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ORNL-3709

UC-25 - Metals, Ceramics, and Materials
TID-4500 (34th ed.)

IRRADIATION TESTING OF FUEL FOR CORE B OF
THE ENRICO FERMI FAST BREEDER REACTOR

W. C. Thurber
F. R. McQuilkin
E. L. Long, Jr.
M. F. Osborne

OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

for the

U.S. ATOMIC ENERGY COMMISSION

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U.S. Department of Commerce, Springfield, Virginia

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ORNL-3709

Contract No. W-7405-eng-26

METALS AND CERAMICS DIVISION

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NOVEMBER 1964

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W. C. Thurber, F. R. McQuilkin,¹ E. L. Long, Jr., and M. F. Osborne²

ABSTRACT

Three instrumented capsules were irradiated in the MTR to assess the in-reactor performance of the fuel proposed for Core B of the Enrico Fermi Fast Breeder Reactor. Each capsule contained two miniature fuel plates, $2.0 \times 0.5 \times 0.116$ in., comprised of an 0.102-in.-thick fuel-bearing section containing 33 wt % UO_2 dispersed in type 347 stainless steel and clad on both surfaces with 0.007-in.-thick type 347 stainless steel. Exposure conditions for the capsules were as follows:

Capsule	Specimen	Maximum Average Surface Temperature (°F)	Burnup (fissions/cm ³) × 10 ²¹
46-2	5	1005	1.7
46-4	1	890	0.36
	10	945	0.39
46-5	8	1000	2.0
	9	1005	2.0

The specimens from capsule 46-4 were virtually unaffected by irradiation, exhibiting density decreases of approximately 0.5%. However, the specimens from capsules 46-2 and 46-5 suffered gross swelling and cracking of the stainless steel matrix, demonstrating that the burnup limits were clearly exceeded. Density decreases of 6 to 13% were measured for the specimens in these two capsules. Correlation of these data with information developed by other experimenters suggests that at a surface temperature of 900°F the maximum tolerable burnup prior to failure will be less than 1.6×10^{21} fissions/cm³.

¹Reactor Division.

²Reactor Chemistry Division.

INTRODUCTION

The fuel element initially considered for Core B of the Enrico Fermi Fast Breeder Reactor consists of 14 fuel plates, each 2.4 in. wide $\times$ 19 in. long $\times$ 0.116 in. thick, assembled by brazing into a rectangular parallelepiped (Fig. 1). Each fuel plate contains a dispersion of 27.5 vol % (34.5 wt %) spheroidal UO_2 dispersed in a type 347 stainless steel matrix and clad on either surface with 0.005 in. of type 347 stainless steel. Design and engineering testing of completed fuel elements were conducted by Atomic Power Development Associates, while development of the components was conducted at the Oak Ridge National Laboratory. Major tasks included in the metallurgical development program were: (1) fabrication of composite fuel plates,³ (2) characterization of spheroidal UO_2 particles,⁴ (3) feasibility studies on nondestructive testing,⁵ (4) establishment of fuel element manufacturing procedures,⁶ (5) mechanical properties measurements on the fuel material,⁷ and (6) irradiation testing.

³J. H. Cherubini, R. J. Beaver, and C. F. Leitten, Jr., Fabrication Development of UO_2 -Stainless Steel Composite Fuel Plates for Core B of the Enrico Fermi Fast Breeder Reactor, ORNL-3077 (April 4, 1961).

⁴W. O. Harms et al., "Characterization of Spheroidal Uranium Dioxide Particles," pp. 354-64 in Meeting on Characterization of Uranium Dioxide held at Oak Ridge National Laboratory, Oak Ridge, Tennessee, December 12-13, 1961, TID-7637 (Oct. 1, 1962).

⁵R. W. McClung, Feasibility Studies - Nondestructive Testing of the Enrico Fermi Core B Fuel Element, ORNL-3221 (Dec. 21, 1961).

⁶R. G. Donnelly, W. C. Thurber, and G. M. Slaughter, Development of Fabrication Procedures for Core B Fuel Elements for the Enrico Fermi Fast Breeder Reactor, ORNL-3475 (July 1964).

⁷J. T. Venard and R. W. Swindeman, Mechanical Properties of the UO_2 -Type 347 Stainless Steel Cermet for Core B of Fermi Reactor, ORNL-TM-665 (Oct. 24, 1963).

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Fig. 1. Core B Fuel Element.

All of this work except the irradiation testing has been reported elsewhere. It is the purpose of this report to describe the irradiation studies carried out in support of the Core B fuel element development program.

The irradiation program involved exposure in the MTR of three instrumented sodium-filled capsules, designated ORNL-MTR-46-2, -4, and -5. Each capsule contained two miniature fuel plates that were fabricated according to procedures developed for the full-size composites.³ The nominal irradiation conditions selected were burnups of 10, 20, and

25% of the ^{235}U atoms at a surface temperature of 925 to 950°F. Nominal center line temperatures were 1100 to 1200°F.

These conditions were selected on the basis that Core B would be used to operate the Fermi Reactor at a maximum core thermal power of 270 Mw. At this power level the calculated throughput burnup would be 20.7% of the initial ^{235}U atoms fissioned. The throughput burnup is the core averaged burnup for all fuel subassemblies at discharge. The hottest subassemblies will operate to less than throughput burnup, while elements cooler than the average will operate to burnups greater than 20.7 at. %. Fuel temperatures at the reactor center would be 750°F for the plate surface and 970°F for the plate center line. The corresponding hot-spot conditions dictate a plate surface temperature of 870°F and a plate center line temperature of 1043°F. These temperatures do not account for hot-channel factors that would increase the maximum plate center line temperature to about 1140°F.

After exposure, the capsules were examined in the hot cell to determine the quality and integrity of the fuel specimens and the conclusion was reached that the burnup capabilities of this fuel were exceeded under certain test conditions. However, these test conditions exceeded the operating parameters expected from the Core B.

MATERIALS SELECTION AND TEST PROGRAM PHILOSOPHY

Early in the fuel element developmental program it was decided to limit the fuel loading to 27.5 vol % (34.5 wt %) of highly enriched UO_2 in the austenitic stainless steel matrix as this represented the upper limit at which significant irradiation experience had been obtained with this type of fuel.⁸ Spheroidal fuel particles were chosen since it was felt that this shape would minimize stress concentrations in the fuel matrix. Such localized stresses could be produced by the irradiation-induced swelling of angular particles.

