IN-1160
Issued: April 1968
Metals, Ceramics, and Materials
TID-4500

IRRADIATION TESTING OF FUEL FOR THE MARK I
CORE OF THE ARGONNE ADVANCED RESEARCH REACTOR

BY

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ACKNOWLEDGEMENTS

The authors wish to acknowledge that these efforts, which include plate fabrications by Martin Nuclear of Baltimore, Maryland, and Battelle Memorial Institute of Columbus, Ohio, as well as the irradiation testing and evaluating by Idaho Nuclear Corporation (INC), were sponsored and coordinated by Argonne National Laboratory (ANL).

Coordination and direction of the irradiation tests were executed by C. V. Pearson of ANL and W. C. Francis of INC. The environmental calculations for reactor sample insertion were performed by M. Zukor under the direction of V. A. Walker. Analysis of the microconstituents was made on the electron microprobe by Dr. W. F. Zelezny. The remote handling and examinations were performed by the Metallurgy and Hot Cells Branch under the direction of D. R. Alvord and K. L. Hoopingarner. Isotopic burnup analyses were performed by the Analytical Chemistry Branch.

In addition to the efforts mentioned above, numerous services and suggestions of others have been utilized. The cooperation of these organizations and individuals is also gratefully acknowledged.

ABSTRACT

The irradiation testing and evaluation of a 37 wt% low-density UO_2 stainless steel dispersion fuel is reported. This fuel was originally proposed for the long fuel cycle operation of the Argonne Advanced Research Reactor. The results show that starting density of the fuel can affect the operating life.

SUMMARY

In the past, the use of porosity in UO_2 for the accommodation of fission products in dispersion fuel plates was limited because crushing during element fabrication was considered detrimental. Since maximization of the mean free path between the particles is considered to be of primary importance, uranium dioxides of high bulk densities which resist fragmentation and retain a high degree of spheroidicity during fabrication have been preferred to the low-density UO_2 which has a tendency to fragment and stringer.

Characterization and evaluation studies at Battelle Memorial Institute (BMI) have recently shown that the crush strength of low-density UO_2 could be improved by small additions of TiO_2 and CaO . The basic UO_2 fuel selected for the stainless steel design of the Argonne Advanced Research Reactor had a low bulk density, nominally 85 percent of theoretical, and contained 0.26 wt% CaO and 0.96 wt% TiO_2 to resist particle fracturing.

This basic fuel was used to produce 37 wt% UO_2 stainless steel fuels by two different fabrication procedures. One procedure, similar to the Advanced Army Reactor techniques developed by Martin Company and using a total roll reduction $\approx 2:1$, resulted in nearly idealized particle dispersion with very little particle fragmentation and an as-fabricated fuel density of 89 percent of theoretical. The other procedure used a 6:1 roll reduction resulting in particle fragmentation with some stringing but with a lower as-fabricated fuel density of 82 percent of theoretical. Both of these low-density fuels exhibited the ability to accommodate growth in the voidage provided, but the 82 percent dense material (even with its less desirable microstructure) exhibited the lower growth rate. The growth rates amounted to 0.40 and 0.26 percent, respectively, for each 10^{20} fission/cc.

The sample fuel plates, measuring $5\text{-}3/4 \times 1\text{-}1/4$ inches and containing 0.030-inch-thick cores with 0.005-inch 347 stainless cladding, were irradiated for two cycles in the Engineering Test Reactor (ETR), or up to 25 percent burn-up of the fully enriched fuel. The ability of the UO_2 to grow into its own voidage, rather than cause swelling to the entire fuel plate, was apparent at these heat fluxes, which reached $1.1 \times 10^6 \frac{\text{Btu}}{\text{hr-ft}^2}$, and at the relatively low center line temperatures of $\approx 200^\circ\text{C}$.

The original fabrication cracks and voids in the UO_2 were filled in during irradiation with a more ductile phase before fission gas agglomeration became evident. When this phase was present in sufficient quantities, the tendency for brittle cracking was reduced.

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I. INTRODUCTION

This is being written to report the results of an irradiation program conducted in support of the Argonne Advanced Research Reactor (AARR) which was performed at the National Reactor Testing Station by first the Phillips Petroleum Company and later the Idaho Nuclear Corporation (INC). The purpose of this program was to investigate the irradiation stability of the fuel and cladding material proposed for use in the fuel elements of the Mark I core.

This reactor, using beryllium as a reflector, is light-water cooled and moderated. It was expected that initially the reactor would produce thermal neutron fluxes in the range of 3 to 5×10^{15} . Plans were made to significantly increase these fluxes in a second stage of the reactor development. In order to provide the flexibility needed for this planned upgrading of the reactor, the original reactor design used 37 wt% UO_2 dispersed in a stainless steel matrix and clad with stainless steel by roll bonding[1]. The fuel loadings proposed for use in the reactor fuel plates and the flux levels expected were somewhat higher than those experienced in other UO_2 stainless steel dispersion-type fuel systems. Moreover, the fuel assembly method proposed was unique in that stainless steel spacer wires were to be brazed over the active portions of the fuel plates, thus creating heat transfer and thermal stress problems not previously encountered in high flux reactors. In view of these new parameters in fuel element operation, it was believed that an irradiation test program was worthwhile. A similar program directed toward the needs of the Advanced Test Reactor (ATR) and the High Flux Isotope Reactor (HFIR) had just been completed[2], and space was available in the irradiation facility used for this test.

The ATR and HFIR fuel elements use aluminum clad dispersion-type fuel elements. When the AARR irradiation test program was nearing completion it was decided that this reactor would also use aluminum elements. Consequently, it was deemed desirable and feasible to change the internal design of the AARR to accommodate the HFIR type core, and the irradiation test program of the stainless steel clad fuel plates was terminated leaving some of the post-irradiation examinations unfinished.

II. FUEL SAMPLES TESTED

Three fuel compositions, made by two manufacturing procedures, were tested in two geometries. Table I shows the compositions of the sample fuel plates. These were made by Martin Nuclear (Martin) of Baltimore, Maryland, and Battelle Memorial Institute (BMI) of Columbus, Ohio.

TABLE I
AARR SAMPLE PROGRAM COMPOSITIONS

Composition Number	A-15	A-16	A-29
Fabricator	Martin Nuclear	Battelle Memorial Institute	Battelle Memorial Institute
Fuel Material	UO ₂	UO ₂	UO ₂
Wt.% Fuel	39	37	37
Roll Reduction Ratio Used	2:1	6:1	6:1
U-235 Loading g/plate	6.75	5.86	6.12
g/in ² core	1.24	1.08	1.12
B(nat.) Loading g/plate	Nil	Nil	0.0013 to 0.041
g/in ² core	Nil	Nil	0.00023 to 0.0075

In Figures 1 and 2 the geometries used for testing the specimens are shown. All of the sample fuel plates were made to the dimensions shown in Figure 1. Three of the samples were brazed together to form a micro fuel assembly as shown in Figure 2. The purpose of the micro assembly was to test the use of the flat wire spacers which were brazed over the active portion of the fuel plate cores. This method of fuel assembly was to be a unique feature of AARR fuel elements.

The three-plate AARR brazed-rib subassembly consisting of fuel plates A-15-932, A-15-934 (Martin, numbers 32, 34), and A-16-954 (BMI, number 54) was irradiated during ETR Cycles 80 through 82 for a total of 13,221 MWd of reactor operation. The three plates of the subassembly were held together by 40-mil-wide 347 stainless steel spacer wires, the wires forming a 100 mil channel between the plates. The spacer wires were brazed to the plates with GE Number 81 brazing alloy (70 percent Ni, 20 percent Cr, 10 percent Si), titanium-aluminum oxide being used as a flux stop. The brazing operation was performed by Pyromet Corporation of San Carlos, California.

The same batch of fully enriched UO₂ was used as fuel in all of the samples. Characterization and evaluation studies at Battelle had shown a high crush strength for this particular batch of UO₂[³]. The chemical and isotopic analysis of this material is shown in Table II.

Most of the procedures used to fabricate the samples manufactured by Martin Nuclear have also been described by Goslee[⁴] and were designed to

Fig. 1 Design of sample fuel plates used for AARR irradiation test program.

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Fig. 2 Design of micro fuel assembly used in AARR irradiation testing program -- end view of 5-3/4-inch-long assembly.

produce an optimum dispersion of UO_2 particles in stainless steel matrix with a minimum of particle fragmentation. In these procedures flame fired stainless steel coated UO_2 balls (37 wt% UO_2) were mixed with a Jaquar (Guar gum) binder and water. The blended slurry was extruded (by means of a hydraulic ram) onto a flat strip of stainless steel. The extruded strips were transferred to "Mylar" sheets and then air-dried at room temperature for 1-1/2 to 2 hours. After this the strips were transferred to flatscreens for drying in an oven operating at 75°C . The green strips were next fired in a hydrogen atmosphere at 1300°C for three hours. The fired strips, after cooling, were further densified by hydrostatic pressing at 60,000 psi followed by annealing at 1300°C for one hour in a hydrogen atmosphere. After cooling, the UO_2 stainless steel strips were sheared to form the fuel plate cores.

The cores were assembled with picture frames and cover plates (in the form of foils) by arc welding. The assemblies were heated to 1180°C in a hydrogen atmosphere for 20 minutes and then hot rolled to $0.038 \begin{smallmatrix} + 0.001 \\ - 0.000 \end{smallmatrix}$ inch using a 2:1 reduction with five-minute reheats between rolling passes.

