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

Phase I Report on AEC Fuel-Cycle Program

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Basic properties of several types and grades of commercial UO_2 have been determined. Compacting characteristics of these powders were evaluated with the objective of obtaining a minimum cold-pressed density of 70 per cent of theoretical prior to gas-pressure bonding. Fused and special dense grades of UO_2 powders were compacted to a density of 85 per cent of theoretical by use of a 50-tsi compacting pressure.

Cold-pressed compacts were simultaneously clad and densified to a maximum density of 99.5 per cent of theoretical. The UO_2 powders that were capable of being pressed to the highest cold-pressed density exhibited the least amount of densification during gas-pressure bonding.

A small stainless steel-clad UO_2 flat-plate assembly was pressure bonded at 2100 F for 3 hr at 10,000 psi. Examination of this assembly indicates that it is feasible to prepare fuel assemblies of this type in a one-step operation by use of the gas-pressure-bonding process.

INTRODUCTION

The Fuel-Cycle Development Program of the U. S. Atomic Energy Commission has been instituted to develop, advance, and improve fuel cycles for power reactors. In research and development in support of this program, Battelle is exploring the application of gas-pressure bonding to reduce manufacturing costs for fuel elements. This report discusses the progress made in Phase I of this study.

The feasibility of this process has been fully demonstrated in the preparation of control, fuel, and moderator elements and assemblies.[1-3] This study will be concerned with the refinement and further development of the gas-pressure-bonding process for the simultaneous densification and cladding with stainless steel of ceramic, cermet, and dispersion fuels. The process has many advantages not possessed by other conventional fabrication techniques now being used: (1) complex shapes can be produced with a minimum of additional development, (2) much lower fabrication costs seem possible, (3) close dimensional control can be achieved, (4) simultaneous densification and cladding of such materials as uranium dioxide can be achieved, and (5) it is adaptable to any core system.

(1) References at end.

Description of the Program

The work will include fabrication development for several basic fuel-element designs. The development work is to be conducted to the point that preliminary process specifications will be established, so that only a minimum amount of additional development work will be needed to undertake production. The additional work will be only that necessary to adapt the process to a specific fuel element.

The fabrication studies will be primarily concerned with the development of uranium dioxide fuel material clad with Type 304 stainless steel. Uranium dioxide was chosen because it is being considered by more power-reactor programs than any other material and because the general results that are obtained with this material should also be applicable to other ceramic fuels. Stainless steel cladding was selected because it is a readily available and low-cost material which is compatible with most gas and liquid-metal coolants and with most fuels. It is also currently being considered as cladding in several of the civilian power-reactor programs.

Description of the Process

Gas-pressure bonding utilizes gas pressure at elevated temperatures for the direct densification, cladding, and joining of reactor components. The components to be bonded are fabricated to final size, cleaned, and then assembled in a metallic container. The container is evacuated, sealed, placed inside a high-pressure-gas autoclave, and subjected to a high external gas pressure at an elevated temperature. A temperature above the recrystallization temperature of the components that are being bonded is used so that the external pressure is transmitted through the container with sufficient force to plastically deform all of the components and bring them into intimate contact.

The autoclaves are of a relatively simple design.

Research and Development Procedures

The greatest effort in this program will be directed toward the development of Type 304 stainless steel-clad ceramic uranium dioxide fuel elements and assemblies of various shapes. The prime objective throughout the study will be to reduce costs.

Preliminary studies in this program have shown that uranium dioxide can be densified to a high theoretical density by pressure bonding at a high temperature. Studies are being conducted to determine the conditions that are required for achieving any desired density. The effects of the bonding parameters of time, temperature, and pressure and the effect of different grades of uranium dioxide and their physical properties on the densities achieved are being evaluated. This is believed important as the specified density may vary for different applications.

The development of fabrication techniques to produce several basic fuel-element and -assembly shapes will be studied. These shapes will include rod, tube, flat-plate, corrugated-plate, radiator-type and cylindrical assemblies with coolant channels.

A subscale flat-plate assembly has been fabricated. It may be desirable to use compartments for reinforcement or to localize damage and minimize contamination in the event of a cladding rupture, so applications of compartmenting are also being investigated.

The fabricated components are being evaluated to determine fuel density, high-temperature stability, and integrity of cladding and bonds. Pressure tests will be conducted to obtain the cladding rupture-strength data that can be used to calculate allowable fission-gas buildup. Limited studies will also be conducted to obtain values of thermal conductivity and expansion.

A small effort will be directed to the development of uranium dioxide cermet and dispersion systems. The gas-pressure-bonding techniques developed for the ceramic uranium dioxide fuel can be applied to these systems with only a small amount of development work.

INITIAL PHASE OF EXPERIMENTAL PROGRAM

The initial phase of this study was mainly concerned with the controlled densification of ceramic uranium dioxide fuel by gas-pressure bonding. Preliminary studies in Phase I have shown that uranium dioxide can be highly densified by this technique. By the proper selection of oxide and gas-pressure-bonding parameters it is believed that it will be possible to achieve almost any desired density. Investigations have been conducted to determine the effects of the following variables on the ultimate density achieved during gas-pressure bonding: (1) bonding parameters, (2) types of UO_2, (3) UO_2 particle size, and (4) cold-pressed density prior to pressure bonding.

Optimum gas-pressure-bonding parameters of time, temperature, and pressure for obtaining acceptable Type 304 stainless steel bonds were developed.

Also investigated were various bonding-container configurations and methods of loading them. A subsize stainless steel-clad UO_2 flat-plate subassembly was pressure bonded to demonstrate process applicability to production operations.

Preparation and Evaluation of UO_2 Core Powders

Basic Properties of Commercial UO_2 Powders

Seven types of uranium dioxide have been studied. These powders include a Mallinckrodt ceramic grade, a Mallinckrodt dense ceramic grade, a Mallinckrodt high-fired grade, a Mallinckrodt special dense grade, a Mallinckrodt spherical grade, a NUMEC high-fired grade, and a Spencer fused grade.

The ceramic grade is a very fluffy compound which is not easily green pressed to a high density. The dense ceramic grade is referred to as Type S-4 oxide by MCW.

It has been reported that it can be densified with very high pressure and not show appreciable cracking. Densities between 63 and 67 per cent of theoretical (7.0 and 7.3 g per cm^3) have been realized by MCW using pressures of 75 to 100 tsi.

The high-fired grades and the spherical grade are primarily designed for use in matrix-type fuel elements. High-fired is prepared from ceramic grade UO_2 and contains approximately 0.3 w/o TiO_2 as an active internal binder. The spherical UO_2 is formed into shot and then fired in much the same way as high fired but does not contain an additive. In producing these bodies a temperature of 1700 C has been used.

The special dense UO_2 is a swaging grade of UO_2. This relatively new material is similar to high-fired UO_2, but has a wide particle-size distribution and contains no TiO_2. The fused grade of UO_2 from Spencer Chemical Company is made by fusing the UO_2 in a modified arc in air.