⁸D. L. Keller, Nucleonics 19(6), 45 (1961).

Type 347 stainless steel was selected as the structural metal. This alloy in the temperature range of 800 to 1000°F has a strength that is superior to the austenitic steels in the 18-8 family. From theoretical analyses⁹ yield strength should be a controlling factor in determining the resistance of the matrix to stresses resulting from fission-gas release by the UO₂ particles. Type 347 stainless steel also met the other requirements for the clad and matrix material, such as fabricability, well-developed technology, and corrosion resistance in flowing sodium. On the basis of information available at the outset of the program,¹⁰ it was postulated that the Core B fuel would withstand the specified burnup at the temperature of interest. This probability was further enhanced by the selection of spheroidal particles homogeneously distributed in the matrix and by the selection of a relatively strong stainless steel matrix and cladding.

Although it was believed that the reference fuel would perform adequately, there are certain features in the Core B design that represented significant deviations from previous tests and thus proof testing was necessary. Included among the deviations are the following:

1. The plates are relatively thick (approx 0.116 in.) and have a rather large temperature gradient (220°F at the reactor center). Thus plastic strain fatigue resulting from the attendant thermal stresses and reactor cycling could play a possible role in inducing premature failure.

2. The ratio of clad-to-meat thickness is small, being approximately 10% rather than 30% frequently encountered in flat-plate fuel element designs, and the relative restraint afforded by the cladding is consequently less. Conditions therefore exist that could promote accentuated fuel-plate swelling under irradiation.

⁹J. R. Weir, A Failure Analysis for the Low-Temperature Performance of Dispersion Fuel Elements, ORNL-2902 (May 27, 1960).

¹⁰V. O. Haynes, F. H. Neill, L. D. Schaffer, Summary of UO₂-Stainless Steel Dispersion Element Irradiation Experiments, ORNL-CF-58-2-71 (March 18, 1958).

It was the purpose of this program to check out the significance of these postulates and to generally demonstrate the burnup capabilities of the Core B fuel.

IRRADIATION CAPSULE DESIGN

The instrumented capsules, designed for exposure in the MTR reflector, are schematically illustrated in Fig. 2. The inner stainless steel capsule contained two tandem-mounted specimens surrounded by two MgO-insulated, type 347 stainless steel-sheathed Kanthal A-1 heater coils. The operating plan was such that in the specified neutron flux, heaters would only be required for shim control; but as burnup progressed additional electrical heat would be supplied to compensate for the reduced fission heat. Each specimen was instrumented with two 1/16-in.-diam stainless steel-sheathed Chromel-P-Alumel thermocouples mounted on the specimen edge and centered at the axial midplane. The thermocouples of B & S gage wire with grounded junctions were calibrated against a standard Pt vs Pt-10% Rh thermocouple prior to test. As shown in Fig. 2 (Section A-A), the thermocouples were bent from vertical runs in U-channels to contact the specimen surface at the selected locations. The control thermocouple was connected to a Speedomax Model H controller, while information from the remaining three thermocouples was conventionally recorded. Auxiliary electrical heat was supplied when the control thermocouple fell below a predetermined set point.

Sodium was used in the inner container as the heat-transfer medium. The largest portion of the total temperature gradient within the capsule occurred in the 0.005-in. helium-filled annulus between the inner and outer containers.

A one-velocity, two-region diffusion theory was used to calculate the flux incident on the fuel specimen surface. In this calculation the inner region was taken as a cylinder of sodium 13/16 in. in diameter and the outer region was a stainless steel container 1 1/8 in. in diameter. It was assumed that the flux incident on the surface of the stainless steel container was the unperturbed flux quoted by the MTR project engineer. To calculate the flux depression through the strongly absorbing fuel sample, the one-velocity transport theory was used assuming an isotropic incident flux and no scattering within the fuel.¹¹

¹¹Private communication from T. S. Lundy to W. C. Thurber.

Fig. 2. Irradiation Capsule for Testing Miniature Fermi Core B Fuel Plates.

Pertinent capsule design parameters are summarized below:

Specimen heat output, kw	2.12
Capsule exterior surface temperature, °F	135
Fuel center line temperature, °F	1260 (max)
Fuel surface temperature, °F	925-950 (nominal)
Fuel perturbation factor (specimen plus capsule)	0.37

SPECIMEN DESIGN AND FABRICATION

Design

As indicated in the previous section, each capsule contained two $2.0 \times 0.50 \times 0.116$ -in. specimens containing highly enriched spheroidal UO_2 dispersed in and clad with type 347 stainless steel. The nominal core dimensions were $1.5 \times 0.35 \times 0.102$ in., thus fixing the nominal cladding thickness at 0.007 in. One specimen in each capsule contained a bushing that penetrated through the fuel-bearing portion of the miniature plate. This fitting was added to simulate a spacer arrangement initially contemplated in the Core B fuel element design. The bushing was sealed into the plate by brazing with a nickel-base alloy (GE-81). This type of fuel plate is isometrically sketched in Fig. 3. The second fuel plate in each capsule was identical except that it contained neither the bushing nor the hole through the matrix.

Fabrication

Fabrication of the miniature plates was by the conventional picture-frame technique. A master compact containing 33.2 wt % of -100 +140 mesh, highly enriched (93.2% ^{235}U) spheroidal UO_2 dispersed in a matrix of prealloyed -100 mesh type 347 stainless steel (modified with 2.6 wt % Si) was prepared by so-called "paste-blending" procedures outlined elsewhere.¹² Briefly, the process was as follows:

1. Place weighed powders in 4-oz jar.
2. Mix for 1/2 hr in oblique blender.

¹²J. H. Cherubini, R. J. Beaver, and C. F. Leitten, Jr., Fabrication Development of UO_2 -Stainless Steel Composite Fuel Plates for Core B of the Enrico Fermi Fast Breeder Reactor, ORNL-3077 (April 4, 1961).

Fig. 3. Isometric Drawing of Miniature Irradiation Sample Containing Bushing Through Fuel Bearing Section.

