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PROGRESS ON THE USE OF GAS-PRESSURE BONDING FOR FABRICATING LOW-COST CERAMIC, CERMET, AND DISPERSION FUELS

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Final Report on AEC Fuel-Cycle Program

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Development of techniques for the fabrication of Type 304 stainless steel-clad uranium dioxide fuel elements by the gas-pressure-bonding process was continued in the final phase of a 2-1/2-year study conducted under the AEC Fuel-Cycle Development Program and discussed earlier in BMI-1372 and BMI-1475. In this phase more definite studies of the process operations directed toward establishing preliminary process specifications were undertaken. Both rod and compartmented flat-plate fuel geometries were considered. Individual operations were optimized with respect to their effects on the characteristics of the final bonded oxide. Prior to completion, these investigations were terminated in favor of fabrication of fuel rods for irradiation in the VBWR. The work during this phase demonstrated that fuel rods can be fabricated by pressure bonding using cold-pressed oxide fuel. Good dimensional control was achieved, and, in the case of the VBWR rods, the general specifications for a fuel rod designed for pelletized fuel were met by the pressure-bonding route. Compartmented flat-plate assemblies as processed by gas-pressure bonding also demonstrated good dimensional control.

INTRODUCTION

The use of the gas-pressure-bonding technique in producing low-cost quality fuel elements has been investigated at Battelle in a three-phase 2-1/2-year study in support of the Fuel-Cycle Development Program of the U. S. Atomic Energy Commission. In the first two phases of this program, conditions were established for achieving both fuel densification and cladding bonding in a single pressure-bonding cycle.[1,2] This effort concerned uranium dioxide ceramics, cermet, and dispersion fuels as clad with Type 304 stainless steel with the major emphasis placed on the ceramic fuel systems. As a result of these studies, mixtures of ceramic and fused uranium dioxides were selected for further consideration since they possessed the desired characteristics in both the initial compacting and pressure-bonding operations.

The present report reviews the progress achieved in Phase III of the Battelle program. In this portion of the investigation, compaction and pressure-bonding studies were continued with the objective of establishing preliminary process specifications for fuel-rod and compartmented flat-plate assemblies. These particular configurations were selected from those considered in previous studies as the most promising elements for further development. In view of the attractiveness of the mixtures of fused and ceramic grade oxides, such fuels were considered in both fuel-element applications. Prior to completion of the Phase III program, the definition of preliminary process specifications was suspended in favor of fabricating experimental fuel rods for irradiation in the VBWR.

(1) References at end.

MATERIALS

In previous studies, the compacting and pressure-bonding behaviors of various commercially available uranium dioxides were established.[1,2] Ultimately, this work led to the investigation of oxide mixtures and the selection of ceramic-fused mixtures as the most promising fuel material for pressure bonding fuel rods. In such mixtures the ceramic oxide imparts the required activity for achieving high bonded densities, whereas the fused oxide imparts a high density in the initial compacting operation, thereby minimizing cladding deformation during the pressure-bonding cycle. The mixtures which appear most desirable consist of those with from 60 to 70 w/o fused oxide. The fused oxide utilized in this work consisted of a minus 20-mesh product with an oxygen/uranium ratio of 2.005. The ceramic oxide component in the mixtures consisted of a minus 325-mesh material with an oxygen/uranium ratio of 2.08 and a 0.6-μ average crystal size.

In the further development of compartmented flat plates, both sintered oxide cores and cold-pressed oxide-mixture cores were considered. The sintered cores were provided as depleted material at approximately 95 per cent of theoretical, whereas the oxide mixtures, which were comprised of depleted fused and normal ceramic grades, were pressed to approximately 70 per cent of theoretical density. With the pressed cores, it was necessary to also use partially sintered Type 304L stainless steel powder spacers to achieve uniform deformation in the assemblies. This stainless steel was supplied as a minus 200-mesh powder by the Vanadium Alloys Steel Company. The cladding in all cases consisted of Type 304 stainless steel.

In fabricating fuel rods for irradiation in the VBWR, it was necessary to consider additional ceramic oxides. Both a GE grade and a superactive grade were evaluated as these where considered the most reproducible types for processing as partially enriched fuel by MCW. These oxides exhibited oxygen/uranium ratios of 2.11 and 2.08 respectively, with a characteristic fluffy appearance. The fused oxide used for this rod fabrication was provided in the same size distribution as that employed in previous portions of the program. The oxygen/uranium ratio was 2.005. Type 304L stainless steel tubing was selected for this fabrication in order to minimize sensitization during the slow cooling characteristic of the pressure-bonding cycle.

GENERAL FUEL-ELEMENT DEVELOPMENT

The development effort in the present phase of this investigation concerned the completion of previously initiated tests and a more detailed study of the operations in the general process. Studies therefore involved both the powder-compaction procedures and the pressure-bonding operation. Fuel geometries of particular interest in these investigations were the rod and flat-plate elements.

Compaction Studies

In this phase of the program, investigations of the compacting behavior of Type 304 stainless steel powder and mixtures of uranium dioxide were continued. With

both studies, efforts were directed to refining powder-metallurgy techniques for the fabrication of fuel rod and compartmented-plate configurations. In the latter case, the assembly and densification of green powder components was considered. More detailed comparisons of binder materials and density-measurement techniques were also included in this phase of development.

Stainless Steel Powders

The approach considered in the preparation of stainless steel components was to provide partially sintered flat spacer stock from which the required components could be machined. In this respect, the compaction and sintering of Type 304 stainless steel in the form of flat plates measuring 0.150 by 2.0 by 10.0 in. were briefly investigated. All powders were initially compacted at 27 tsi to yield a green density of approximately 70 per cent of theoretical. Subsequent sintering at 2000 and 2200 F produced densities of 72 and 74 per cent of theoretical, and additional densification to 84 per cent of theoretical was attainable through cold rolling. The results of these tests are summarized in Table 1. With the range of densities demonstrated, it was possible to provide stainless steel spacer stock consistent with the density of various cold-pressed fused-ceramic uranium dioxide mixtures. Through the matching of initial densities, control of relative deformation on pressure bonding can be exerted.

Uranium Dioxide

The effectiveness of various binders in compacting 60 w/o fused-40 w/o ceramic uranium dioxide mixtures was briefly studied to select an optimum binder for such mixtures. The blended powders and binder in each case were compacted at 50 tsi and the compacted density and handling characteristics noted. The binders were further related to the final stoichiometry of the oxide mixtures after pressure bonding for 3 hr at 2100 F and 10,000 psi. The data for these tests are summarized in Table 2 where small differences in compacted density are noted. Of the binders considered in this study only polyvinyl alcohol adversely affected the final stoichiometry of the bonded oxide. It is noteworthy that this binder also produced the lowest compacted density. On the basis of these tests and previous use of several of the binders considered, 1 w/o Ceremul "C" was selected as the optimum binder addition.

In view of the variability experienced in density measurements, representative specimens were subjected to quantitative metallographic analysis to determine the more reliable method. Several fused and ceramic oxide mixtures were considered along with special dense oxide specimens for lower density comparisons. In each case, the density was determined by radiographic techniques and pycnometer measurement. Metallographic analysis consisted of superimposing a grid on photomicrographs of the subject specimens at 250 diameters and utilizing the interstices of the grid for point counting relative to contained porosity and the UO_2. Photomicrographs were taken on two different fields in each specimen, and the metallographic density on each field was determined with three grid orientations to provide a total of six determinations. The results of these tests, given in Table 3, indicated a reasonably good correlation between the pycnometer determinations and the densities obtained by the metallographic analysis. In this comparison, radiographic determinations generally yielded lower values, most likely due to poor edge definition in subsequent film measurements. Metallographic measurements with different grid orientations at different fields within the same sample showed good agreement.

TABLE 1. SINTERING AND ROLLING OF STAINLESS STEEL POWDER PLATES[a]

Compact	Sintering[b] Temperature, F	Thickness, in.	Sintered Density		Cold-Rolled Density	
			G per Cm^3	Per Cent of Theoretical	G per Cm^3	Per Cent of Theoretical
A	2000	.1465	5.66	71.6	--	--
B	2000	.143	5.83	73.8	--	--
C	2000	.144	5.64	71.4	--	--
D	2000	(c)	--	--	--	--
E	2000	.146	5.75	72.9	6.55	82.7
F	2000	.144	5.82	73.9	--	--
G	2000	.144	5.79	73.4	6.50	82.1
H	2000	(c)	--	--	--	--
I	2000	.145	5.78	73.3	6.49	82.0
J	2000	(d)	--	--	--	--
K	2000	(d)	--	--	--	--
L	2000	.145	5.78	73.3	(e)	--
M	2200	.146	5.75	72.9	6.57	82.9
N	2200	.143	5.87	74.4	--	--
O	2200	.146	5.75	72.9	6.48	81.8
P	2200	.147	5.81	73.6	6.51	82.2
Q	2200	.145	5.83	73.8	--	--
R	2200	.146	5.80	73.5	6.61	83.4
S	2200	.141	5.87	74.4	--	--
T	2200	.143	5.85	74.0	--	--

(a) Compacts pressed at 27 tsi to 70 per cent theoretical density, degassed at 700 to 900 F.
(b) Sintering time was 2 hr in hydrogen.
(c) Samples badly chipped on ends.
(d) Broken during handling as a green core.
(e) Broken during rolling.

TABLE 2. EFFECT OF BINDER ADDITIONS ON COMPACTING DENSITY AND STOICHIOMETRY OF UO_2 MIXTURES[a]

Binder Material	Amount	Average Density[b]		Oxygen/Uranium Ratio After Pressure Bonding[c]	Final Density, per cent of theoretical	Handling Characteristics
		G per Cm^3	Per Cent of Theoretical			
Ceremul C	1 dr/3 gm powder	7.82	71.2	2.017	97.6	Very good
Ceremul C + water	1 w/o solution	8.36	76.3	2.01	97.6	Good
Ceremul C + MEOH	1 w/o solution	8.28	75.5	2.01	97.6	Good
Carbowax 6000 + MEOH	1/2 w/o solution	7.99	72.8	2.013	94	Fragmented
Carbowax 6000 + MEOH	2 w/o solution	7.93	72.3	2.014	96	Fair
Carbowax 6000 + MEOH	2-1/2 w/o solution	8.45	77.1	--(d)	--	Very good
Carbowax 6000 + water	2-1/2 w/o solution	8.39	76.5	2.02	98.5	Very good
Polyvinyl alcohol	2 w/o solution	7.26	66.2	2.065	96.8	Very good
Paraffin + MEOH	2 w/o solution	8.27	75.5	2.01	96.7	Very good
Stearic acid * MEOH	2 w/o solution	8.34	76.1	2.01	95.8	Good
Camphor + MEOH	2 w/o solution	8.20	74.8	--(e)	--	Fragmented

(a) 60 w/o fused-40 w/o ceramic grade UO_2 mixture.
(b) Average of seven compacts
(c) Pressure bonded 3 hr at 2100 F and 10,000 psi.
(d) Failed during pressure bonding.
(e) Not loaded for pressure bonding.

TABLE 3. COMPARISON OF DENSITY-MEASUREMENT TECHNIQUES FOR PRESSURE-BONDED URANIUM DIOXIDE[a]

Material	Density Value Obtained by Method Shown, per cent of theoretical				
	Metallographic Counting[b]			Radiograph Measurement	Pycnometer Determination
	Field A	Field B	Average		
70 w/o fused UO_2-30 w/o ceramic UO_2	96.2	95.3	95.8	92.1	97.8
50 w/o fused UO_2-50 w/o ceramic UO_2	98.2	98.7	98.5	90.1	99.5
Special dense UO_2 (ideal cubic distribution)[c]	89.3	92.7	91.0	80.8	90.7
Special dense UO_2 (-140 + 200 mesh)	85.5	85.4	85.5	87.0	89.7

(a) Pressure bonded 3 hr at 2100 F and 10,000 psi.
(b) Three grid orientation measured on each field.
(c) 1.25 w/o minus 325 mesh, 6.25 w/o minus 150 plus 120 mesh, and 92.5 w/o minus 60 plus 80 mesh.

A limited study of vibratory packing of as-received fused and ceramic oxide mixtures was conducted to obtain a brief appraisal of such compacting methods. To minimize potential costs, the materials considered were limited to those size distributions representative of normal production oxides and slight modifications. These sizings are given in Table 4 along with other pertinent data characterizing the oxides.

TABLE 4. URANIUM DIOXIDE POWDERS USED IN VIBRATORY PACKING[a]

Oxide	Size Distribution		Tap Density, per cent of theoretical	Press Density at 50 TSI, per cent of theoretical	Oxygen/Uranium Ratio
	Mesh Fraction	w/o			
Fused, minus 6 mesh	-6+20	96.4	57.1	87.3	2.00
	-20+40	2.1			
	-40+60	0.6			
	-60+100	0.7			
	-100+200	0.1			
	-200+270	Nil			
	-270+325	Nil			
	-325	0.1			
Fused, minus 20 mesh	-20+80	63.4	64.4	88.6	2.00
	-80+100	5.7			
	-100+140	8.6			
	-140+200	6.1			
	-200+325	8.2			
	-325	7.9			
Ceramic, minus 325 mesh	-200+325	70.6[b]	16.0	56	2.08
	-325	29.4			

(a) All powders represent as-received sizings.
(b) Minus 200 plus 325-mesh fraction due to agglomeration; screening with a washer results in all minus 325 mesh.

The experiments were performed on Calidyne Model 174 unit which permitted a maximum force of 1500 lb at 75 G and vibration frequencies up to 3600 cps. A special aluminum fixture was fabricated to permit fastening the 12-in.-long by 0.5-in.-diameter tubes to the vibrating table. The tubes were positioned in this fixture by set screws. The operations employed in these tests were similar to the basic techniques developed by Hanford for vibratory packing of fused oxides.[3-6] At the outset, resonance frequencies for the subject 12-in.-long tube configurations were established. The loading and frequency variations were related to the packing density as measured by the change in height of the UO_2 column. Visual examination of the powders after packing and removal from the tubes did not reveal any gross size segregation.