The hot rolled plates were washed in acetone and then cleaned in an aqueous solution of 15 percent HNO_3 and 5 percent HF by volume. After being rinsed in flowing water the plates were dried with warm air.

The cleaned plates were heat treated at 1180°C in a hydrogen atmosphere for one hour and flattened by roll leveling. After ultrasonic inspection, the plates were sheared to final size.

The fabrication procedures used by BMI have been reported by Acunius et al[5]. In these procedures the UO_2 powders were coated with 1-1/2 wt% of paraffin by blending with CCl_4 as a solvent. The coated UO_2 powder was chilled to -10°C and screened through a 60 mesh screen onto a weighed amount of stainless steel powder. The powders were blended in a rotating glass jar and then compacted at 50 tsi in a double acting steel die.

The green compacts were sintered at 1200°C for 75 minutes in hydrogen. After cooling the sintered compacts were coined in a double acting die at 50 tsi. The core compacts were assembled into packs with picture frames and cover plates by welding. After preheating for 30 minutes in a hydrogen atmosphere furnace at 1200°C the packs were hot rolled for a 6:1 reduction until the plate thickness was 0.045 inch. The hot rolled plates were cleaned of surface oxide by immersing in hot aqueous solution of 15 percent HNO_3 and 5 percent HF . The sintering and hot rolling temperature was lowered to 1150°C when a burnable poison was used.

The excess stock was removed from the ends of the rolled plates, and the plates were cold rolled to the final thickness using reduction of less than 1 percent per pass. The cold rolled plates were flattened by annealing at 1175°C in a hydrogen atmosphere while being clamped between stainless steel platens.

After being radiographed and cleaned, the plates were machined to final size.

Prior to irradiation the fuel plates were inspected to ensure reactor-worthiness and to establish the pre-irradiation properties of the samples. The plates were dimensioned, radiographed, ultrasonic inspected, and weighed. The parameters determined by the inspection on the irradiation specimens are shown in Table III.

Examination of the data in Table III indicates that the parameters of the plates made without a boron addition (compositions A-15 and A-16) are reasonably uniform from plate to plate within the batch. However, in the A-29 compositions, two plates (A-29-938 and A-29-939) have considerably different properties than the other plates in the batch.

Plate A-29-938 was over heated during fabrication. This produced a more dense core. The core area was increased and the core thickness decreased.

Boron was added to plate A-29-939 as 6 vol% of boro-silicate glass beads. This addition had the effect of lowering the core density.

TABLE II

ANALYSIS OF UO_2 USED IN
AARR IRRADIATION PROGRAM

Wt% U in UO_2	=	87.2
Wt% U-234 in U	=	1.0556
Wt% U-235 in U	=	93.0693
Wt% 236 in U	=	0.2584
Wt% 238 in U	=	5.6167
Wt% CaO in UO_2	=	0.26
Wt% T_iO_2 in UO_2	=	0.96
O/U	=	1.99

TABLE III

PARAMETERS OF AARR IRRADIATION PROGRAM FUEL SPECIMENS

Composition No.	Plate No.	Fabricator	Fabricator's Plate No.	Sample Plates									Average Core						
				Length (in.)	Width (in.)	Thickness (mils) PF Area	Core Area	Gross Wt. (g)	Density (g/cc)	Core Wt. g	U-235 Wt. g	B Wt. g	Calculated Density (g/cc)	Length (in.)	Width (in.)	Area (in.)	Calculated Volume (cc)	Calculated Volume (in. ³)	Calculated Thickness ^[a] (in.)
A-15	923	Martin	23	5.760	1.257	39.3	39.8	38.043	8.301	20.976	6.752	---	8.564	5.37	1.00	5.37	2.449	0.1491	0.0278
"	925	"	25	5.763	1.258	39.1	39.7	37.787	8.309	20.870	6.718	---	8.576	5.70	0.98	5.59	2.434	0.1485	0.0266
"	926	"	26	5.760	1.260	39.8	40.4	38.229	8.308	20.962	6.748	---	8.578	5.39	1.00	5.39	2.444	0.1491	0.0277
"	933	"	33	5.758	1.253	39.6	39.8	37.871	8.310	21.067	6.781	---	8.547	5.43	1.00	5.43	2.457	0.1499	0.0267
A-16	943	BMI	43	5.750	1.251	40.0	40.9	38.903	8.150	19.619	5.874	---	8.304	5.23	0.98	5.13	2.363	0.1442	0.0281
"	945	"	45	5.750	1.251	39.9	40.9	38.948	8.140	19.581	5.863	---	8.291	5.23	0.98	5.13	2.362	0.1441	0.0281
"	947	"	47	5.749	1.251	40.0	40.8	38.913	8.147	19.615	5.873	---	8.298	5.23	0.98	5.14	2.364	0.1443	0.0281
"	952	"	52	5.748	1.251	39.8	40.9	38.856	8.147	19.480	5.833	---	8.304	5.25	0.98	5.15	2.346	0.1431	0.0278
A-15 ^[d]	934	Martin	34	5.755	1.252	39.0	39.6	37.880	8.312	20.987	6.756	---	8.581	5.36	1.00	5.36	2.446	0.1493	0.0279
" ^[d]	932	"	32	5.751	1.252	39.0	39.6	37.686	8.314	21.005	6.762	---	8.581	5.38	1.00	5.38	2.448	0.1494	0.0278
A-16 ^[d]	954	BMI	54	5.749	1.250	40.0	40.8	38.939	8.166	19.548	5.853	---	8.311	5.19	0.98	5.09	2.352	0.1435	0.0282
A-29	927	BMI	48	5.752	1.251	40.6	40.7	38.849	8.154	21.403	6.19	0.026 ^[b]	8.283	5.49	1.01	5.55	2.584*	0.1577	0.0284
"	928	"	49	5.750	1.251	40.8	41.0	39.304	8.162	21.487	6.22	0.026 ^[b]	8.338	5.47	1.02	5.58	2.577	0.1573	0.0282
"	929	"	50	5.752	1.250	41.2	41.2	39.477	8.151	21.397	6.19	0.026 ^[b]	8.322	5.40	1.01	5.45	2.571	0.1569	0.0288
"	936	"	77	5.751	1.251	40.8	41.0	39.278	8.163	21.391	6.188	0.041 ^[b]	8.349	5.50	1.01	5.55	2.562	0.1563	0.0282
"	938	"	23	5.750	1.252	41.0	41.0	39.459	8.211	21.391	6.15	0.0397 ^[b]	8.399	5.21	1.20	6.25	2.547	0.1554	0.0249
"	939	"	68	5.752	1.251	40.9	41.2	38.435	7.954	19.892	5.755	0.0013 ^[c]	7.951	5.40	1.00	5.40	2.502	0.1527	0.0283

[a] Calculated from core area as determined by radiography and plate density as determined by water immersion.

[b] In the form of ZrB_2 .[c] In the form of glass beads containing B_2O_3 .

[d] These samples were brazed together to form a brazed rib micro-fuel assembly.

One plate of each type was prepared for metallographic examination -- these microstructures are shown in Figures 3 through 6.

50X

VILELLA'S ETCH

Fig. 3 Microstructure of UO_2 stainless steel fuel plate made by Martin Nuclear Company (composition A-15). This is a section through the core end of the plate and shows the small amount of core stringering produced by the 2:1 reduction ratio used.

50X

VILELLA'S ETCH

Fig. 4 Microstructure of UO_2 stainless steel fuel plate made by Battelle Memorial Institute (composition A-16). This is a section through the fuel core end of the plate and shows somewhat greater amount of core stringering than that observed in the composition A-15 plates.

50X

VILELLA'S ETCH

Fig. 5 Microstructure of UO_2 stainless steel fuel plate containing boron as ZrB_2 . This plate was made by Battelle Memorial Institute (composition A-29). This is a section through the core end of the plate and shows the end effects produced.

Fig. 6 Microstructure of UO_2 stainless steel fuel plate (composition A-29 plates in which boron was added as boro-silicate glass beads).

The composition A-15 plates, made by Martin Nuclear, had fuel plate cores that were appreciably more dense than the other plates. The microstructures of these plates also show less stringering of the fuel particles.

III. TEST ENVIRONMENT

Irradiation testing was conducted in the PAED G-12 water loop of the Engineering Test Reactor (ETR). The PAED G-12 loop, which provides an irradiation facility in the ETR for fuels and materials testing in a high flux, pressurized water environment, is located in the southwest corner (G-12 position) of the ETR and utilizes a Zircaloy-2 in-pile tube. During ETR Cycles 70 and 71, a once-through mode of loop operation was employed. A bypass stream of ETR coolant is pressurized, pumped through the sample train, and returned to the ETR primary coolant stream. Bulk inlet temperature is limited to that of the ETR primary coolant temperature, 43°C.

Commencing with ETR Cycle 76 the PAED G-12 loop was converted to a feed-and-bleed mode of operation to permit higher bulk coolant temperatures. After passing through the sample train, the coolant flow is split, the major fraction being recirculated through the loop. The nominal loop operating parameters for each mode of operation are listed in Table IV.