3. Add 0.3 cm^3 of 5% paraffin- CCl_4 solution with an atomizer and mix 1 1/2 hr.
4. Place resultant paste in cavity of double-acting die and press at 33 tsi.
5. Bake at 200°C for 1 1/2 hr.
6. Sinter at 1225°C for 1 1/4 hr in dry hydrogen.
7. Coin at 33 tsi.

Miniature fuel cores of the proper size were machined from the master compact, and four such cores were assembled in a type 347 stainless steel picture frame. Cover sheets of type 347 stainless steel were attached to the frame by fusion welding, and the billet was evacuated through a stem prior to final sealing.

The assembled billets were initially preheated to 1120°F in air then transferred to another furnace with a hydrogen muffle and preheated an additional 45 min at 2200°F. The plates were then hot rolled to the final thickness. To minimize the particle degradation no cold rolling was performed. The total reduction of 8:1 yielded a sound metallurgical bond, as indicated in Fig. 4. The microstructure of the fueled region is shown in Fig. 5 demonstrating that reasonable homogeneity and little oxide fracturing were obtained.

Mill scale was removed by pickling the plates in an aqueous room-temperature bath of 15 vol % HNO_3 -5 vol % HF.

Radiographs were used to locate the fuel-bearing regions, and thus delineate the fuel-plate perimeter. Typical radiographs are included in Fig. 6. Varying amounts of flashover may be noted at the ends of the fuel-bearing section of each specimen.

For those plates with the bushing (see Fig. 3), an 0.188-in.-diam hole was drilled through the fueled region. The samples were leached for 15 min with a 15% HNO_3 aqueous solution to remove the exposed UO_2 . Bushings were then pressed into the holes, and GE-81 brazing alloy (66 wt % Ni-19 wt % Cr-10 wt % Si-4 wt % Fe-1 wt % Mn) was placed around the bushing periphery. Microbraz cement fixed the alloy in position and Microbraz Green Stop-Off was used to prevent flow on the plate surface. Brazing was conducted in dry hydrogen (inlet dew point of -60°F) at 2160°F. A typical joint, as shown in Fig. 7, attests to the high quality of the seal.

IRRADIATION HISTORY

At the outset of the program two MTR reflector positions, A-11-NW and A-7-SE, were available. Consequently, it was decided to simultaneously initiate the short (10%) and the intermediate burnup (20%) tests; at the completion of the short burnup experiment the high burnup (25%) capsule would be inserted into the same position. Because of slightly different fluxes in the two positions, exposure of the intermediate and long burnup tests would be completed at about the same time.

The irradiation history of these capsules is summarized in Table 1. On the basis of the advertised MTR flux of 5.0×10^{13} neutrons $\text{cm}^{-2} \text{ sec}^{-1}$ and a calculated flux perturbation factor for specimen plus capsule of 0.37,

Fig. 4. Section of Irradiation Specimen Roll Bonded at 2200°F to a Reduction in Thickness of 87.5%. Etchant: glyceria regia 200X.

Fig. 5. Transverse Section of Typical Fuel Irradiation Sample Containing 33.2 wt % -100 +140 Mesh Spheroidal UO₂ Dispersed in Type 347 Stainless Steel. Etchants: 10% HNO₃, 20% H₂O₂ (30%), and 70% H₂O for UO₂ structure; glyceria regia for matrix structure. 50X.

Fig. 6. Radiographs of Typical Miniature Fuel Plate Irradiation Specimens. Plates 1, 5, 6, and 8 were actually tested as were 9 and 10 (not shown). Positive print approximately 1.7X.

Fig. 7. Plate Section with Spacer Bushing Showing GE-81 Brazed Joint. As polished. 100X.

Table 1. Operating History of ORNL-MTR-46 Program Capsules

	Capsule ORNL-MTR-		
	46-2	46-4	46-5
MTR position	A-7-SE	A-11-NW	A-11-NW
Insertion date	10-24-60	10-24-60	2-27-61
Discharge date	5-14-62	2-27-61	8-27-62
Insertion cycle number (beginning of)	147	147	153
Discharge cycle number (end of)	173	152	178
Number of cycles	27	6	26
Peak thermal flux, unperturbed, neutrons $\text{cm}^{-2} \text{sec}^{-1}$	$3 \times 10^{13}^a$	5×10^{13}	5×10^{13}
Reactor operation, Mwd	17,700	4,110	17,000
Total dose, neutrons/ cm^2	11.5×10^{20}	4.45×10^{20}	18.4×10^{20}
Maximum clad temperature, °F	1165	1010	1130
Number of thermocouples operable at discharge	0	4	3
Desired burnup (% of ^{235}U)	20	10	25

^aCapsule retracted from peak flux position of 6×10^{13} neutrons $\text{cm}^{-2} \text{sec}^{-1}$.

ORNL-MTR-46-4 was discharged after an estimated burnup of 9.5% of the ^{235}U atoms. Subsequent measurements revealed that the actual fission burnup was only 6.30% (average for both samples) of the ^{235}U atoms. Consequently, an experience factor of $\frac{6.3}{9.5}$ or 0.66 was applied in predicting the discharge dates for the other two capsules. Using this experience factor, capsules ORNL-MTR-46-2 and -5 were discharged after estimated burnups of 16 and 26% of the ^{235}U atoms, respectively. As will be pointed out subsequently, application of the experience factor

to capsule 46-2 produced a significant error (approx 50%), while application of this factor to capsule 46-5 produced an estimated burnup within about 20% of the measured values. This difference is not unreasonable since the experience factor was obtained for the same MTR position (A-11-NW) in which capsule ORNL-MTR-46-5 was subsequently irradiated.

Upon initial reactor insertion, capsule ORNL-MTR-46-2 was operating at a temperature higher than desired. It was hotter at each power level during MTR startup than was capsule 46-4 which was being simultaneously brought to temperature. Consequently, during the first two days the capsule was retracted from the peak-flux position to a position such that the capsule resided in a region from 6 to 17 in. below the top of the reflector. During this period of adjustment, the surface temperature never exceeded 980°F. In this position the capsule was estimated by the MTR project engineer assigned to the program to receive a thermal flux of 3×10^{13} neutrons $\text{cm}^{-2} \text{sec}^{-1}$. The estimate was based on the axial flux map in similar experimental holes in the MTR. Irradiation continued unaltered until discharge. It is perhaps noteworthy that the flux estimates were based on a flux map that was made during MTR cycle 146 - the cycle previous to that in which capsules 46-2 and -4 were inserted.

