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Grain Growth Behavior of DOP-26 Iridium Clad Vent Set Cups Used in Radioisotope Thermoelectric Generators

April 2002

C. G. McKamey, E. H. Lee, G. B. Ulrich, J. L. Wright, and E. P. George

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RADIOISOTOPE POWER SYSTEMS PROGRAM

GRAIN GROWTH BEHAVIOR OF DOP-26 IRIDIUM CLAD VENT SET CUPS USED IN RADIOISOTOPE THERMOELECTRIC GENERATORS

C. G. McKamey, E. H. Lee, G. B. Ulrich, J. L. Wright, and E. P. George

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GRAIN GROWTH BEHAVIOR OF DOP-26 IRIDIUM CLAD VENT SET CUPS USED IN RADIOISOTOPE THERMOELECTRIC GENERATORS*

C. G. McKamey, E. H. Lee, G. B. Ulrich, J. L. Wright, and E. P. George

ABSTRACT

This report summarizes results of studies conducted to date under the Iridium Alloy Characterization and Development subtask of the Radioisotope Power System Materials Production and Technology Program to characterize the grain growth characteristics of DOP-26 iridium clad vent set cups produced at the Y-12 plant and at Oak Ridge National Laboratory (ORNL). These cups are currently used by the National Aeronautics and Space Administration (NASA) for cladding and post-impact containment of the radioactive fuel in radioisotope thermoelectric generator (RTG) heat sources which provide electric power for interplanetary spacecraft. The conclusions from this study are: (1) Grain growth rates in the clad vent set cups are very low for temperatures below 1400-1500°C. (2) The grain growth characteristics of clad vent set cups produced at the Y-12 plant appear to be the same as the blanks from which the cups were fabricated. (3) The grain growth characteristics of qualification-production clad vent set cups produced at ORNL are similar to those produced at the Y-12 plant and to the blanks from which the cups were fabricated. (4) Non-homogeneous grain growth was observed in two ORNL cups produced during pilot production. However, this type of grain growth was not observed in several qualification-production and production-maintenance cups and is therefore believed to have been a one-time event that should not occur in future flight-quality cups. (5) It was shown that small strains (less than ~2.5% tensile strain or ~1% bending strain) produced in the cups during fabrication that are not relieved by annealing can result in non-homogeneous grain growth upon subsequent annealing for 100 h at 1500°C. (6) Specimens from DOP-26 blanks were bent to strains of 2.5 and 5% and then subjected to heat treatments that were intended to simulate cup fabrication, shelf storage for one month, life in space for three months, and reentry into the earth's atmosphere. Analyses of these specimens showed that significant grain growth occurred only in the bend region of the specimen strained to 5%. (7) An equation to describe grain growth in DOP-26 iridium was determined and used to predict the grain size of the F-5 generator which has been in storage at Mound Laboratories for approximately 15 years. It was found that, if the F-5 generator has stayed below ~960°C, then little or no grain growth should have occurred.

INTRODUCTION

Iridium-based alloys have been developed at Oak Ridge National Laboratory (ORNL) for applications in radioisotope thermoelectric generators (RTG) [1-3]. The currently used alloy (designated DOP-26) is Ir+0.3wt% W to which 60 wppm thorium has been added to improve the alloy's high-temperature

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impact ductility [1]. Auger electron spectroscopy has indicated that thorium segregates strongly to the grain boundaries in this alloy [4,5], resulting in increased cohesive strength of the boundaries and a change in fracture mode from intergranular in the undoped alloy to ductile transgranular fracture in the thorium-doped alloy. In addition to segregating to grain boundaries, thorium in excess of the solubility limit (which may be as low as 5-10 wppm [6]) forms a second phase with a composition of Ir_5Th [5,7-9], which pins the grain boundaries and results in grain size refinement. Together these two factors contribute to an improvement in the high-temperature tensile impact ductility [1]. A recent review paper summarizes the effects of alloying elements on the physical metallurgy and mechanical properties of iridium [10].

Since grain size is an important factor in determining high temperature impact ductility [5], several studies have been completed in recent years to determine the grain growth characteristics of old- and new-process DOP-26 material [3,11], in both vacuum and low-pressure oxygen atmospheres [11-13]. All of these studies were conducted on samples taken from blanks (0.65 mm thick, 52 mm diameter) or rolled sheets and were performed as a function of time and temperature.

The DOP–26 blanks are used to manufacture clad vent set (CVS) cups (see Fig. 1) which contain the fuel for the RTG. Cups for the Cassini mission to Saturn were produced at the Y-12 Plant in Oak Ridge. In 1996 production was moved to ORNL and pilot production of the first set of ORNL cups began in 1997. Because grain size is such an important factor in obtaining good impact ductility, it was considered important to determine the grain growth characteristics also of the completed cups (and not just the blanks). Grain size was measured as a function of the heat treating conditions and position in the cup with respect to both the rolling direction and the cup geometry. These studies have been conducted on cups produced at both the Y-12 Plant and ORNL. The purpose of this report is to consolidate all of the data on the grain growth behavior of clad vent set cups produced at both Y-12 and ORNL and to compare their grain growth characteristics to that of the DOP-26 iridium sheet material from which the cups were fabricated.

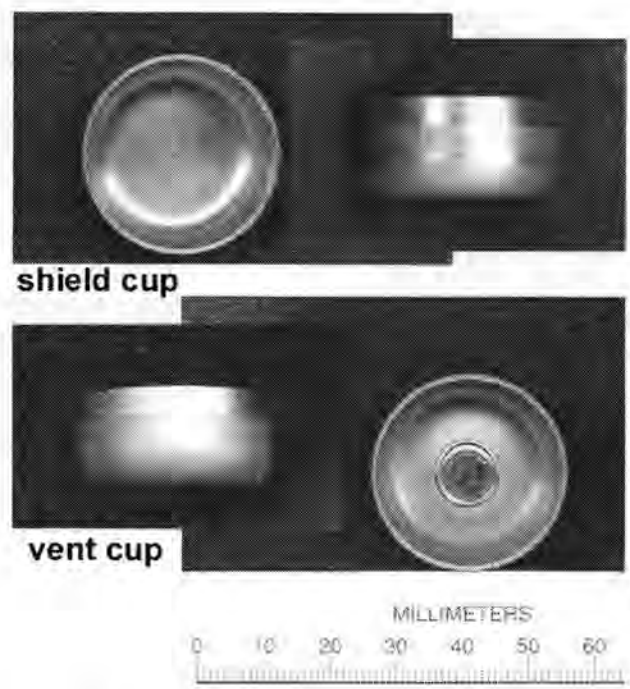


Fig. 1. Photograph of iridium clad vent set cups used for containment of fuel in radioisotope thermoelectric generator power sources: (a) shield cup, (b) vent cup.

EXPERIMENTAL PROCEDURES

The specimens for this study were cut from clad vent set cups which had previously been designated as noted in Table I. All of these cups were fabricated either at ORNL or the

Table I. Details of Clad Vent Set Cups and Heat Treatments Used for Grain Growth Studies

Cup/Segment Identification Number[a]	Heat Identification	Heat treatment for grain growth study	Orientation with respect to RD
3624-01-2783 -- P	ER8	1h/1200-1800°C	0°,45°,90°
3624-00-1805 -- P	D2	1h/1200-1800°C	0°,45°,90°
3624-30-2045 -- P	E4	time/1400°C	0°,45°,90°
3625-00-2409 -- P	D2	time/1500°C	0°,45°,90°
3625-92-0036[b] -- QP	G1	As recrystallized	0°
3625-91-0033[b] -- QP	G1	As recrystallized	90°
3625-NC-5011 -- T for QP	D1	1h/1200-1800°C	0°,45°,90°
9753-NC-0005 -- PP	D2	time/1400°C	0°,45°,90°
9753-NC-0004 -- PP	CR4	time/1500°C	0°,45°,90°
9753-30-4000 -- QP	AR2	100h/1500°C	0°
9754-02-A5NV -- QP	G1	100h/1500°C	0°
3624-05-4013 -- PM	G2	100h/1500°C	crease, 180° to crease
3625-05-5026 -- PM	G2	100h/1500°C	crease, 180° to crease
3625-91-0053[c] -- PM	G2	As recrystallized	crease
TC33 -- T for PM	RS11	100h/1500°C	0°,90°
TC38 -- T for PM	RS11	100h/1500°C	0°,90°

[a]P=Production at Y-12, all others were fabricated at ORNL; QP=qualification production; PP=pilot production; PM=production maintenance; T=training.
[b]Segments from cup 3625-03-A5NT.
[c]Segments from cup 3625-05-5022.