Many of the properties of the powders have been studied and physical values were measured for each type.

In Table 1 the screen analyses of the UO_2 powders are listed. These analyses were obtained on as-received batches from commercial vendors. The apparent or top densities of these UO_2 powders were measured and are reported in Table 2. As would be expected from the sieve analyses in Table 1, the Spencer fused UO_2 and MCW special dense UO_2 appeared to have the highest apparent density. The physical properties of each, however, greatly influence the final density. Table 3 gives an insight of the microstructure of the various oxide powders as determined by preliminary petrographic examination. Further investigations along this line will include a direct count of the porosity, and the determination of crystal size.

Evaluation of Specimen-Charging Methods

Green Compacting. The compacting characteristics of these powders will be evaluated on the basis of their particle shape, size, and distribution, their porosity and their method of preparation. In Table 4, the results of the cold-compacting studies of 1/2-in.-diameter pellets are listed with the compacting pressure varied between 20 and 55 tsi. Below 20 tsi, the green strength of the pellets was not adequate to permit either rapid ejection from the die or subsequent handling during assembly. Above 55 tsi, it is thought that the process would become uneconomical because of excessive die wear and breakage.

In Figure 1, the data obtained with various commercial powders are plotted as a function of density and compacting pressure. The superiority of several of the UO_2 grades from the standpoint of densification during compacting is quite apparent.

The UO_2 powders were mixed for 1 hr to insure uniformity of size of particles. Then 0.5 w/o binder was added to the powder mixture and was mixed for 2 hr in a Kelly V-type blender. These powders were then placed into a die and pressed at a given pressure. Die walls were lubricated with camphor-alcohol solutions. The compacts were then assembled into the tubular pressure-bonding container, and the container was welded. The container was degassed through an evacuation tube at 1112 F until a constant vacuum was obtained. The container was then evacuated to a pressure of 5×10^{-3} mm of mercury or less for 2 hr, and the evacuation tube was sealed.

TABLE 1. SIEVE ANALYSIS OF UO_2 POWDERS AS RECEIVED[a]

Mesh Size	Size Distribution of Type of Uranium Dioxide Powder Shown, w/o			
	MCW Ceramic UO_2	MCW Dense Ceramic UO_2	MCW Special Dense UO_2	Spencer Fused UO_2
+80	--	37.17	58.54	63.43
-80+100	--	8.31	6.12	5.68
-100+140	--	16.98	9.00	8.62
-140+200	--	13.41	5.80	6.10
-200+325	70.58	19.02	8.56	8.25
-325	29.42	5.10	11.98	7.92

(a) NUMEC high-fired UO_2 is minus 325 mesh.
MCW high-fired UO_2 is minus 400 mesh.
MCW spherical is minus 200 plus 325 mesh.

TABLE 2. TAP DENSITIES OF THE VARIOUS URANIUM DIOXIDE POWDERS

Type of Powder	Mesh Size	Tap Density, g per cm^3	Density, per cent of theoretical
MCW ceramic grade	-200	1.75	16.0
MCW dense ceramic grade	-40	3.74	34.1
MCW spherical grade	-200+325	5.72	52.2
MCW high-fired grade	-400	5.93	54.1
MCW special dense grade	-40	6.94	63.3
Spencer fused grade	-20	7.06	64.4

TABLE 3. MICROSCOPIC EXAMINATION OF SIX DIFFERENT SAMPLES OF UO_2 POWDERS AS RECEIVED FROM COMMERCIAL SUPPLIERS

Sample	Visual Volume Phase Composition by Direct-Area-Count Method: Phase Identity	Average, volume per cent	Variation in the Index of Refraction of Phases
Mallinckrodt ceramic grade (minus 200 mesh) UO_2	Extremely fine grained, very finely cryptocrystalline, nodular, grape-cluster type of UO_2 aggregates. The absence of crystal-structure development and the presence of a uniform, oolitic, granular particle structure was noteworthy. Between three and seven tiny grains were observed in each oolite. The average grain size was approximately 0.8μ in diameter.	99.5 plus	Isotropic $N = 2.34_{Li} \pm 0.01_{Li}$ $N > 2.33_{Li}$ but below 2.35_{Li}
	Scattered traces of impurities. No enriching secondary phase appeared to be present.	Traces	--
Mallinckrodt dense ceramic grade UO_2	Isotropic, coarsely crystalline grains and particles of UO_2 phase. This appeared to be a high-temperature calcined type of UO_2. It did not appear to be an electric-furnace type.	87.7	Isotropic $N = 2.34_{Li} \pm 0.01_{Li}$
	Isotropic, tiny "pin points" of a secondary, bonding phase appeared to be present. The tiny pin points appeared to be uniformly distributed as inclusions through the particles and grains of UO_2 phase. The secondary phase was probably glass in structure.	12.3	Isotropic $N > 2.35_{Li}$
Mallinckrodt high-fired grade (minus 400 mesh) UO_2	Isotropic, coarsely crystalline grains and particles of UO_2 phase. This appeared to be a high-temperature calcined UO_2 powder. It was not an electric-furnace type of UO_2 powder. Many of the grains were broken portions of a single crystal.	92.3	Isotropic $N = 2.34_{Li} \pm 0.01_{Li}$
	Black, opaque, interstitial stringers of a secondary, cementing phase appeared to be present. No birefringence was observed in the secondary-phase portion.	7.7	Black, opaque, in transmitted light
NUMEC high-fired grade (minus 325 mesh) UO_2	Isotropic, coarsely crystalline grains and particles of UO_2 phase. Temperature of calcination was high enough to produce anhedral crystal grains. Rectangular shapes, polygon shapes, and octahedrons were present. This appeared to be a calcined high-temperature type of UO_2 powder. It was not an electric-furnace product.	95.4	Isotropic $N = 2.34_{Li} \pm 0.01_{Li}$
	Tiny "pin points" of a secondary, cementing phase were present. They were translucent in transmitted light and uniformly dispersed through the aggregates. The secondary phase was probably glass in structure.	4.6	Isotropic $N > 2.35_{Li}$
Mallinckrodt special dense grade UO_2	Isotropic, microcrystalline type of crystalline UO_2 powder. The internal microstructure was better developed than a cryptocrystalline product, but not as well developed as a coarsely crystalline product.	89.1	Isotropic $N = 2.34_{Li} \pm 0.01_{Li}$
	Isotropic, "pin points" of a secondary cementing phase were present. A few black, opaque, inclusions of secondary phase were also present. The secondary phase appeared to be uniformly distributed. No birefringence was seen.	10.9	Isotropic $N > 2.35_{Li}$

TABLE 3. (Continued)