Operating temperatures for the three capsules are presented in Figs. 8, 9, and 10. The temperature ranges plotted here are the high and low temperatures observed for each operational thermocouple during the indicated MTR cycle. The particular data from which these measurements were selected were the thermocouple readings recorded by the shift technician three times daily. It may be noted that the surface temperatures as indicated by all four thermocouples in capsule ORNL-MTR-46-4 generally ranged from 800 to 950°F. It is further apparent that all thermocouples remained operational throughout the test and that reasonable agreement existed among the thermocouples. Thermocouple No. 15, attached near the center of the bottom specimen, served as the control thermocouple in this experiment with a controller set point of 950°F. The capsule was fully inserted during its six-cycle exposure, and auxiliary heat was supplied to maintain the desired temperature.

Fig. 8. Temperature History of Capsule ORNL-MTR-46-2.

In capsule ORNL-MTR-46-5 the surface temperatures generally ranged between 950 and 1050°F for cycles 153 through 164 and between 900 and 1000°F for cycles 165 through 178. Here, again, agreement among the thermocouples was reasonable. Failure of thermocouple No. 6 occurred during the fifth cycle, but performance of the other three thermocouples was satisfactory. From startup through cycle 171, the control function was vested in thermocouple No. 7; and from cycle 172 to completion of the irradiation, thermocouple No. 8 was the controlling thermocouple.

A detailed examination of the actual temperature recordings of thermocouple No. 7 for cycle 155 and thermocouples Nos. 5, 7, and 8 in capsule ORNL-MTR-46-5 for cycle 160 was made to determine during what fraction of the cycle the surface temperatures exceeded 1050°F. These cycles were selected from the temperature plots as being those during which unusual conditions were detected. Thermocouple No. 7 was above 1050°F approximately 18% of cycle 155. Since this temperature was higher than desired, the capsule was retracted 3 in. from the peak-flux position

Fig. 9. Temperature History of Capsule ORNL-MTR-46-4.

at the conclusion of the cycle. The high readings of the thermocouples in cycle 160 persisted only during a brief period, approximately 1.8% of the cycle.

Thermocouple performance in capsule ORNL-MTR-46-2 was less satisfactory. Thermocouple No. 9 (that on the edge of the top specimen) was never operative during the experiment. During the first two days of operation the top center thermocouple (No. 10) behaved erratically but then appeared to settle down and apparently remained functional until failure during cycle 167. Because of the questionable behavior during startup, the control function was shifted from this thermocouple to thermocouple No. 12 on the edge of the lower specimen. It should also

Fig. 10. Temperature History of Capsule ORNL-MTR 46-5.

be noted from Fig. 10 that the thermocouple on the edge of the bottom specimen yielded consistently higher readings than did the thermocouple located at the center of the specimen. This may have resulted from the bead of thermocouple No. 11 being moved from intimate contact with the cladding during handling prior to startup. Thermocouples 11 and 12 failed during cycles 172 and 173, respectively, and the capsule was discharged at the end of cycle 173. In light of the foregoing, it is felt that thermocouple No. 12 is the best measure of temperatures experienced by capsule ORNL-MTR 46-2. Temperature measurements made with this thermocouple indicate that for the first 17 cycles (147 to 163) the surface temperature generally ranged from 850 to 1000°F, while during the remainder of the experiment the surface temperature was higher, ranging from 900 to 1150°F.

POSTIRRADIATION EXAMINATION

Preliminary Operations

Visual examination of both the outer and inner containers from all three experiments revealed no flaws attributable to irradiation. A slight darkening of the inner cans was observed at positions approximating

the fuel-plate locations. After visual examination, each inner can was heated to about 250°F and held in a vertical position to ensure that all of the sodium was at the lower end. A transverse cut was then made near the top, and the can was inverted and reheated to 250°F to drain the bulk of the sodium. Any sodium that remained after drainage was reacted with isobutyl and methyl alcohols. The specimen rack was then removed and the specimens prepared for further examination.

The solution remaining after reaction of the sodium with alcohol in capsule ORNL-MTR-46-5 was assayed for fission-product activity, since it was suspected that an imperceptible breach in the cladding integrity had developed during irradiation. Radiochemical analyses revealed 1.5×10^{20} atoms of ^{137}Cs , 3.6×10^{14} atoms of ^{144}Ce , and 3.0×10^{12} atoms of ^{95}Zr . As will be described subsequently, no flaws were detected that could have provided a path for fission-product release; however, comparison of burnup values based on ^{144}Ce -to-uranium and ^{137}Cs -to-uranium ratios suggests that undetectable flaws did exist in both the solid fuel plate (No. 8) and the fuel plate with brazed bushing (No. 9).

Gamma Scanning

A gross gamma activity or relative burnup profile was obtained by gamma scanning the fuel plates. The scan was obtained by passing the specimens over an 0.02- × 0.75-in. slit in a 3-ft-thick hot-cell wall plug. The discriminator of the scintillation detector unit was set in such a manner that all energies below 0.65 Mev were excluded.

The fuel specimens from capsule ORNL-MTR-46-4 were scanned in the specimen holder after removal from the inner container; the specimens in capsules ORNL-MTR-46-2 and -5 were scanned while still contained in the inner can. These gamma scans are presented in Fig. 11a, b, and c. The scan of the specimens in capsule 46-4 indicates a uniform activity profile along the length of each specimen. The dip in the trace for the bottom specimen results from the bushing in that fuel plate. The erratic profile for plate 6 in capsule 46-2 is suggestive of damage and fuel redistribution; a normal profile was observed for plate 5. The gamma scans for plates 8 and 9 in capsule 46-5 appear about as expected. Vicissitudes in those scans may be the result of some fission product redistribution since fission-product leakage from the plates was detected.

Fig. 11. Gross Gamma Activity Scans for Capsules in ORNL-MTR-46 Experiment.

Visual Examination of Fuel Plates

Capsule ORNL-MTR-46-4

Visually, plates 1 and 10 from this capsule were unaffected by irradiation. This condition is illustrated for plate 10 in Fig. 12.