Oak Ridge Y-12 Plant according to approved procedures from full-size ingots produced using the "new process" [3,11,14]. Cup forming begins with 52-mm diameter, 0.65-mm thick DOP-26 iridium blanks that have been electrical discharge machined (EDM) from rolled sheet, ground, electrolytically cleaned in KCN, vacuum stress relieved, acid cleaned, and sandwiched between two tantalum barrier discs. The sandwiches are then encapsulated between two 304L stainless steel sheets with an electron beam circumferential closure weld. This "blank assembly" is deep drawn at 925°C in a two draw operation. The stainless steel and tantalum are chemically stripped from the formed iridium cup with solutions of $3HCl:1HNO_3$ and HF/HNO_3, respectively. Once the cups have been stripped and fully acid cleaned, they are annealed for 1 h at 1375°C in vacuum in order to achieve full recrystallization. The cups are sized to the required contour dimensions and then a number of detailed mechanical features are fabricated, resulting in two different kinds of cups, the shield cup (Fig. 1a) and the vent cup (Fig. 1b). The mechanical features include a thin weld shield on the inside of the shield cup (not shown in Fig. 1a) and a frit vent and decontamination cover (as shown in Fig. 2) in the bottom of the vent cup (Fig. 1b). Each cup is 100% dye penetrant inspected. The outer surfaces of the cups excluding the weld zones are grit blasted for functional (emissivity) purposes and then each cup is acid cleaned in heated solutions of $3HF:7HNO_3$,

$3HCl:1HNO_3$, and HNO_3. In some cases, individual cups may require resizing or must otherwise be reworked to meet specifications. Table II lists any extra forming treatments that were done to the cups used in this study and the annealing treatments that each received during fabrication. The air burn off (ABO) and vacuum outgas (VO) heat treatments were performed after the final electron beam weld assembly operations required to attach the weld shields and frit vent/ decontamination cover assemblies to the shield and vent cups, respectively. For those produced at ORNL, cups from pilot production, qualification production, training, and production maintenance were included in this study.

The first two sets of cups listed in Table I were used to determine the grain growth behavior as a function of time

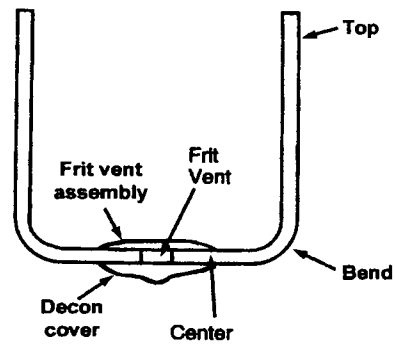


Fig. 2. Schematic drawing (not to scale) of a vent cup showing the top, bend, and center regions where grain sizes were measured. Also shown are the locations of the frit vent assembly and decontamination cover.

Table II. As-Received Condition of CVS Cups Used for Grain Growth Studies

Cup ID#	Contained frit vent or decon cover weld	Reworking	Heat treatment received prior to grain growth study[a]
3624-01-2783	Yes	No	RX,ABO-2,VO-2
3624-00-1805	No	No	RX
3624-30-2045	No	No	RX
3625-00-2409	No	No	RX
3625-92-0036[b]	No	No	RX
3625-91-0033[b]	No	No	RX
3625-NC-5011	No	sized 4X	RX
9753-NC-0005	Yes	sized twice	RX,ABO,VO
9753-NC-0004	Yes	sized twice	RX,ABO,VO
9753-30-4000	Yes	No	RX,ABO,VO-2
9754-02-A5NV	No, weld shield	sized twice	RX,ABO,VO-2
3624-05-4013	No	No	RX
3625-05-5026	No	sized twice	RX,ABO,VO
3625-91-0053[c]	No	sized twice	RX
TC33	No	sized, intentionally distorted, then sized 2nd time	RX,ABO,VO
TC38	No	sized, intentionally distorted, then sized 2nd time	RX

[a]RX= recrystallization (1 h at 1375°C); ABO= air burn-off (4 h at 635°C); VO= vacuum outgas (1 h at 1250°C in vacuum of 1.3 mPa).
[b]Segment from cup 3625-03-A5NT.
[c]Segment from cup 3625-05-5022.

and temperature for cups produced at the Y-12 Plant and ORNL, respectively. Figure 2 shows a schematic (not to scale) of a cup and identifies the areas that were analyzed for grain size. These areas have been designated as top, bend, and center regions. Through visual inspection, the rolling direction (RD) was determined, and the cups were sliced and numbered with respect to the rolling direction according to the schematic diagram shown in Fig. 3. Grain size data were determined not only for the top, bend, and center regions of the cup, but also for specimens cut 0, 45, and 90° to the rolling direction. The stars in Fig. 3 denote the edge of each cut specimen that was mounted and polished for grain size measurements. Specimens were annealed in a vacuum of approximately 10^{-4} Pa for 1 h at temperatures from 1200 to 1800°C and for various times at temperatures of 1400 or 1500°C. After heat treatment, the specimens were mounted, polished, and then electrolytically etched in a solution of 400 ml H_2O, 100 ml HCl, and ~2.5 g NaCl using a current of 0.9 A at an applied potential of approximately 10 V AC for 4-5 min. Grain sizes were measured by the method of linear intercepts in the direction perpendicular to the thickness of the cut specimen. The numbers of intercepts counted are included in parentheses in the tables as a measure of the accuracy of the grain size data.

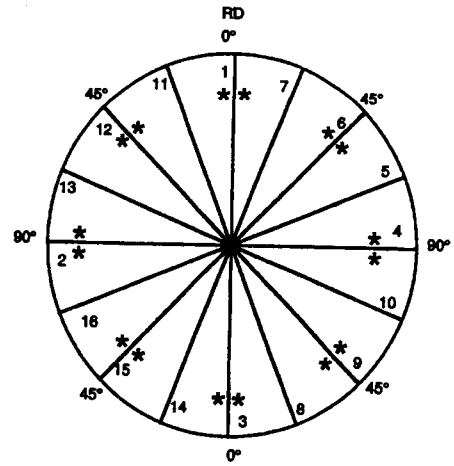


Fig. 3. Schematic drawing showing the sectioning and labeling scheme for clad vent set cup material.

Some characteristics of the grain growth of the ORNL pilot-production cups were not observed in either cups from the Y-12 Plant or in qualification-production cups from ORNL. Heat treating conditions of 100 h at 1500°C in vacuum were chosen for samples taken from these cups. Therefore, ORNL qualification-production cups 9753-30-4000 and 9754-02-A5NV, as well as the TC33 and TC38 cups produced at ORNL for training during production maintenance, were used to further study features observed in the grain growth study described above.

Cups 3624-05-4013, which received only a recrystallization heat treatment of 1 h at 1375°C, and 3625-05-5026, which was recrystallized then given air burnoff (4 h at 635°C) and vacuum outgas (1 h at 1250°C) heat treatments (see Table II), were ORNL production-maintenance cups. Each had a small crease down the side that was produced during the second forming operation when a small spot of dried and hardened lubricant was built up on the die. This built-up spot formed a shallow U-shaped impression in the sidewall of the cup. In the as-formed condition the impression measured approximately 2 mm wide by a maximum of 0.11 mm deep. After recrystallization and sizing the maximum depth was 0.035 mm. These cups were heat treated for 100 h at 1500°C in a vacuum of 10^{-4} Pa. After annealing, a section of each cup (about 1 cm wide) encompassing the crease was cut from the top of the cup, around the bend, and into the flat section at the bottom of the cup. This section was then sliced perpendicularly (as shown by #1-4 in Fig. 4) into 4 pieces which were mounted, polished, and etched for optical metallography as described above. In order to determine the general microstructures and average grain sizes of the cups, a

similar set of samples were cut from the unaffected area of the cups opposite the crease (#5-8 in Fig. 4). Each piece was mounted in such a way as to study the grain size and structure of the edge marked with a star in Fig. 4. In addition, a pie-shaped cup segment, 3625-91-0053, containing a crease or impression was removed from recrystallized cup 3625-05-5022 (not resized). This segment was cut into four smaller pieces and metallography was performed in the same way in order to determine the as-recrystallized grain size.

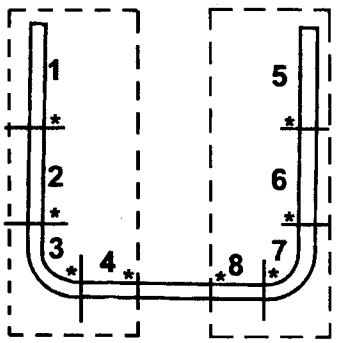


Fig. 4. Schematic drawing (not to scale) showing the areas where grain sizes were measured in the 3624-05-4013 and 3625-05-5026 cups.

For a study of induced strain and its effect on grain growth, specimens were obtained from a DOP-26 blank designated as CR4-9 (fabricated by the "new-process" [11]) which had been recrystallized at 1300°C for 1 h. From this single blank, specimens for the study of tensile, compression, and bending strains were obtained. Specimens for bending were cut with dimensions of approximately 0.7 by 3 by 20-30 mm using EDM. They were bent at room temperature around mandrels with radii of 25.4, 19.05, 12.7, and 6.35 mm to produce bending strains of approximately 1.3, 1.7, 2.5, and 4.9%, respectively. Tensile specimens were machined with a gage size of approximately 9.5 by 0.7 by 1.5 mm and were tensile tested in air at room temperature to strains of 1.75, 2.5, 3.75, and 5% (determined by measuring the specimen before and after tensile straining). Sheet specimens measuring approximately 0.7 by 7 by 20 mm were cut using EDM and rolled at room temperature to strains of 1.8, 2.7, 3.85, and 5.6%. Additional specimens were cut from blank AR2-9 (received after only a stress relief anneal of 1 h at 900°C) and bent to produce approximately 2.5 and 5% strain. The specimens from blank AR2-9 were given the same thermal treatments as are given to CVS cups during fabrication, i.e., recrystallization at 1375°C for 1 h, an air-burnoff at 635°C for 4 h, and a vacuum outgas for 1 h at 1250°C. They were then heat treated for one month at 950°C (in an argon-filled quartz tube) to simulate a short shelf life after fuel has been loaded plus three months at 1330°C to simulate short-term operation in space. One each of the 2.5 and 5% bend-strained specimens were also heat treated to simulate reentry from orbit into the earth's atmosphere [15]. A thermocouple was attached and they were heated to 1300°C and held for a few minutes to reach equilibrium. Then they were heated to 1580°C at a rate of 85°/min, held for 3 minutes, cooled back to 1300°C at a rate of 38°/minute, and then furnace cooled to room temperature. After heat treatment, the specimens were mounted, polished, and etched for optical metallography as described above.