The results of these tests are given in Table 5. It is evident from the data that considerable variations in packing densities were experienced within the limited range of powder mixtures and vibration parameters studied. From the standpoint of uniformity of deformation on pressure bonding, only those materials yielding densities of 70 per cent of theoretical or greater would be considered usable. The brief experiments conducted in this study indicate that such densities are feasible; however, additional tests will be required to define the limitations of ceramic additions and an optimized procedure.

TABLE 5. RESULTS OF VIBRATORY-COMPACTION EXPERIMENTS WITH URANIUM DIOXIDES

UO_2 Mixture[a], w/o			Vibration Frequency, cps			Density[b]	
Fused		Ceramic					
Minus 6 Mesh	Minus 20 Mesh	(Minus 325 Mesh)	Load	Sweep	Load, G	G per Cm^3	Per Cent of Theoretical
50[c]	45	5	780-810	--	40	8.0	73
			780-810	1300-1340	40	8.0	73
			1300-1340	3200	40	7.67	69.9
			780-810	3200	40	8.35	75.6
			780-810	3200-780	40	8.17	74.6
			780-810	3200	20	7.92	72.2
			780-810	3200	60	8.46	77.0
100	--	--	1300-1340	3200	40	6.86	62.5
--	85	15	780-810	3200	60	7.10	64.0
--	80	20	780-810	3200	60	6.83	62.3
			780-810	3200	60	7.27	66.3
75	--	25	780-810	3200	40	6.61	60.3
			780-810	3200	60	6.63	60.4
--	75[d]	25	780-810	3200	60	6.33	57.8
--	75	25	780-810	3200	60	6.05	55.1
--	75[e]	25	780-810	3200	60	6.16	56.2
--	50	25[f]	780-810	3200	60	6.18	56.4
37.5	37.5	25	780-810	3200	60	6.46	59.0

(a) All mixtures according to size distribution given in Table 4 except where noted.
(b) Single measurements on single samples.
(c) Minus 6 plus 20 mesh.
(d) Minus 20 plus 60 mesh.
(e) Minus 20 plus 325 mesh.
(f) 25 w/o remainder made up of minus 80 plus 200-mesh fused UO_2.

The cold pressing of UO_2 platelet cores was briefly investigated during this phase of the program. These components, which consisted of mixtures of fused and ceramic oxides, were prepared for the compartmented flat-plate development effort utilizing both green oxide platelets and partially sintered stainless steel components. Mixtures containing from 60 to 75 w/o fused oxide were blended with either 2 w/o polyvinyl alcohol or 1 w/o Ceremul "C" prior to compacting at 50 tsi. It was found that the Ceremul "C" binder yielded the higher pressed densities, which ranged from 72 to 85 per cent of theoretical for the mixtures investigated. In contrast, the polyvinyl alcohol yielded densities of 62 to 66 per cent of theoretical. In general, the handling characteristics of the compacts produced were good.

Pressure-Bonding Studies

The bonding studies during this phase of the program were directed toward a more complete definition of the effects of process variables and a detailed study of the fabrication of larger scale elements. The process variables concerned those other than the compaction of the initial powder materials, whereas the fabrication efforts were devoted to the rod and flat-plate element designs. These investigations were conducted in order to gain a more complete evaluation of the processing for the formulation of preliminary specifications.

Uranium Dioxide

The effects of bonding conditions on the final stoichiometry and density of ceramic grade UO_2 were further defined. The range of times and temperatures investigated in these experiments represented only slight modifications of the conditions used extensively in previous developments. To preclude the effects of binders and their removal, all specimens consisted of tamp-packed powders as contained in Type 304 stainless steel tubes. The results of these tests are listed in Table 6, where only slight differences in oxygen/uranium ratio and density are noted. The differences in stoichiometry are within the range of measurement, while the densities reported constitute a range of 97 to 99.5 per cent of theoretical. On the basis of these tests, it would appear that slight variations in bonding parameters have little effect on the final oxide characteristics.

In view of the variability experienced previously in the final oxide stoichiometry and structures, a brief study of the effects of different binders and methods of binder removal was undertaken. The binders considered in this series included polyvinyl alcohol, Carbowax 6000, and Ceremul "C". Removal techniques consisted of direct heating to 1200 F after evacuation and an extremely slow heating to 1200 F in vacuum. The results, which are given in Table 7, demonstrate the effects of the removal techniques and binders. It is evident that the polyvinyl alcohol binder resulted in high final oxygen/uranium ratios after pressure bonding, with the highest ratio related to the extremely slow binder-removal treatment. The remaining binder materials resulted in more acceptable oxygen/uranium ratios with similar final densities.

TABLE 6. EFFECTS OF GAS-PRESSURE-BONDING CONDITIONS ON THE STOICHIOMETRY AND DENSITY OF UO_2

Type of UO_2[a]	Pressure-Bonding Conditions			Final Oxygen/Uranium Ratio	Final Density[b], g per cm^3
	Time, hr	Temperature, F	Pressure, psi		
Ceramic grade	3	2100	10,000	2.011	10.6
Ceramic grade	3-1/2	2100	10,000	2.006	10.9
Ceramic grade	4	2100	10,000	2.014	10.7
Ceramic grade	3	2150	10,000	2.016	10.8
Ceramic grade	3	2200	10,000	2.002	10.9
75 w/o ceramic-25 w/o fused	3	2150	10,000	2.008	10.6
75 w/o ceramic-25 w/o fused	3-1/2	2100	10,000	2.000	10.9
50 w/o ceramic-50 w/o fused	3	2150	10,000	2.006	10.7
50 w/o ceramic-50 w/o fused	3-1/2	2100	10,000	2.006	10.8
Ceramic grade	3	2050	10,000	2.008	10.7

(a) Oxide was tamp-packed into tubes without binders.
(b) Pycnometer measurements.

TABLE 7. EFFECTS OF BINDERS AND METHODS OF BINDER REMOVAL ON THE STOICHIOMETRY AND DENSITY OF UO_2 MIXTURES[a]

Binder	Method of Binder Removal	Oxygen/Uranium Ratio	Density[b], g per cm^3
2 w/o polyvinyl alcohol semislurry	(c)	2.09	10.5
Ceremul "C", 1 drop per 3 g of UO_2	(d)	2.017	10.7
1/2 w/o of saturated solution of Carbowax 6000 and methyl alcohol	(d)	2.013	10.3
2 w/o saturated solution of Carbowax 6000 and methyl alcohol	(d)	2.014	10.5
2 w/o polyvinyl alcohol semislurry	(d)	2.065	10.6

(a) Mixtures consisted of 60 w/o fused-40 w/o ceramic UO_2 pressed at 50 tsi and pressure bonded at 2100 F for 3 hr and 10,000 psi.
(b) Densities by pycnometer measurement.
(c) Specimen was evacuated to 5 μ prior to heating. Successive evacuations to 5 μ were performed at temperatures of 400 to 1200 F with temperature intervals of 100 F. At 1200 F, the sample was held under vacuum for 6 hr. Throughout the heating, the vacuum did not exceed 70 μ.
(d) Specimen was evacuated to 5 μ prior to heating. Sample was then heated to 1200 F and held for 1-1/2 hr under vacuum.

The tendency for the polyvinyl alcohol binder to yield high oxygen/uranium ratios was further demonstrated in a series of experiments concerning a wide range of UO_2 mixtures. These mixtures, containing 2 w/o polyvinyl alcohol, were pressure bonded for 3 hr at 2100 F and 10,000 psi and then evaluated on the basis of final oxygen/uranium ratio and density. Binder removal in this case consisted of heating to 1200 F under vacuum in temperature intervals of 100 F, resulting in a vacuum of from 5 to 80 μ throughout the treatment. The results of these tests are listed in Table 8. High final oxygen/uranium ratios with each of the UO_2 mixtures considered are evident. In view of this consistent effect with the polyvinyl alcohol binder, it was excluded from consideration in further work.

A more detailed study of the effects of the binder removal and pressure-bonding operations on the oxygen/uranium ratio of uranium dioxide was conducted in conjunction with a 1 w/o addition of Ceremul "C" binder. The mixture considered in this comparison consisted of 60 w/o fused oxide and 40 w/o ceramic grade oxide. Comparisons were made with tap-packed and cold-pressed materials after binder removal and after pressure bonding. The results are given in Table 9. It is apparent from the data that outgassing at 300 F has little effect on the oxygen content, whereas removal of the Ceremul "C" at 1200 F results in a slight oxygen decrease. In both cases, further oxygen reductions are achieved on pressure bonding at 2100 F.

Stainless Steel

A brief series of corrosion tests was conducted to establish the effects of pressure bonding on the corrosion resistance of Types 304 and 304L stainless steel. Specimens in the as-received condition and bonded for 3 hr at 2050 F and 3-1/2 hr at 2100 F were represented in this study. Small stainless steel couples were encased in Ti-Namel for this study. The test environments considered included 750 F steam at 1500 psi and 680 F degassed water. All specimens were degreased prior to testing with the pressure-bonded samples for the 680 F tests subjected to a subsequent pickle in a nitric-hydrofluoric aqueous solution. The weight-change data recorded in these tests are given in Table 10. Surface films were noted to be characteristically blue-gray with some brown streaking in visual examinations after each test interval. In general, slightly higher weight changes were observed in the specimens which had been pressure bonded, particularly with the Type 304 stainless steel specimens. Metallographic examination of the materials after corrosion testing revealed a slight intergranular attack as shown in Figures 1 and 2.

Possible contamination of stainless components from spacer mat rials used in flat-plate pressure-bonding assemblies was also briefly examined. For these tests, the effects of contact with Ti-Namel and Armco iron spacers, bare and coated with waterglass, on the carbon content and corrosion behavior of stainless steel were considered. Multiple specimen packs were assembled and pressure bonded for 3 hr at 2100 F and 10,000 psi. After bonding, the individual samples were pickled in a 10 volume per cent HNO_3-2 volume per cent HF aqueous solution for 2 min at 120 F, degreased in acetone, and then corrosion tested for 12 weeks in 750 F steam (1500 psi). These results along with final carbon analyses are listed in Table 11. It is apparent that the Armco iron spacers coated with waterglass yielded the lowest weight change in corrosion and the lowest carbon content in the Type 304L stainless steel.

TABLE 8. PRESSURE-BONDED CHARACTERISTICS OF UO_2 MIXTURES[a]

Mixture[b]	Oxygen/Uranium Ratio	Density, g per cm^3
75 w/o fused-25 w/o ceramic	2.08	10.2
70 w/o fused-30 w/o ceramic	2.08	10.2
65 w/o fused-35 w/o ceramic	2.08	10.3
60 w/o fused-40 w/o ceramic	2.06	10.5
60 w/o fused-40 w/o ceramic	2.09	10.3
55 w/o fused-45 w/o ceramic	2.06	10.5
50 w/o fused-50 w/o ceramic	2.10	10.4
45 w/o fused-55 w/o ceramic	2.09	10.6
40 w/o fused-60 w/o ceramic	2.10	10.5

(a) All mixtures were compacted with 2 w/o polyvinyl alcohol binder and pressure bonded for 3 hr at 2100 F and 10,000 psi.

(b) Minus 20-mesh as-received fused oxides; minus 325-mesh ceramic grade oxide.

TABLE 9. THE EFFECTS OF PROCESSING ON THE OXYGEN/URANIUM RATIOS OF UO_2 MIXTURES[a]

Sample Condition[b, c]	Average Pressed Density, g per cm^3	Oxygen/Uranium Ratio	Bonded Density, g per cm^3
Mixture of as-received oxides	--	2.029	--
Tap-packed powders evacuated 1 hr at 300 F	--	2.03	--
Tap-packed powders evacuated 1 hr at 300 F and pressure bonded	--	2.016	10.4
Cold pressed and evacuated 1-1/2 hr at 1200 F	8.12	2.02	--
Cold pressed and evacuated 1-1/2 hr at 1200 F and pressure bonded	8.02	2.011	10.5

(a) 60 w/o as-received minus 20-mesh fused oxide-40 w/o minus 325-mesh ceramic grade oxide.

(b) Samples pressure bonded 3 hr at 2100 F and 10,000 psi where indicated.

(c) Cold-pressed pellets contained 1 w/o Ceremul "C" binder.

TABLE 10. EFFECT OF PRESSURE BONDING ON THE CORROSION RESISTANCE OF TYPE 304 AND TYPE 304L STAINLESS STEEL

Stainless Steel	Condition	Test Environment	Specimen Surface, cm^2	Weight Change, mg per cm^2			
				4 Weeks	8 Weeks	12 Weeks	16 Weeks
304L	As received	750 F steam at 1500 psi	51.7	0.009	0.11	0.099	0.085
304	As received	750 F steam at 1500 psi	83.3	0.032	0.10	0.091	0.090
304L	3-1/2 hr at 2100 F and 10,000 psi	750 F steam at 1500 psi	47.8	-0.014	0.036	0.021	0.01
304	3-1/2 hr at 2100 F and 10,000 psi	750 F steam at 1500 psi	74.8	0.18	0.33	0.35	0.35
304L	3 hr at 2050 F and 10,000 psi	750 F steam at 1500 psi	46.0	0.12	0.29	0.31	0.32
304	3 hr at 2050 F and 10,000 psi	750 F steam at 1500 psi	73.5	0.081	0.17	0.18	0.187
304L	As received	680 F degassed water	56.3	--	--	--	0.037
304	As received	680 F degassed water	84.3	--	--	--	0.040
304L	3-1/2 hr at 2100 F and 10,000 psi[a]	680 F degassed water	55.2	--	--	--	0.17
304	3-1/2 hr at 2100 F and 10,000 psi[a]	680 F degassed water	74.8	--	--	--	0.24
304L	3 hr at 2050 F and 10,000 psi[a]	680 F degassed water	53.6	--	--	--	0.046
304	3 hr at 2050 F and 10,000 psi[a]	680 F degassed water	73.5	--	--	--	0.040

(a) Samples vapor blasted and pickled in 10 volume per cent HNO_3 -2 volume per cent HF aqueous solution for 1 hr at room temperature prior to testing.