TABLE IV

PAED G-12 PRESSURIZED WATER
LOOP OPERATING PARAMETERS

	<u>"Once-Through"</u> <u>System</u>	<u>"Feed-and-Bleed"</u> <u>System</u>
ETR Cycles:	70, 71	78, 80, 81, 82
Loop Flow:	114 gpm	105 gpm
Inlet Temperature:	43°C	90°C
Inlet Pressure:	462 psig	760 psig
In-pile Differential Pressure:	86 psig	82 psi
pH	5.0 ± 0.2	5.0 ± 0.2

The G-12 sample assembly (shown in Figure 7) consists of six 6-inch-long sample holders, each holder containing space for four sample fuel plates 1.25 x 5.75 inches. Each tier can accommodate a 0.150-inch-thick fueled, instrumented thermocouple plate and three 0.050-inch-thick sample fuel plates separated by 0.100-inch-wide flow channels. The three-plate sample holder is detachable from the thermocouple section. Quarter-inch-wide hafnium flux suppressor plates separate plates in adjacent tiers.

Tiers 1 and 6, located near the top and bottom respectively of the reactor core, exhibit sharp flux gradients down the tier. The neutron flux varies by a factor of two down these tiers. Tier 1 exhibits the lowest thermal neutron flux ($\approx 3 \times 10^{14}$ n/cm²sec) followed by tier 6 ($\approx 3.5 \times 10^{14}$ n/cm²sec). The maximum nominal thermal flux ($\approx 5.5 \times 10^{14}$ n/cm²sec) occurs in tier 4 with tiers 3 and

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Fig. 7 G-12 sample holder assembly.

5 slightly lower. Because of the high fuel loading in the AARR samples, it was necessary in some instances to shroud portions of the sample holders with hafnium to reduce the thermal flux (see Figure 8).

Fig. 8 Three-plate sample holders with hafnium shroud on tiers 5 and 6 to reduce neutron flux in tier 6.

During ETR Cycles 70 and 71, a total of eight composition A-15 and A-16 UO_2 -SS fuel plates were irradiated up to 5881 MWd (33.6 full power days) of reactor operation. Four A-15 (Martin) plates each containing 6.75-g U-235 were irradiated in tier 1, while four 5.86-g U-235 A-16 (BMI) plates were irradiated in tier 6 of the G-12 sample assembly. Due to the higher flux in tier 6 it was

necessary to shroud the bottom 2-1/2 inches of tier 5 and top 2-1/2 inches of tier 6 with 0.010 inch of hafnium. One plate of each composition was changed out after Cycle 70. Thus, two plates of each composition were irradiated for one cycle and two plates for two cycles. Heat fluxes of 0.8 to $1.6 \times 10^6 \frac{\text{Btu}}{\text{hr-ft}^2}$ and plate center line temperatures of up to $\approx 250^\circ\text{C}$ were obtained.

For ETR Cycle 80, AARR fuel plates A-29-927, -928, and -929 containing 6.2 g U-235 and 0.026 g natural boron as zirconium diboride (ZrB_2) were irradiated in tier 1 of the sample assembly. For Cycle 81 these three plates were moved to tier 2, the lower 4-1/2 inches being shrouded with 0.035-inch-thick hafnium. The hafnium shroud reduced the flux near the bottom of the holder to such an extent as to reverse the flux profile in this tier, resulting in a uniform plate burnup over the two cycles. These samples were irradiated for a total of 50.9 full power days, under average heat fluxes of 0.9 to $1.0 \times 10^6 \frac{\text{Btu}}{\text{hr-ft}^2}$ and average plate center line temperatures of up to $\approx 200^\circ\text{C}$.

During Cycles 81 and 82, AARR fuel plates A-29-936, -938, and -939 were irradiated in tier 1 for 50.2 full power days of reactor operation. Plates A-29-936 and -938 contained boron in the form of ZrB_2 , whereas sample A-29-939 contained 6 vol% boro-silicate glass beads containing 1.13 wt% boric oxide, B_2O_3 . Average heat fluxes of 0.6 to $1.1 \times 10^6 \frac{\text{Btu}}{\text{hr-ft}^2}$ and core center line temperatures to $\approx 215^\circ\text{C}$ were obtained.

The three-plate brazed-rib subassembly, consisting of fuel plates A-15-932, A-15-934, and A-16-954, were irradiated in tier 6 during ETR Cycles 80, 81, and 82 for a total of 75.5 full power days. The three plates of the subassembly were held together by 0.040-inch-wide 347 stainless steel spacer wires, the wires forming 0.100-inch-wide channels between the plates of the subassembly. Shrouding of part of the sample holders in tiers 5 and 6 was necessitated by the reduced heat transfer brought about by the spacer wires being located over the fuel core area. During all three cycles, the top 2-1/2 inches of tier 6 and bottom 2-1/2 inches of tier 5 were shrouded with hafnium to reduce the thermal flux, the hafnium thickness being reduced after each cycle to compensate for fuel burnup. Average heat fluxes of 0.9 to $1.1 \times 10^6 \frac{\text{Btu}}{\text{hr-ft}^2}$ and fuel center line temperatures of up to $\approx 215^\circ\text{C}$ were achieved.

Three nonfueled 347 stainless steel plates, with a 0.40-inch-square 347 stainless steel spacer wire brazed down the center of each plate, were irradiated in tier 6 during Cycle 78 and in tier 5 during Cycle 80. These ribs were also brazed with GE Number 81 brazing alloy. A thermal neutron fluence up to 20×10^{20} n/cm² was achieved to show that the braze would withstand irradiation.

IV. POST-IRRADIATION EXAMINATION PROCEDURES

The purpose for post-irradiation examination of these sample fuel plates was to detect the changes which take place as a function of radiation exposure and as a function of temperature. Selection of the tests and property measurements was based on the likely changes anticipated. It was essential to know the history and properties of the starting material in order to detect the changes which occur. The tests which were performed on the various plates are shown in condensed form in Table V.

Overall photography was used to record the general appearance of the plates and detect any discoloring, warping, blistering, or handling defects. The pictures were taken through the TRA hot cell periscopes which can achieve up to ten power.

When uranium fissions, the fission products occupy more volume than the uranium did^[6]. If these fission products are not accommodated in the fuel core they will cause the fuel plate to swell. This swelling is detected by water immersion techniques employing the Archimedes principle. The accuracy of these data in terms of plate density is 0.01 g/cc.

It is important that any swelling which occurs is in the thickness direction. Changes in length or width could result in plate bowing. If the cladding material is strong enough, length and width swelling will be restrained. The plates were dimensioned before and after irradiation to ensure proper swelling direction. The readings are taken to the nearest 0.001 inch. This accuracy could detect about 3 percent swelling if it is in the thickness direction or 0.1 percent swelling in the width direction. Plate bowing was not observed, so there has been no further reporting of this data.

There was a sharp neutron flux gradient in the longitudinal direction on compositions A-15 and A-16. To determine the variation in U-235 burnup, a series of five samples (A through E) were taken along the plates for isotopic burnup analysis (see Figure 9). Metallography samples, F through J, of known burnup could then be selected.

A different technique for U-235 burnup analysis was used on plates of composition A-29. First the plates were gamma scanned. Then isotopic burnup analysis was made on selected samples of known gamma activity. Burnup at any position on the plate could then be determined as a function of gamma activity.

Some of the fission products are gaseous. These gaseous atoms may agglomerate in the fuel particle and exert a pressure. The pressure is resisted by a shell of stainless steel matrix material which surrounds each fuel particle. At some threshold temperature, the matrix shells will rupture causing blistering of the fuel plate. This condition was purposely produced as part of the test procedure by post-irradiation, incremental heat treatments. The samples were heated for 30 minutes at a particular temperature, air cooled, and weighed in water (as previously described). These incremental heat treatments were continued until blistering occurred.

Since all the tests could not be made on each portion of each fuel plate, the material was divided, some for the as-irradiated examination and some for as-blistered examination. The A-15 and A-16 compositions were divided by designating certain whole plates for the as-irradiated information. The composition A-29

TABLE V

POST-IRRADIATION TESTS PERFORMED ON THE VARIOUS FUEL PLATES

<u>Plate Number</u>	<u>Photography</u>	<u>Dimension</u>	<u>Gamma Scan</u>	<u>Dry Weight</u>	<u>Δ Volume</u>	<u>As-Irradiated Metallography</u>	<u>Burnup</u>	<u>Blister Test</u>	<u>Blister Metallography</u>	<u>Micro-Hardness</u>
A-15-923	X	X		X	X	X	X			X
A-15-925	X	X		X	X	X	X			X
A-15-926	X	X		X	X			X	X	X
A-15-933	X	X		X	X	X	X			X
A-16-943	X	X		X	X			X	X	X
A-16-945	X	X		X	X	X	X			X
A-16-947	X	X		X	X	X	X			X
A-16-952	X	X		X	X	X	X			X
A-15-932	X			X	X					
A-15-934	X			X	X					
A-16-954	X			X	X					
A-29-927	X	X	X	X	X	1/2		1/2	X	X
A-29-928	X	X	X	X	X	1/2	X	1/2	X	X
A-29-929	X	X	X	X	X	1/2	X	1/2	X	X
A-29-936	X	X	X	X	X	1/2	X	1/2		X
A-29-938	X	X	X	X	X	1/2	X	1/2	X	X
A-29-939	X	X	X	X	X	1/2		1/2		X

Fig. 9 Sectioning scheme for composition A-15 and A-16 plates which were used for burn-up and as-irradiated metallography.

plates were each divided into longitudinal halves, half for as-irradiated information and half for as-blistered information. Metallography and microhardness tests were taken in these states to ascertain the effects of the irradiation test.

Plates A-15-932, A-15-934, and A-16-954 were joined together in a sub-assembly which was treated as a unit and was examined for plate warping and swelling. The pertinent data are reported in the various sections of this report as needed to illustrate a point or property change.

