. C. E. Dickerman, R. D. Johnson, and J. Gasidlo, *Kinetics of TREAT Used as a Test Reactor*, ANL-6458 (May 1962).
27. R. Armani, Argonne National Laboratory, private communication.

28. R. Larson, Argonne National Laboratory, private communication.
29. A. DeVolpi and C. H. Freese, *Fast Neutron Transient Detections Hodoscope for Reactor Fuel Meltdown Studies*, IEEE Trans. Nucl. Sci. NS-13(1), 623 (1965).
30. D. F. Schoeberle, J. Heestand, and L. B. Miller, *A Method of Calculating Transient Temperatures in a Multiregion, Axisymmetric, Cylindrical Configuration. The ARGUS Program*, 1089/RE-248, Written in Fortran II, ANL-6654 (Nov 1963).
31. R. A. Hein, L. H. Sjodahl, and R. Szwarc, *Heat Content of Uranium Dioxide from 1200 to 3100°K*, J. Nucl. Mat. 25, 99 (1968).
32. W. D. Kingery, F. Vasilos, A. L. Loeb, F. H. Norton, J. Franil, and R. L. Cable, *The Measurement of Thermal Conductivity of Refractory Materials*, NYO-3647 (1953).
33. J. C. Hedge and J. B. Fieldhouse, *Measurement of Thermal Conductivity of Uranium Dioxide*, AECU-3381 (1956).
34. R. D. Reiswig, *Thermal Conductivity of Uranium Dioxide to 2100°C*, J. Am. Ceram. Soc. 36, 48 (1961).
35. J. Belle, R. M. Berman, W. F. Bourgeois, I. Cohen, and R. C. Daniel, *Thermal Conductivity of Bulk Oxide Fuels*, WAPD-TM-586 (revised) (1967).