Using grain size data from DOP-26 blanks [11] and from two plutonia-fueled clads that have been shelf-aged for 50,000 h at Los Alamos National Laboratory, an equation for grain growth in DOP-26 was determined and used to predict the grain size of the F-5 generator which currently has been in storage at Mound Laboratories with full fuel complement for approximately 15 years.

RESULTS AND DISCUSSION

AS-RECEIVED MICROSTRUCTURE

Table III shows the starting grain sizes for DOP-26 cups measured perpendicular to the wall thickness of the cups. Grain sizes at the top, bend, and center regions were all comparable for both the Y-12- and ORNL-produced cups and ranged from 20 to 26 μm in diameter. Figure 5 shows the typical as-received microstructure (in this case from ORNL qualification production cups 3625-92-0036 and 3625-91-0033). Specimens cut along the rolling direction (0° to RD) showed a slight elongation of the grains along the rolling direction (Fig. 5a), while those cut at 45 or 90° to the rolling direction (Fig. 5b) were more equiaxed. These micrographs are typical of the as-received cups produced both at Y-12 and ORNL. As noted above, all cups were received in the as-recrystallized condition, so Fig. 5 represents the as-recrystallized microstructure of all the cups used in these studies

CUPS PRODUCED AT THE Y-12 PLANT

Table IV and Figure 6 show the variation in grain size as a function of heat treatment temperature for specimens taken from the top, bend, and center regions of the cups produced at Y-12. Data for specimens cut at 0, 45, and 90° to the rolling direction (RD) are included in each plot. Also included in each plot is the grain growth data determined earlier [11] for specimens taken from a blank fabricated from the D2 ingot (new process) of the DOP-26 alloy. For 1-h anneals, grain growth was not observed at temperatures below approximately 1600°C. At 1500°C the average grain diameter was about 30 μm for all specimens. Grain growth began at 1600°C and the growth rate increased dramatically at higher temperatures. The larger amount of scatter in the data for heat treatment temperatures above 1600°C is attributed to the larger grain sizes at these high temperatures. Larger grains mean that there are fewer grains through the thickness of the specimen for counting, and therefore statistically more scatter in the data. Within the expected experimental error of the grain-size data, no difference related to the position in the cup could

Table III. Grain Size of As-Recrystallized Clad Vent Set Cups

Produced at	Position	Grain Size[a] (μm)		
		0° to RD	45° to RD	90° to RD
Y-12	top	--	22	--
(cups 3624-30-2045	bend	--	20	--
and 3625-00-2409)	center	--	22	--
ORNL -- Qualification Production	top	20 (399)	--	22 (360)
(cup segments 3625-92-0036	bend	23 (345)	--	26 (316)
and 3625-91-0033)	center	23 (346)	--	22 (359)

[a]Numbers in parentheses indicate number of intercepts counted for grain size determinations.

be determined. Likewise, no difference could be detected between the grain growth characteristics of the two cups used for this study (cups 3624-01-2783 and 3624-00-1805) and the D2 blank (sheet) material which is typical of the sheet material from which cups were deep-drawn.

Table V and Figures 7-8 show the variation in grain size as a function of annealing time at 1400 (cup 3624-30-2045) and 1500°C (cup 3625-00-2409), respectively, for specimens taken from the top, bend, and center regions of cups produced at Y-12. Data for specimens which were cut 0, 45, and 90° to the rolling direction (RD) are included in each plot. Also included for comparison are grain growth data for specimens taken from sheets of the D2 ingot heat treated at the same temperatures [11]. Examination of the data plotted in Figs. 7 and 8 shows that, within the expected experimental error, no difference in grain size related to the position in the cup with respect to the rolling direction could be determined; grain sizes at 0, 45, and 90° to the rolling direction were comparable at both temperatures. In general, the data show initially a grain size of approximately 20 μm which increased to 40-50 μm after 520 h at 1400°C and to 125-150 μm after 240 h at 1500°C. Figures 9 and 10 show that the grain sizes of specimens taken from the top, bend, and center regions of the cup are also comparable. These results support the conclusions noted above for 1-h heat treatments.

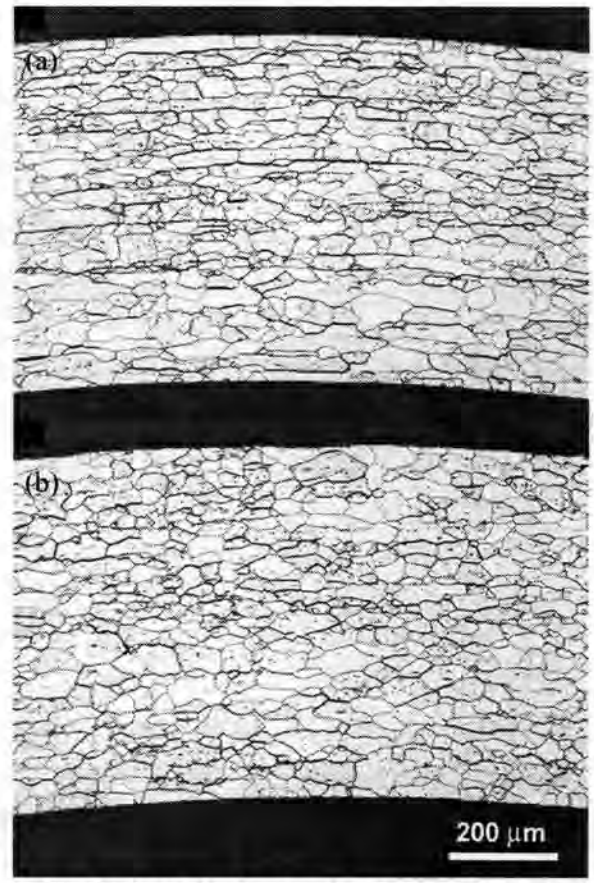


Fig. 5. Optical micrographs showing the typical as-recrystallized microstructure of DOP-26 iridium clad vent set cups for specimens cut (a) parallel (0°) and (b) perpendicular (90°) to the rolling direction. These particular micrographs are of the bend regions of segments 3625-92-0036 and 3625-91-0033, respectively, from ORNL qualification-production cup 3625-03-A5NT.

A comparison of data in Figs. 8 and 10 to the data reported earlier for D2 sheet material [11] shows that cup material has approximately the same grain growth behavior as the sheet material at 1500°C. Since the earlier data for sheet material goes only to 100 h annealing time, it is not possible to determine if the grain growth rate for longer times at 1500°C was the same for the cup material versus the sheet material. The data at 1400°C (Figs. 7 and 9) indicates that the grain growth rate at longer times is approximately the same for sheet and cup material, but the initial grain growth rate is much higher for the D2 sheet resulting in larger grain sizes. This difference in grain size for annealing times at 1400°C can be attributed to the difference in cold work present in the cup versus sheet material. The blank sheet material was not recrystallized before the heat treatments for grain growth studies were performed (blank material is typically received after only a stress relief heat treatment of 1 h at 900°C). It therefore con-

Table IV. Dependence of Grain Size on Annealing Temperature and Cup Location in Clad Vent Set Cups Produced at the Y-12 Plant
(all anneals for 1 h at the indicated temperatures)

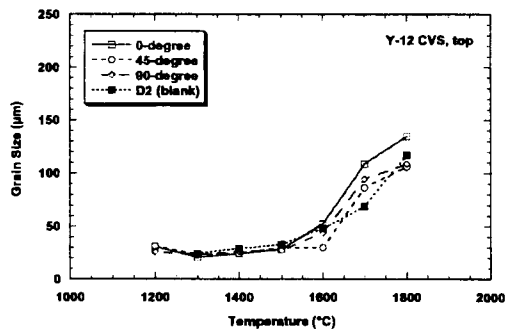
Position	Temperature (°C)	Grain Size (μm)		
		0° to RD	45° to RD	90° to RD
Top	1200	31	31	26
	1300	21	23	23
	1400	24	24	26
	1500	28	30	27
	1600	52	30	44
	1700	108	86	95
	1800	135	106	108
Bend	1200	31	28	22
	1300	25	23	21
	1400	24	24	27
	1500	26	29	27
	1600	29	29	43
	1700	67	64	73
	1800	145	171	93
Center	1200	29	27	25
	1300	23	21	20
	1400	25	23	28
	1500	22	20	22
	1600	42	35	39
	1700	61	76	144
	1800	211	135	175

tained considerable cold work (and retained energy) from the rolling operations and therefore recrystallized easily and grew large grains at 1400°C. The cup material, on the other hand, had already been recrystallized and did not contain enough energy to produce large grains at this temperature. The 1500°C temperature is high enough above the recrystallization temperature that differences in levels of cold work do not affect the grain growth kinetics. Therefore, at this higher temperature, grain sizes of cup material were more comparable to the sheet material. This conclusion is also illustrated in Fig. 11 which shows the large increase in grain growth rate between the 1400 and 1500°C anneals.