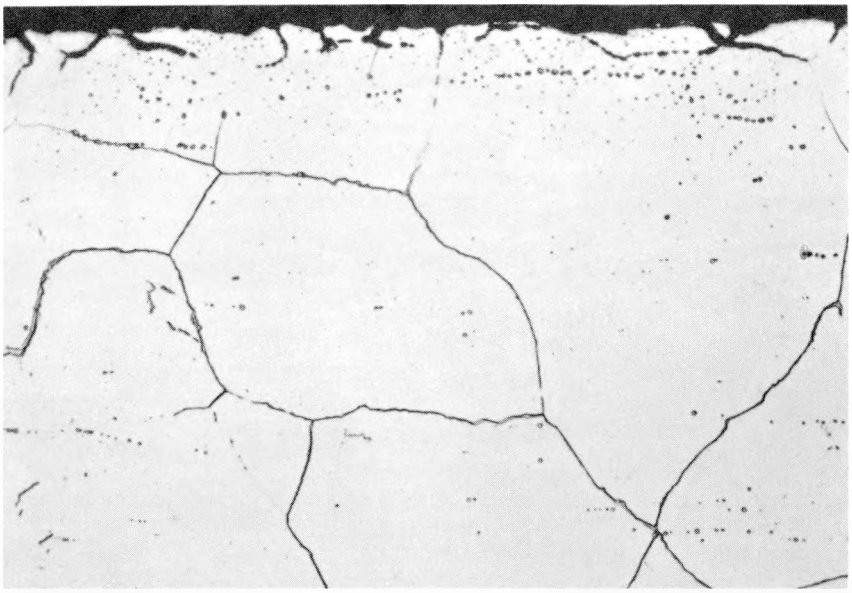

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FIGURE 1. EFFECT OF 16-WEEK 750 F STEAM CORROSION TEST ON TYPE 304 STAINLESS STEEL GAS-PRESSURE BONDED 3-1/2 HR AT 2100 F AND 10,000 PSI

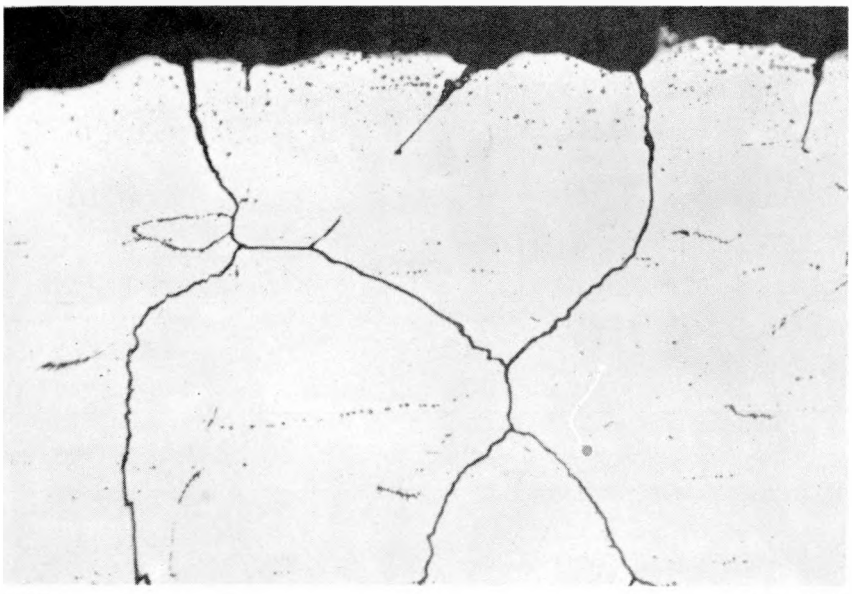

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FIGURE 2. EFFECT OF 16-WEEK 750 F STEAM CORROSION TEST ON TYPE 304 STAINLESS STEEL GAS-PRESSURE BONDED 3 HR AT 2050 F AND 10,000 PSI

TABLE 11. THE EFFECT OF VARIOUS SPACER MATERIALS ON THE CARBON CONTENT AND CORROSION RESISTANCE OF PRESSURE-BONDED STAINLESS STEEL[a]

Stainless Steel	Spacer Material	Carbon Content After Bonding, w/o	Specimen Surface Area, cm^2	Weight Change, mg per cm^2[b]		
				4 Weeks	8 Weeks	12 Weeks
304L	Ti-Namel coated with waterglass	0.05	20.8	0.28	0.26	0.24
304	Ti-Namel	0.08	18.7	0.26	0.22	0.20
304L	Armco iron	0.07	18.6	0.23	0.19	0.15
304L	Armco iron coated with waterglass	0.04	17.1	0.13	0.11	0.11
304	Armco iron	0.08[c]	19.6	0.18	0.18	0.20
304L	Ti-Namel	0.10[c]	18.0	0.30	0.29	0.27

(a) Specimens pressure bonded 3 hr at 2100 F and 10,000 psi.
(b) Specimens corrosion tested in 750 F steam (1500 psi).
(c) Pack leaked during pressure bonding.

Pressure-Bonded Assemblies

Further studies with rod and plate elements were directed toward larger scale designs with particular emphasis on dimensional control. This work also provided the opportunity to devise means to attain greater process control since a greater number of components were involved in each design. Fuel rods approximately 24 in. long and multiplate subassemblies were considered in this work.

Flat-Plate Elements

An investigation to determine the feasibility of pressure bonding green-pressed UO_2 platelets into flat-plate elements by the use of stainless steel powdered frames was conducted during the early part of this phase. From previous studies with rod-type specimens using green-pressed UO_2 and stainless steel pellets, it was shown that through a matching of initial densities, uniform deformation could be achieved during pressure bonding. It was believed that this same uniformity of deformation would apply to flat plates to produce plates of uniform thickness.

For this study, the cold-pressed UO_2 platelets consisted of a mixture of 60 w/o fused (minus 20 mesh) and 40 w/o ceramic (minus 325 mesh) oxide and a mixture of 75 w/o fused (minus 20 mesh) and 25 w/o (minus 325 mesh) oxide both with a 1 w/o Ceremul "C" binder addition. The platelets, measuring 0.100 by 1.0 by 1.0 in., were pressed at 50 tsi and ranged in density from 64.0 to 77.0 per cent of theoretical.

The stainless steel frames were machined from the partially sintered powder stainless steel flat plates described previously in this report. The frames were machined in four pieces, as shown in Figure 3, to provide for ease of fabrication and assembly.

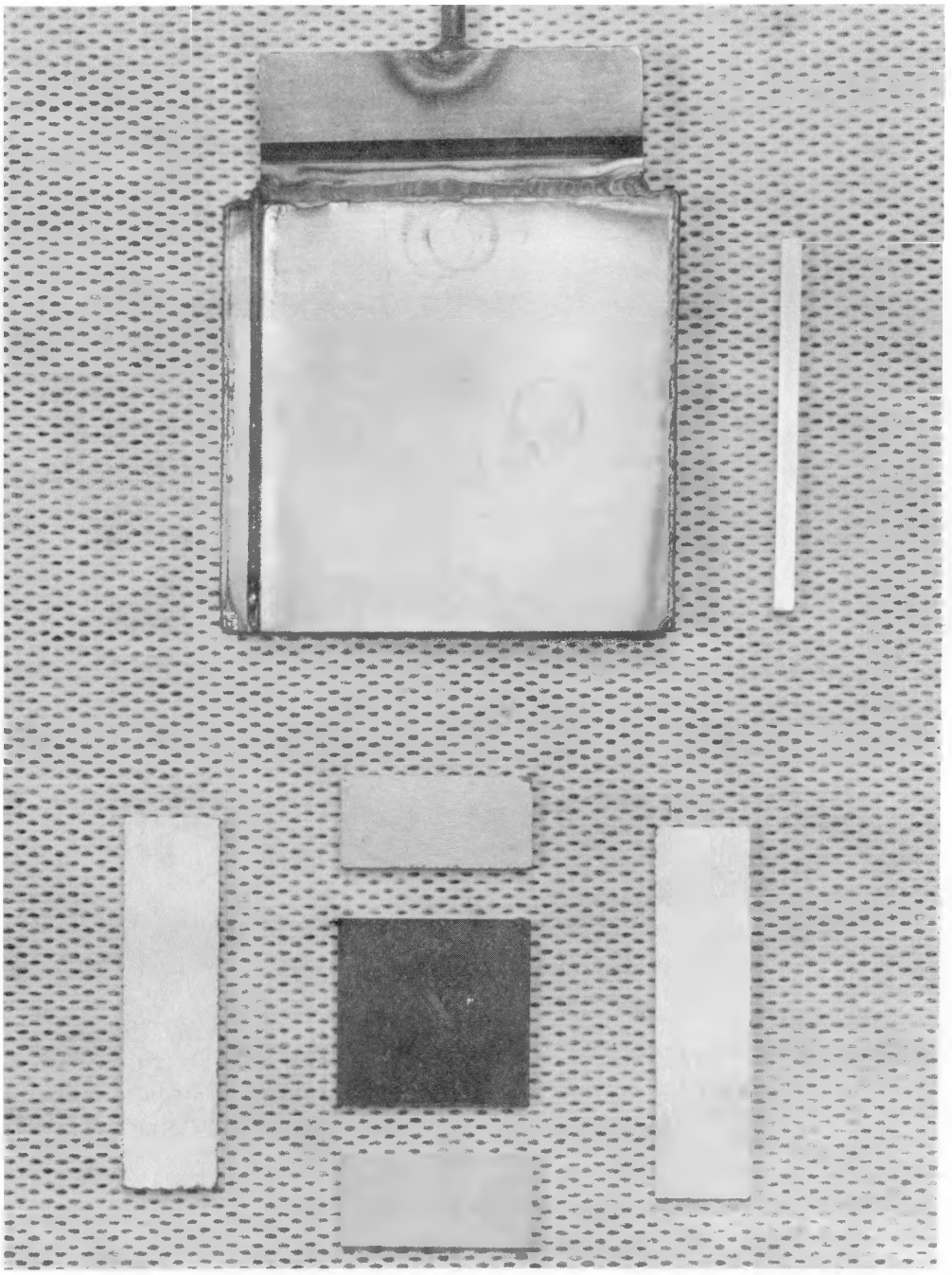

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FIGURE 3. COMPONENTS FOR A FLAT-PLATE SPECIMEN UTILIZING A GREEN-PRESSED UO_2 PLATELET AND STAINLESS STEEL POWDERED FRAMES

The components were treated with the standard pressure-bonding wash cycle and assembled into a flanged-type container. After the end seal was made by welding, the plate was degassed at 1200 F for 1-1/2 hr under a vacuum, allowed to cool, and the evacuation stem sealed. The 20-mil container served as the top and bottom cladding while the powdered stainless steel formed a 1/2-in. frame.

Of the six specimens prepared, two failed during pressure bonding through the cladding at the location of the evacuation hole in the end cap. This was corrected by decreasing the diameter of this hole in the last four specimens. All plates were pressure bonded for 3 hr at 2100 F and 10,000 psi with no external jigging. Table 12 presents a dimensional survey of the four plates that were successfully bonded. In general, these experimental plates demonstrated that, through the application of stainless steel powdered frames, green-pressed UO_2 platelets could be utilized for flat-plate elements. One of these plates is shown in Figure 4. It would, however, require closer control over the selection of starting densities for both the UO_2 platelets and the stainless steel powdered frames. Also, a jigging technique would be required in bonding these assemblies to prevent warpage. Further studies were not undertaken in order that the major effort could be applied to fabricating fuel rods for irradiation tests.

Flat-Plate Assemblies

The gas-pressure bonding of flat-plate fuel assemblies containing high-density ceramic platelets and fabricated stainless steel components was demonstrated in Phase II of the present study. Composites measuring 1.698 by 2.000 by 6.875 in. which contained nine fuel plates and eight coolant channels were successfully fabricated, as illustrated in Figure 5. Initial examination of these assemblies revealed good dimensional stability and excellent stainless steel self-bonds. It was necessary, however, to conduct further dimensional studies upon the flat-plate assemblies to define permissible tolerances for the stainless steel components and ceramic platelets.

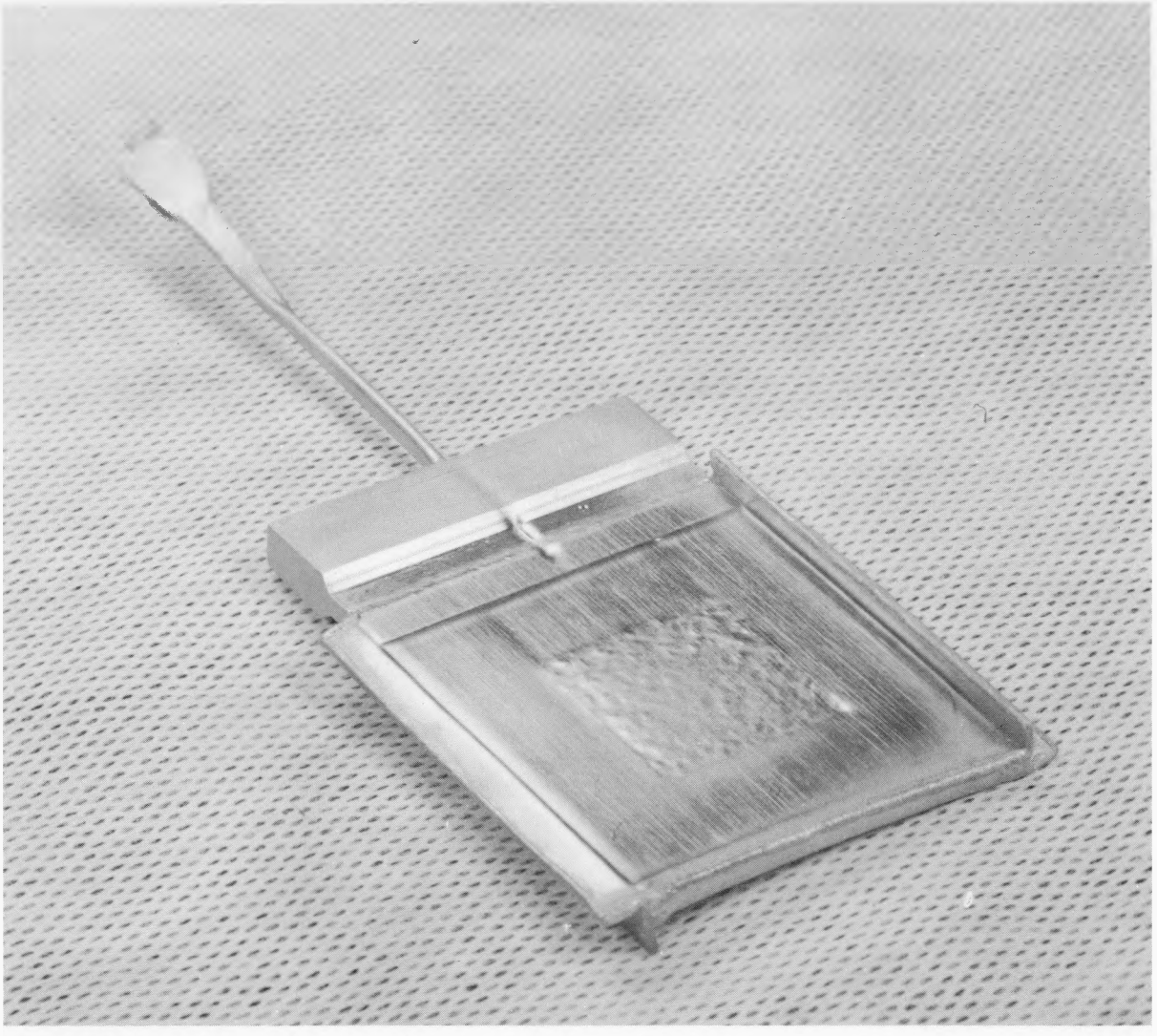
For this study, two compartmented flat-plate assemblies containing five fuel plates with over-all intended dimensions of 1.000 by 2.232 by 6.000 in. were bonded. A diagram of the assembly is shown in Figure 6, and a list of components is given in Table 13. The components, after being put through a standard wash cycle, were assembled into a flanged container. The container was then welded closed, evacuated, sealed, and pressure bonded for 3 hr at 2100 F and 10,000 psi.