V. EXPERIMENTAL RESULTS

1. FUEL BURNUP, HEAT GENERATION, AND PLATE TEMPERATURE

The composition A-15 and A-16 plates irradiated during ETR Cycles 70 and 71 were sectioned according to the sectioning scheme previously shown in Figure 9. The five longitudinal samples (Sections A through E) from each of plates A-15-923, -925, -933, and A-16-945, -947, -952 were sent to the Analytical Chemistry Branch for isotopic and Cs-137 burnup analysis. A burnup profile down these plates was obtained. The burnup for plates A-15-926 and A-16-943 was obtained from the U-235 burnup of adjacent AARR fuel plates and from the ETR Critical Facility (ETRC) flux measurements made on a nuclear mock-up of the G-12 loop. A sharp flux gradient, exhibiting a burnup variation of up to 80 percent along the 5-1/2-inch fuel core is observed. The burnups for these plates are shown schematically in Figure 10.

The AARR fuel plates irradiated during ETR Cycles 80 through 82 (A-29-927, -928, -929, -936, -939) were gamma scanned in the TRA Hot Cells using a 0.50-inch-long 0.040-inch-wide aperture. Five horizontal scans across and one vertical scan down each plate were made. The isotopic burnup values at positions of known gamma activity were used to calibrate the gamma scans so as to obtain the burnup profile along each plate. Plates A-29-927, -928, and -929 exhibited a flat burnup profile, with the average U-235 being 23.2 percent (12.0×10^{20} fiss/cc), 21.3 percent (11.1×10^{20} fiss/cc), and 23.9 percent (12.5×10^{20} fiss/cc), respectively, as shown in Figure 11. Plates A-29-936, -938, and -939 were irradiated in tier 1 during both Cycles 81 and 82 and exhibited the sharp burnup profiles typical of this tier. The burnup ranged from 15 to 28 percent down a given plate, averaging 20.1 percent (10.5×10^{20} fiss/cc) 21.8 percent (11.5×10^{20} fiss/cc) and 21.1 percent (10.5×10^{20}), respectively. The burnup profiles on these plates are shown in Figure 12.

The AARR brazed-rib subassembly, consisting of plates A-15-932, A-15-934, and A-16-954 was irradiated in tier 6 during Cycles 80, 81, and 82. No chemical burnup analysis was made on the brazed rib subassembly plates. Based on the analysis of the ETRC G-12 nuclear mock-up data, the fuel burnup was calculated for these plates. The plates of the AARR subassembly received an average U-235 burnup of ≈ 32 to 35 percent.

The irradiation data for the UO₂-SS fuel plates irradiated during ETR Cycles 70 and 71 and 80 and 82 are expanded in Table VI. With the exception of the plates comprising the subassembly, U-235 burnup was determined from gamma scans and isotopic burnup analysis. Based on these burnup analyses, average heat fluxes and plate surface and center line temperatures were calculated. These are values averaged over the total irradiation -- one, two, or three cycles.

Fig. 10 Cycles 70 and 71 fuel burnup shown schematically.

Ave U^{235} Burnup (%)

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Fig. 11 Cycles 80 and 81 fuel burnup shown schematically.

Fig. 12 Cycles 81 and 82 fuel burnup shown schematically.

TABLE VI

AARR FUEL PLATE IRRADIATION DATA -- BURNUP,
HEAT GENERATION, TEMPERATURE

Plate Number	Cycle	U-235 (g)	BU (percent)	Fissions cc of core	Heat Flux Btu hr-ft ²	Average Temperature Surface (°C)	Average Temperature Center line (°C)
A-15-923	70	6.75					
A			5.1	3.0 x 10 ²⁰	0.66 x 10 ⁶	80	132
B			4.8	2.9	0.64	78	127
C			5.8	3.5	0.77	84	143
D			7.1	4.2	0.92	92	165
E			8.1	4.8	1.06	104	188
Average			6.2	3.7	0.85	88	152
A-15-925	70 and 71	6.72					
A			11.3	6.7 x 10 ²⁰	0.72 x 10 ⁶	84	143
B			11.5	6.9	0.74	84	145
C			14.1	8.4	0.90	92	166
D			16.6	9.9	1.07	100	187
E			19.8	11.8	1.27	109	213
Average			14.7	8.7	0.95	94	171
A-15-926	70 and 71	6.75					
A			9.8	6.0 x 10 ²⁰	0.65 x 10 ⁶	76	129
B			9.9	6.1	0.66	77	130
C			12.2	7.5	0.81	85	154
D			14.3	8.7	0.94	96	172
E			17.1	10.4	1.12	101	196
Average			12.7	7.7	0.84	87	156
A-15-933	71	6.76					
A			5.6	3.5 x 10 ²⁰	0.71 x 10 ⁶	83	142
B			5.2	3.3	0.67	80	134
C			6.5	4.1	0.83	88	156
D			7.8	4.9	1.00	97	179
E			9.4	5.9	1.20	107	204
Average			6.9	4.3	0.88	91	163
A-16-943	70 and 71	5.87					
A			21.9	11.8 x 10 ²⁰	1.23 x 10 ⁶	130	235
B			20.2	10.8	1.13	126	222
C			19.5	10.5	1.09	124	217
D			16.3	8.7	0.91	118	196
E			15.3	8.2	0.86	115	188
Average			18.6	10.0	1.04	124	210
A-16-945	70 and 71	5.86					
A			24.8	13.3 x 10 ²⁰	1.39 x 10 ⁶	137	253
B			22.9	12.3	1.29	132	241
C			22.1	11.8	1.23	131	237
D			18.5	9.9	1.04	123	212
E			17.4	9.3	0.97	122	206
Average			21.1	11.3	1.18	129	230
A-16-947	70	5.87					
A			11.1	6.0 x 10 ²⁰	1.27 x 10 ⁶	134	242
B			10.4	5.6	1.19	130	231
C			9.9	5.3	1.13	128	224
D			8.2	4.4	0.93	120	200
E			8.0	4.3	0.91	119	197
Average			9.5	5.1	1.09	126	219
A-16-952	71	5.83					
A			12.0	6.5 x 10 ²⁰	1.33 x 10 ⁶	132	245
B			11.3	6.1	1.25	128	234
C			10.0	5.4	1.11	126	219
D			8.6	4.6	0.94	121	201
E			8.0	4.3	0.88	118	192
Average			10.0	5.4	1.10	125	218
A-29-927	80 and 81	6.19	23.2	12.0 x 10 ²⁰	0.93 x 10 ⁶	127	193
A-29-928	80 and 81	6.22	21.3	11.0 x 10 ²⁰	0.86 x 10 ⁶	124	185
A-29-929	80 and 81	6.19	23.9	12.5 x 10 ²⁰	0.98 x 10 ⁶	130	198

TABLE VI (Contd.)

AARR FUEL PLATE IRRADIATION DATA -- BURNUP,
HEAT GENERATION, TEMPERATURE

Plate Number	Cycle	U-235 (g)	BU (percent)	Fissions cc of core	Heat Flux Btu hr-ft ²	Average Temperature Surface (°C)	Average Temperature Center line (°C)
A-29-936	81 and 82	6.19					
Top			15.3	8.0×10^{20}	0.62×10^6	115	160
Middle			19.2	10.1	0.78	122	177
Bottom			26.4	13.8	1.07	133	209
Average			20.1	10.5	0.82	123	180
A-29-938	81 and 82	6.15					
Top			14.9	7.8×10^{20}	0.60×10^6	114	156
Middle			21.6	11.3	0.87	125	186
Bottom			27.8	14.6	1.12	135	214
Average			21.8	11.5	0.88	115	177
A-29-939	81 and 82	5.76					
Top			15.4	7.7×10^{20}	0.58×10^6	114	155
Middle			20.0	10.1	0.77	121	175
Bottom			27.0	13.5	1.02	132	204
Average			21.1	10.5	0.80	122	178
AARR Braze Rib Subassembly							
A-15-932	80, 81, 82	6.76	32.2 ^[a]	19.3×10^{20}	$0.95^{[a]} \times 10^6$	170	240 ^[a]
A-15-934	80, 81, 82	6.76	32.2 ^[a]	19.32	$0.95^{[a]}$	170	240 ^[a]
A-16-954	80, 81, 82	5.85	35.2 ^[a]	19.0	$0.90^{[a]}$	165	230 ^[a]
[a] Estimated Values							

2. FUEL CORE GROWTH AND SWELLING

At these relatively low test temperatures, it is expected that swelling is produced primarily by the solid fission products. A list of fission yields and atomic volumes, Table VII[6], shows that during the fissioning process there is a ΔV of 3.04 if all fission products are considered in their solid elemental form. Of course these fission products may form alloys or compounds that when produced give a ΔV slightly different from the elemental form. Examining the swelling rates of three test cases, it can be seen what this difference might be. Dense UO_2 swells 1.7 percent per atom percent burnup; uranium has a value of 3 percent and uranium-aluminum alloy 0.09 percent. If these values are normalized by putting them in terms of fiss/cc as shown on the graph, Figure 13, it is found that the low-temperature swelling values for dense material[7, 8, 9], whether associated with the alloy (uranium metal) or the oxide, are in good agreement with the calculated swelling based on the formation of the solid elemental fission products.

Uranium dioxide is usually produced with some porosity, and early experiments with bulk UO_2 showed a low initial swelling rate (also shown in Figure 13). But, as these experiments were carried to higher burnup, it was found when the voidage was filled, the expected swelling rate of the solid UO_2 was resumed.

Likewise (as reported for other systems)[2], UAl alloy which was powdered to facilitate the distribution of boron as a burnable poison, and thereby retained a certain amount of porosity in the finished plate, exhibited this delayed swelling effect (also shown in Figure 13).