Sample	Visual Volume Phase Composition by Direct-Area-Count Method		Variation in the Index of Refraction of Phases
	Phase Identity	Average, volume per cent	
Mallinckrodt spherical shot (minus 200 plus 325 mesh)	Isotropic, coarsely crystalline grains and coalescent crystalline particles of UO_2 phase were seen. Nodular, grape-cluster type of aggregate particles were also seen. The UO_2 would be classified as a moderately high-temperature calcined product.	90.6	Isotropic $N = 2.34_{Li} \pm 0.01_{Li}$
	Isotropic, tiny, "pin-point" translucent grains and a few angular, black and opaque, inclusions of a secondary phase were present. The secondary phase appeared to be distributed uniformly through the solid-body aggregates.	9.4	Isotropic when translucent; also opaque $N > 2.35_{Li}$

TABLE 4. GREEN DENSITIES OF UO_2 COMPACTS

Binder	Specimen	Pressure, tsi	Height-to-Diameter Ratio	Density	
				G per Cm^3	Per Cent of Theoretical
	Prepared With NUMEC Minus 325-Mesh High-Fired UO_2				
Camphor-alcohol slurry	U-1044	20	0.5	7.40	67.5
	U-1045	25	0.5	7.86	71.8
	U-1046	30	0.5	7.84	71.5
	U-1047	35	0.5	7.96	72.9
	U-1048	40	0.5	8.14	74.3
	U-1049	50	0.5	8.06	73.6
Ceremul "C"	U-1052	20	0.5	7.71	70.4
	U-1053	25	0.5	7.80	71.3
	U-1054	30	0.5	7.90	72.1
	U-1055	35	0.5	8.07	73.7
	U-1056	40	0.5	8.05	73.5
	U-1057	50	0.5	8.12	74.1
	U-1058	20	1	7.72	70.5
	U-1059	25	1	7.85	71.7
	U-1060	30	1	8.00	73.1
	U-1061	35	1	8.07	73.7
	U-1062	40	1	8.15	74.4
	U-1063	50	1	8.22	75.0
	Prepared With MCW As-Received Depleted Special Dense Grade UO_2				
Ceremul "C"	U-1094	20	1	8.33	76.0
	U-1093	25	1	8.52	77.8
	U-1092	30	1	8.60	78.5
	U-1089	35	1	8.64	78.8
	U-1091	40	1	8.75	80.0
	U-1090	50	1	9.34	85.3
Camphor-alcohol slurry	U-1095	20	1	7.78	71.0
	U-1096	25	1	8.29	75.6
	U-1097	30	1	8.35	76.3
	U-1098	35	1	8.48	77.4
	U-1099	40	1	8.53	77.9
	U-1100	50	1	8.67	79.1
	U-1101	55	1	8.75	80.0
	Prepared With MCW Depleted Dense Ceramic Grade UO_2				
Camphor-alcohol slurry	U-1102	20	1	6.17	56.3
	U-1103	25	1	6.31	57.6
	U-1104	30	1	6.38	58.2
	U-1105	35	1	6.49	59.2
	U-1106	40	1	6.59	60.2
	Prepared With MCW Minus 400-Mesh High-Fired UO_2				
Camphor-alcohol slurry	U-1107	20	1	7.54	68.8
	U-1108	25	1	8.04	73.3
	U-1109	30	1	7.89	72.0
	U-1110	35	1	8.05	73.5
	U-1111	40	1	8.11	74.0
	U-1112	50	1	8.29	75.6
	U-1113	55	1	8.34	76.1

TABLE 4. (Continued)

Binder	Specimen	Pressure, tsi	Height-to-Diameter Ratio	Density	
				G per Cm^3	Per Cent of Theoretical
	Prepared With MCW Depleted Special Dense Grade UO_2				
Ceremul "C"	U-1114	20	1	8.33	76.0
	U-1115	25	1	8.44	77.0
	U-1116	30	1	8.54	78.0
	U-1117	35	1	8.70	79.4
	U-1118	40	1	8.75	79.9
	U-1119	50	1	9.36	85.5
	U-1120	55	1	9.47	86.4
	Prepared With MCW Depleted Dense Ceramic Grade UO_2				
Ceremul "C"	U-1121	20	1	6.08	55.5
	U-1122	25	1	6.24	56.9
	U-1123	30	1	6.33	57.8
	U-1124	35	1	6.38	59.3
	Prepared With MCW Minus 400-Mesh High-Fired UO_2				
Ceremul "C"	U-1127	25	1	7.93	72.4
	U-1128	30	1	8.04	73.4
	U-1129	35	1	8.14	74.2
	U-1130	40	1	8.15	74.4
	U-1131	50	1	8.34	76.1
	Prepared With Spencer Fused Minus 20-Mesh UO_2				
Ceremul "C"	U-1158	55	1	9.75	88.9
	U-1159	50	1	9.70	88.6
	U-1160	45	1	9.61	87.9
	U-1161	40	1	9.62	87.9
	U-1162	35	1	9.50	87.2
	U-1163	25	1	9.38	85.6
	Prepared With Spencer Fused Minus 100-Mesh UO_2				
Ceremul "C"	U-1157	55	1	8.85	80.7
	U-1156	50	1	8.82	80.6
	U-1155	45	1	8.68	79.3
	U-1154	40	1	8.64	78.9