After bonding, the end plugs of both assemblies were removed by machining. In one unit, the Ti-Namel spacers were etched out in an aqueous acid solution, whereas the remaining unit contained these spacers intact. Both assemblies were sectioned as illustrated in Figure 6c. The sections of each of the assemblies were ground square, and tracks were marked, as shown in Figure 6d, for measurement. All measurements were made by an optical comparator on the lettered faces. A compilation of the measurements is given in Table 14. It can be concluded that flat-plate assemblies fabricated by the gas-pressure-bonding process achieve excellent uniformity, and the assembly tolerances are extremely close to the variations of the contributing components.

TABLE 12. RESULTS OF EXAMINATION OF GAS PRESSURE BONDED[a] FLAT-PLATE ELEMENTS UTILIZING GREEN-PRESSED UO_2 PLATELETS AND STAINLESS STEEL POWDER FRAMES

	Density, per cent of theoretical				Average Thickness, in.				
	Frame		Platelet		Frame		Platelet		
Plate	Starting	Final	Starting	Final	Starting	Final	Starting	Final	Comments
1	71.5	99[b]	63.9	96.0[b]	.100	.117	.101	.120	Plate slightly bowed and the surface of cladding over core very rough; uniform thickness of platelet and frame within ±0.003 in.
2	77.0	--	77.0	95.0[c]	.100	.121	.100	.129	Plate flat and cladding surface smooth; uniform thickness of platelet and frame within ±0.003 in.
3	77.0	--	74.5	94.0[c]	.100	.124	.102	.130	Plate badly warped; however, surface of cladding smooth; uniform thickness of platelet and frame within ±0.004 in.
4	74.0	--	74.1	94.0[c]	.100	.118	.103	.130	Plate flat and cladding surface smooth; uniform thickness of platelet and frame within ±0.002 in.

(a) Plates pressure bonded at 2100 F for 3 hr at 10,000 psi.
(b) Pycnometer measurement.
(c) Calculated density.



N72511

FIGURE 4. GAS-PRESSURE BONDED STAINLESS STEEL-CLAD UO_2 FLAT-PLATE ELEMENT

This plate contained a green-pressed UO_2 platelet and stainless steel powder frame components and was bonded for 3 hr at 2100 F and 10,000 psi.

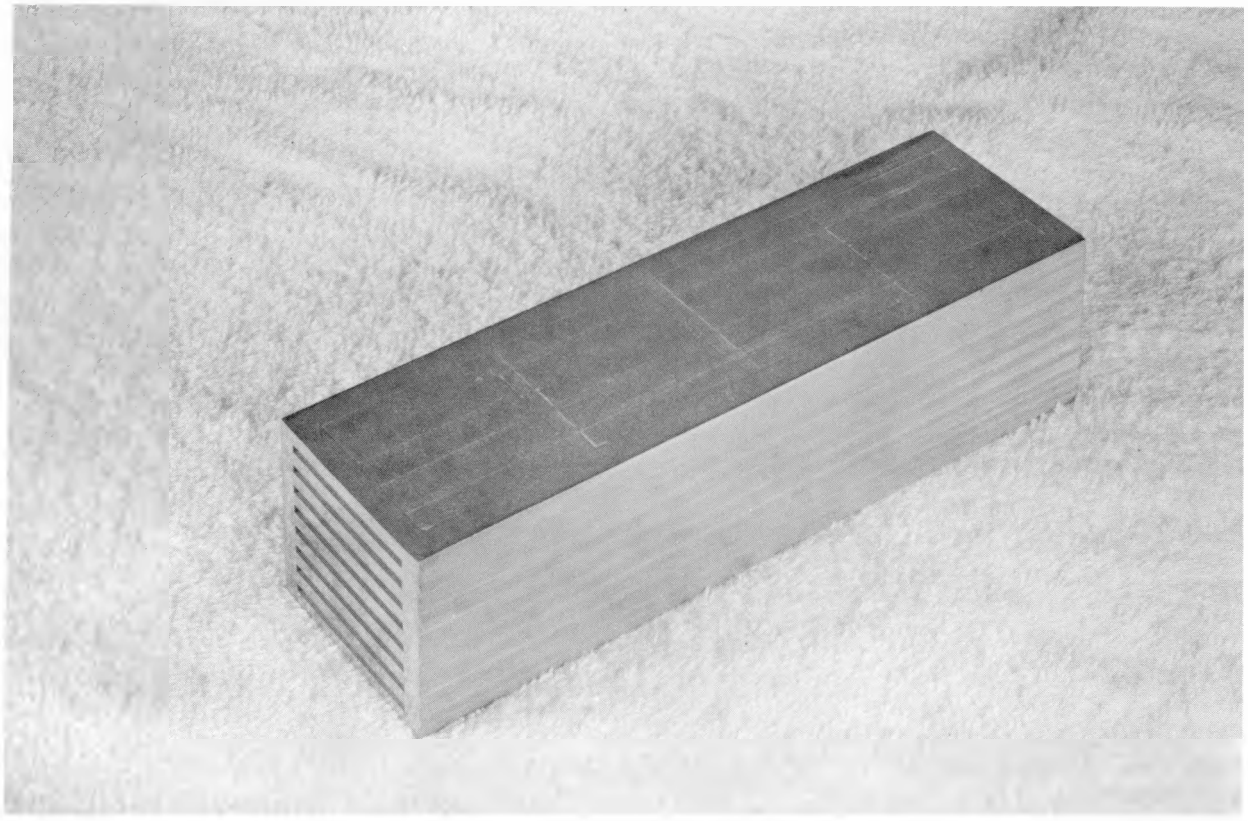


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FIGURE 5. STAINLESS STEEL-CLAD UO_2 COMPARTMENTED FLAT-PLATE ASSEMBLY PRESSURE BONDED FOR 3 HR AT 2100 F AND 10,000 PSI

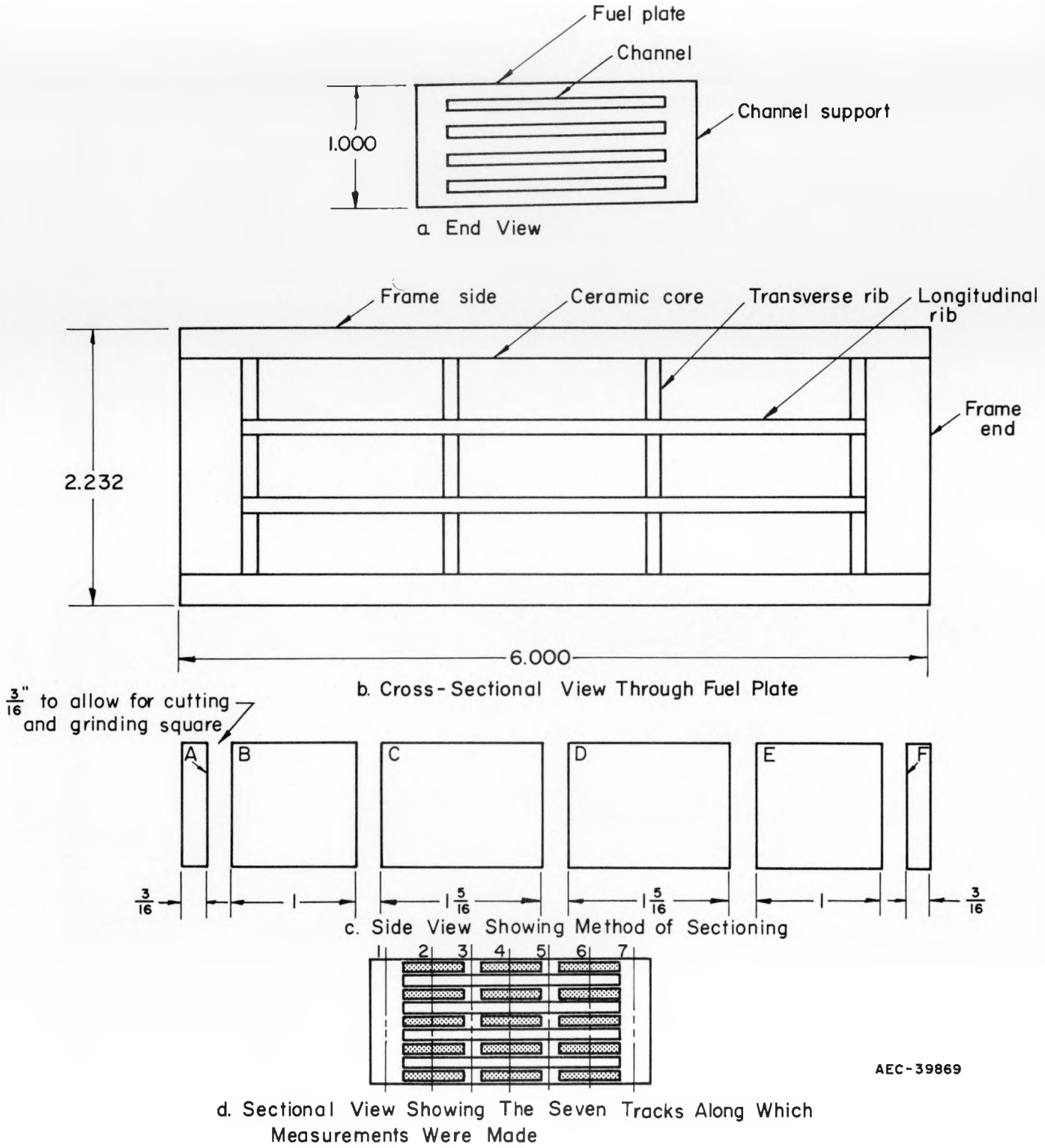

FIGURE 6. DIAGRAM OF THE COMPARTMENTED FLAT-PLATE ASSEMBLY USED FOR DIMENSIONAL STUDY

TABLE 13. COMPONENTS FOR THE 1 BY 2.232 BY 6 IN. FLAT-PLATE ASSEMBLY USE FOR DIMENSIONAL-CONTROL STUDIES[a]

Number of Components	Type of Component	Material[b]	Size, in.		
			Thickness[c]	Width[d]	Length[e]
10	Covers	Type 304 stainless	0.010	2.232	5.998/6.000
10	Frame sides	Type 304 stainless	0.100	0.250	6.000
10	Frame ends	Type 304 stainless	0.100	0.500	1.732
10	Longitudinal rib	Type 304 stainless	0.100	0.125	5.000
60	Transverse rib	Type 304 stainless	0.100	0.130 0.135 0.140 0.150	0.494/0.493
8	Channel support	Type 304 stainless	0.100	0.250	6.000
4	Channel spacer	Ti-Namel	0.100	1.732	6.000
2	Top and bottom pressure plates	Ti-Namel	0.100	2.232	6.000
3	End pressure plates	Ti-Namel	0.100	1.210	2.232
45	Platelets	UO_2 (95 per cent of theoretical density)	0.099/0.100	0.493/0.494	1.488/1.498

(a) The assembly has 157 components plus 5 pressure plates.
(b) All flat surfaces on the stainless steel and Ti-Namel components have a 100 to 125-μin. rms belt-abraded surface except for the covers, which have a cold-rolled surface. Edges on all metal components have a 125 to 150-μin. rms shaper-machined surface.
(c) Thickness tolerance ±0.0005 in. unless otherwise specified.
(d) With tolerance ±0.001 in. unless otherwise specified.
(e) Length tolerance ±0.002 in. unless otherwise specified.

TABLE 14. DIMENSIONAL EVALUATION OF GAS-PRESSURE-BONDED FLAT-PLATE ASSEMBLIES

Source of Variation	Intended Size, in.			Actual Size, in.		
	Print Dimension	Maximum	Minimum	Average Dimension	Maximum Deviation	Minimum Deviation
Thickness of assembly over the stainless steel components	1.0000	1.0095	0.9905	0.9971	1.0027	0.9932
Thickness of assembly over the core area	1.0000	1.0070	0.9880	0.9896	0.9960	0.9872
Thickness of fuel plate over the stainless steel components	0.1200	0.1215	0.1185	0.1185	0.1220	0.1145
Thickness of fuel plate over the core area	0.1200	0.1210	0.1180	0.1185	0.1206	0.1168
Thickness of channel[a]	0.1000	0.1010	0.0990	0.0991	0.1006	0.0974
Thickness of cladding	0.0100	0.0105	0.0095	0.0096	0.0106	0.0092
Width of assembly	2.2320	2.2390	2.2250	2.2310	2.2360	2.2265

(a) Channel alignment was within ±0.005 in.