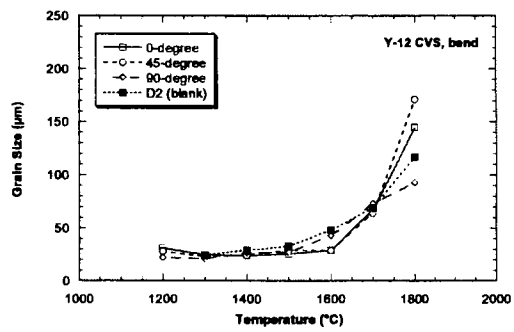
CUPS PRODUCED AT ORNL

Table VI and Figure 12 show the variation in grain size as a function of heat treatment temperature for specimens taken from the top, bend, and center regions of cup 3625-NC-5011 produced for training during qualification production at ORNL. Data for specimens which were cut at 0, 45, and 90° to the rolling direction are included in each plot. Also included in each plot is the grain size data determined

(a)



(b)



(c)

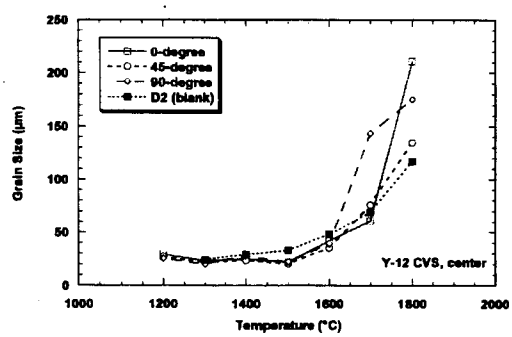
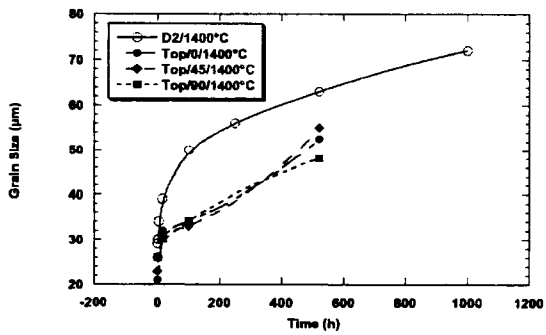
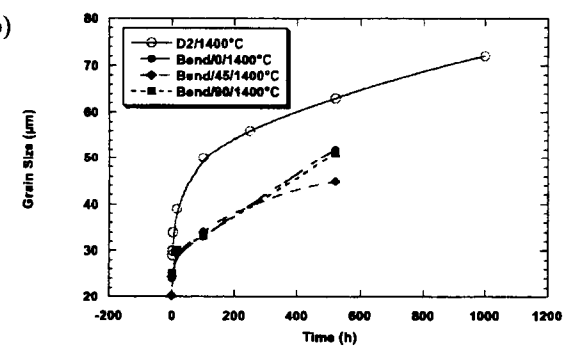


Fig. 6. Effect of heat treatment temperature on grain size near the (a) top, (b) bend, and (c) center regions of clad vent set cups produced at the Y-12 Plant. Data for specimens cut 0, 45, and 90° to the rolling direction are included. For comparison, data for D2 sheet material [11] are also included.

(a)



(b)



(c)

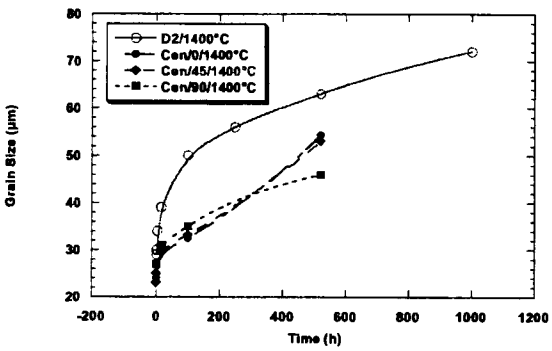


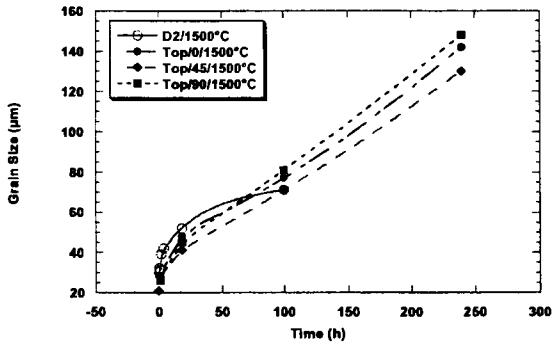
Fig. 7. Effect of annealing time at 1400°C on grain size near the (a) top, (b) bend, and (c) center regions of clad vent set cup 3624-30-2045 produced at the Y-12 Plant. Data for specimens cut 0, 45, and 90° to the rolling direction are included. For comparison, data for D2 sheet material [11] are also included.

Table V. Average Grain Size as a Function of Location in the Cup and Time of Heat Treatment at 1400 and 1500°C for Clad Vent Set Cups Produced at the Y-12 Plant

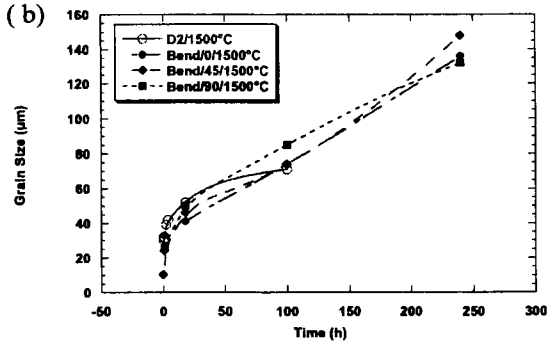
Position	Temperature (°C)	Length of Anneal (h)	Grain Size (μm)		
			0° to RD	45° to RD	90° to RD
Top	1400	0	--	23	--
		1	21	26	26
		18	32	31	30
		100	34	33	34
		520	52	55	48
	1500	0	--	21	--
		1	28	28	26
		18	48	41	45
		100	77	71	81
		240	142	130	148
Bend	1400	0	--	20	--
		1	24	24	25
		18	29	30	30
		100	33	34	33
		520	52	45	51
	1500	0	--	20	--
		1	33	24	26
		18	41	46	50
		100	74	74	85
		240	136	148	132
Center	1400	0	--	23	--
		1	24	25	27
		18	30	30	31
		100	32	33	35
		520	54	53	46
	1500	0	--	21	--
		1	31	26	24
		18	42	45	43
		100	65	67	86
		240	127	150	125

earlier [11] for specimens fabricated from sheet of the new-process D2 material. Within the experimental scatter of the grain-size data, no difference related to the position in the cups could be determined. Also, there is no difference between the grain sizes of the cups used for this study and the grain size of D2 blank (sheet) material. Grain sizes of approximately 17-22 μm were maintained up through the heat treatment temperature of 1500°C, regardless of orientation with respect to the rolling direction or position in the cup. These specimens all had microstructures similar to that of the as-recrystallized material shown

(a)



(b)



(c)

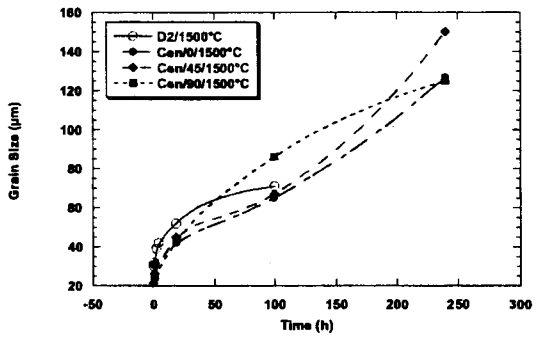


Fig. 8. Effect of annealing time at 1500°C on grain size near the (a) top, (b) bend, and (c) center regions of clad vent set cup 3625-00-2409 produced at the Y-12 Plant. Data for specimens cut 0, 45, and 90° to the rolling direction are included. For comparison, data for D2 sheet material [11] are also included.

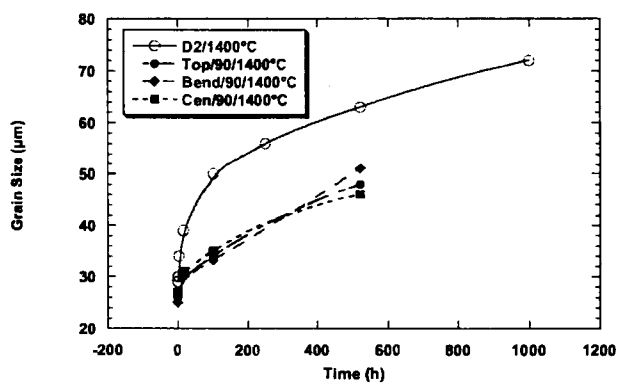


Fig. 9. Effect of annealing time at 1400°C on grain size at 90° to the rolling direction for specimens taken from the top, bend, and center regions of clad vent set cup 3624-30-2045 produced at the Y-12 Plant. For comparison, data for D2 sheet material [11] are also included.