Capsule ORNL-MTR-46-2

Examination of the specimens from this capsule confirmed the hypothesis developed from the gamma scan that a fuel plate failure had occurred. As may be seen in Fig. 13, the upper specimen in the capsule had developed completely penetrating holes in two locations as well as a large crack near one edge. Blobs of fuel plate material indicative of melting are also obvious. A readily visible thickness increase was noted in the bottom specimen in the capsule. This is depicted in Fig. 14.

Capsule ORNL-MTR-46-5

Obvious swelling in the thickness direction also occurred in plates 8 and 9 from capsule 46-5, as illustrated in Fig. 15. However, no clad ruptures could be visually detected in these plates.

Since significant ^{137}Cs activity had been detected in the sodium-alcohol residue from capsule 46-5, further attempts were made to locate the site of fission-product leakage. To this end, a dye-penetrant examination of the fuel plate surfaces was performed using standard "Zyglo" florescent penetrant inspection procedures. The source of leakage could not be found although it must be pointed out that inspection of the region around the brazed bushing was questionable.

Dimensional and Density Measurements

Length, width, and thickness dimensions were determined for each fuel plate except plate 6, which had been severely damaged during test. Length and width measurements were read to ± 0.001 in. or better, while the thickness measurements were read to ± 0.0002 in. Density of the samples after irradiation was determined by a conventional carbon tetrachloride immersion technique with a reproducibility of ± 0.02 g/cm³. Duplicate measurements were taken on each sample.

Results of density and dimensional measurements are listed in Table 2. Changes in plate length and width were small with the maximum observed increases being only 1.4 and 1.8%, respectively. Thus,

R-4548

Top View

R-4549

Edge View

R-4545

Bottom View

Fig. 12. Fuel Plate 10 from Capsule ORNL-MTR-46-4.

R-11566

R-11567

R-11568

R-11569

Fig. 13. Fuel Plate 6 from Capsule ORNL-MTR-46-2.

R-11573

R-11575

R-11572

Fig. 14. Fuel Plate 5 from Capsule ORNL-MTR-46-2.

R-11598

R-11599

R-11600

R-11601

Fig. 15. Fuel Plates 8 and 9 from Capsule ORNL-MTR-46-5.

Table 2. Dimensional and Density Measurements for Fermi Core B Miniature Fuel Plates.

Plate	Length				Width				Thickness				Density			
	Pre-Irradiation (in.)	Post-Irradiation (in.)	Change (in.)	Change (%)	Pre-Irradiation (in.)	Post-Irradiation (in.)	Change (in.)	Change (%)	Pre-Irradiation (in.)	Post-Irradiation (in.)	Change (in.)	Change (%)	Pre-Irradiation (g/cm ³)	Post-Irradiation (g/cm ³)	Change (g/cm ³)	Change (%)
1 A	2.002	2.005	+0.003	+0.14	0.4972	0.499	+0.002	+0.40	0.1165	0.1152	-0.0013	-1.1	8.16	8.11	-0.05	-0.61
B					0.4987	0.500	+0.001	+0.20	0.1160	0.1170	+0.0010	+0.86				
C					0.5005	0.497	-0.004	-0.80	0.1153	0.1162	+0.0009	+0.77				
10 A	2.002	2.003	+0.001	+0.05	0.4996	0.500		0	0.1212	0.1212		0	8.14	8.11	-0.03	-0.36
B					0.5007	0.502	+0.002	+0.40	0.1214	0.1221	+0.0007	+0.57				
C					0.5001	0.500		0	0.1210	0.1215	+0.0005	+0.41				
5 A	2.002	2.003	+0.001	+0.05	0.4994	0.5015	+0.0021	+0.40	0.1175	0.1391	+0.0216	+18	8.19	7.16	-1.03	-13
B					0.4998	0.5071	+0.0078	+1.60	0.1112	0.1601	+0.0489	+44				
C					0.4994	0.5009	+0.0015	+0.30	0.1177	0.1229	+0.0052	+4.4				
8 A	2.001	2.0017	+0.0007	+0.035	0.4994	0.5007	+0.0013	+0.20	0.1170	0.1243	+0.0073	+6.2	8.15	7.47	-0.68	-8.3
B					0.5004	0.5075	+0.0071	+1.4	0.1176	0.1498	+0.0322	+27				
C					0.5000	0.5019	+0.0019	+0.40	0.1178	0.1257	+0.0079	+6.7				
9 A	2.001	2.0023	+0.0013	+0.065	0.4986	0.5003	+0.0017	+0.34	0.1194	0.1358	+0.0164	+14	8.12	7.62	-0.50	-6.1
B					0.4998	0.5087	+0.0089	+1.8	0.1197	0.1573	+0.0376	+31				
C					0.4996	0.5036	+0.0040	+0.80	0.1186	0.1262	+0.0076	+6.4				

swelling was manifested almost entirely by changes in plate thickness. In capsule 46-4 containing plates 1 and 10 only very slight dimensional density effects were noted. However, the specimens in capsules 46-2 and -5 exhibited gross thickness increases of up to 44% and similar large density decreases ranging from 6 to 13%.

Nondestructive Inspection for Flaws

To check the bond quality of plates 1 and 10 after irradiation, an ultrasonic test was performed on a Branson Instruments, Inc., "Vidigage." Scanning was performed on both sides of these two plates, and by comparison with an intentionally defective control sample it was demonstrated that no detectable nonbonds were present in plates 1 and 10. Because of obvious visual damage, ultrasonic inspection of plates 5, 6, 8, and 9 was not performed.

Burnup Analyses

The plates were then sectioned as indicated in Fig. 16 using a small abrasive wheel driven by an air motor. Burnup samples were dissolved in 1:1 $\text{HNO}_3\text{-HCl}$ using a dissolution system consisting of a thermostatically controlled hot plate, refluxing condenser, liquid trap, and gas scrubber. Aliquot solutions were analyzed by radiochemical analysis for ^{95}Zr , ^{137}Cs , and ^{144}Ce , and by fluorometric analysis for total uranium. In one instance (plate 5), where significant discrepancies existed among the various radiochemically determined values, a mass spectrometric analysis for uranium isotopes was made. This isotopic analysis agreed closely with that determined from the ^{144}Ce nuclide. Burnup in three plates was also checked by measuring the ratio of ^{60}Co to total cobalt and using this value to calculate the effective flux received by the plates. Burnup measurements are summarized in Table 3. The low burnup results obtained for capsule ORNL-MTR-46-5 using the ^{137}Cs nuclide and the fast-fission products detected in the sodium reaction residue strongly suggest that an undetectable flaw existed in both fuel plates in this capsule.