Fuel Rods

Two rod element assemblies measuring 26 in. long were pressure bonded to demonstrate the degree of dimensional control attainable in scaled-up fuel-element geometries. The uranium dioxide consisted of a mixture of 60 w/o fused (minus 20 mesh) oxide and 40 w/o ceramic (minus 325 mesh) oxide with a 1 w/o Ceremul "C" binder addition. Pellets 0.540 in. in diameter were cold pressed to 72 per cent of theoretical density and then loaded into Type 304 stainless steel tubes measuring 0.542 in. in inside diameter with an 0.023-in. wall. A follower rod 0.539 in. in diameter was used to facilitate loading and insure intimate contact between individual oxide pellets. After seal welding of the end plugs and insertion of the evacuation stems, the rods were degassed at 1200 F for 1-1/2 hr under vacuum. The evacuation stems were then forge welded shut.

Two autoclave loading techniques were involved in pressure bonding the assembled fuel rods to establish their effects on the final straightness of the bonded rods. Rod 118 was loaded in the usual manner, being placed in the furnace cavity with a refractory packing material. The other, Rod 119, was first loaded into a stainless steel tube with a fine-grained alumina, and then this assembly was loaded into the furnace cavity. The bonding cycle consisted of 3 hr at 2100 F and 10,000 psi. Examination of the bonded assemblies revealed both a diametral and axial shrinkage corresponding to that noted previously in short assemblies. This effect is readily seen in Figure 7, where a final rod is compared with the initial stainless steel tube component. Both rods were found to be fairly straight, with a slight improvement attributable to the technique in which the rod was first loaded into an oversized tube. A dimensional survey of these rod assemblies is summarized in Table 15 along with the appropriate density and stoichiometry information. In general, good dimensional control was experienced with the scaled-up rod assemblies. Surface irregularities were more difficult to control because the cladding tended to indent at the UO_2 pellet interfaces, particularly where the UO_2 cores apparently chipped on loading.

TABLE 15. RESULTS OF EXAMINATION OF GAS-PRESSURE-BONDED STAINLESS STEEL-CLAD UO_2 ROD ELEMENTS

Rod	Core Length, in.		Tube OD, in.			Final Wall Thickness, in.	Average Density, g per cm^3		Average Final Oxygen/Uranium Ratio	Comments
	Starting	Final	Starting	Final Maximum	Final Minimum		Starting	Final		
118	25.375	23.204	0.588	0.547	0.542	0.026	8.0	10.51[a] 10.6[b]	2.01	Rod very uniform except for two indentations 10 mil deep; a bow of approximately 1/16 in. over a 1-ft length noted
119	25.375	22.985	0.588	0.547	0.542	(c)	7.7	10.45[a]	(c)	Very uniform except for one indentation (related to a chipped core); less than 1/32 in. bow over entire rod length

(a) Calculated density.

(b) Pycnometer measurement.

(c) Rod 119 was not sectioned for these measurements.

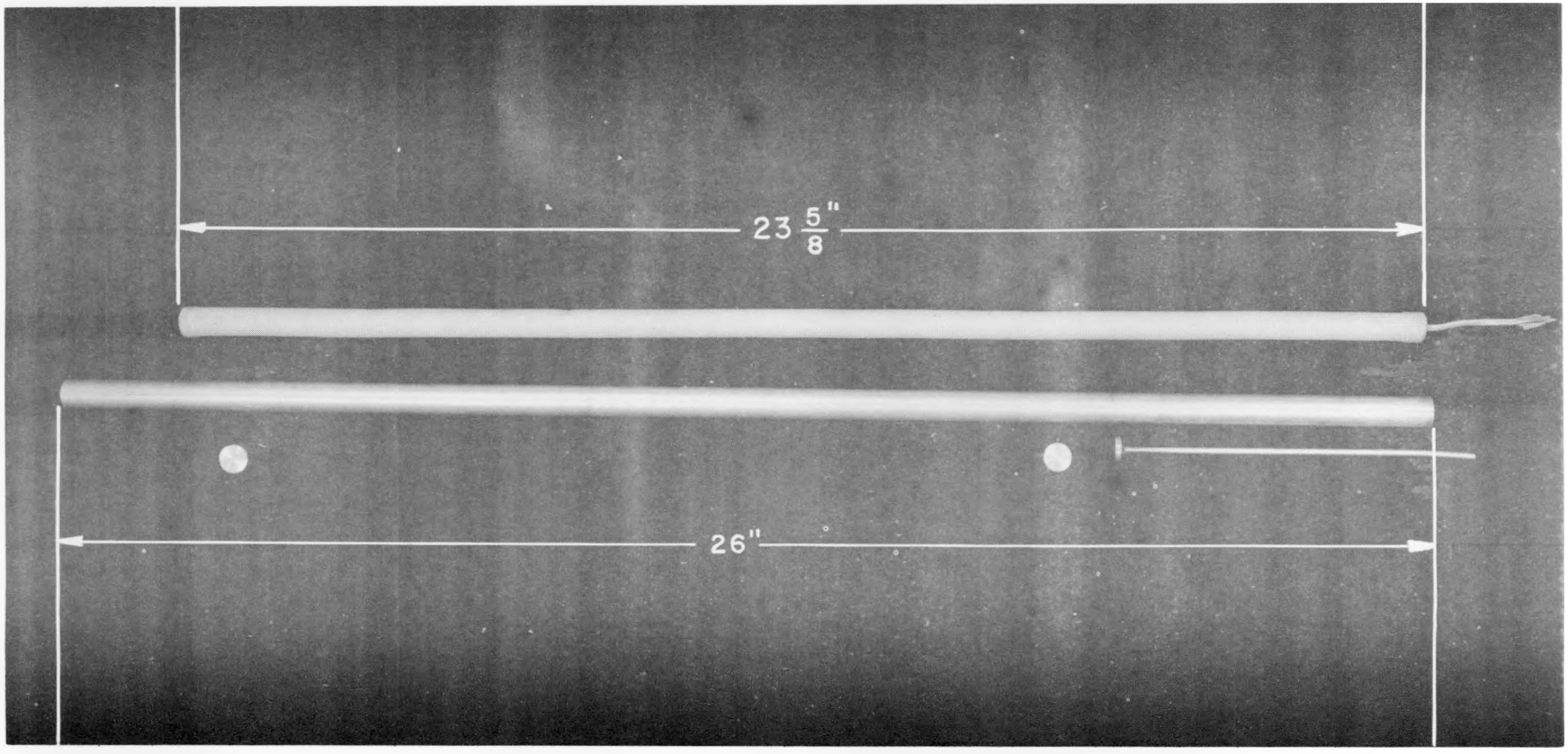

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FIGURE 7. GAS-PRESSURE-BONDED FUEL ROD AND INITIAL COMPONENTS

Note that both diametral and longitudinal shrinkage occurred during bonding.

Although the foregoing fabrication procedure does not represent an optimized low-cost process, it does afford the proper control of deformation during pressure bonding. On this basis, this procedure was selected as the method for the subsequent fabrication of fuel rods for irradiation. Since cold pressing as included in the above processing is inherently expensive, an optimized low-cost process would require a cheaper fuel loading method. Loading techniques such as vibration packing would offer a distinct cost advantage in this case; however, further development with regard to uranium dioxide mixtures would be required.

FABRICATION OF FUEL RODS FOR IRRADIATION TESTING IN THE VBWR

The investigation to define process specifications for the fabrication of fuel elements of various configurations by the gas-pressure-bonding technique was terminated before completion. This was done in favor of fabricating fuel rods for the irradiation testing of gas-pressure bonded UO_2 in the VBWR.

During the course of this program, the gas-pressure-bonded UO_2 appeared consistent with that of pressed and sintered oxide of equivalent density, as based on permeability and thermal-conductivity results. However, by the gas-pressure-bonding process, UO_2 has been produced over a range of densities, as high as 99 per cent of theoretical with intimate cladding contact, at temperatures considerably below those of conventional techniques. From these results, it appeared desirable to establish if any differences exist between the irradiation properties of gas-pressure bonded UO_2 and conventionally processed UO_2.

Specifications for a gas-pressure-bonded stainless steel-clad UO_2 fuel rod for irradiation testing were formulated with the General Electric Company, Atomic Products Division. The decision was made to prepare these fuel rods by loading cold-pressed pellets into stainless steel tubes and pressure bonding. It was considered that a minimum amount of investigation would be required to fabricate the fuel rods in this manner, even though it was apparent from previous studies that this would not give the optimum surface and dimensional uniformity. Such assemblies, however, would provide a good test of pressure-bonded UO_2. Four pressure-bonded stainless steel-clad UO_2 fuel-rod assemblies were furnished to the General Electric Company for irradiation and postirradiation evaluation of the pressure-bonded UO_2 under the AEC Fuel-Cycle Development Program.

Design Development

Specifications and drawings of the standard pelletized fuel-rod assembly for irradiation testing in the VBWR were furnished by the General Electric Company.[7,8] It was necessary, however, to make certain modifications in the design of this pelletized fuel rod to adapt it for the gas-pressure-bonding process. The standard pelletized fuel-rod assembly and its component parts are illustrated in Figure 8. It was desirable to hold the outer dimensions given in this drawing to permit easy installation of the rods into the present fuel-bundle design.

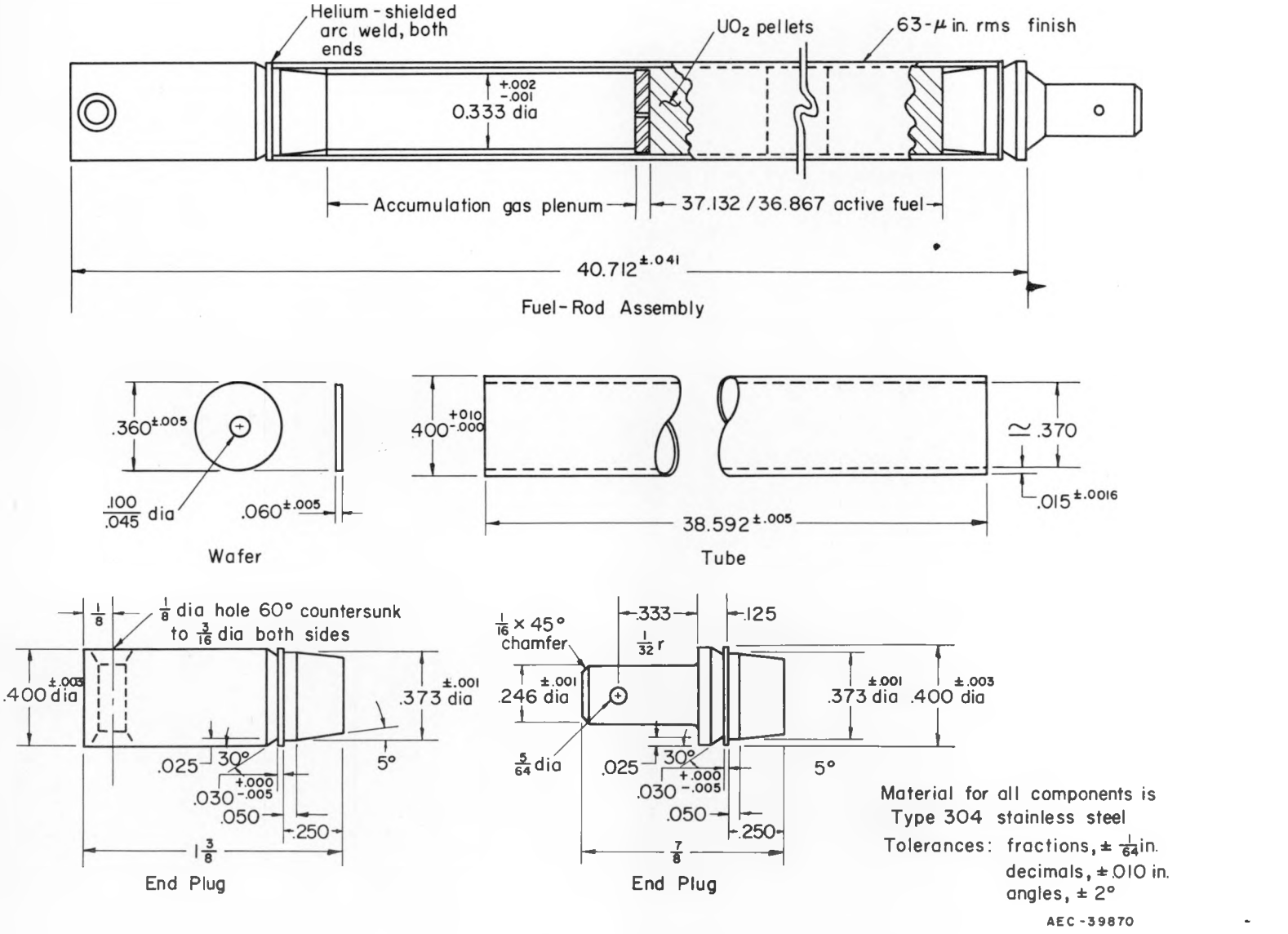


FIGURE 8. DRAWINGS FOR PELLETIZED UO_2 FUEL ROD FOR IRRADIATION TESTING IN THE VBWR

The specification required for the fuel enrichment was set at 5.5 per cent, assuming a final UO_2 density of 99 per cent of theoretical. The cladding was specified as being weld-drawn Type 304 stainless steel tubing manufactured to conform with ASTM designation A-269-58, referring to seamless and welded austenitic stainless steel tubing for general service, Grade TP 304. Supplemental requirements, as to mechanical properties, finish, and packing, were also given for this tubing. Both the 5.5 per cent enriched UO_2 and stainless steel tubing were supplied by the General Electric Company.

The modifications that were required to adapt the pelletized rod design for gas-pressure-bonding fabrication are illustrated in Figures 9 and 10 and summarized as follows:

(1) The gas-pressure bonding equipment available for fabricating these rods had a 30-in. hot zone for the bonding conditions required. It was therefore necessary to produce the rods in two segments. This brought about the following changes:

   (a) The two fuel-rod segments were to contain 24- and 12-in.-long fuel sections, respectively.