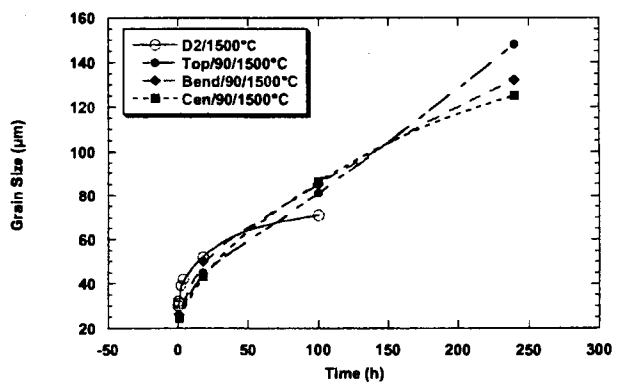


Fig. 10. Effect of annealing time at 1500°C on grain size at 90° to the rolling direction for specimens taken from the top, bend, and center regions of clad vent set cup 3625-00-2409 produced at the Y-12 Plant. For comparison, data for D2 sheet material [11] are also included.

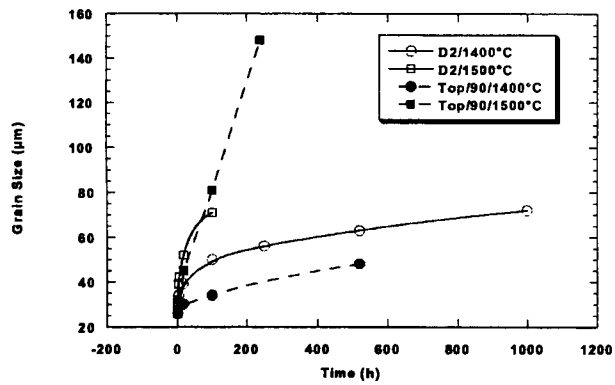


Fig. 11. Effect of annealing time at 1400 and 1500°C on grain size of specimens taken from the top region and 90° to the rolling direction in clad vent set cups produced at Y-12. Data from D2 sheet material [11] are included for comparison.

Table VI. Dependence of Grain Size on Annealing Temperature and Cup Location in ORNL-Produced Clad Vent Set Cup 3625-NC-5011
(all anneals for 1 h at the indicated temperatures)

Position	Temperature (°C)	Grain Size[a] (μm)		
		0° to RD	45° to RD	90° to RD
Top	1200	18 (1144)	21 (1028)	20 (1048)
	1400	18 (1153)	22 (950)	22 (959)
	1600	36 (575)	36 (577)	35 (601)
	1800	109 (387)	107 (310)	119 (385)
Bend	1200	17 (1061)	20 (904)	18 (977)
	1400	18 (993)	20 (885)	19 (923)
	1600	37 (485)	34 (509)	33 (555)
	1800	146 (284)	121 (312)	116 (332)
Center	1200	18 (1053)	19 (999)	18 (1032)
	1400	19 (1012)	20 (961)	21 (954)
	1600	38 (511)	36 (529)	34 (542)
	1800	95 (417)	97 (407)	111 (366)

[a]Numbers in parentheses indicate number of intercepts counted for grain size determination.

in Fig. 5. A comparison of the grain size data presented in Fig. 12 to data generated for CVS cups fabricated at the Y-12 Plant (data from Fig. 6 above) indicates that, as a function of temperature for 1-h anneals, the grain size of the ORNL-fabricated DOP-26 cups is comparable to that of the cups fabricated at Y-12.

At approximately 1600°C, grain growth began, resulting in grain sizes of 30-40 µm for the 1600°C heat treatment (see Fig. 13a) and grain sizes of over 95-146 µm for the 1800°C heat treatment (see Fig. 13b). More scatter in the data was observed for heat treatment temperatures above 1600°C due to the smaller number of intercepts available for counting. In addition, a random distribution of much larger diameter grains, as shown in Fig. 13c, was observed in samples heat treated at the higher temperatures.

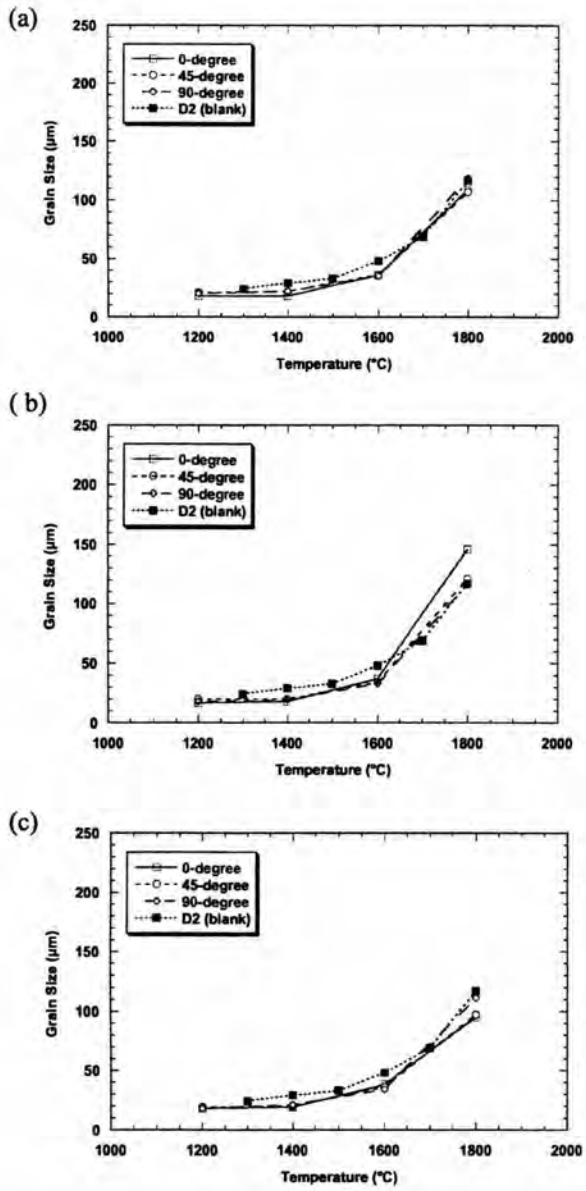


Fig. 12. Effect of heat treatment temperature on grain size near the (a) top, (b) bend, and (c) center of ORNL-produced cup 3625-NC-5011 at 0, 45, and 90° to the rolling direction. Data for specimens taken from D2 sheet material (11) are included for comparison.

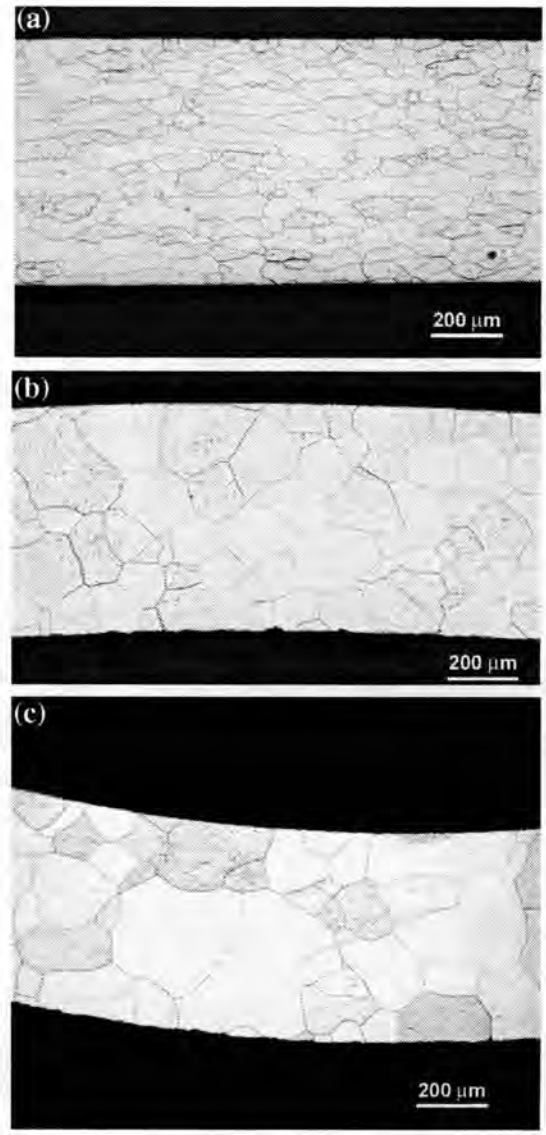


Fig. 13. Optical micrographs showing the microstructures of ORNL-produced cup 3625-NC-5011 after annealing for one hour at (a) 1600°C in a top region specimen cut at 0° to RD, (b) 1800°C in a center region specimen cut 45° to RD, and (c) 1800°C in a bend region specimen cut 0° to RD.