TABLE VII

RELATIVE VOLUMES OF U-235 FISSION PRODUCTS
60 DAYS AFTER IRRADIATION

Element	Fission Yields Per 100 U-235 Atoms	Atomic Volume (cc/g atom)	Relative Volumes
Se	0.399	16.48	6.576
Br	0.133	25.6	3.405
Kr	5.163	28.0	144.564
Rb	2.717	55.87	151.799
Sr	10.556	34.0	358.904
Y	5.182	19.89	103.070
Zr	31.860	14.06	447.952
Nb	1.343	10.8	14.504
Mo	21.859	9.39	205.256
Tc	6.100	8.6	52.460
Ru	11.689	8.29	96.902
Rh	2.509	8.27	20.749
Pd	1.282	8.88	11.384
Ag	0.028	10.28	0.288
Cd	0.059	13.00	0.767
In	0.011	15.71	0.173
Sn	0.074	16.26	1.203
Sb	0.046	18.4	0.846
Te	2.623	20.45	53.640
I	1.234	25.7	31.714
Xe	21.198	37.6	797.045
Cs	18.560	69.84	1296.230
Ba	5.872	39.00	229.008
La	6.005	22.44	134.752
Ce	17.278	17.03	294.244
Pr	5.398	20.82	112.386
Nd	17.835	20.61	367.579
Pm	2.376	21.0	49.896
Sm	2.406	20.07	48.288
Eu	0.176	28.98	5.100
Gd	0.025	20.01	0.500
Total relative volume of fission products			5041.184
U-235	100.0	12.48	1248.00

Fig. 13 The swelling of various fuels as induced by solid fission product formation—dense fuels versus fuels with voids.

So the calculated solid fission product swelling is used as a base line to indicate to what extent fission products are accommodated in the voids.

The core swelling observed for the AARR plates tested is shown in Table VII together with the calculated UO₂ fuel densities and average plate fuel burnup. The core swelling is obtained from plate density measurements before and after irradiation. Knowing the initial core weight and the density value of 8.00 g/cc for the 347 stainless steel cladding material, the pre- and post-irradiation core densities can be calculated. If it is assumed the 347 SS matrix material is fully dense, the UO₂ fuel particle densities in the as-fabricated plates can be calculated. This is desirable since the as-fabricated UO₂ fuel density can differ from the bulk UO₂ density prior to fabrication in that densification or breakup of the fuel particles during fabrication can occur. A density value of 8.00 g/cc was used for the 347 SS matrix and 6.71 g/cc for the coated ZrB₂ poison. The average as-fabricated UO₂ densities of the six composition A-15 Martin plates were 9.65 ± 0.04 g/cc as compared to 8.87 ± 0.03 g/cc for the five composition A-16 BMI plates. The as-fabricated UO₂ densities of the composition A-29 plates, with the exception of A-29-938 and A-29-939, all agree within ± 1.5 percent of the average value of 9.03 g/cc. Plate A-29-938 was overheated during fabrication and some fuel densification occurred, increasing the calculated UO₂ density to 9.42 g/cc. Plate A-29-939 contained 6 wt% glass beads. The difference between the Martin and BMI plates is indicative of the difference in fabrication methods.

The theoretical swelling rate for theoretically dense UO₂, considering only the U-235 fission products, is 6.38 percent for each 10^{21} fiss/cc. The observed core swelling in Table VIII is the core swelling caused by fission products plus the amount caused by the B¹⁰ neutron reaction. The composition A-15 samples exhibited a significantly higher swelling rate than the A-16 plates,

TABLE VIII

AARR UO₂-SS FUEL PLATE CORE SWELLING DATA

Plate Number	As-Fabricated UO ₂ Density	Theoretical UO ₂ Density (percent)	Initial Core Volume (cc)	Average U-235 Burnup (percent)	Average Fissions cc of core	Observed Core Swelling $\frac{\Delta V}{V} \times 100$	Measured Swelling $\frac{10^{20} \text{ fissions/cc}}$
A-15-923	9.61	88.7	2.499	6.2	3.7×10^{20}	2.13	0.58
A-15-925	9.65	89.1	2.434	14.7	8.7	3.16	0.36
A-15-926	9.66	89.2	2.444	12.7	7.6	3.35	0.44
A-15-932 ^[b]	9.67	89.3	2.448	32.2est	19.3	----	----
A-15-933	9.65	89.1	2.457	6.9	4.3	0.91	0.21
A-15-934 ^[b]	9.67	<u>89.3</u> 89.1	2.446	32.2est	19.3	----	<u>----</u> 0.40
A-16-943	8.88	82.0	2.363	18.6	10.1×10^{20}	2.45	0.24
A-16-945	8.84	81.6	2.362	21.1	11.3	4.00	0.35
A-16-947	8.86	81.8	2.364	9.5	5.1	-0.28	----
A-16-952	8.88	82.0	2.346	10.0	5.4	0.95	0.18
A-16-954 ^[b]	8.90	<u>82.2</u> 81.9	2.352	35.2est	19.0	----	<u>----</u> 0.26
A-29-927	8.87	81.9	2.584	23.2	12.0×10^{20}	3.27 ^[a]	0.27
A-29-928	9.04	83.5	2.577	21.3	11.0	2.92 ^[a]	0.27
A-29-929	8.99	83.0	2.571	23.9	12.5	3.40 ^[a]	0.27
A-29-936	9.12	84.2	2.562	20.1	10.5	3.25 ^[a]	<u>0.31</u> 0.28
A-29-938	9.42 (overheated)	87.0	2.534	21.8	11.5	5.17 ^[a]	(0.45)
A-29-939	8.68	80.1	2.502	21.1	10.5	2.90	(0.28)
AARR Brazed Rib Subassembly			7.246		19.1×10^{20}	6.07	0.32
A-15-932	9.67	89.3	2.448	32.2	19.3		
A-15-934	9.67	89.3	2.446	32.2	19.3		
A-16-954	8.90	<u>82.2</u> 86.9	2.352	35.2	19.0		

[a] Includes swelling due to B⁻¹⁰ reaction.

[b] These three plates comprise the AARR brazed rib subassembly.

as can be seen by comparing the swelling produced per 10²⁰ fiss/cc (Table VII). An average swelling per 10²⁰ fiss/cc of 0.40 was recorded for the A-15 plates as compared to 0.26 for the A-16 samples. This increased swelling is inversely proportional to the percent fuel voids.

Plate A-16-947 exhibited a negative swelling (-0.28 percent). Both plates A-16-947 and A-16-952 received the same burnup and should have exhibited equal swelling. It is felt that an error in the post-irradiation density measurements resulted in the negative swelling rate; hence, this data point was discarded.

The A-29 plates contained coated ZrB₂ as a burnable poison, and part of the swelling was due to the presence of helium and lithium, formed from the B¹⁰ (α) Li⁷. The effect of boron on the swelling behavior of the fuel plates can be calculated. For fuel burnups in excess of ≈ 15 percent, essentially all of the B¹⁰ is converted to He⁴ + Li⁷. Table IX shows the swelling due to the formation of helium and lithium on an atomic volume basis. Since the poison compounds did not provide porosity for the accommodation of the neutron reaction products, the helium and lithium are considered to be 100 percent effective in promoting swelling. On an atomic volume basis, Li⁷ + He⁴ occupy 4.81 times the volume of a B¹⁰ atom. Based on a density of 2.13 g/cc for B¹⁰, 1 mg of B¹⁰ will occupy 0.000469 cc. Twenty-six mg of natural boron, containing 5.14 mg B¹⁰, result in a net volume increase of 0.00918 cc. In plates A-29-927, -928, and -929, which

TABLE IX

SWELLING OF COMPOSITION A-29 CORRECTED
FOR THE B-10-NEUTRON REACTION

Plate Number	Natural B (mg)	Measured Core ΔV (cc)	Swelling Due to $Li^7 + He^4$ Formation (cc)	Core Swelling due to $Li^7 + He^4$ Formation (percent)	Core Swelling Due to U-235 Fission Products Alone (percent)	Measured Fission Product Swelling per 10^{20} Fissions per cc (percent)
A-29-927	26.0	0.085	0.0092	0.34	2.93	0.24
A-29-928	26.0	0.076	0.0092	0.33	2.59	0.24
A-29-929	26.0	0.088	0.0092	0.34	3.06	0.24
A-29-936	41.0	0.084	0.0145	0.54	2.71	0.26
A-29-939	1.3	0.073	0.005	0.01	2.89	<u>0.28</u>
						Average 0.26
A-29-938 (overheated)	39.7	0.132	0.0140	0.54	4.63	0.40

contain 26 mg natural boron, 19.78 percent of which is B^{10} , ≈ 0.35 percent core swelling is due to the B^{10} reaction. In plate A-29-936 which contained 41 mg natural boron as ZrB_2 , ≈ 0.55 percent core swelling is attributed to helium and lithium formation.

After subtracting the effect of helium formation from the A-29 plate swelling, a comparison between the swelling behavior of the A-16 and A-29 BMI fabricated plates could be made. Neglecting plate A-29-938, which was overheated during fabrication, the average fission product swelling 0.26 percent per 10^{20} fissions per cc are the same for both A-16 and A-29 composition fuel plates.