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Fig. 16. Sectioning Diagram for Miniature Fuel Plates.
"B.U." is burnup sample and "Met." is metallographic sample.

Table 3. Burnup Analyses for Fuel Plates in ORNL-MTR-46 Capsules

Capsule	Plate	Fission Burnup (% ^{235}U)				
		$^{137}\text{Cs}/\text{U}$	$^{144}\text{Ce}/\text{U}$	$^{95}\text{Zr}/\text{U}$	$^{235}\text{U}/^{236}\text{U}$	$^{60}\text{Co}/\text{Co}$
46-4	1	6.20	6.06			7.87
	10	6.69	6.52			8.72
46-2	5	9.20 ^a	29.1	21.5	27.8	29.6
	6					
46-5	8	22.3 ^a	32.9	29.4		
	9	22.2 ^a	33.4	33.4		

^aProbably low because of the volatility of ^{137}Cs , and its volatile precursors; ^{137}Cs detected in sodium of capsule 46-5.

Using the ^{144}Ce burnup values, which appear to be internally consistent, the fission densities and average heat fluxes were calculated and the results are given in Table 4.

Table 4. Burnup and Heat Flux Summary for ORNL-MTR-46 Capsules

Plate	Fission Density (Fissions per Cubic Centimeter of Fuel-Bearing Matrix)	Heat Flux ^a (Btu/hr-ft ²)
	$\times 10^{21}$	
1	0.36	540,000
10	0.39	590,000
5	1.7	590,000
8	2.0	720,000
9	2.0	720,000

^aBased on uniform volumetric heat generation and an average core thickness of 0.102 in.

The basis for the initial flux perturbation calculations was discussed previously in the "Irradiation Capsule Design" section. Because obvious differences existed between the measured and calculated burnups, the perturbation calculations were reexamined using a revised calculational model. This model considered not only the flux variations in the capsule but also the flux variation from the reactor environment into the capsule. An overall perturbation factor of 0.32 was computed with this model. A comparison of the measured burnups was made with those predicted (see Table 5) on the following bases:

Method A. Assuming $\phi_{\text{eff}} = 0.37 \phi_0$ and fission rate independent of time, (i.e., $\text{FR} = N_0 \phi_{\text{eff}} \sigma_f$).

Method B. Application of 0.66 experience factor from capsule 46-4 to capsules 46-2 and -5 and using burnups calculated as in A above.

Method C. Assuming $\phi_{\text{eff}} = 0.32 \phi_0$ and burnup defined as

$$N_t/N_0 = \sigma_f/\sigma_t [1 - \exp(-\sigma_t \phi_{\text{eff}} t)]$$

The symbols are defined as follows:

FR = fission rate in fissions $\text{cm}^{-3} \text{ sec}^{-1}$

ϕ_0 = MTR quoted value of unperturbed thermal neutron flux

ϕ_{eff} = effective or perturbed thermal neutron flux

t = time in seconds

N_0 = initial ^{235}U concentration in atoms cm^{-3}

N_t = atoms of ^{235}U remaining at time t

σ_f = microscopic fission cross section

($577 \times 10^{-24} \text{ cm}^2$ for 2200-m/sec neutrons)

σ_t = microscopic total cross section

($683 \times 10^{-24} \text{ cm}^2$ for 2200-m/sec neutrons)

Thus, Method A employs the originally calculated flux perturbation factor (see page 6) and makes no correction for ^{235}U depletion with time.

Method B merely applies an experience factor to Method A, and Method C employs the flux perturbation factor calculated from the revised model and takes into account ^{235}U depletion with time.

Table 5. Comparison of Calculated and Measured Burnups for
ORNL-MTR-46 Irradiation Capsules

Capsule	Fuel Plate	Measured Fission Burnup ^a (% ²³⁵ U)	Calculated Fission Burnup (% ²³⁵ U)		
			Method A	Method B	Method C
46-4	1	6.06	9.5		7.6
	10	6.52			
46-2	5	29.1	24.0	16.0	19.0
46-5	8	32.9	39.0	26.0	28.0
	9	33.4			

^aBased on ¹⁴⁴Ce-to-uranium ratio.

Agreement between measured and calculated burnups is poor. It is further disconcerting that the calculations are not biased either to higher or lower burnups than actually experienced. All of the calculations use the quoted MTR flux which can vary from cycle to cycle depending upon the fuel loading and changes in adjacent experiments. Significant flux shifts can be anticipated over the 26 or 27 cycle exposures of capsules 46-2 and -5.

Annealing Tests

Several annealing tests were conducted on the fuel plates in capsule ORNL-MTR-46-4. A series of anneals was made on plate 1 prior to sectioning. In the first heat treatment the plate was annealed in argon for 4 hr at 1100°F. After annealing, the plate was photographed, measured, and the bond integrity checked by ultrasonic testing. During the anneal the argon cover gas was sampled for gross fission-product activity and none was detected. Subsequent anneals were performed at 1200, 1300, 1400, and 1500°F. No thickness changes or disruptions of the core-clad bond were detected throughout this series of experiments.

After fuel plate 10 was sectioned, as illustrated in Fig. 16, two coupons labeled "Sample A" and "Sample B" were annealed at 1800°F for 50 hr using a procedure similar to that outlined above. Changes were assessed by visual, density, and dimensional observations. In addition, the argon cover gas in the annealing furnace was sampled for ^{85}Kr activity. Gas analysis indicated that about 2.5% of the total fission gases was released during the 50-hr exposure at 1800°F. Presumably, the release was from the exposed sample edges. Dimensional and density changes are summarized in Table 6. Visually, there appeared to be some surface roughening of the edges containing exposed fuel as illustrated in Fig. 17. This condition was not characterized in detail by metallography.

Metallography

Metallographic samples were taken from each miniature fuel plate at the locations indicated in Fig. 16. Generally, both longitudinal and transverse specimens were prepared. Results for each specimen are summarized below:

Plate 10.