Whereas ORNL qualification-productions cups were used for the 1-h anneals described above, pilot-production cups were used to study the effect of annealing time on grain size. Table VII and Figs. 14-15 show the effect of annealing time at 1400 and 1500°C, respectively, on grain size of ORNL pilot-production cups. Data obtained previously for the CVS cups produced at Y-12 (Figs. 7 and 8) are included for comparison. In general, grain sizes for ORNL-produced cup specimens annealed at 1400°C were slightly lower than the values measured for the cups produced at Y-12. At least some of this difference is be-

Table VII. Average Grain Size as a Function of Location in the Cup and Time of Heat Treatment at 1400 and 1500°C for Pilot-Production Clad Vent Set Cups 9753-NC-0004 and -0005 Produced at ORNL

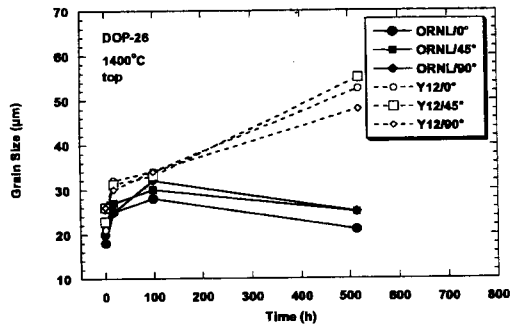
Position	Temperature (°C)	Length of Anneal (h)	Grain Size[a] (μm)		
			0° to RD	45° to RD	90° to RD
Top	1400	1	22 (297)	--	26 (254)
		18	25 (267)	27 (264)	25 (283)
		100	28 (250)	30 (239)	32 (221)
		515	21 (309)	25 (285)	25 (263)
	1500	1.5	24 (274)	26 (252)	24 (276)
		18	19 (339)	24 (273)	44 (303)
		100	19 (325)	77 (266)	46 (288)
		250	22 (280)	25 (265)	21 (307)
Bend	1400	1	21 (264)	--	22 (252)
		18	25 (227)	26 (213)	25 (219)
		100	27 (203)	30 (183)	28 (202)
		515	31 (358)	31 (178)	40 (420)
	1500	1.5	22 (258)	30 (188)	27 (206)
		18	26 (217)	45 (248)	45 (251)
		100	60 (296)	62 (186)	57 (309)
		250	23 (245)	23 (237)	20 (271)
Center	1400	1	23 (266)	--	23 (258)
		18	26 (230)	27 (222)	25 (239)
		100	30 (204)	33 (182)	28 (215)
		515	38 (327)	32 (189)	40 (306)
	1500	1.5	25 (241)	25 (240)	25 (242)
		18	35 (351)	40 (152)	36 (350)
		100	55 (113)	59 (211)	56 (332)
		250	100 (63)	--	112 (121)

[a]Numbers in parentheses indicate number of intercepts counted for grain size determination.

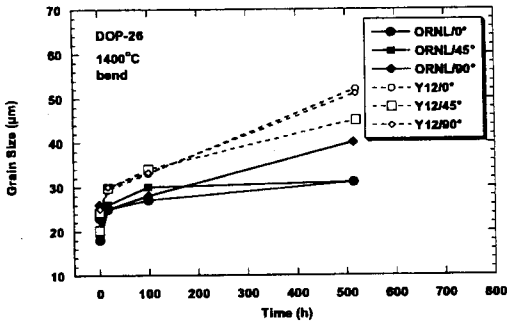
lieved to be due to a slight change in the technique used to count intercepts during the measurement of the grain sizes. This change in technique results in a more accurate determination of the number of intercepts counted. Taking this into consideration, almost all of the grain sizes measured at 1400°C for the ORNL-produced cup specimens are comparable to those produced at Y-12. The only discrepancy is for the longest anneal of 515 h in which the grain sizes for the top region of the cup are much smaller than those measured previously for the cups produced at Y-12 (see Fig. 14a). The data show no dependence on the orientation to the rolling direction.

Except for the samples annealed for 1.5 h, the samples annealed for various times at 1500°C (Fig. 15) also showed non-homogeneous grain growth similar to that noted for the 515 h anneal at 1400°C. For

(a)



(b)



(c)

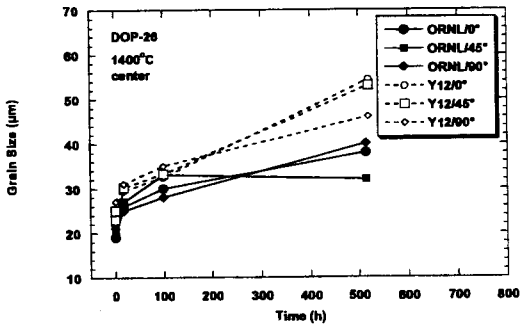
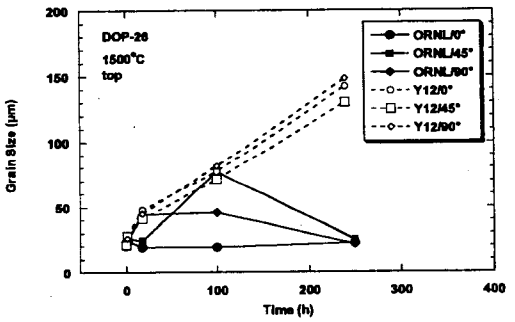
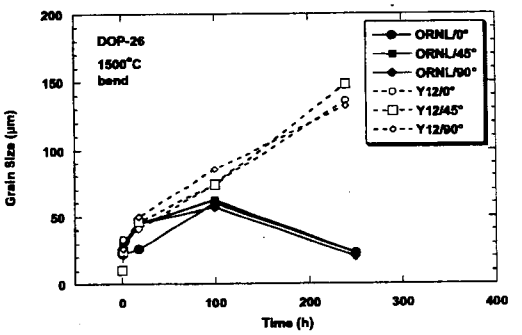


Fig. 14. Effect of annealing time at 1400°C on grain size near the (a) top, (b) bend, and (c) center of ORNL-pilot-production cup 9753-NC-0005 at 0, 45, and 90° to the rolling direction. Data for CVS cups produced at Y-12 (from Table V) are included for comparison.

(a)



(b)



(c)

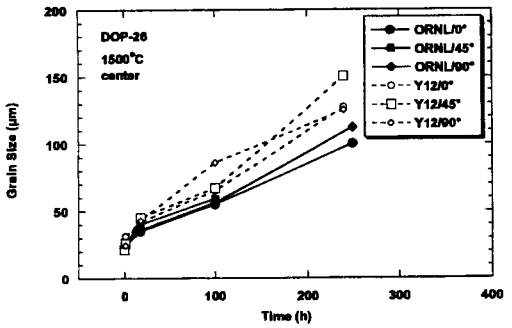


Fig. 15. Effect of annealing time at 1500°C on grain size near the (a) top, (b) bend, and (c) center of ORNL-pilot-production cup 9753-NC-0004 at 0, 45, and 90° to the rolling direction. Data for CVS cups produced at Y-12 (from Table V) are included for comparison.

example, Fig. 16 shows the microstructure of the ORNL cup annealed for 250 h at 1500°C. The first 1-2 mm of the top region of that sample (left half of Fig. 16a) had very large grains (150-300 μm). But from there through the bend region (Fig. 16b), the grain sizes averaged only 20-25 μm. The grains in the center region (Fig. 16c) were about 100 μm in diameter which, in comparison to the Y-12 cup data, is closer to the size one would expect for this heat treatment temperature and time. On averaging 3-4 fields in each specimen, the grain sizes in the ORNL-produced cups were comparable to or smaller than grain sizes in the cups produced at Y-12 (see Fig. 15). However, many of the samples contained areas like that shown in Fig. 16a where the grains were much larger. There appeared to be no correlation between the non-homogeneous grain growth and different regions in the cup. Nor did the grain size results depend on orientation with respect to the rolling direction of the sheet from which the cups were produced.

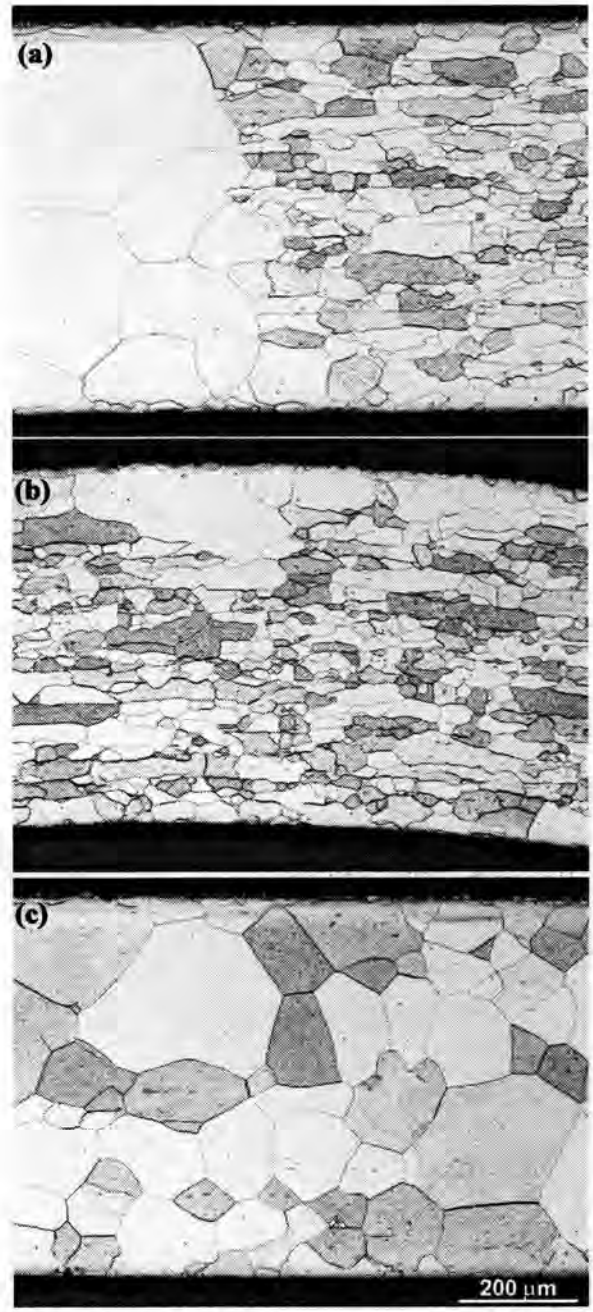


Fig. 16. Optical micrographs showing the grain structure in the (a) top, (b) bend, and (c) center regions of ORNL-pilot-production cup 9753-NC-0004 after a heat treatment of 250 h at 1500°C.