   (b) A screw-and-pinned type connection was designed to join the two segments. A maximum length of 1 in. of stainless was allowable for this joint.

   (c) Each of the two segments was to contain an accumulation gas plenum. The combination of the two should be 1-3/4 to 2 in. in length.

(2) Because of the amount of cladding deformation that is associated with the gas-pressure bonding of green-pressed cores, it was necessary to make the following changes:

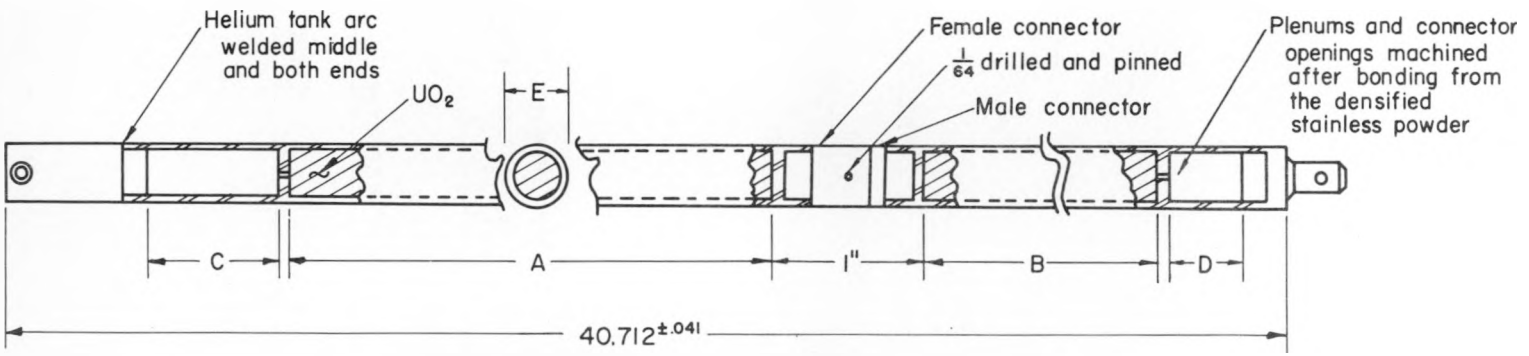
   (a) The tolerance of the rod diameter was relaxed as shown in Figures 8 and 9.

   (b) It was necessary to use green-pressed stainless steel powdered pellets in the plenum and connection areas, which were machined out after densification by pressure bonding.

(3) The surface finish was to be equal to that achieved on Rods 118 and 119.

Materials Development

The 5.5 per cent enriched oxide requirements and specifications were established from previous studies. This consisted of the use of a 40 w/o ceramic-60 w/o fused mixture, which had demonstrated the best pressure-bonding results. The fused oxide specified for the enriched rods consisted of a minus 20-mesh material with an oxygen/uranium ratio of 2.005 procured from the Spencer Chemical Company. The ceramic oxide consisted of a minus 325-mesh material with an oxygen/uranium ratio of 2.08 and a 0.6-μ average crystal size procured from the Mallinckrodt Chemical Works.



Source of Variation	Dimension, in.
Core Length, A	$24^{\pm\frac{1}{2}}$
Core Length, B	$12^{\pm\frac{1}{2}}$
Total Core Length, A+B	$36^{\pm\frac{1}{2}}$
Gas Plenum, C	1.332/1.166
Gas Plenum, D	0.666/0.583
Total Gas Plenum, C+D	$2/1\frac{3}{4}$
Outside Diameter, E	$.400^{+.005}_{-.010}$ maintain ±.005 for given diameter
Cladding Thickness Over Core	$.015^{\pm.0016}$
Plenum Wall Thickness	.040 approximate
Plenum Spacer Thickness	.060 approximate

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FIGURE 9. MODIFICATIONS REQUIRED TO ADAPT THE PELLETIZED ROD FOR THE GAS-PRESSURE-BONDING PROCESS

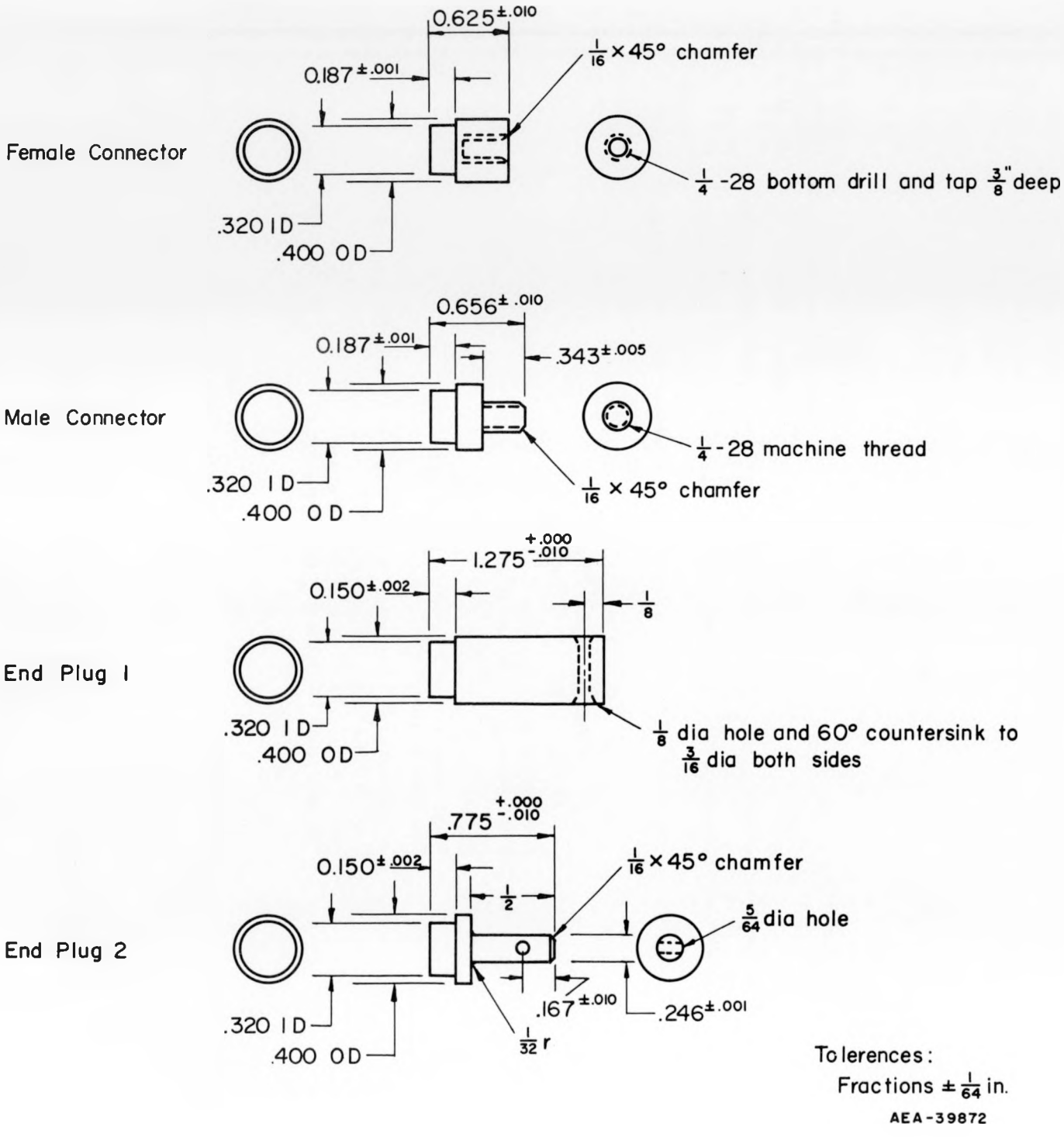


FIGURE 10. COMPONENTS FOR THE GAS-PRESSURE-BONDED UO_2 FUEL ROD

Due to a change in MCW production techniques over the past several years, it was necessary to evaluate samples of an approximate duplication of the oxide used previously in the program. Tests conducted on these oxides revealed that they all demonstrated less activity than that previously used (Type A-150). Further and more detailed evaluations were made of the GE-type and superactive-type oxides, as these were considered the more reproducible types. These included studies of the effect of outgassing temperatures and binders upon the two oxides, as shown in Table 16. After completion of these studies, the superactive oxide was selected as an alternate enriched ceramic oxide.

During these evaluation studies, difficulties with varied particle size distribution and extensive fragmentation on cold pressing were experienced with the fused oxide received from the Spencer Chemical Company. It was determined that the method of manufacture of this product had been slightly altered over the past year to produce a more friable oxide for the swaging and vibratory-compaction processes. As a result a tendency toward a higher percentage of coarse particles in the more recent material was noted. These difficulties were resolved with the Spencer Chemical Company, and an enriched oxide was ordered to duplicate that used during the earlier development studies.

Process Development

Utilizing the information obtained from the development studies on the compartmented rod and the fabrication by gas-pressure bonding of the two 26-in.-long rod assemblies presented in this report, details were further developed for the irradiation fuel-rod assemblies.

During the previous development studies, tubing with a nominal 0.580-in. inside diameter and a 0.020-in. wall thickness was used almost exclusively. From the pressure-bonding results obtained with this tubing in the fabrication of rod-type elements, it was calculated that tubing with a 0.425-in. outside diameter and 0.013 ± 0.0016-in. wall thickness should be used to meet the dimensional requirements for the present fuel rods. Dies 0.394 in. in diameter were designed and made for cold pressing the UO_2 cores and stainless steel pellets. To verify these computations, several 6-in.-long pieces of tubing were machined to the calculated size, loaded with UO_2 cores of the desired material specifications, and pressure bonded at 2100 F for 3 hr at 10,000 psi. The results from these assemblies demonstrated that cores and pellets produced from the 0.394-in.-diameter die were easily loaded and that the final desired diameter dimension could be achieved.

It had been shown during the study[2] of the compartmented rods that by proper selection of green-pressed densities between the fuel material and minus 100-mesh Type 304L stainless steel powder fuel rods with uniform diameters along the length could be produced by pressure bonding. Investigations showed that, utilizing the 0.394-in.-diameter die, UO_2 cores of the specified mixture pressed at 50 tsi yielded densities of 73 to 76 per cent of theoretical, while stainless steel pellets pressed at 40 tsi represented a density range of 78 to 80 per cent of theoretical. This difference in starting densities provided the uniform deformation of the respective components during pressure bonding.

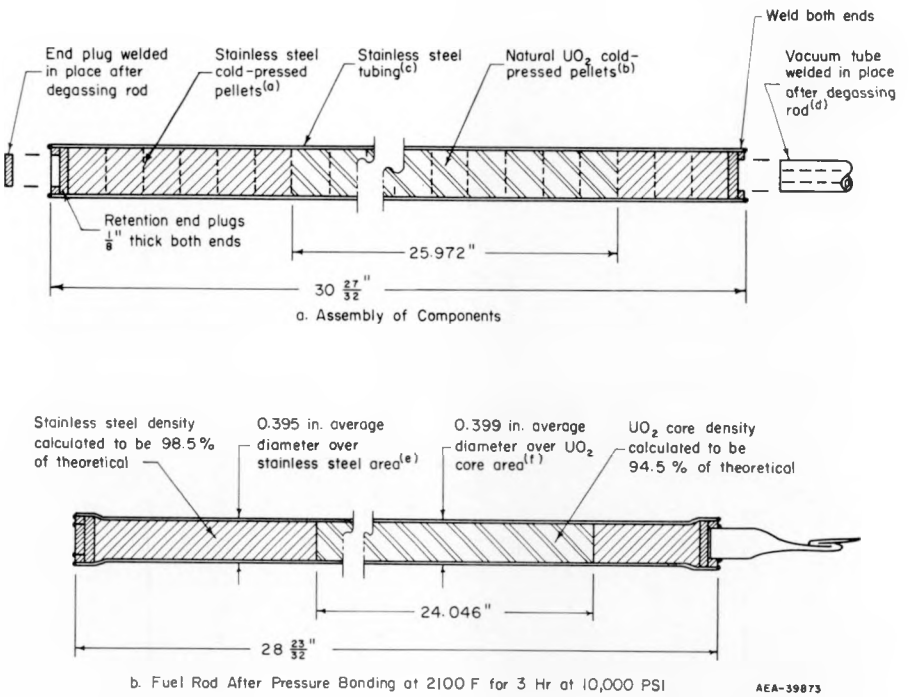
TABLE 16. COMPARISON OF DIFFERENT MCW CERAMIC GRADE OXIDES IN PRESSURE BONDING[a]

Material[b]	Initial Oxygen/Uranium Ratio	Packing Method	Binder	Outgassing Method	Average Starting Density[c], g per cm^3	Oxygen/Uranium Ratio After Pressure Bonding	Final Density[c], g per cm^3
A-150 type	2.08	Cold pressed at 50 tsi	1 w/o Ceremul "C"	1 hr at 1200 F	6.65	2.03	10.78
PWR type	2.071	Cold pressed at 50 tsi	1 w/o Ceremul "C"	1 hr at 1200 F	7.85	2.012	9.51
Approximate duplication of A-150 type	2.093	Ditto	Ditto	Ditto	7.7	2.015	10.00
GE type	2.118	Ditto	Ditto	Ditto	7.45	2.021	10.20
Superactive type	2.085	Ditto	Ditto	Ditto	6.45	2.042	10.70
40 w/o A-150 type – 60 w/o fused	--	Tamp packed	None	1 hr at 300 F		2.012	10.50
40 w/o GE type – 60 w/o fused	--	Tamp packed	None	1 hr at 300 F		2.010	10.40
40 w/o superactive type – 60 w/o fused	--	Tamp packed	None	1 hr at 300 F		2.006	10.20
40 w/o GE type – 60 w/o fused	--	Cold pressed at 50 tsi	1 w/o Ceremul "C"	1 hr at 400 F	7.63	2.034	9.84
40 w/o superactive type – 60 w/o fused	--	Ditto	Ditto	1 hr at 400 F	7.7	2.042	9.72
40 w/o GE type – 60 w/o fused	--	Ditto	Ditto	1 hr at 800 F	7.65	2.020	10.40
40 w/o superactive – 60 w/o fused	--	Ditto	Ditto	1 hr at 800 F	7.7	2.019	9.93
40 w/o GE type – 60 w/o fused	--	Ditto	Ditto	1 hr at 1200 F	7.6	2.012	10.40
40 w/o superactive – 60 w/o fused	--	Ditto	Ditto	1 hr at 1200 F	7.85	2.016	10.30
40 w/o GE type – 60 w/o fused	--	Ditto	1-1/2 w/o Carbowax 6000	1 hr at 400 F	7.85	2.010	9.84
40 w/o superactive – 60 w/o fused	--	Ditto	Ditto	1 hr at 400 F	8.06	2.009	9.98
40 w/o GE type – 60 w/o fused	--	Ditto	Ditto	1 hr at 800 F	7.72	2.005	10.3
40 w/o superactive – 60 w/o fused	--	Ditto	Ditto	1 hr at 800 F	8.01	2.007	10.3
40 w/o GE type – 60 w/o fused	--	Ditto	Ditto	1 hr at 1200 F	7.76	2.004	10.3
40 w/o superactive – 60 w/o fused	--	Ditto	Ditto	1 hr at 1200 F	7.74	2.005	10.4

(a) Pressure bonding conditions were 3 hr at 2100 F and 10,000 psi.