Typical as-polished cross sections through the braze and the matrix are presented in Figs. 18 and 19. The high quality of the braze joint, showing only a few small pores, and of the fuel-clad bond are attested in these photomicrographs. Absence of extensive UO_2 stringering in these longitudinal sections is also apparent. Etching of the specimens revealed a zone 6.3 μ thick around the UO_2 particles typically attributed to alteration of the matrix by fission-fragment recoils into the stainless steel (Fig. 20). Upon protracted etching a second zone 2.5 μ thick adjacent to the UO_2 particles was detected. Some of the UO_2 particles exhibited a sharply defined grain structure; in others a grain pattern could not be detected.

Table 6. Results of Postirradiation Annealing of Samples from Plate 10

Time: 50 hr
 Temperature: 1800°F
 Atmosphere: Argon

Sample	Length				Width				Thickness				Density			
	Pre-Irradiation (in.)	Post-Irradiation (in.)	Change (in.)	Change (%)	Pre-Irradiation (in.)	Post-Irradiation (in.)	Change (in.)	Change (%)	Pre-Irradiation (in.)	Post-Irradiation (in.)	Change (in.)	Change (%)	Pre-Irradiation (g/cm ³)	Post-Irradiation (g/cm ³)	Change (g/cm ³)	Change (%)
A	0.4015	0.4045	+0.003	0.7	0.5000	0.5015	+0.0015	0.3	0.1221	0.1257	+0.0035	0.3	8.11	8.00	-0.11	1.3
B	1.0190	1.0225	+0.0035	0.3	0.2195	0.2200	+0.0005	0.2	0.1232	0.1268	+0.0036	0.3	8.11	8.00	-0.11	1.3

R-5914

Before Annealing

R-5925

After Annealing

Fig. 17. Sectioned Edge of Fuel Plate 10 Before and After Annealing at 1800°F.

R-5973

R-5974

R-5975

R-5976

R-5977

Fig. 18. Bushing to the Cladding Brazed Joint in Fermi Plate 10. As polished. 100X.

Fig. 19. Typical Cross Section Through Fermi Plate 10. As polished. 100X.

Fig. 20. Central Region of Plate 10 Showing Typical Particles of UO_2 in the Stainless Matrix. Etchant: Aqua regia. 250X.

Plate 1.

Observations on this sample were generally similar to those made on plate 10, although it will be recalled that plate 1 had been step-annealed in 4-hr increments at temperatures from 1100 to 1500°F. The inner 2.5- μ -thick damaged zone detected in the type 347 stainless steel matrix of plate 10 after heavy etching was not well defined in this plate. Typically etched microstructures observed at the edge and central regions are shown in Fig. 21.

Plate 5.

Macrographs of the longitudinal and transverse sections (see Fig. 16) from plate 5 are presented in Fig. 22. Extensive matrix cracking generally parallel to the plate surfaces is apparent as is the gross thickness increase of the specimen. Typical cross sections from the transverse metallographic sample are shown in Fig. 23 in both the polished and etched condition. Etching uncovered an unidentified second phase in the stainless steel matrix, which was concentrated near the core-clad interface. Many microcracks emanating from the UO_2 particles and propagating into the matrix are apparent. One should also note that the porosity in

Fig. 21. Microstructure of the (a) Edge and (b) Center of Plate 1 from Capsule ORNL-MTR-46-4. Etchants: 10% HNO_3 , 20% H_2O_2 (30%), and 70% H_2O for UO_2 structure; glyceria regia for matrix structure. 250X.

R-13091

Plate 5 Transverse Section

R-13125

Plate 5 Longitudinal Section

R-13148

Plate 6 Transverse Section

R-13184

Plate 6 Longitudinal Section

Fig. 22. Photomicrographs of the Metallographic Specimens of Plates in Capsule ORNL-MTR-46-2.

Fig. 23. Composite Photomicrograph Across the Thickness of Plate 5 from Capsule ORNL-MTR-46-2. (a) As polished. (b) Etchants: 10% HNO_3 , 20% H_2O_2 (30%), and 70% H_2O for UO_2 structure; glyceric regia for matrix structure. 100X

the fissile phase is greater in those particles near the surface. This situation may be the result of higher burnup near the fuel plate surface as the neutron flux at the fuel plate center line is calculated to be only about 35% of the surface flux. Examination of the UO_2 particles revealed that some contained a bright metallic second phase that was not attacked by either glycerine or the UO_2 etchant. This phase also has been observed by Richt¹³ and others.

Plate 6.

Macrophotographs of this plate are also shown in Fig. 22. The stainless steel matrix showed definite evidence of melting and the UO_2 particles had fused together into larger agglomerates. Many stainless steel inclusions were noted in the UO_2 (Fig. 24).

Plate 8.

Both longitudinal and transverse sections that were made through plate 8 are shown in Fig. 25. Swelling, as manifested by a thickness increase of 27%, is apparent in the transverse section. A 12-mil-thick separation at the core-cladding interface is also obvious in the longitudinal section. Metallographic results were generally similar to plate 5.

Plate 9.

Transverse and longitudinal sections of this plate are presented in Fig. 25, which again documents the existence of matrix cracking in specimens receiving high burnups. An indication of what appears to be intergranular cracks in the cladding associated with heavy strain in the outer fibers is shown in Fig. 26, as is the highly porous structure of the UO_2 . Large amounts of a metallic phase within the UO_2 were also observed in this plate, as illustrated in Fig. 27.

¹³A. E. Richt and L. D. Schaffer, Army Reactors Program Ann. Progr. Rept. Oct. 31, 1962, ORNL-3386, pp. 73-74.

Fig. 24. Panorama of the Molten Zone in Fuel Plate 6 from Capsule ORNL-MTR-46-2. (a) As polished. (b) Etchants: 10% HNO_3 , 20% H_2O_2 (30%), and 70% H_2O for UO_2 structure; glyceria regia for matrix structure. 100X

R-14283

Plate 8 Transverse Section

R-14261

Plate 8 Longitudinal Section

R-14325

Plate 9 Transverse Section

R-14303

Plate 9 Longitudinal Section

Fig. 25. Photomicrographs of the Metallographic Specimens from Capsule ORNL-MTR-46-5.