To further study the non-homogeneous grain growth in the ORNL-pilot-production cups, the 100 h anneal at 1500°C was repeated with new samples (45° to RD) from both the 9753-NC-0004 and -0005 DOP-26 CVS cups. The results are shown in Table VIII. Included in this table are the data from the first anneal (from Table VII). The data from the repeat anneal showed that the non-homogeneous grain growth is truly anomalous; many of the values for a similar region of the same cup were drastically different from the first anneal. For example, Fig. 17 shows micrographs of the top and bend regions of the specimen from cup -0004 used in the repeat anneal. As can be seen, the microstructures for these two areas of the same specimen were quite different. In addition, the micrograph for the bend region shown in Fig. 17b contains grains with a much larger diameter than did the first anneal whose microstructure looked more like Fig. 17a. A piece of the DOP-26 cup produced at Y-12 that was used previously for the Y-12 cup grain growth study (cup # 3625-00-2409) was included in this repeat anneal and it showed

Table VIII. Repeat of Grain Size Measurements in ORNL-Pilot-Production CVS Cups
(samples were 45° to RD; heat treatment was 100 h at 1500°C)

Cup #	Fabricated at	Grain Size[a] (μm)		
		top	bend	center
9753-NC-0004 (1st anneal)	ORNL	77 (266)	62 (186)	59 (211)
9753-NC-0004	ORNL	23 (283)	78 (220)	60 (210)
9753-NC-0005	ORNL	90 (242)	75 (228)	72 (87)
3625-00-2409	Y-12	78 (158)	70 (81)	74 (84)

[a]Numbers in parentheses indicate number of intercepts counted for grain size determinations.

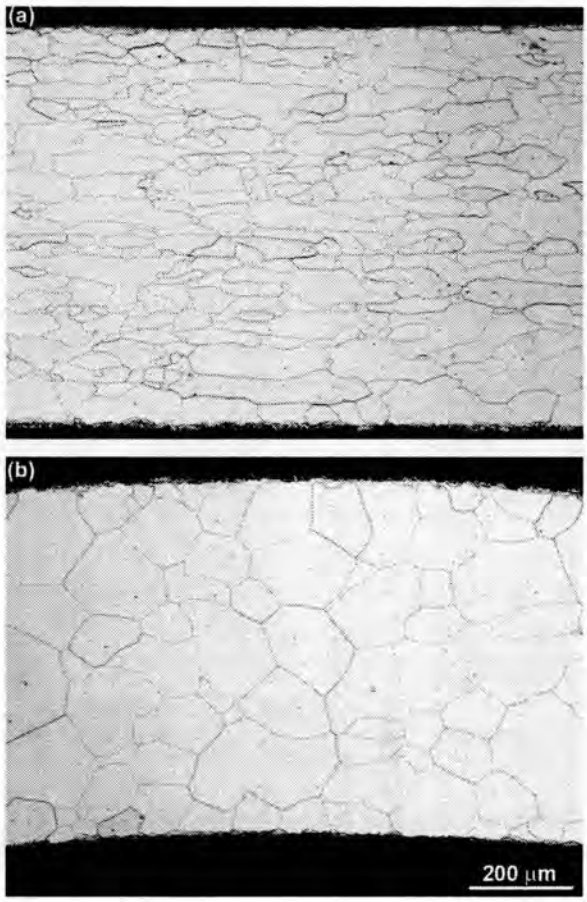


Fig. 17. Optical micrographs showing non-homogeneous grain size distribution in the (a) top and (b) bend regions of a piece of ORNL-produced cup 9753-NC-0004 included in the second anneal of 100 h at 1500°C.

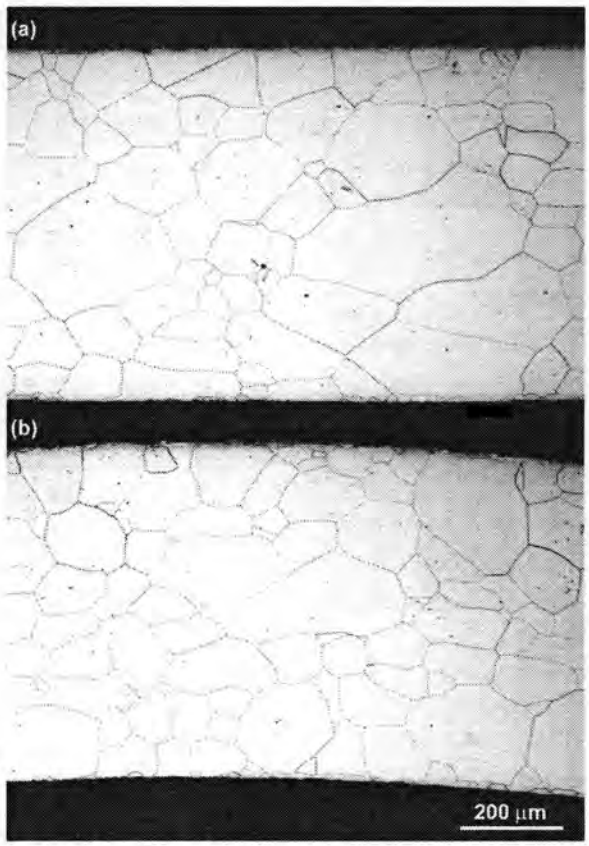


Fig. 18. Optical micrographs showing homogeneous grain size distribution in the (a) top and (b) bend regions of a piece of cup 3625-00-2409 (produced at Y-12) included in the second anneal of 100 h at 1500°C.

homogeneous grain growth along the entire length of the specimen (see Fig. 18). In addition, its grain sizes were comparable to the grain sizes measured previously for the same cup (compare data in Table VIII for cup -2409 to that in Table V for this heat treatment). These results suggest that the anomalous grain growth is not related to anomalies in the heat treatment procedures (e.g., problems with the furnace) but is a characteristic only of the ORNL pilot-production cups.

In the past, grain size studies have been conducted on material taken from several ingots, including one (D2 ingot) from which one of the cups for this study was fabricated [11]. None of the sheet material has shown this type of non-homogeneous grain-growth behavior. Since the repeat anneal described above included specimens from cups produced both at Y-12 and ORNL, and the Y-12 cup specimen showed homogeneous grain growth, faulty heat treatment procedures are not believed to be a factor. One possibility is that since the pilot-production cups were made from non-flight-quality sheet material, inhomogeneities in composition and/or distribution of Ir_5Th particles could have produced these results. Another possibility is that sometime during pilot-production of cups at ORNL a critical amount of strain was introduced into the cups, which then caused this non-homogeneous grain growth upon subsequent heat treatment. Three avenues were investigated to study this possibility. (1) The grain growth behaviors of several cups produced during qualification production and production maintenance at ORNL were determined. The results would show whether a stress relief heat treatment, which is done on the qualification cups but was not done on the pilot production cups, would eliminate the possibility of non-homogeneous grain growth. (2) During pilot production too much hold-down force was applied for the scribing operation and these cups had to be resized. This could have provided residual stress that led to the non-homogeneous grain growth. To investigate this possibility, two cups were formed from blank assemblies in an attempt to duplicate the operations used during pilot production. Their grain growth behavior was determined and compared to the previous pilot production data. (3) The effects on grain growth of small amounts of strain induced in DOP-26 sheet by bending, tensile, and compression loading were determined. This study would show how much residual stress would be required to produce the kind of anomalous grain growth discussed above and would shed some light on how that strain could have been introduced into the cup during fabrication.