(b) Fused oxide used in mixtures was a minus 20-mesh material with an oxygen/uranium ratio of 2.005.

(c) Pycnometer measurements.



(a) Minus 100-mesh Type 304L stainless steel powder cold pressed at 40 tsi into pellets 0.395 in. in diameter by 0.500 in. high to a density of 78 per cent of theoretical.

(b) 60 w/o fused (friable type minus 20-mesh) and 40 w/o ceramic (superactive type, minus 325 mesh) UO_2 mixture with a Ceremul "C" binder (1 w/o of total) cold pressed at 50 tsi into pellets 0.397 in. in diameter by 0.400 in. high to a density of 74 per cent of theoretical.

(c) Type 304L stainless steel tubing 0.426 in. in OD with a 0.014-in. wall thickness.

(d) After loading the rod was degassed in a vacuum at 1200 F for a period of 2 hr to remove binder.

(e) The maximum and minimum deviation from this average diameter was 0.396 and 0.393 in., respectively.

(f) The maximum and minimum deviation from this average diameter was 0.404 and 0.396 in., respectively.

FIGURE 11. DIAGRAM OF THE GAS-PRESSURE-BONDED STAINLESS STEEL-CLAD NATURAL UO_2 PROTOTYPE VBWR IRRADIATION FUEL ROD

A prototype fuel-element segment with a fuel length of 24 in., as shown in Figure 11, was prepared using the Type 304L stainless steel tubing provided for the enriched fuel rods and natural uranium dioxide core material. This rod was processed to further check the gas-pressure-bonding procedure anticipated for the enriched fuel rods. In this case, the tubing was cut to a length providing for the solid stainless end plugs and 7.5 per cent core calculated length shrinkage. On loading, the pellets were inserted individually and seated with a 0.390-in.-diameter rod. Radiographs were taken after loading to check for possible void areas and also provide a measurement of core length. Outgassing for binder removal consisted of loading the unit into a stainless tube which was subsequently evacuated to 10 μ and then heated for 2 hr at 1200 F under vacuum. The techniques employed in plugging and sealing the assembly are evident from the detailed sketch in Figure 11, where other particulars of the fuel rod are also listed.

The completed assembly was then gas-pressure bonded for 3 hr at 2100 F and 10,000 psi. Examination of the rod showed good dimensional control, as illustrated in Figure 11; however, it was apparent that the density of the core, surface roughness of the cladding, and straightness of the rod should be improved through modifications in the subsequent fabrication of the enriched fuel rods.

Fabrication of VBWR Elements

Four gas-pressure-bonded fuel-rod assemblies were fabricated for irradiation testing in the VBWR. The processing in this case was based on previous process developments and, where necessary, minor modifications were made. The fuel in these rods consisted of a mixture of 60 w/o fused and 40 w/o superactive (ceramic) UO_2, as listed in Table 17. In view of the somewhat lower activity demonstrated by the superactive oxide as compared to the Type A-150 ceramic, a final density of 96 per cent of

TABLE 17. UO_2 MATERIAL USED IN THE GAS-PRESSURE BONDED VBWR FUEL-ROD ASSEMBLIES

Oxide Type	Supplier	Lot	Mixture, w/o	Enrichment, per cent	Net Weight, g	Oxygen/ Uranium Ratio	Mesh Size	Water-Immersion Density, g per cm^3
Fused	Spencer	359-55-1	60	5.54	3,479.3	2.000	-20[a]	10.9
Superactive[b] (ceramic)	Mallinckrodt	KO1LD	40	5.5	2,862.19	2.06	-325	--

(a) Sieve analysis of fused UO_2:

Mesh	Amount, w/o
+80 mesh	60.8
-80+100	6.45
-100+140	8.32
-140+200	7.54
-200+325	6.16
-325	10.07

(b) This material was selected as an alternate for the Type A-150 ceramic previously used.

theoretical was expected with this mixture. This estimate was based on the fact that the individual superactive oxide pressure bonded to 96 per cent of theoretical at 2100 F as compared to 99 per cent of theoretical for the Type A-150. The cladding consisted of Type 304L stainless steel tubing which was selected in place of the Type 304 stainless steel to reduce the possibility of sensitization due to the slow cool during the bonding cycle. This tubing measured 0.425 in. in outside diameter with a 0.014-in. wall thickness.

During the course of fabricating the VBWR fuel-rod assemblies, it was found necessary to make several slight modifications in the intended procedure. A slight growth of the cold-pressed uranium dioxide pellets was noted in storage after pressing; therefore, the pellets were loaded directly into tubes to facilitate this operation and insure a minimum pellet-cladding gap. Also, a swage sizing operation was employed after bonding to increase the surface smoothness and straightness of the fuel segments. Since the possibility existed that the fuel may have been slightly fragmented, the fuel sections were rebonded. Also, the rods were not subjected to any pickling treatments to preclude the possibility of stress corrosion in subsequent testing. The fabrication of these rods involved five major areas of effort which are detailed with respect to the individual steps in the following:

(1) Preparation of UO_2 pellets

Weigh fused and superactive oxides for blending.
Add 1 w/o of total mixture Ceremul "C" binder to superactive UO_2.
Blend 3 hr in a V-mixer.
Size blended oxide through a 40-mesh screen.
Add appropriate weight of fused oxide.
Blend 3 hr in a V-mixer.
Cold press at 50 tsi in a 0.394-in.-diameter die (72 to 76 per cent of theoretical density).
Measure and weigh each pellet.
Load pellets directly into stainless steel tube.

(2) Preparation of Type 304 stainless steel pellets

Use powder directly as supplied (minus 100 mesh).
Cold press at 40 tsi in a 0.394-in.-diameter die (77 to 80 per cent of theoretical density).
Measure and weigh each pellet.
Store in a desiccator until required.

(3) Fuel-segment assembly

Machine Type 304 stainless steel end caps, end plugs, and connectors.
Cut Type 304L stainless steel tubing to lengths of 30-7/8 and 17-7/8 in.
Select and weigh tubes and corresponding stainless steel components.
Identify.
Clean components according to the standard cleaning cycle.[2]
Affix tube in retaining jig.
Load bottom end caps.

Load six stainless steel pellets and seat each with a 0.390-in.-diameter rod.
Load UO_2 pellets and seat each with a 0.390-in.-diameter rod.
Load and seat three stainless steel pellets.
Insert end caps and evacuation stem.
Weigh assembly.
Radiograph.
Weld retainer end caps.
Place assembled rod into a stainless steel vacuum tube.
Evacuate and heat to 1200 F for 2 hr; slow cool.
Weld solid end caps and evacuation tube in place.
Evacuate to 5 μ and seal.

(4) Pressure bonding

Pressure check for leaks.
Load into autoclave.
Pressure bond for 3 hr at 2100 F and 10,000 psi.
Make visual postbonding inspection.
Swage size (reduce from 0.404 to 0.400 in. in diameter).
Pressure bond for 2 hr at 2100 F and 10,000 psi.
Make visual postbonding inspection.
Radiograph.

(5) Fuel-rod assembly

Machine fission-gas plenums and connector openings.
Radiograph.
Clean all components according to standard cleaning cycle.[2]
Drill 1/16-in.-diameter hole from plenum into UO_2.
Insert end plugs and connectors.
Radiograph.
Weld end plugs and connectors in a helium tank (see Figure 12).
Dress welds.
Leak check.
Clean completed segments and connector.
Fasten segments into complete rod assemblies (see Figure 9).
Make dimensional survey.

Evaluation of comparison fuel rods revealed a final average UO_2 density of 10.55 g per cm^3 (96.3 per cent of theoretical) and an average oxygen/uranium ratio of 2.002. A typical microstructure of the bonded oxide and oxide-cladding interface is shown in Figure 13. In this photomicrograph the intimate contact between cladding and core is demonstrated. Residual cracking in the oxide is also evident, indicating an incomplete healing of cracks apparently initiated during the swage sizing operation. Examination of the cladding material showed a relatively large grain size, as demonstrated in Figure 14. It should be noted that neither the tubing as supplied nor the pressure-bonded rods were subjected to any acid to preclude the possibility of stress corrosion.

To further establish the integrity of the cladding on the pressure-bonded fuel rods, further tests were conducted on representative samples of this material. A 1.650 by 0.580 by 0.025-in. section of bonded cladding from a comparison fuel rod

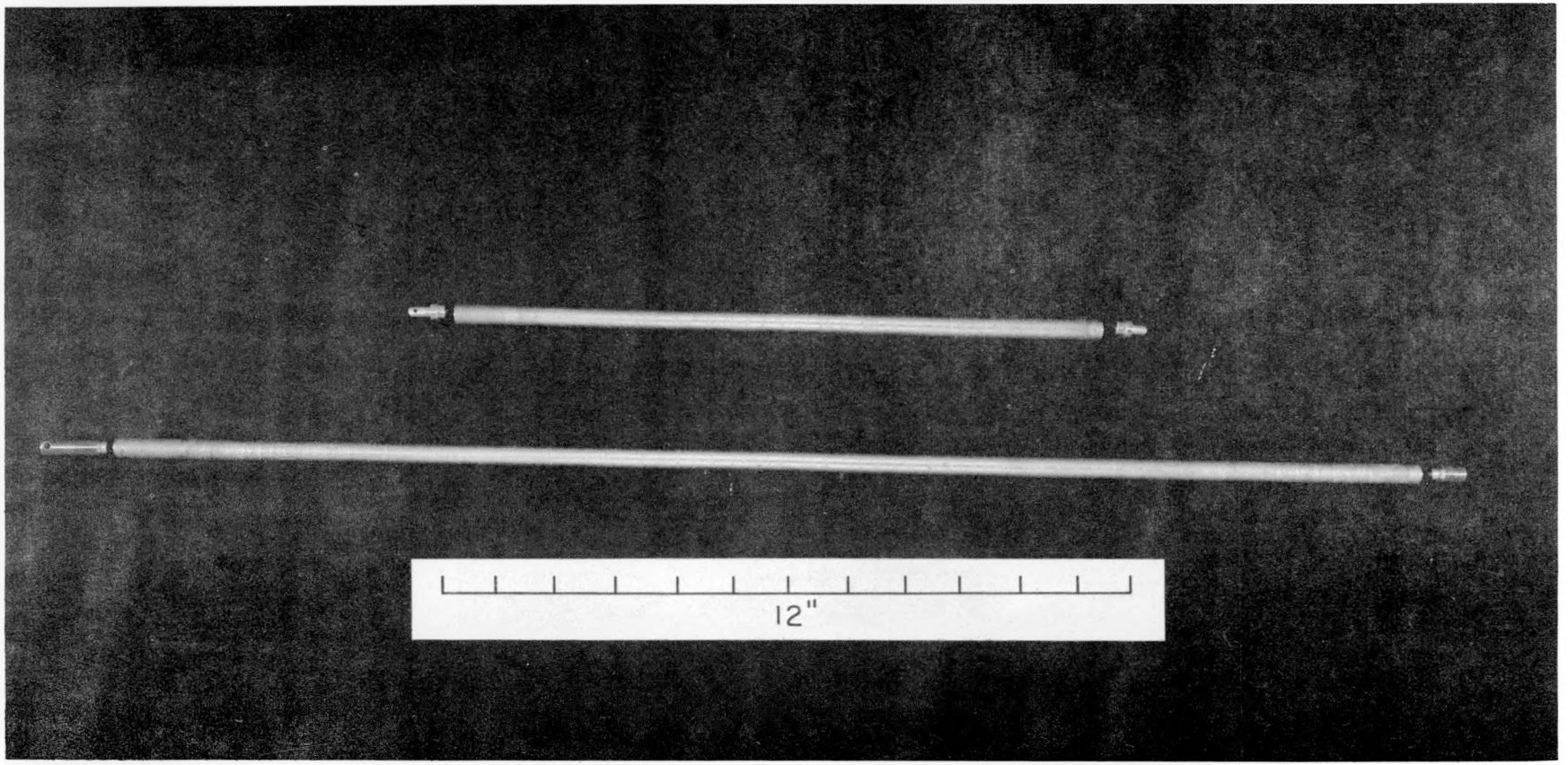


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FIGURE 12. 12- AND 24-IN. FUEL-ROD SEGMENTS AFTER MACHINING END PLUGS AND CONNECTORS IN PREPARATION FOR WELDING