Fig. 26. Fuel-Cladding Interface of Plate 9 from Capsule ORNL-MTR-46-5, Showing Evidence of Intergranular Cracking in the Cladding and Highly Porous UO_2 . (a) As polished. (b) Etchants: 10% HNO_3 , 20% H_2O_2 (30%), and 70% H_2O for UO_2 structure; glyceric regia for matrix structure. 500X.

(a)

(b)

Fig. 27. Metallic Phase in the Fuel Particles of Plate 9 from Capsule ORNL-MTR-46-5. (a) As polished. (b) Etchants: 10% HNO_3 , 20% H_2O_2 (30%), and 70% H_2O for UO_2 structure; glyceria regia for matrix structure. 500X.

DISCUSSION

As noted earlier, the maximum fuel-plate center line temperature specified for the Core B hot-spot condition is 1043°F (870°F surface). This increased to 1140°F, if statistical (3- σ) hot-channel factors are considered. The maximum surface and center line temperatures experienced by the various plates in ORNL-MTR-46 experiment are listed in Table 7. The "maximum" temperature is the highest single value recorded by the indicated thermocouples, and, as such, represents an isolated experience. The "maximum average" temperature is the maximum cycle temperature averaged over in-reactor exposure. For obvious reasons, plate 6 was not included in this analysis.

Both the "maximum average" and the "maximum" surface temperatures experienced by the three capsules in the ORNL-MTR-46 program exceeded the Fermi Core B hot-spot condition. The deviations from the designed Core B surface temperatures were small for capsule 46-4 but were in excess of 100°F for capsules 46-2 and -5. In view of demonstrated temperature control problems with small in-reactor experiments, these are not unreasonable results.

The center line temperatures listed in Table 7 were calculated on the following bases:

1. The temperature drop through the cladding was computed using a 0.007-in.-thick sheath, a thermal conductivity of 12 Btu/(hr ft² °F/ft), and the heat fluxes quoted previously.

2. The temperature drop through the fuel assumed nonuniform volumetric heat generation governed by the calculated flux distribution, a fuel thickness of 0.102 in., and a fuel thermal conductivity of 10 Btu/(hr ft² °F/ft)(ref. 14). The unperturbed neutron fluxes in each capsule were those listed in Table 1.

¹⁴D. L. McElroy, unpublished work.

Table 7. Comparison of Fermi Core B Design Parameters and Experimental Results from ORNL-MTR-46 Program

Capsule	Specimen	Thermocouple	Surface Temperature (°F)			Center Line Temperature (°F)				Fission Burnup (at. % ²³⁵ U)		Specimen Condition
			Specimen Maximum	Specimen Maximum Average	Core B Hot Spot	Specimen Maximum	Specimen Maximum Average	Core B Hot Spot	Core B Hot Spot and Hot Channel	Measured	Core B Throughput	
46-2	5	TC-12	1165	1005	870	1270	1110	1043	1140	29.1	20.7	Severe swelling ($\Delta V = -13\%$) and matrix cracking.
46-4	1	TC-13	935	890	870	1085	1040	1043	1140	6.06	20.7	Virtually unaltered.
	10	TC-15	1010	945	870	1165	1100	1043	1140	6.52	20.7	Virtually unaltered.
46-5	8	TC-7	1130	1000	870	1290	1160	1043	1140	32.9	20.7	Severe swelling ($\Delta V = 8\%$) and matrix cracking.
	9	TC-8	1100	1005	870	1260	1165	1043	1140	33.4	20.7	Severe swelling ($\Delta V = 6\%$) and slight matrix cracking.

It is apparent from Table 7 that the "maximum average" center line temperatures experienced by all samples, except plate 1, in the ORNL-MTR-46 program closely approximated the Fermi Core B hot-spot plus hot-channel conditions.

As discussed in an earlier section, agreement between measured and calculated burnups was not good. Thus the burnup in capsules 46-5 and -2 exceeded the Core B throughput burnup by 60 and 40%, respectively, while the burnup in capsule 46-4 was, as planned, lower than the throughput burnup. An analysis of available burnup data, from several UO₂-stainless steel irradiation programs, presented as Fig. 28, suggests that for a surface temperature of 900°F, the UO₂-stainless steel dispersions should be capable of burnups of $16 \text{ to } 18 \times 10^{20}$ fissions/cm³ without gross failure. Gross failure is here defined as blistering or extensive matrix cracking. The lower figure probably is more realistic for thick fuel plates with large temperature gradients. This fission density is equivalent to a burnup of 27% of the ²³⁵U atoms for a 33 wt % UO₂-stainless steel dispersion.

This limit deals only with gross failure of fuel. It must also be remembered that a controlled linear rate of swelling with burnup occurs at burnups less than those required to produce failure. For example, 26 wt % UO₂-stainless steel dispersions irradiated at temperatures of 300 to 700°F underwent a volume increase of 4.6% at a burnup of 1×10^{21} fissions/cm³. (Ref. 15)

CONCLUSION

Three instrumented capsules containing miniature fuel plates were irradiated in the MTR to burnups of 6, 29, and 33% of the ²³⁵U atoms at maximum average surface temperatures of about 900 to 1000°F. The test specimens, containing 33 wt % UO₂ dispersed in and clad with type 347 stainless steel, represented fuel proposed for Core B of the Enrico Fermi Reactor. No significant changes occurred in the lowest burnup

¹⁵A. E. Richt, Metals and Ceramics Div. Ann. Progr. Rept. May 31, 1963, ORNL-3470, pp. 225-26.

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Fig. 28. Irradiation Stability of UO_2 -Stainless Steel Dispersions.

samples during irradiation. At the two higher exposures, severe fuel-plate swelling and intergranular fracturing of the matrix were observed, indicating that the burnup limitations of this fuel had been exceeded for these irradiation conditions. Correlation of these results with other work suggests that at a surface temperature of 900°F the maximum tolerable burnup will be less than 27%.

ACKNOWLEDGMENT

The authors wish to acknowledge the services of the large group of people who participated in this program. Particular recognition is afforded to:

J. H. Cherubini	Specimen Fabrication
R. C. Williams (APDA)	Specimen Fabrication
J. I. Federer	Capsule Fabrication
T. S. Lundy	Capsule Design
M. C. Ziemke	Capsule Design

The services of R. W. Darmitzel, who supervised the postirradiation work conducted at General Electric Company's Vallecitos Atomic Laboratory hot cells, are also gratefully acknowledged.

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