Plate A-29-938 was overheated during fabrication, resulting in densification of the fuel. This increased the UO_2 density from 9.03 to 9.42 g/cc, which is approaching the fuel density of the A-15 plates (9.65 g/cc). It was expected and observed that plate A-29-938 would behave more like an A-15 plate. Plate A-29-938 exhibited a 5.17 percent core swelling, 70 percent greater than the other A-29 plates with equivalent burnup. Subtracting the effect of B-10 yields 4.63 percent swelling due to fission products. On a swelling per 10^{20} fiss/cc basis, this gives a value of 0.40 in agreement with the average for the A-15 plates. Thus, A-29-938 with its lower void content is more typical in behavior to the A-15 composition plates than to the A-29 plates.

Plate A-29-939 contained 1.3 mg natural boron in the form of boro-silicate glass beads. The small amount of boron present did not allow for an evaluation of how well the glass beads retained the helium gas. Swelling was near the average of the A-29 plates.

The three-plate AARR brazed-rib subassembly which was irradiated for three cycles in tier 6 received a higher burnup than the other plates. An average core swelling value of 6.1 percent was obtained for the subassembly. Since the subassembly did not contain boron, swelling was due to fission products alone. The measured core swelling amounts to 50 percent of that for theoretically dense UO_2 . The AARR irradiation test program was terminated before isotopic burnup analysis could be taken on the subassembly plates. Consequently, the burnup had

to be calculated from ETRC flux measurement data. The reduction in hafnium shrouding around part of tier 5, between cycles, complicated the situation. Thus, there is some doubt concerning the accuracy of the calculated burnup values obtained for the AARR subassembly. Also, the two Martin (A-15) plates in the subassembly contained a higher fuel particle density and lower void fraction than the other plate, fabricated by BMI (A-16). Thus the swelling rate of the subassembly would be a weighted average of the two compositions tested. In addition, there was a sharp burnup profile along the subassembly plates resulting in nonuniform swelling. For these reasons the swelling data for the AARR subassembly are not included in Figure 14, discussed below.

The average fission product core swelling versus average fiss/cc of core for the BMI and Martin plates is shown in Figure 14. The calculated swelling base line for fully dense material, 6.38 percent for each 10^{21} fiss/cc, is shown as a dotted line. At lower burnups, there are fuel particle voids available to accommodate the fission products, and a lower swelling rate results. Once all the voids are filled in, the swelling curve is expected to become parallel to that for theoretically dense UO_2 . From Figure 14, it can be seen that there is a considerable difference in swelling between the Martin (A-15) and BMI (A-16, A-29) fabricated samples. The A-15 plates which contain 3.8 percent core voids show an average swelling rate of 60 percent of theoretical compared to 40 percent for the A-16, A-29 BMI plates which contained 6.2 and 5.8 percent core voids.

Fig. 14 Effect of low-density UO_2 on core swelling.

The fuel assembly which received the highest burnup, 19×10^{20} fission/cc (estimated), showed a swelling of six percent. This falls in the area predicted by the dotted line extensions in the plot (also shown in Figure 14).

The effect of fuel particle voids on 37 to 39 wt% UO_2 can be summed up by comparing the core swelling at 10^{21} fission/cc as in Table X.

TABLE X
EFFECT OF VOID ON SWELLING

	Fuel Particle Voids (percent)	Voids in Core (percent)	Core Swelling (percent)
Theoretically dense UO_2	0	0	6.4
Comp 15 (Martin)	10.9	3.8	4.0
Average Comp, 16, 29 (BMI)	17.3	6.0	2.6

Heat transfer and coolant flow considerations allow for a maximum nominal 2 mil decrease in channel width. This would be six percent swelling in the fuel. At that level of swelling the fuel life would be doubled by the incorporation of six percent core voids.

3. BLISTER RESISTANCE

Blistering of irradiated fuel plates is due to the accumulation of gaseous fission product, mainly Xe and Kr, in the fuel particle voids. As the gases accumulate, the internal pressure of the gas inside the voids increases. When the internal gas pressure exceeds the strength of the material, the void expands. This expansion relieves the pressure in the void. As more gas collects in the voids, the internal pressure again exceeds that of the strength of the matrix, and creep or yielding again occurs. As the voids expand, particle cracking, matrix cracking, and interconnecting may occur and liberate the gas from small bubbles and surface tension control. In fuel plates containing boron, helium gas is produced by the $\text{B}^{10} + n \rightarrow \text{Li}^7 + \text{He}^4$ reaction. Helium may diffuse through the matrix, collecting in the voids and increasing the internal pressure in the void. The analysis of the gas collected from an irradiation-produced blister was determined in a previous test on a 42 wt% UO_2 aluminum plus 0.19 wt% B_4C (8.1 mg natural B) plate [10]. The gas was 41 percent helium.

Even though the gas pressure forces were restrained in the AARR samples during irradiation, it is known that at some elevated temperature this equilibrium will break down. Blistering of selected samples was intentionally induced by post-irradiation incremental heat treatments to indicate the maximum operating temperature at various fission densities. Blister testing in this manner has been used previously on aluminum dispersion systems and found to correlate with in-pile blister resistance.

The plates selected for blister testing were sectioned half-longitudinally. One half of the plate was used for blistering and the other half for other purposes. The blister plates were placed in a furnace at $400 \pm 10^\circ\text{C}$ (750°F) for 30 minutes, after which time the plates were removed for visual inspection and density determinations. The temperature of the furnace was increased in 28°C (50°F) increments up to 677°C (1250°F), at which time all but one of the plates (A-29-939) had blistered. The blister data for the above plates are recorded in Table XI.

TABLE XI
AARR PLATE BLISTER TEMPERATURES

<u>Plate No.</u>	<u>Boron (mg)</u>	<u>No. of Blisters</u>	<u>Blister Interval $^\circ\text{C}$</u>	<u>Fiss/cc 10^{20}</u>
A-15-926	--	10	570-630	8.3-10.8 6 @ 10.8
A-16-943	--	3	450-510	10.7 11.0 11.8
A-29-927	26	1	425-455	11.9
A-29-928	26	1	425-455	11.3
A-29-929	26	2	425-455	12.6 14.0
A-29-936	41	2	400-425	13.2 13.8
A-29-938 (overheated)	39.7	1	Below 400	14.6
A-29-939	1.3	Rapid Swelling	540-680	13.4

Plate A-15-926, which did not contain boron, showed ten blisters forming between 570 and 630°C , at points of burnup ranging from 8.3 to 10.8×10^{20} fiss/cc, six blisters forming at 10.8×10^{20} fiss/cc. Blisters, typical of those produced, are shown in Figure 15. Plate A-16-943, which also did not contain boron, exhibited three blisters forming between 450 and 510°C at points of burnup of 10.7, 11.0, and 11.8×10^{20} fiss/cc.

Plates A-29-927 and -928 exhibited one blister each, whereas A-29-929 blistered twice. All blisters occurred at 455°C . After blistering occurred, these three plates were blister tested at 480°C (900°F) and 510°C (950°F) to determine if further blistering would occur. No new blisters were formed. Of the four blisters, three occurred near but not at the core-picture-frame interface. Blistering occurred at or near points of maximum burnup, which for these plates is from 11 to 14×10^{20} fiss/cc. Plates A-29-927 through -929 all contained 26

SIDE 1

SIDE 2

Fig. 15 Plate A-15-926 after irradiation and heat treating to 630°C. The arrows indicate typical plate blistering.

mg of natural boron, which would produce 14.0 ml of helium at standard temperature and pressure (STP). Plate A-29-936, containing 41 mg boron, blistered twice at 425°C (800°F) at points of maximum burnup (13.2 and 13.8×10^{20} fissions/cc).

Plate A-29-938 was overheated during fabrication which led to densification of the UO_2 fuel, reducing the number of voids available for fission product accommodation. This plate blistered at 400°C (750°F) at a burnup of 14.6×10^{20} fissions/cc. It contained 39.7 mg natural boron which would yield 17.6 ml of helium at STP. Plate A-29-939 did not exhibit any visual blistering even at 677°C, at which time blister anneal testing was terminated. However, from the plate density determinations taken after each temperature increment, at 538°C a sudden decrease in density was noted, which accelerated with succeeding temperature increments indicating that rapid or breakaway swelling had occurred. Between 400 and 510°C there was a total density decrease of 0.4 percent. At 538°C (1000°F) the density decreased to 1.6 percent and decreased to 5.3 percent at 677°C (1250°F). Plate A-29-939 contained only a minor amount of boron (1.3 mg) in the form of boro-silicate glass beads. Since this was the only plate containing glass beads and only a minor amount of helium was produced (0.6 ml at STP), the effect of the glass beads in retaining helium gas could not be evaluated.

A plot of the blistering temperature as a function of radiation exposure, Figure 16, compares the blister resistance of the low-density fuels to previously tested fuels reported by D. L. Keller [11]. Keller established two curves which mark off three areas, a no-blister area, a some-blister area, and an all-blister area.

Fig. 16 Blister resistance of low-density 37 wt% UO_2 stainless steel fuel compared to previously tested 18 to 30 wt% fuel.

With the exception of plates A-29-927 and A-29-928 and plate A-29-939, all the plates blistered in the some-blister area of the stability curves. A blister temperature shift to the left (the difference between the solid points of A-16-943 and the open points A-29-927 and A-29-928) is owed to a 25 mg addition of boron as ZrB_2 . Even though plate A-29-939 did not blister, rapid swelling began at 540°C and continued until the termination of the heat treatments at 680°C . This is shown on the graph by two points connected with an arrow.