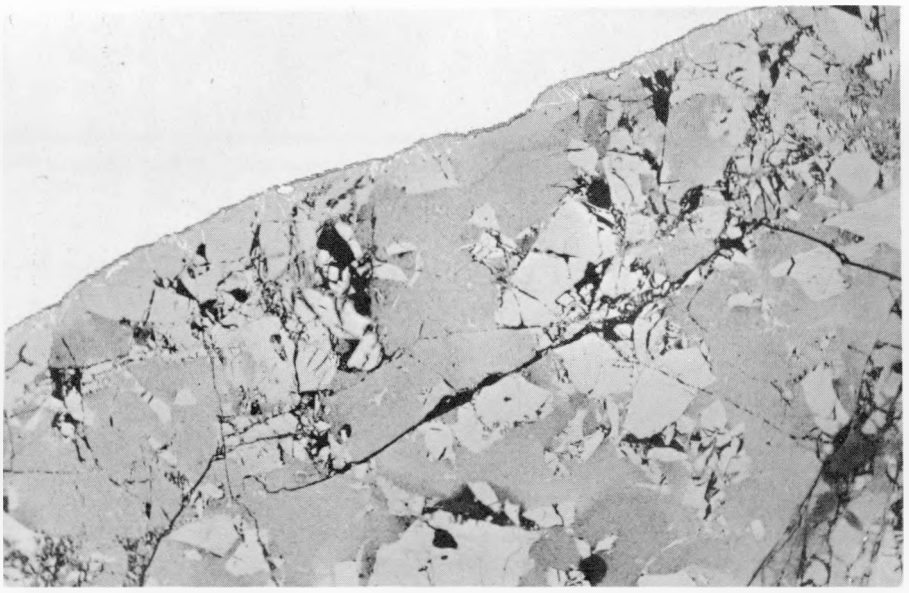

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FIGURE 13. TYPICAL STRUCTURE OF 5.5 PER CENT ENRICHED UO_2 PRESSURE BONDED FOR IRRADIATION IN THE VBWR

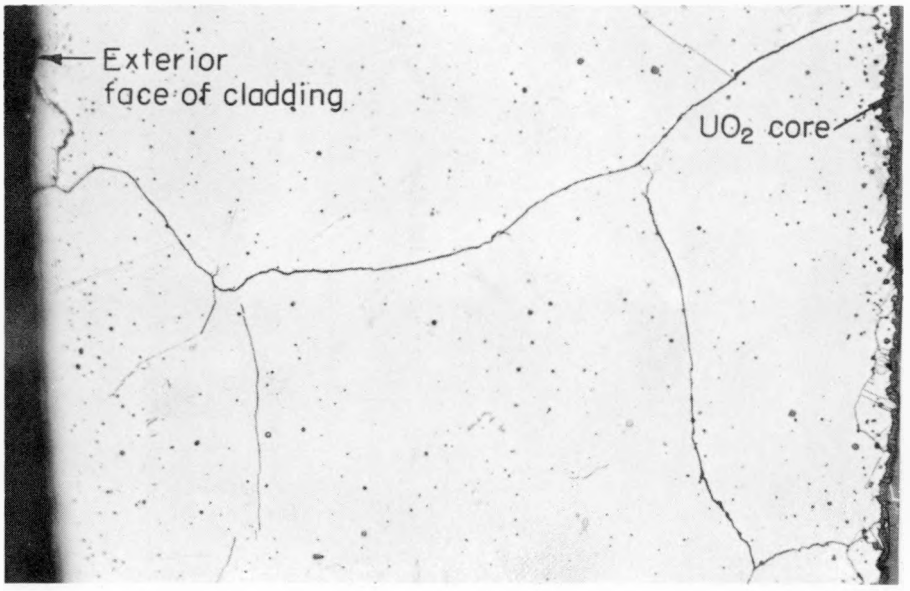

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FIGURE 14. TYPE 304L STAINLESS STEEL CLADDING AFTER PRESSURE BONDING FOR CYCLES OF 3 AND 2 HR AT 2100 F AND 10,000 PSI

corrosion tested for 48 hr in 750 F steam demonstrated a weight change of from 2.2760 to 2.2763 g and no abnormal surface effects. The structure of the cladding after corrosion is shown in Figure 15, where a uniform exterior surface is evident. Examination of representative indented areas in the final rod surface revealed no thinning of the cladding. A comparison 24-in. fuel-rod segment previously fabricated with depleted oxide materials was thermal cycled 50 times between room temperature and 1000 F. Inspection of this rod at various intervals and at the end of the 50 cycles revealed no growth or change in surface conditions.

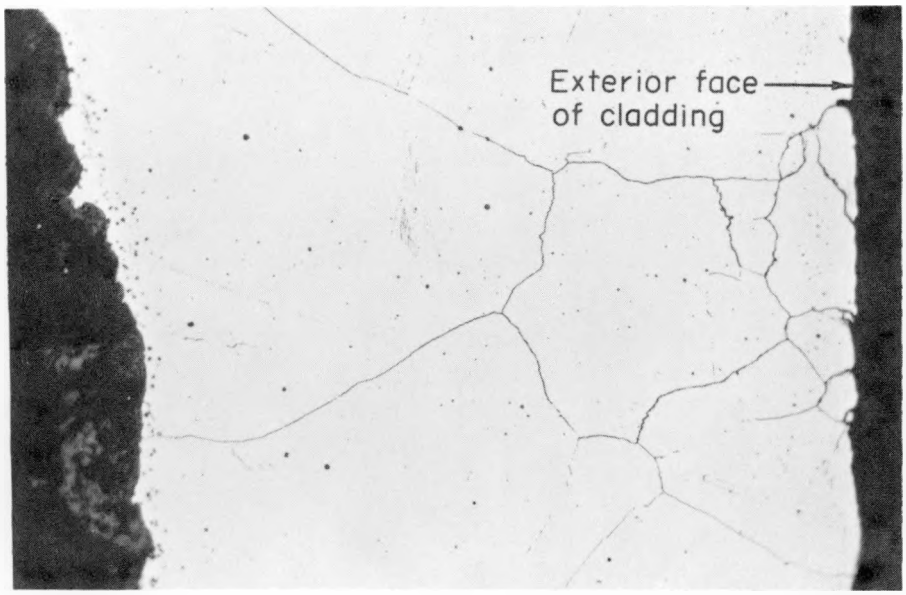

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FIGURE 15. PRESSURE-BONDED TYPE 304L STAINLESS STEEL AFTER CORROSION TESTING FOR 48 HR IN 750 F STEAM

Dimensional data for the four fuel-rod assemblies supplied to GE for irradiation testing of the gas-pressure bonded UO_2 are given in Table 18. The total rod lengths were maintained uniform; however, there were slight variations in the fuel-segment lengths. The four gas-pressure-bonded fuel-rod assemblies are shown in Figure 16.

In view of the initial goals of this phase of the program, the fuel rods fabricated for VBWR irradiation are not considered optimum. The process as employed in this case utilized the cold pressing of pellets, which is an inherently expensive operation. The use of these pellets led to slight surface irregularities in the final bonded assemblies because of chipped cores. It is believed that these problems could be overcome by the use of the vibration packing technique as the fuel-loading method. The rods, however, will provide a good test for pressure-bonded UO_2 under irradiation. The rods as supplied contain a high-density oxide, processed at temperatures below those of conventional sintering, with an intimate contact between the core and cladding. The general specifications for a fuel rod designed to contain pelletized fuel have been met by the pressure-bonding route.

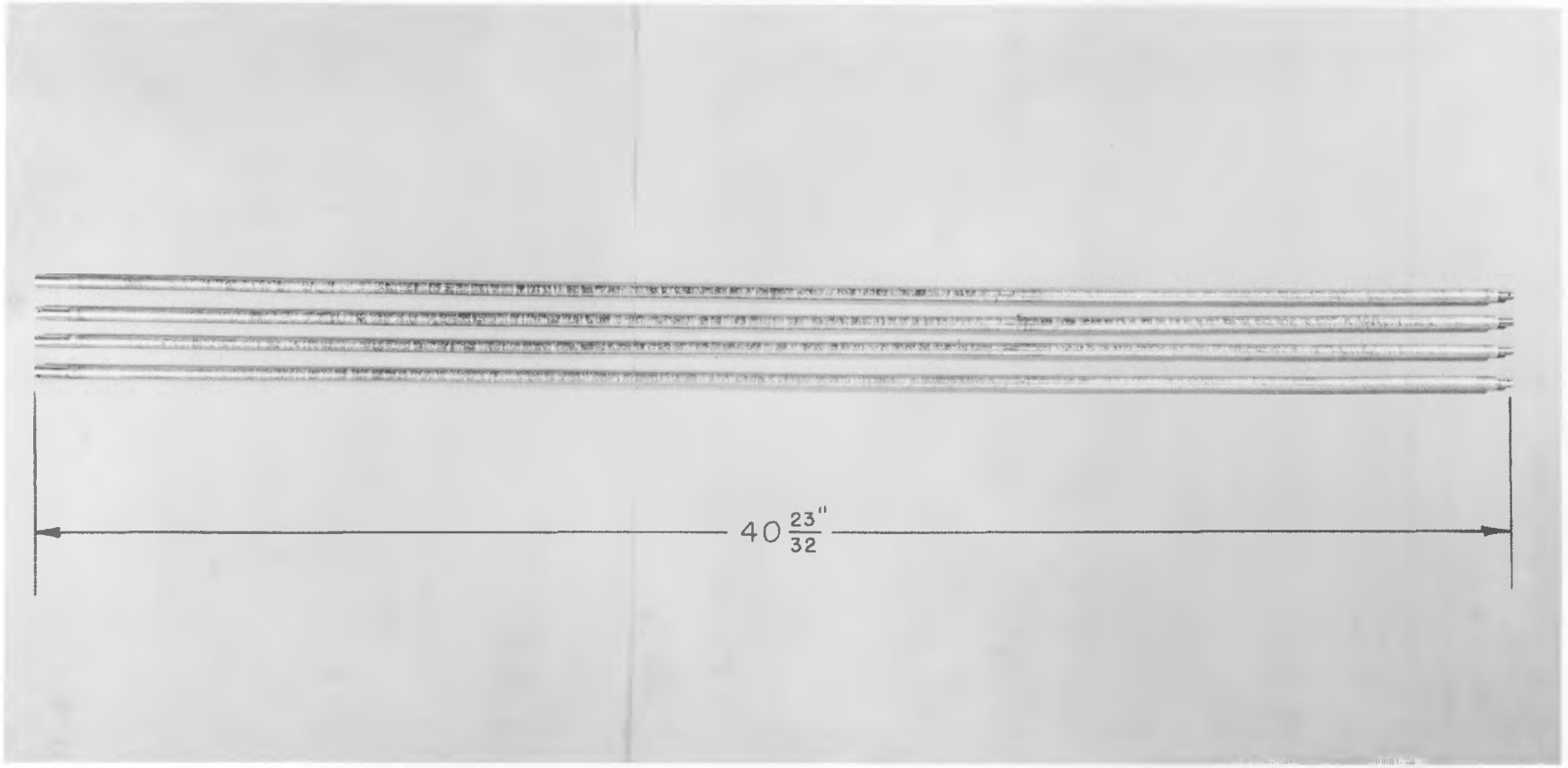

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FIGURE 16. FOUR GAS-PRESSURE-BONDED VBWR FUEL-ROD ASSEMBLIES

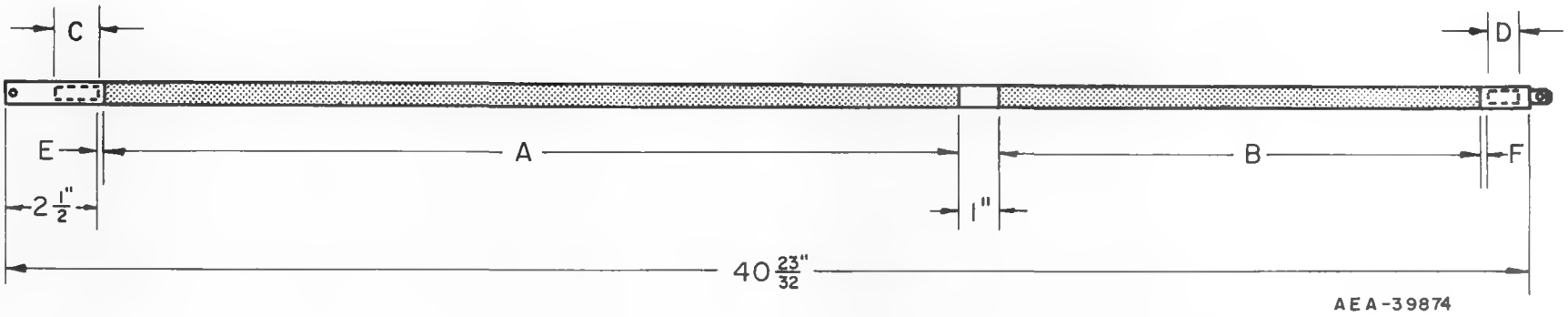

TABLE 18. FINAL DIMENSIONS ON FOUR PRESSURE-BONDED UO_2 FUEL RODS FABRICATED FOR VBWR IRRADIATION PROGRAM

Rod	Average[a] Diameter Over A, in.	Average[a] Diameter Over B, in.	Core Length, A, in.	Core Length, B, in.	Total Length of Core, in.	Total Core Weight, g	Plenum[b] Length, C, in.	Plenum[b] Length, D, in.	Spacer[c] Thickness, E, in.	Spacer[c] Thickness, F, in.
1	0.3965	0.3965	24-3/8	11-11/16	36-1/16	636	1-3/16	41/64	1/16	1/16
2	0.397	0.397	24-5/16	11-1/2	35-13/16	634	1-13/64	31/64	3/64	3/64
3	0.3975	0.397	24-7/32	12-1/16	36-9/32	640	1-3/16	33/64	1/16	1/16
4	0.3975	0.397	24-5/32	12-1/8	36-9/32	642	1-3/16	33/64	1/16	3/64

(a) Average cladding thickness is 0.017 in.
(b) Average Plenum wall thickness is 0.037 in.
(c) Plenum spacers contain a 0.062-in.-diameter hole.

CONCLUSIONS

Although the originally intended work for this phase of the program was not completed in order to fabricate the irradiation test rods, the following conclusions can be drawn:

(1) Flat-plate compartmented assemblies as fabricated by gas-pressure bonding exhibit close dimensional control and high bond integrity.

(2) Fuel rods can be fabricated by the gas-pressure-bonding technique utilizing cold-pressed UO_2 cores. Four fuel rods were successfully fabricated for irradiation in the VBWR. However, this method is not considered optimum with respect to the cost and the final characteristics of the fuel rod. It is recommended that a combined vibration packing-pressure bonding approach be investigated.

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