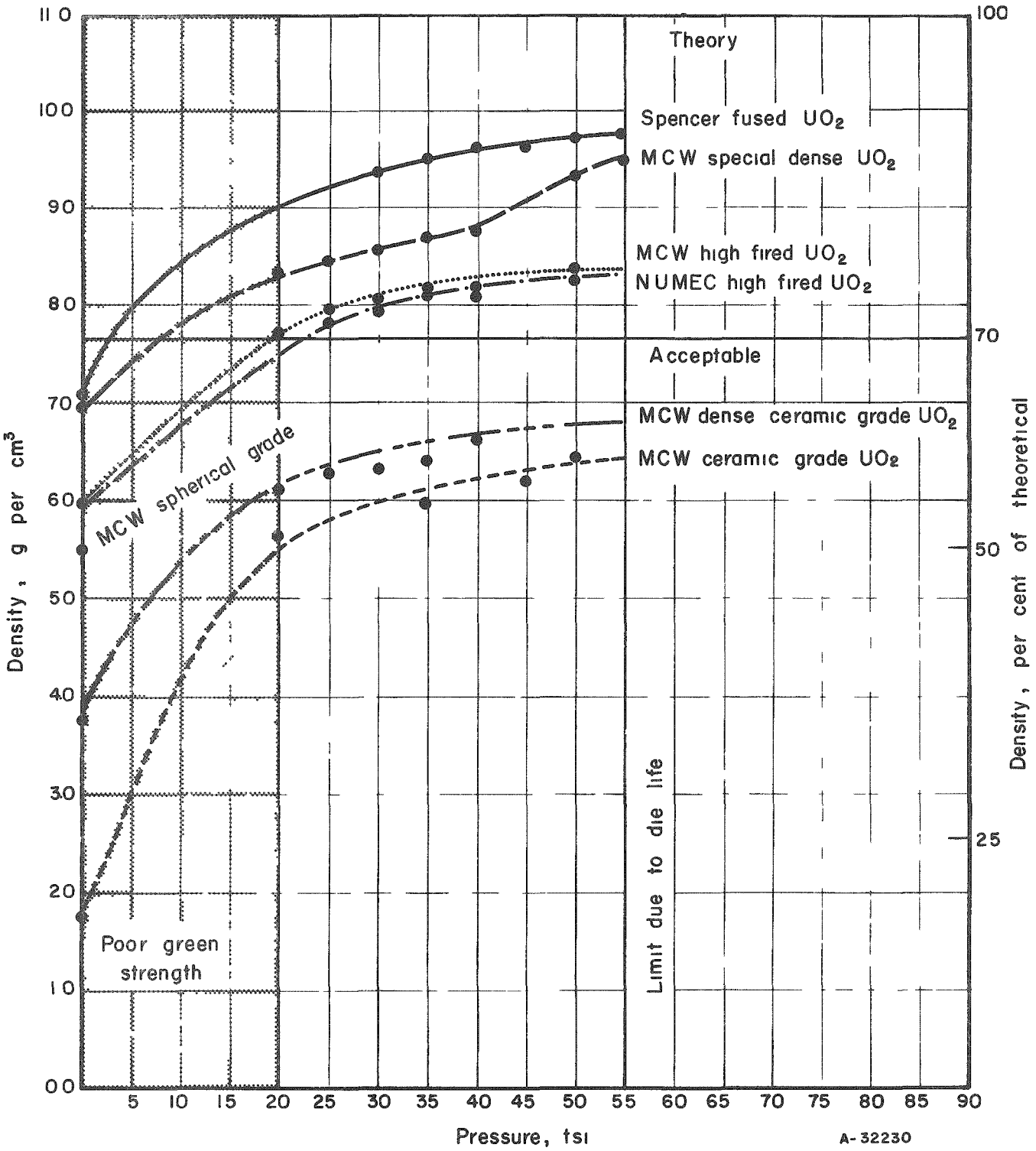


FIGURE 1. EFFECT OF COMPACTING PRESSURE ON GREEN DENSITY OBTAINED WITH VARIOUS UO_2 POWDERS

Compacts produced by various pressures and using various types of UO_2 were selected from those listed in Table 4 on the basis of their initial green density. Those possessing a high density and good green strength were submitted for pressure bonding. These results are tabulated in Table 5.

Tamp-Packing Procedure. To prevent excessive cladding wrinkling during the pressure-bonding operation, an arbitrary minimum cold-pressed density of 70 per cent of theoretical was selected. It is apparent from the tap-packing data as shown in Table 2 that the objective of 70 per cent of theoretical density prior to pressure bonding might be accomplished by loading loose UO_2 powder directly into the bonding container, thereby eliminating the need for cold compacting with binders and subsequent degassing. This would remove two steps from the fabrication procedure and reduce costs. A single tube was filled with MCW special dense UO_2 by adding small amounts of powder and repeatedly tamping and tapping by hand until the tube was completely filled. The UO_2 density was approximately 77 per cent of theoretical. The specimen was then pressure bonded. Radiography demonstrated that this core was uniform throughout, and there were no indications of cleavage or other defects. The resultant core density was 86 per cent of theoretical density. Additional specimens are planned in which selective sizing of particles is used and in which pressure-bonding conditions are varied.

Pressure-Bonding Studies

Pressure-Bonding Experimental Procedures

Gas-Pressure-Bonding Process and Equipment. Gas-pressure bonding, a process utilizing relatively high gas pressures at elevated temperatures, is being utilized for the direct densification and cladding of ceramic, cermet, and dispersion fuels. The components to be bonded are assembled into a container which is evacuated, sealed, and then heated under gas pressure in an autoclave. The differential gas pressure forces the components into the intimate contact necessary for solid-phase diffusion to occur. If the temperature and pressure are sufficiently high, powder-process materials of low density are also densified during the operation.

The components for bonding or densification are fabricated to approximate size. The only deformation that occurs is the amount necessary to bring the components into intimate contact and the amount resulting from densification.

The pressure-bonding equipment utilized in this study is of a relatively simple design but incorporating several unique features. The vessels are sealed at both ends by the use of a modified Bridgman closure. The units have pressure-sealed openings for electrical and thermocouple leads, a pressure inlet, a pressure-control outlet, and a safety-disk assembly. A resistance heater is located within the center of the vessel and ceramic-fiber insulating material is tightly packed around the heater to prevent loss of heat to the autoclave wall. There are three cold-wall high-pressure autoclaves available for this study. Two of these are 9 in. in ID by 4 ft long and the other is 14 in. in ID by 6 ft long.

TABLE 5. PRESSURE-BONDING STUDIES

Type of UO_2 Powder	Binder	Specimen	Compacting Pressure, tsi	Height, in.	Diameter, in.	Height-to-Diameter Ratio	Density, g per cm^3 Green	Density, g per cm^3 Pressure Bonded	Density, per cent of theoretical Green	Density, per cent of theoretical Pressure Bonded	Tube
			Bonded 2 Hr at 2100 F and 10,000 PSI								
MCW ceramic grade UO_2	Ceremul "C"	U-1028	35	0.516	0.539	1	5.97	--	54.5	--	1
		U-1029	45	0.517	0.539	1	6.12	10.9	55.9	99.5	1
			Bonded 2 Hr at 2100 F and 10,000 PSI								
NUMEC high-fired UO_2	Ceremul "C"	U-1058	20	0.432	0.538	1	7.72	--	70.5	--	2
		U-1060	30	0.467	0.538	1	8.00	--	73.1	--	2
		U-1062	40	0.526	0.538	1	8.15	--	74.4	--	2
		U-1063	50	0.489	0.539	1	8.22	10.5	75.0	95.9	2
			Bonded 1 Hr at 2100 F and 10,000 PSI								
NUMEC high-fired UO_2	Camphor-alcohol slurry	U-1064	35	0.445	0.539	1	7.83	--	71.5	--	3
		U-1065	35	0.503	0.539	1	7.89	--	72.0	--	3
		U-1069	35	0.510	0.539	1	7.78	10.4	71.0	95.0	3
NUMEC high-fired UO_2	Ceremul "C"	U-1074	35	0.510	0.539	1	7.79	--	71.1	--	3
		U-1076	35	0.510	0.539	1	7.80	--	71.1	--	3
		U-1078	35	0.505	0.539	1	7.78	--	71.1	--	3
			Bonded 2 Hr at 2200 F and 10,000 PSI								
NUMEC high-fired UO_2	Ceremul "C"	U-1081	35	0.507	0.539	1	7.80	--	71.2	--	4
		U-1082	35	0.505	0.539	1	7.85	10.5	71.7	95.9	4
		U-1083	35	0.506	0.539	1	7.81	--	71.2	--	4
		U-1084	35	0.507	0.539	1	7.80	--	71.2	--	4
		U-1085	35	0.504	0.539	1	7.90	--	72.1	--	4
		U-1086	35	0.500	0.539	1	7.91	--	72.2	--	4
			Bonded 2 Hr at 2150 F and 10,000 PSI								
NUMEC high-fired UO_2	Camphor-alcohol slurry	U-1068	35	0.513	0.539	1	7.78	--	71.0	--	5
		U-1072	35	0.516	0.539	1	7.71	10.4	70.4	95.0	5
		U-1073	35	0.514	0.539	1	7.75	--	70.7	--	5
NUMEC high-fired UO_2	Ceremul "C"	U-1075	35	0.513	0.539	1	7.76	--	70.8	--	5
		U-1077	35	0.513	0.539	1	7.77	10.5	71.0	95.9	5
		U-1079	35	0.513	0.539	1	7.70	--	70.4	--	5
		U-1089	35	0.500	0.539	1	7.94	--	72.5	--	6
			Bonded 2 Hr at 2100 F and 10,000 PSI								
MCW special dense UO_2	Ceremul "C"	U-1120	55	0.424	0.539	1	9.47	10.30	86.4	94.1	7
		U-1090	50	0.431	0.539	1	9.34	10.37	85.3	94.6	7
		U-1091	40	0.470	0.539	1	8.75	10.32	80.0	94.4	7
MCW special dense UO_2	Camphor-alcohol slurry	U-1099	40	0.470	0.539	1	8.53	10.27	77.9	93.6	7
		U-1100	50	0.462	0.539	1	8.67	10.26	79.1	93.5	7
		U-1101	55	0.458	0.539	1	8.75	10.27	80.0	93.6	7
			Bonded 2 Hr at 2100 F and 10,000 PSI								
MCW high-fired UO_2	Ceremul "C"	U-1129	35	0.493	0.539	1	8.14	10.26	74.2	93.5	8
		U-1130	40	0.492	0.539	1	8.15	10.20	74.4	93.2	8
		U-1131	50	0.477	0.539	1	8.34	10.24	76.1	93.5	8
MCW high-fired UO_2	Camphor-alcohol slurry	U-1111	40	0.491	0.539	1	8.11	10.17	74.0	92.8	8
		U-1112	50	0.481	0.539	1	8.29	10.24	75.6	93.5	8
		U-1113	55	0.476	0.540	1	8.34	10.31	76.1	94.2	8