It would appear that the blister resistance of these high wt% (37 wt%) low-density fuels with improved dispersion is equal to the low wt% (18 to 30 percent) dense fuels previously tested. It is also indicated that if the fabrication voidage is present in larger amounts, as in plate A-29-939, that blister resistance is enhanced further. This is discussed in detail in the next section on metallographic observations.

4. RELATION OF GROWTH, SWELLING, AND BLISTER RESISTANCE TO MICROSTRUCTURES

The mechanical and physical changes which take place in fuel materials during fabrication and irradiation generally result in corresponding changes in the microconstituents. This section records those changes which are considered pertinent to the stability of this fuel.

As discussed in other sections of this report, the Martin fabrication process attained the goal of low amounts of stringering and low amounts of particle fracturing. The BMI process resulted in a larger degree of particle fracturing (increasing the void content) and some stringering. A comparison of the two end products representative of compositions A-15 and A-16 can be seen in Figure 17. Based on density data and verified by these photomicrographs, the in situ particle void space in the BMI fuel particle is about 1.5 times that of the Martin.

Figure 18 is a series of photomicrographs which show the changes in microstructure as a function of fissions per cc of core. Figures 18A through 18H show the BMI composition A-16 plates, with increasing irradiation exposure, and are arranged for comparison with the Martin composition, A-15 plates, also with an exposure range, in Figures 18AA through 18GG. Cracking of the fuel is evident in both 18A (A-16) and 18AA (A-15) in the as-fabricated condition but is greater in the A-16. At very low radiation exposure, 2.9×10^{20} fiss/cc, the small pores and fine cracks show evidence of being filled with growth material (Figure 18BB). The large cracks and voids still remain vacant at this time. At 3.4×10^{20} fiss/cc (Figure 18CC), essentially all the fracture cracks have been filled in with the growth material, and very little change occurs from this exposure level through the 6.8×10^{20} fiss/cc exposure (Figures 18E and 18EE).

At 9 to 10×10^{20} fiss/cc (Figures 18F and 18FF), fission gas porosity is initiated. The large fill-in areas have fewer but larger gas bubbles than the remaining particle. The gas bubbles grow rapidly in size from 9 to 12×10^{20} fiss/cc as shown in Figures 18G and 18GG. Observations of Figure 18 show that the void areas incorporated in the fuel particles during fabrication are being utilized for fill-in of solid growth material (accommodation of fission products)

64 4 2380A

50X

VILELLA'S ETCH

Martin Process, as-fabricated, A-15 composition average of 3.8 percent core voids.

64 4 2380B

50X

VILELLA'S ETCH

Battelle Process, as-fabricated, A-16 comparison average of 6.4 percent core voids.

Fig. 17 Photomicrographs of fuel plates made by the two different processes.

and account for the reductions in swelling rate as a function of void content. Fission gas agglomeration is not significant before 9×10^{20} fission/cc and comes after fabrication voids have been filled with solid growth material. This shows that the initial voidage does not act as sinks for the collection of detrimental amounts of fission gas.

At 12×10^{20} fission/cc, a considerable number of the gas bubbles are 2 microns and a few are 4 microns and above. The relation between pressure and volume can be determined by the Beattie-Bridgeman equation of state since the constants for Kr and Xe are known^[12]. If the gas volume is expressed in terms of atomic volumes, it is possible to tell at what pressure the atomic volume will be approached. See Table XII.

The pressure that exists in a 4μ bubble, in equilibrium with the external pressure, the compressive strength of UO_2 , and the surface tension, is ≈ 4000 atmospheres. Based on these considerations, the kinetic volume approaches the atomic volume, suggesting very little kinetic effect. However, the measured bubble volume at 12×10^{20} fission/cc is 2.5 times the combined atomic volumes of the Kr and Xe produced. The difference between the calculated and the measured is owed to the apparent plastic behavior (which is effectively a loss of compression strength) of UO_2 during irradiation.

On a volume basis, there is 15 percent gas bubbles in a given fuel particle or 5 percent gas bubbles in the fuel plate core at 12×10^{20} fission/cc. From fission yield considerations, the sum of the atomic volumes of Kr and Xe produced amounts to ≈ 2 percent of the core. If these atoms are considered to be in the bubbles, this would leave ≈ 3 percent voids (remainder of the bubble volume) due to the kinetic effect of the gases. Also, at this time ≈ 5.5 percent growth would be expected from the formation of other fission products. A summation of these growth units and their relation to core voids and core volume increase is shown in Table XIII. The 10.5 growth units are partially accommodated by fabrication voids and partially appear as fuel core growth. A balance discrepancy of 1.5 units amounts to ≈ 15 percent of the total 10.5 growth units. On this basis, the kinetic effect accounts for only ≈ 30 percent of the volume increase.

The fill-in zones which appeared as a function of irradiation in Figure 18 have different properties than the remainder of the fuel particle even though microprobe analysis could detect little difference in composition. (The "second phase" regions are about 5 percent lower in uranium than the surrounding UO_2 . Determinations of fission products are less definite, but higher Zr and lower Mo concentrations are indicated.) Figure 19 shows a difference in etch characteristics. The fill-in portion is not affected by the etchant, but the remainder of the particle undergoes a rapid attack. The fill-in zones also have lower hardness than the remainder of the particle as shown in Figure 20 and Table XIV. The data also show that the final hardness of the stainless steel matrix material is consistent with values found for radiation damage^[13]. Some reduction in hardness was detected in the anneal samples. A longer exposure of the particle in Figure 19 reveals that a recoil zone does exist around the fuel particle in the matrix as shown in Figure 21. This damage zone is very narrow, 0.00025 cm (0.0001 inch), at these irradiation conditions and need not be considered a factor of matrix deterioration.

The blisters produced during the post-irradiation incremental heat treatments were also examined by metallography. Typically, fracturing occurred at the

edge of the core but was not a core-clad separation (Figure 22). This blister zone would have higher burnup than the center of the core because of neutron self-shielding effects. The cracking in the UO_2 appears to be selective and generally avoids the softer areas which were filled in. Further evidence of greater ductility in the fill-in zone is seen in the particles at the edge of the blister. The gas bubbles in the fill-in material expanded and deformed in the presence of the blister gap, rather than causing fracturing as in the balance of the particle. An example of this is indicated by the arrow in Figure 22. An enlarged view of this particle is shown in Figure 23. It is evident that brittle cracking occurs first in the UO_2 as gas pressure increases with increased temperature and then extends into the matrix and finally interconnects with only localized deformation of the stainless steel. Density determinations after incremental heat treatments reflect the typical low-deformation cracking (see Figure 24). One plate, A-29-939, was an exception. This plate, which contained the poison in the form of glass beads, had the lowest in situ fuel particle density (80 percent) of theoretical, compared to an average of 82 percent for composition A-29 (see Figure 25). These larger void areas were filled during irradiation with the more ductile growth material, which apparently offered greater resistance to brittle fracturing of the UO_2 particle. Consequently, blistering never did occur during the incremental post-irradiation heat treatments. The heat treatment of this sample was terminated at 680°C , or 200°C higher than the blistering temperature of the other A-29 samples.

Some of the fuel plates contained between 0.025 and 0.040 gram of natural boron in the form of ZrB_2 . As discussed in detail in other portions of this report, the ZrB_2 had the effect of lowering the blister temperature slightly. Although the poison for all practical purposes was totally burned up in these samples, blistering was associated with the fuel particles rather than the poison particles. There was evidence that a slight amount of helium agglomerated in the ZrB_2 particle

67 4 2375

550X

VILELLA'S ETCH

Fig. 18A Microstructure of plate fabricated by BMI process, composition A-16, before irradiation. Particle cracking is greater than that in 18AA.

500X

VILELLA'S ETCH

Fig. 18AA Microstructure of plate fabricated by Martin process, composition A-15 before irradiation.

3991

500X

UNETCHED

Fig. 18BB Microstructure of A-15 plate irradiated to 2.9×10^{20} fiss/cc. The small cracks and fine pores have been filled in with growth material.

(Figure 26) but not a sufficient amount to initiate matrix cracking. This would seem to indicate that early blistering was brought about either by diffusion of helium into voids initiated at the fuel particles or by effects that ZrB_2 had on the matrix.

One of the plates, A-29-938, was fabricated (hot rolled) at an excessively high temperature (1300 to 1400°C). This high temperature caused melting at the ZrB_2 stainless steel interface and at the grain boundaries of the stainless steel. The increased fluidity caused intrusion of the matrix into the UO_2 fuel particles. This can be seen in Figure 27. Since part of the fabrication cracks and voids were filled in, they were not available for the accommodation of fission products, and a corresponding increase in swelling was detected. It also should be noted that even though ZrB_2 is volatile at elevated temperatures, poison particles were retained in plate A-29-938.

Fig. 18D Microstructure of A-16 plate irradiated to 4.3×10^{20} fiss/cc. Total fill-in area is larger than in composition A-15.

3988

Fig. 18CC Microstructure of A-15 plate irradiated to 3.4×10^{20} fissions/cc. All fabrication cracks appear filled with growth material.

3986

Fig. 18DD Microstructure of A-15 plate irradiated to 4.2×10^{20} fissions/cc.

Fig. 18E Microstructure of A-16 plate irradiated to 6.1×10^{20} fissions/cc.

Fig. 18F Microstructure of A-16 plate irradiated to 9.3×10^{20} fissions/cc. Fission gas agglomeration is initiated.

Fig. 18EE Microstructure of A-15 plate irradiated to 6.8×10^{20} fissions/cc. Very little change is evident when compared to 18DD and 18CC.

Fig. 18FF Microstructure of A-15 plate irradiated to 9.9×10^{20} fissions/cc. Fission gas porosity is initiated.