Fabrication of Containers for Pressure Bonding. The gas-pressure-bonding process requires a pressure differential between the inside of the evacuated container and the outside. The container may be of two types. One becomes the cladding for the element or assembly, while the other is removed from the element or assembly after bonding and is discarded. The container must be fabricated of a material that will become plastic at bonding temperature to allow the external pressure to be transmitted uniformly through it to bring the components into intimate contact.

Type 304 stainless steel tubes have been used as a combined container and cladding for all rod-type specimens. These containers consist of 1/2-in.-ID tubes with 10- or 20-mil walls and end plugs 1/8 in. thick. One of these end plugs is drilled to accommodate an evacuation tube. All parts are degreased and first the end plug with the evacuation tube is assembled into the tube. After Heliarc welding this end plug with one diffusion pass and one filler pass, the tube is put through the wash cycle and loaded with pellets or powder. Then the other end plug is welded into place.

Containers for flat-plate specimens were fabricated of Type 304 stainless steel. In cross section these containers resemble a swastika. Other container designs are being studied to achieve the best results for this application. The side plates for these containers were fabricated from 0.018-in.-thick sheet by shearing and forming a 90-deg flange 0.125 in. wide on one edge. The end plugs were sheared from 0.080-in. sheet. A form block slightly larger than the intended compact or assembly size is fabricated of mild steel to close tolerances. The parts are then assembled and clamped into position on the form block after being degreased. They are Heliarc welded in the same manner as the tubes. When the form block is removed, the container is put through a wash cycle. A compact or assembly is then loaded into the container, and the end plug is welded into place.

After loading, all containers are leak tested under water with 50 to 75 psi of helium pressure. Provided there are no leaks, the container is placed on a vacuum system and evacuated to a pressure of 5×10^{-3} mm of mercury, or less, prior to sealing the evacuation tube. Those containers which contain green-pressed cores with a binder are placed into a furnace at 1200 F and degassed for 2 hr or until a constant vacuum is reached. They are then allowed to cool, are evacuated to a pressure of 5×10^{-3} mm of mercury, or less, and sealed.

Cleaning Techniques. After machining the stainless steel components to the desired size, they were pickled in a 10 volume per cent nitric acid-2 volume per cent hydrofluoric acid aqueous solution at a temperature of 120 to 140 F for 2 min, and rinsed in cold running water.

Prior to assembly the metal components and containers were scrubbed with a brush in alcohol, and transferred immediately into a rinse of cold running water. After degreasing, all components were subjected to a series of washing and rinses as outlined below:

(1) Scrubbed in a hot Alconox solution (180 F)

(2) Rinsed in cold running water

(3) Scrubbed in alcohol

(4) Rinsed in cold running water

(5) Rinsed in hot water (200 F)

(6) Dried in air.

Clean rubber gloves were worn during the cleaning, drying, and assembling operations to prevent recontamination of the cleaned surfaces. The components were immediately assembled after cleaning.

Gas-Pressure-Bonding Densification Studies

Fourteen rod-type specimens containing a total of 62 compacts of UO_2 with initial green densities as high as 86 per cent of theoretical have been pressure bonded. The specification for the compacts in eight of these specimens are given in Table 5. The other six specimens all contain compacts of the MCW special dense UO_2 with the Ceremul "C" binder with green densities ranging between 79.6 and 80.4 per cent of theoretical. The specimens are being evaluated to determine the final densification of each compact and dimensional tolerances of each specimen.

The Type 304 stainless steel pressure-bonding containers for these specimens measured 0.580 in. in OD by 0.540 in. in ID, giving a wall thickness of 0.020 in. These tubes were of various lengths, depending upon the number of compacts contained in each. In several of the tubes disks 1/8 in. thick were used to compartmentalize the specimen.

In order to prevent excessive cladding deformation with accompanying wrinkling during the pressure-bonding operation, a minimum cold-pressed density of 70 per cent of theoretical was arbitrarily selected. Many of the initial compacts that were subjected to the pressure-bonding treatment possessed a density that was considerably higher than this minimum value. The results of densification for the initial eight specimens are also shown in Table 5. These results tend to point out, as in conventional sintering operations, that an initial high green density does not always result in the highest fired density. The characteristic of the initial UO_2 powder is a much more important factor.

A plate compact 0.250 by 1.0 by 1.0 in. was placed into a flat-plate swastika-Type 304 stainless steel container and pressure bonded at 2100 F for 2 hr at 10,000 psi. The plate compact was compacted of a high-fired UO_2 powder using a camphor-alcohol binder and had an original green density of 61.13 per cent of theoretical (6.70 g per cm^3). The compact was utilized to check the design of the pressure-bonding container, as well as methods of loading and degassing the compacts. The container was uniformly but excessively deformed. This pointed out the fact that in order to prevent excessive deformation of the elements, a minimum UO_2 cold-pressed density of 70 per cent of theoretical or better will be required. The compact after pressure bonding had a density of 98.9 per cent of theoretical (10.83 g per cm^3).

A single tube specimen was filled with Mallinckrodt special dense grade UO_2 by successive hand tapping and tamping to an approximate 77 per cent of theoretical density. This specimen was pressure bonded at 2100 F for 2 hr at 10,000 psi to determine process feasibility. The final density was 85.8 per cent of theoretical.

Additional rod- and plate-type specimens are being prepared using different types of UO_2 and various particle-size distributions.

Effect of Bonding Parameters on Type 304 Stainless Steel Bonds. In the cladding study the attainment of metallurgical bonding between stainless components is as essential as the densification of the UO_2. A study was conducted to determine the optimum bonding conditions necessary for Type 304 stainless steel.

Cylindrical specimens consisting of two 1/2-in.-diameter by 1-3/4-in.-long Type 304 stainless steel rods assembled into a 0.020-in.-wall stainless steel tube, as shown in Figure 2, were pressure bonded at different bonding parameters. Two distinct types of specimens were bonded at the same conditions. In one, the rods were butted directly together in the tube; in the other, two 0.005-in. disks of as-rolled Type 304 stainless steel were placed between the two rods. This gave a comparison of machined surface versus as-rolled surfaces.

The components were placed through a standard wash cycle after being pickled in an aqueous solution consisting of 10 volume per cent nitric acid and 2 volume per cent hydrofluoric acid. They were then assembled and the tubes were evacuated and sealed.

After the gas-pressure-bonding cycle, each bonded assembly was machined into a standard 1/4-in.-round tensile-test specimen. The specimens were tested to failure to obtain the ultimate strength. The tensile-test results as a function of bonding parameters and surface conditions are given in Table 6. A metallographic examination was performed on all of the test specimens. Typical photomicrographs of the two types of specimens are shown in Figures 3 and 4.

Conclusions drawn from this study are as follows:

(1) Strong metallurgical bonds were achieved in 3 hr at 2000 F, 1-1/2 hr at 2100 F, or 3 hr at 2100 F at 10,000 psi.

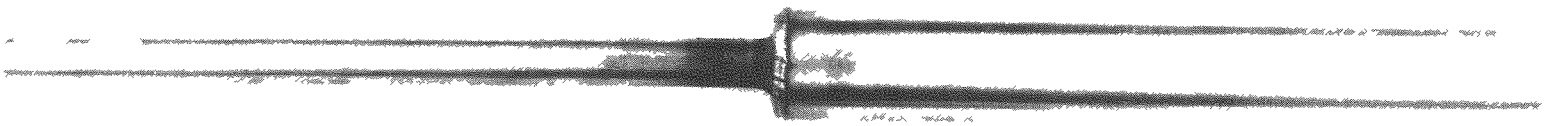
(2) It appears that further refinement of the surface-preparation techniques may be required for obtaining strong consistent stainless steel bonds.

(3) The ultimate load compared favorably with both machined and as-rolled surfaces. However, parting at or near the bond line occurred more often with the as-rolled surfaces.

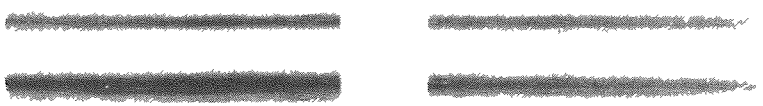
(4) The ultimate load for the starting material and pressure-bonded material compares favorably.

Development of a Flat-Plate Assembly

A cursory investigation was conducted to demonstrate the feasibility of pressure bonding a flat-plate stainless steel-clad UO_2 assembly. Components were machined from as-rolled Type 304 stainless steel and Ti-Namel sheet as shown in Figure 5. The cover plates were fabricated from 10-mil sheet, the spacers, center spacers, frame ends, and frame sides were fabricated from 40-mil sheet, and the side supports were



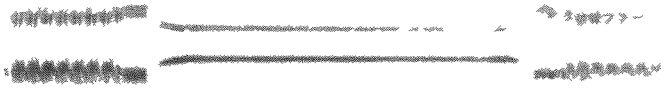
Stainless steel
pressure-bonding tube



Type 304 stainless steel
$\frac{1}{2}$-in.-diameter by $1\frac{3}{4}$-in.-long rods

Stainless steel spacer

Stainless steel end plug



$\frac{1}{4}$-in. tensile specimen

Specimen 2 after fracture

1X

N58906

16

FIGURE 2. ASSEMBLY OF COMPONENTS FOR TYPE 304 STAINLESS STEEL BOND TEST TENSILE SPECIMENS AND SPECIMEN 2 AFTER TESTING

Specimen 2 was pressure bonded at 2100 F for 3 hr at 10,000 psi.

TABLE 6. TENSILE DATA FOR PRESSURE-BONDED TYPE 304 STAINLESS STEEL BOND-TEST SPECIMENS

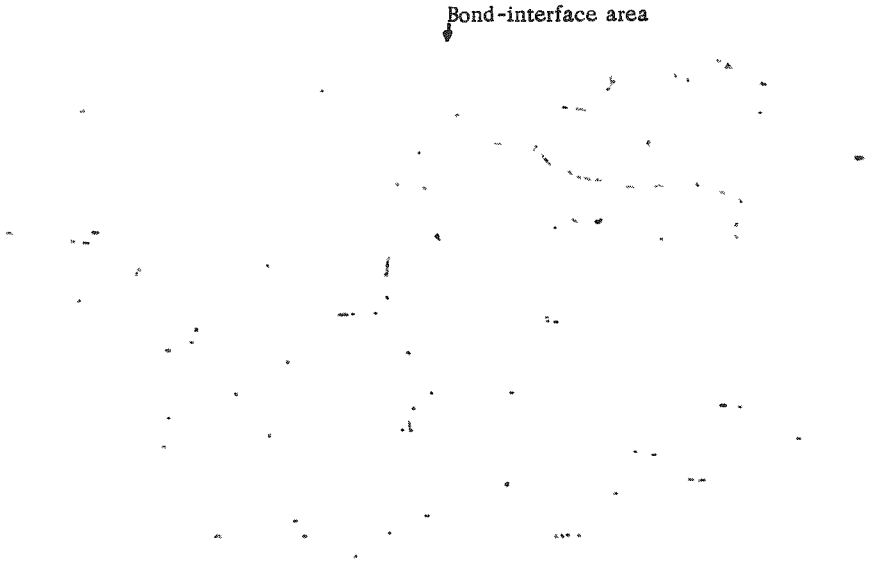
Specimen	Type and Condition	Pressure-Bonding Conditions			Approximate Yield Point, lb	Ultimate Load, lb	Comments
		Time, hr	Temperature, F	Pressure, psi			
1	Machined from Type 304 stainless steel stock	--	--	--	2000	4460	Used as a comparison for the pressure-bonded specimens
2	Butted rods with rms surfaces of 115-135	3	2100	10,000	1700	4100	Specimen necked down and broke at a point away from the bond; complete grain growth, as shown in Figure 3
3	Butted rods with rms surfaces of 110-130	3	2100	10,000	1700	4100	Specimen necked down and broke at a point away from the bond; complete grain growth
4	Butted rods with rms surfaces of 120-150	1-1/2	2100	10,000	1700	4160	Specimen necked down and broke at a point away from the bond; complete grain growth
5	Machined from Type 304 stainless steel stock after the pressure-bonding cycle	1-1/2	2100	10,000	1700	4100	Used as a comparison for the bond-test specimens
6	Butted rods with rms surfaces of 30-40 with two 0.005-in. disks in between	3	2000	10,000	2000	4240	Specimen did not neck down all the way but parted between the disk and rod on one end; pieces of the disk were pulled away leaving a jagged surface; complete grain growth across the disks and 70 per cent grain growth across the rod-to-disk interface, as shown in Figure 4
7	Butted rods with rms surfaces of 50-60 with two 0.005-in. disks in between	3	1900	10,000	1620	3500	Specimen did not neck down all the way but parted between the disk and rod on one end; no metal was torn from either surface; 10 per cent grain between the disks
8	Butted rods with rms surfaces of 5-60	3	1900	10,000	1600	3750	Specimen did not neck down all the way but parted at the bond interface; small pieces of metal were torn from both surfaces
9	Butted rods with rms surfaces of 50-60	3	2000	5,000	1520	3640	Specimen did not neck down all the way but parted at the bond interface; only small pieces of metal were torn from both surfaces
10	Butted rods with rms surfaces of 50-60 with two 0.005-in. disks in between	3	2000	5,000	1450	3700	Specimen did not neck down all the way and parted between the disks; there appeared to be no metal torn from either surface; approximately 30 per cent grain growth between the disks and rods