4039

Fig. 18G Microstructure of A-16 plate irradiated to 11.8×10^{20} fiss/cc.

3975

Fig. 18GG Microstructure of A-15 plate irradiated to 11.8×10^{20} fiss/cc. Gas bubbles have developed which are larger than those in 18FF.

TABLE XII

THE RELATION BETWEEN PRESSURE AND VOLUME
FOR Xe AND Kr AND FOR GAS IN A 4 MICRON BUBBLE

$T = 250^{\circ}\text{C}$

<u>p Xe</u> <u>(Atm.)</u>	<u>p Kr</u> <u>(Atm.)</u>	<u>4μ Bubble</u> <u>(Atm.)</u>	<u>Kinetic Volume/</u> <u>Atomic Volume</u>
100	150	---	10
200	310	---	5
640	1080	---	2
1100	1790	---	1.5
		4100	<1.5

TABLE XIII

A BALANCE OF THE GROWTH UNITS EXPRESSED AS VOLUME % OF THE CORE,
ASSUMING Kr AND Xe ATOMS ARE IN THE BUBBLES, AT 12×10^{20} fiss/cc

<u>Battelle Process</u>		<u>Martin Process</u>	
5.5	solids	5.5	solids
2	Kr + Xe	2	Kr + Xe
3	Kinetic effect	3	Kinetic effect
<u>10.5%</u>	growth units	<u>10.5%</u>	growth units
- 6	voids (Fabrication)	- 4	voids (Fabrication)
<u>4.5%</u>	cal. growth	<u>6.5%</u>	cal growth
- 3 %	observed growth	- 5 %	observed growth
1.5%	discrepancy	1.5%	discrepancy

500X

OXALIC ACID
ELECTROLYTIC ETCH

Fig. 19 Etched fuel particle after irradiation to 11.3×10^{20} fiss/cc of core. Fill in zone is indicated by the arrow.

500X

Fig. 20 Hardness indentation in the matrix, fill-in zone and body of the particle. The relative size of the indentation indicates relative hardness. All indentations were made with the same load and represent hardnesses of DPN 240 (matrix), DPN 430 (fill in zone), and DPN 650 (body of particle).

TABLE XIV

HARDNESS IN THE UO_2 PARTICLES AND FILL-IN ZONES
AS A FUNCTION OF RADIATION AND ANNEAL TEMPERATURE

Plate No./ Mount No.	Fiss/cc of Core	DPN Hardness				
		Fuel Particle			Matrix	
		Pre- <u>irradiated</u>	Post- <u>irradiated</u>	Fill-in <u>Zone</u>	Pre- <u>Pre-</u>	Post- <u>Post-</u>
<u>A-15-923</u>						
T-294	3.03	516	1100		141	265
T-295	2.86		1127			254
T-296	3.45		1052			249
T-297	4.22		1031			266
T-298	4.82					
T-299	4.95		1126			259
T-300	4.52		1102			263
T-336	3.15		1019	748		251
T-337	3.15	516	976	835	141	252
<u>A-15-925</u>						
T-301	6.73	516	1052		141	262
T-302	6.85		1058	961		275
T-303	8.40		1018	961		267
T-304	9.89		961	645		281
T-305	11.80		992	623		283
T-306	6.79		961	633		272
T-307	10.84		1074	585		281
T-338	7.63		992	804		266
T-339	7.63	516	954	817	141	266
<u>A-15-933</u>						
T-308	3.50	516	1068	594	141	254
T-309	3.25		1093	609		250
T-310	4.06		1074	633		265
T-311	4.88		954	868		256
T-312	5.88		954	880		263
T-313	4.37		1052	626		265
T-314	5.38		1051	655		261
T-340	3.66		1057	865		250
T-341	3.66	516	1033	941	141	243

TABLE XIV (Contd.)

HARDNESS IN THE UO_2 PARTICLES AND FILL-IN ZONES
AS A FUNCTION OF RADIATION AND ANNEAL TEMPERATURE

Plate No./ Mount No.	Fiss/cc of Core	DPN Hardness				
		Fuel Particle			Matrix	
		Pre- irradiated	Post- irradiated	Fill-in Zone	Pre-	Post-
<u>A-16-945</u>						
T-315	13.23	460	940	777	167	300
T-316	12.26		932	697		333
T-317	11.83		939	514		290
T-318	9.90		961	800		290
T-319	9.31		977	811		290
T-320	12.77		976	788		290
T-321	9.60		933	805		279
T-342	12.0		976	788		301
T-343	12.0	460	954	728	167	288
<u>A-16-947</u>						
T-322	5.95	460	954	853	167	288
T-323	5.57		969	823		285
T-324	5.31		973	888		288
T-325	4.39		1015	853		282
T-326	4.29		969	919		282
T-327	5.76		1554	818		282
T-328	4.34		992	885		288
T-344	5.44		922	876		275
T-346	5.44	460	961	843	167	280
<u>A-16-952</u>						
T-329	6.84	460	1024	842	167	281
T-330	6.06		947	737		287
T-331	5.36		1028	788		290
T-332	4.61		933	592		290
T-333	4.29		957	788		272
T-334	6.27		956	825		283
T-335	4.45		1033	836		284
T-345	5.71		1007	812		265
T-347	5.71	460	954	847	167	276

TABLE XIV (Contd.)

HARDNESS IN THE UO_2 PARTICLES AND FILL-IN ZONES
AS A FUNCTION OF RADIATION AND ANNEAL TEMPERATURE

Plate No./ Mount No.	Fiss/cc of Core	DPN Hardness					Maximum Anneal Temperature (if any)
		Fuel Particle			Matrix		
		Pre- irradiated	Post- irradiated	Fill-in Zone	Pre-	Post-	
<u>A-29-927</u>							
T-583	11.9	468	625	582	173	201	510°C
		468	697	501	173	243	
T-587			665	540		240	
<u>A-29-928</u>							
T-584	11.3	468	595	474	173	184	510°C
T-588		468	686 659	453 513	173	217 221	
<u>A-29-929</u>							
T-585	12.6	468	634	488	173	190	510°C
		468	291	575	173	216	
T-586		468	609	408	173	246	
T-589			691	460		230	
<u>A-29-936</u>							
T-601	13.2	468	607	415	173	222	425°C
			661	500		240	
<u>A-29-938</u>							
T-600	14.6	468	530	500	173	228	400°C
		468	597	576		231	
T-602			597			251	
<u>A-29-939</u>							
T-603	13.4	468	669	495	173	224	680°C
			593	429		198	

66 12 1667

OXALIC ACID
ELECTROLYTIC ETCH

500X
LONG EXPOSURE

Fig. 21 Recoil zone around fuel particle (arrow) revealed by long exposure of etched sample. This fuel was irradiated to 11.3×10^{20} fission/cc of core.

100X

Fig. 22 UO_2 stainless steel fuel plate after irradiation to 11.8×10^{20} fissions/cc and post-irradiation heat treating to 510°C . The arrow indicates the voids in a fill-in zone which have expanded and deformed in the presence of the blister gap.

Fig. 23 Enlarged view of UO_2 fuel particles at the edge of a blister. Note that the holes in the diffused zone are enlarged and are deformed in the presence of the blister gap.

Fig. 24 Density determinations after incremental heat treatment.

Microstructure of an unirradiated composition A-29 (939 type) plate. Note the large void volume in the fuel particles.

Microstructure of plate A-29-939 after irradiation to 13.4×10^{20} fissions/cc and heat treated to 680°C . Note the large void areas have been filled in, and brittle cracking did not occur.

Fig. 25 Before and after irradiation of A-29-939 type plates.

Fig. 26 UO_2 stainless steel containing ZrB_2 as a burnable poison. This fuel plate was irradiated to 11.3×10^{20} fiss/cc and heated to blister temperature at 455°C . Only a slight amount of helium agglomerated in the poison particle (ARROW).

Fig. 27 Microstructure of plate A-29-938 after irradiation. Part of the voids and cracks was filled in with matrix during fabrication.

VI. CONCLUSIONS

The incorporation of porosity in a nearly idealized dispersion of UO_2 in stainless steel resulted in improved fission product accommodation and a reduction in fuel growth rate. Some fragmentation of the fuel particles, giving a less than "ideal" dispersion and producing voidage, results in even greater fission product accommodation. However, the authors believe that some idealization is obviously beneficial. The control of stringering and particle fracturing has served to indicate the relative importance of voidage.

Idealization should have its greatest benefit in suppressing compressible-type swelling as would be caused by the gases or other volatilized fission products. Since these gases appear to be contained in small bubbles at high pressures, their kinetic contribution to fuel volume increase is not the largest effect at low temperatures.

Growth from the fission products, considered as solids, does not have compressibility and cannot be contained by idealization alone. It is here that some voidage is particularly beneficial. To accommodate the solid fission product volumes when fuel voids are present, the matrix needs sufficient strength to resist external growth and promote internal growth. Since the UO_2 exhibits an apparent plasticity, even at low temperatures during irradiation, the pressure produced in both structures was sufficient for internal growth.

The fuel has demonstrated its capabilities to perform according to the Mark I requirements, if attention is given to total growth and maximum temperature. The test fuels indicated that the more idealized structure could be irradiated to 15×10^{20} fission/cc before the limiting 2 mil decrease in channel is reached and the fragmented structure could be irradiated to 19×10^{20} fission/cc. The blister resistance of both of these 37 wt% stainless steel fuels was found to be similar to that of 18 to 30 wt% fuels previously tested.

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