TABLE 6. (Continued)

Specimen	Type and Condition	Pressure-Bonding Conditions: Time, hr	Temperature, F	Pressure, psi	Approximate Yield Point, lb	Ultimate Load, lb	Comments
11	Butted rods with rms surfaces of 50-60	1	2100	5,000	1420	3800	Specimen parted at the bond interface and did not neck down all the way; no apparent metal torn from either surface
12	Butted rods with rms surfaces of 30-40 with two 0.005-in. disks in between	1	2100	5,000	1444	3440	Specimen did not neck down all the way but parted at the bond interface between the disks; no apparent metal torn from either surface; approximately 10 per cent grain growth between the disks and rods
13	Butted rods with rms surfaces of 30-40	1	2100	10,000	1580	4060	Specimen necked down all the way and broke at a point away from the bond; approximately 90 per cent grain growth
14	Butted rods with rms surfaces of 30-40 with two 0.005-in. disks in between	1	2100	10,000	1630	4100	Specimen did not neck down all the way but parted at the bond interface between the disk and rod on one end; metal pulled away from both surfaces, leaving a jagged face; approximately 80 to 90 per cent grain growth between the disks and 50 per cent grain growth between the disks and rods
15	Butted rods with rms surfaces of 30-40	1	2200	5,000	1620	4120	Specimen necked down all the way but broke at the bond interface; large chunks of metal were torn from both surfaces
16	Butted rods with rms surfaces of 30-40 with two 0.005-in. disks in between	1	2200	5,000	1520	4170	Specimen necked down part way and parted at the bond interface between the disks; metal pulled away from both surfaces, leaving a jagged surface; approximately 80 per cent grain growth between the disks and rods
17	Butted rods with rms surfaces of 30-40	3	2100	5,000	1600	4100	Specimen necked down all the way and broke at a point away from the bond; approximately 95 per cent grain growth
18	Butted rods with rms surfaces of 30-40 with two 0.005-in. disks in between	3	2100	5,000	1680	4090	Specimen did not neck down all the way but broke at the bond interface between the disks; metal was pulled from both surfaces, leaving a jagged surface; approximately 95 per cent grain growth between the rods and disks

TABLE 6. (Continued)

Specimen	Type and Condition	Pressure-Bonding Conditions: Time, hr	Temperature, F	Pressure, psi	Approximate Yield Point, lb	Ultimate Load, lb	Comments
19	Butted rods with rms surfaces of 30-40	1/2	2200	10,000	1560	4090	Specimen did not neck down all the way but parted at the bond interface; small pieces of metal were pulled from both surfaces
20	Butted rods with rms surfaces of 30-40 with two 0.005-in. disks in between	1/2	2200	10,000	1580	4070	Specimen did not neck down all the way but parted at the bond interface between the disks; small pieces of metal were pulled from both surfaces; approximately 30 per cent grain growth between the disks and rods
21	Butted rods with rms surfaces of 20-30	3	2200	2,000	1430	3840	Specimen necked down all the way and broke at a point away from the bond; complete grain growth
22	Butted rods with rms surfaces of 40-50 and 30-40 with two 0.005-in. disks in between	3	2200	2,000	1480	3870	Specimen necked down all the way and broke at a point away from the bond; complete grain growth



100X RM12972

FIGURE 3. BOND INTERFACE OBTAINED WITH AS-MACHINED TYPE 304 STAINLESS SURFACES AFTER TENSILE TEST

Specimen 2, which contained two butted rods with surfaces of 115 to 135 rms, is shown here. The assembly was gas-pressure bonded 3 hr at 2100 F and 10,000 psi.

100X RM13324

FIGURE 4. BOND INTERFACE OBTAINED WITH TYPE 304 STAINLESS SURFACES AS ROLLED AND MACHINED AFTER TENSILE TEST

Specimen 6, which contained two butted rods with surfaces of 30 to 40 rms with two 0.005-in. disks in between, is shown in the photomicrograph. The assembly was gas-pressure bonded 3 hr at 2000 F and 10,000 psi.

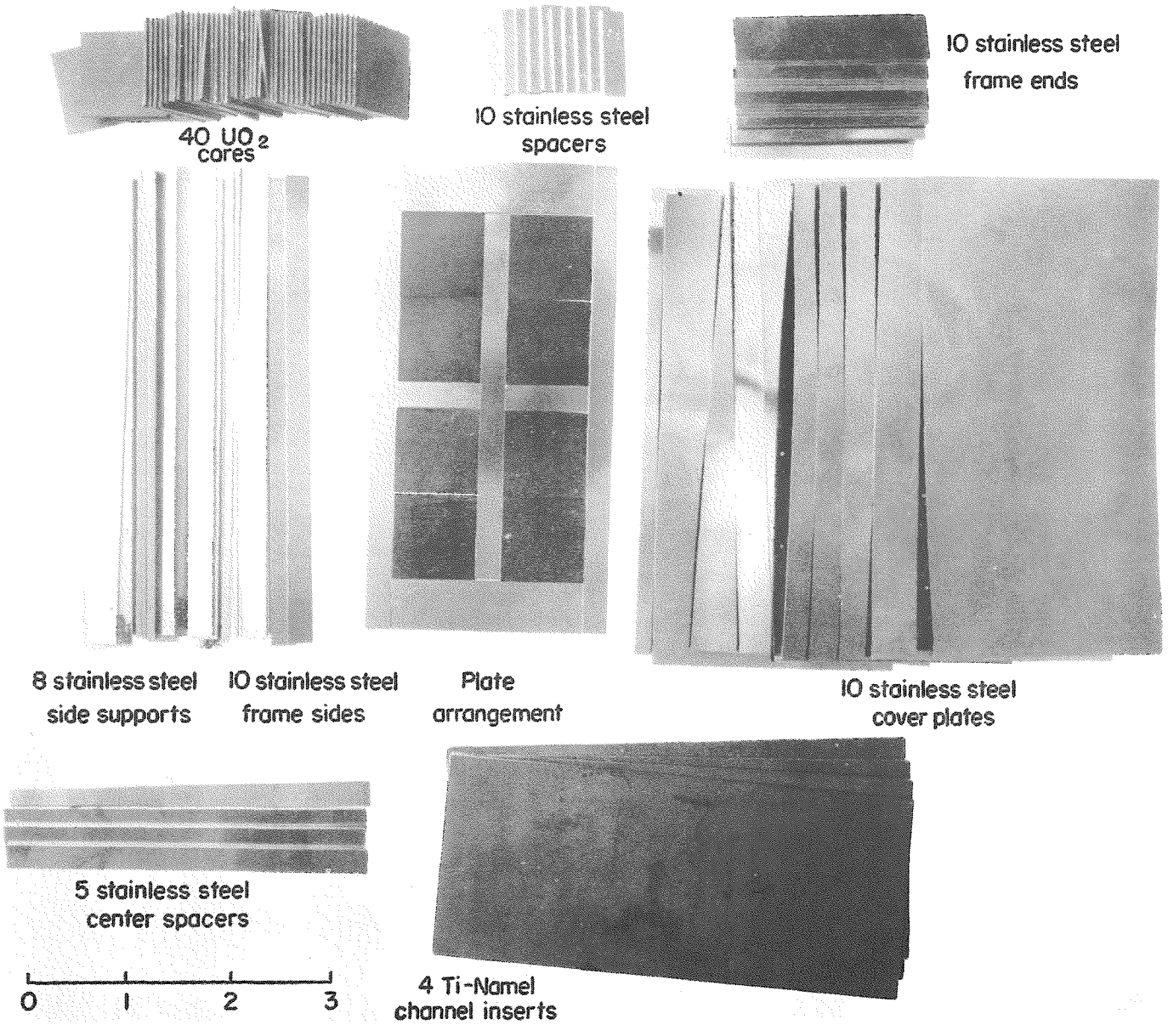


FIGURE 5. ASSEMBLY OF COMPONENTS FOR THE FLAT-PLATE STAINLESS STEEL-CLAD UO_2 ASSEMBLY

fabricated from 75-mil sheet. The Ti-Namel channel inserts were also machined from 75-mil sheet. The UO_2 cores were 0.040 by 0.8305 by 0.8305 in. and possessed a high initial density.

The metal components were put through the standard pickle and wash cycle and then assembled into a Ti-Namel pressure container. The container was evacuated and sealed. Bonding conditions for the assembly were at 2100 F for 3 hr at 10,000 psi. After bonding, the Ti-Namel was dissolved by nitric acid pickling leaving the clad assembly with five plates and four coolant channels. The subassembly was sectioned for examination. A section of the subassembly is shown in Figure 6.

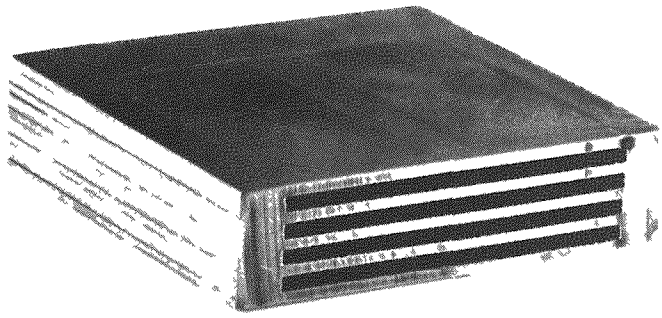


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FIGURE 6. HALF SECTION OF THE 0.600 BY 2.411 BY 4.572-IN. FLAT-PLATE STAINLESS STEEL-CLAD-UO_2 ASSEMBLY CONTAINING FIVE FUEL PLATES

The dark area in the upper right corner is a discoloration produced during cutting.

Examination of this subassembly revealed that such assemblies can be prepared with excellent dimensional control. The bonds were fair but a general evaluation indicated that with further development the pressure-bonding process can be utilized for the bonding of stainless steel-clad UO_2 flat-plate assemblies in a one-step operation.

DISCUSSION AND EVALUATION OF RESULTS

An evaluation of the results obtained during this phase of the program conclusively proves that UO_2 fuel can be simultaneously clad and densified by use of the gas-pressure-bonding process. With additional development the process has potential of becoming the most economic and versatile process for the preparation of stainless-clad high-density UO_2 fuel elements and assemblies.

Uranium dioxide powder was readily cold pressed to densities above 70 per cent of theoretical. Special dense and fused grades of UO_2 were pressed to densities above 80 per cent with the use of relatively low compacting pressures. However, the most significant result involved the attainment of a density above 70 per cent of theoretical by the direct charging and hand tamping of untreated UO_2 powders. This is an extremely low-cost operation that is amenable to automation. There is also an indication that low-cost commercial powders can be utilized without pretreatment for activation or for combination with binder materials.

Several types and grades of oxide were gas-pressure bonded to a density approximately 95 per cent of theoretical. In all instances the highest density was not achieved with the oxide powders capable of being compacted to the highest cold-pressed density. Additional study is required to determine the effects of variables such as particle size, stoichiometry, and green density on the ability of the powders to flow and densify during gas-pressure bonding.

FUTURE WORK

During Phase II of this program studies will be conducted to develop conditions necessary for controlling the UO_2 density. The investigations concerned with relating the type of UO_2 powder, stoichiometry, and particle-size distribution to the cold-pressed density and ultimate pressure-bonded density will be continued. It is anticipated that, at the conclusion of this phase of work, the specifications for UO_2 powder to achieve a desired density will be established.

A constant effort will be maintained to reduce costs. Methods of fabricating UO_2 by the elimination of cold compacting, mixing, binders, die lubricants, and degasification will be investigated. These will include centrifuging and packing by use of air pressure and vibration.

Fabrication of several different basic fuel-element shapes incorporating UO_2 fuel will be undertaken. In conjunction with this work effort will be directed toward developing techniques to accomplish the following:

(1) Providing new and improved container designs

(2) Improving the surface-preparation procedure for stainless components

(3) Optimizing dimensional control

(4) Improving methods for charging of the specimens.

The formulation of process specifications will be developed for these basic elements as the studies progress. The primary objectives of the program, developing fuel-element production techniques that give a superior product at reduced cost, will be the controlling criteria in evaluating the progress of this work.

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