INVESTIGATION OF NON-HOMOGENEOUS GRAIN GROWTH IN CVS CUPS PRODUCED AT ORNL

Qualification-Production Cups 9753-30-4000 and 9754-02-A5NV

Two ORNL-qualification-production cups (listed in Table I as cups 9753-30-4000 and 9754-02-A5NV) fabricated from flight-quality material and each having an initial grain size of approximately 25 μm were received in the as-recrystallized condition (they also received air burn off and vacuum outgas heat treatments). A specimen was cut from each cup in the 0° direction with respect to the rolling direction and, along with a piece of cup 3625-00-2409 produced at Y-12, they were annealed for 100 h at 1500°C in vacuum. The results of grain size measurements are shown in Table IX. Optical micrographs

Table IX. Grain Sizes of Specimens Taken from Clad Vent Set Cups Annealed for 100 h at 1500°C
(samples were approximately parallel to RD)

Cup #	Grain Size[a] (μm)		
	top	bend	center
3625-00-2409 (Y-12)	67 (101)	51 (111)	59 (105)
9753-30-4000 (ORNL)	68 (98)	68 (83)	52 (117)
9754-02-A5NV (ORNL)	69 (98)	56 (103)	62 (109)
TC33 -- parallel to vent notch	59 (315)	62 (293)	57 (324)
TC33 -- perpendicular to vent notch	55 (337)	61 (283)	53 (355)
TC38 -- parallel to vent notch	68 (276)	68 (259)	59 (318)
TC38 -- perpendicular to vent notch	69 (270)	57 (298)	65 (291)

[a]Numbers in parentheses indicate number of intercepts counted for grain size determinations.

of specimens from the three cups are shown in Figs. 19-21. Average grain sizes for all regions of all three cups were 51-69 μm. Within expected experimental variability, these values were comparable to the values obtained earlier for DOP-26 cups produced at the Y-12 Plant. Homogeneous grain growth was observed throughout all three specimens.

Production Maintenance Cups 3625-05-5022 (segment 3625-91-0053), 3624-05-4013 and 3625-05-5026

Production maintenance cups 3625-05-5022 (segment 3625-91-0053), 3624-05-4013, and 3625-05-5026, fabricated from flight-quality material, each contained a crease down the side produced by the presence of dried and hardened lubricant on the deep-drawing second-form die. Figure 22 shows the presence of the crease on the outside surface of cup segment 3625-91-0053 after post-recrystallization sizing. Metallography of this cup showed an average grain size of 24 μm which is a typical grain size for as-recrystallized DOP-26 sheet and cups [11]. Similar creases occurred on cups 3624-05-4013 and 3625-05-5026. Cup 3624-05-4013 had been recrystallized at 1375°C but received no air burn-off or vacuum-outgas heat treatments, while cup 3625-05-5026 received all heat treatments intended for flight-quality cups. These two cups were annealed in vacuum for 100 h at 1500°C, and then specimens for grain size determination were cut from both the creased region and from an uncreased region 180° away from the crease. On examination of the optical microstructures of specimens cut from cup 3624-05-4013, large grains were observed only in the specimens containing the creased region (see Fig. 23). These large grains tended to occur on the inner surface of the cup and were as large as half the wall thickness (~300-350 μm in diameter compared to the cup wall thickness of 630-730 μm). In these same specimens, much smaller grain sizes of 60-70 μm (see Table X and Fig. 24) were determined for areas at least 3-5 mm away from the crease. In order to determine if the crease observed in cup 3624-05-4013 was responsible for the large diameter grains shown in Fig. 23, another series of four samples were cut in a similar manner

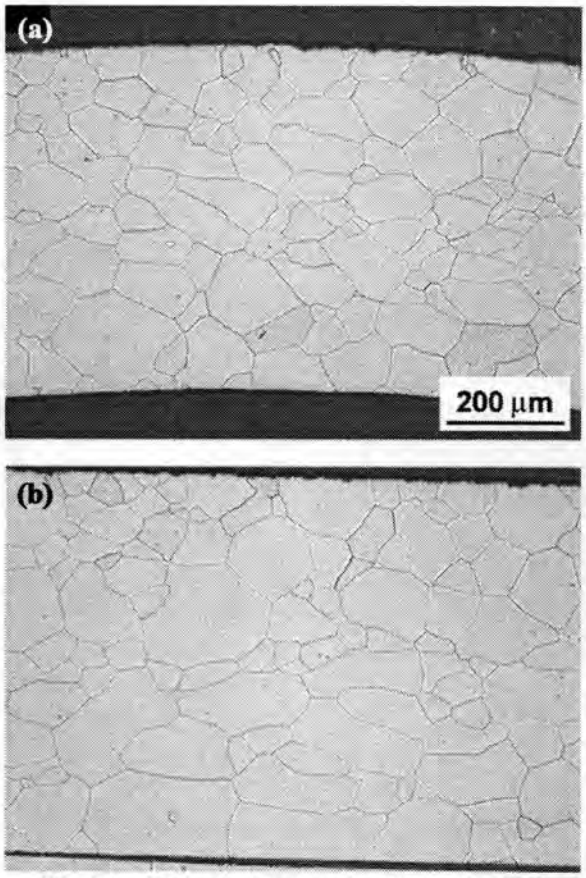


Fig. 19. Optical micrographs of the (a) bend and (b) center regions of cup 3625-00-2409 (produced at Y-12) after annealing along with cups 9753-30-4000 and 9754-02-A5NV for 100 h at 1500°C.

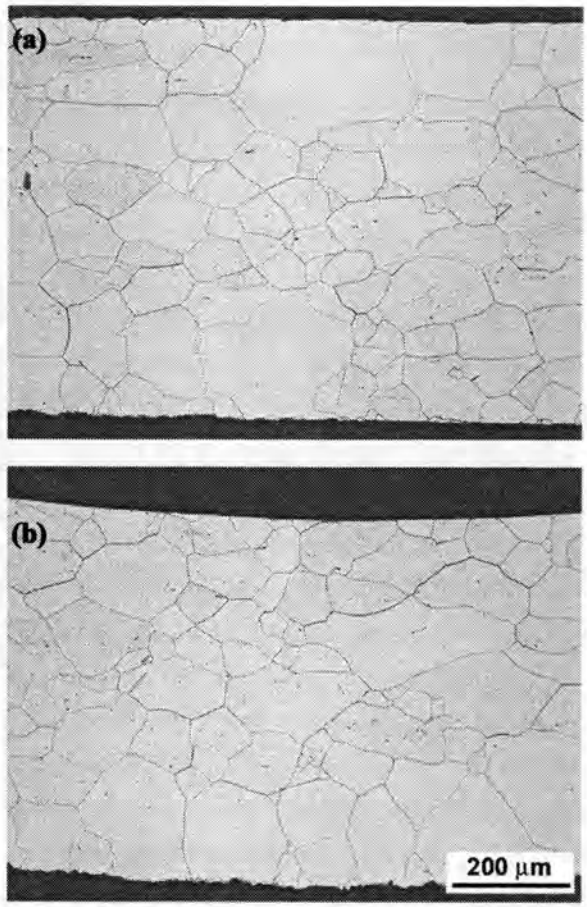


Fig. 20. Optical micrographs of the (a) top and (b) bend regions of ORNL-produced cup 9753-30-4000 after annealing for 100 h at 1500°C.

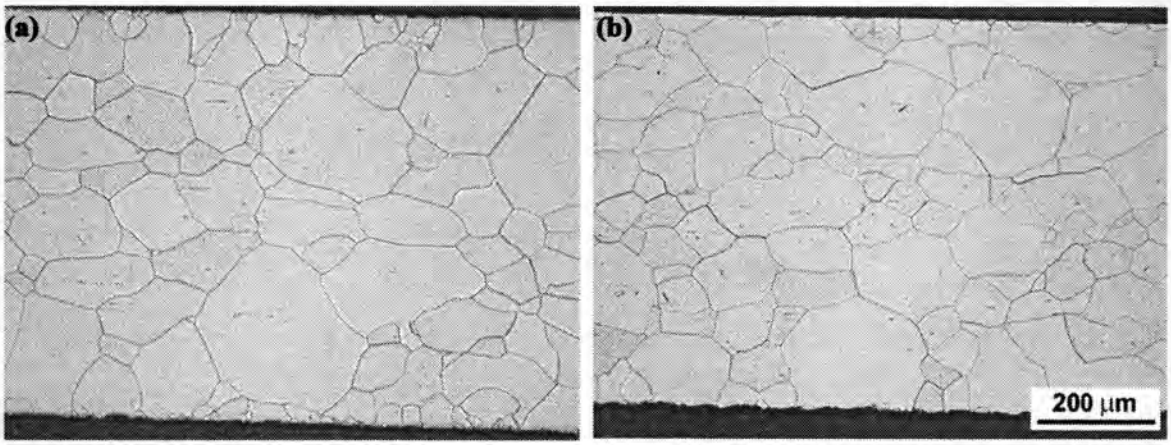


Fig. 21. Optical micrographs of the (a) top and (b) bend regions of ORNL-produced cup 9754-02-A5NV after annealing for 100 h at 1500°C.

from the region of the cup 180° away from the crease (specimens #5-8 in Fig. 4). Although each of these four samples contained a few larger than average grains (as shown in Fig. 25), all four had an average grain size of 60-70 μm (see Table X). This value is comparable to data from samples of the DOP-26 cups produced at the Y-12 Plant which were annealed for 100 h at 1500°C. Non-homogeneous grain growth was not observed in any of these specimens.

In addition to being recrystallized at 1375°C, cup 3625-05-5026 also received air burnoff (4 h at 635°C) and vacuum outgas (1 h at 1250°C) heat treatments before being received for grain growth studies. This cup was annealed for 100 h at 1500°C and specimens were cut from regions around the crease and from 180° to the crease, as was done for the 3624-05-4013 cup. These additional heat treatments did not appear to change the tendency for large grains to form near the crease, as shown in Fig. 26. The average grain size of regions away from the crease (Fig. 27) was comparable to that of the 3624-05-4013 cup and to other DOP-26 specimens (see Table X).

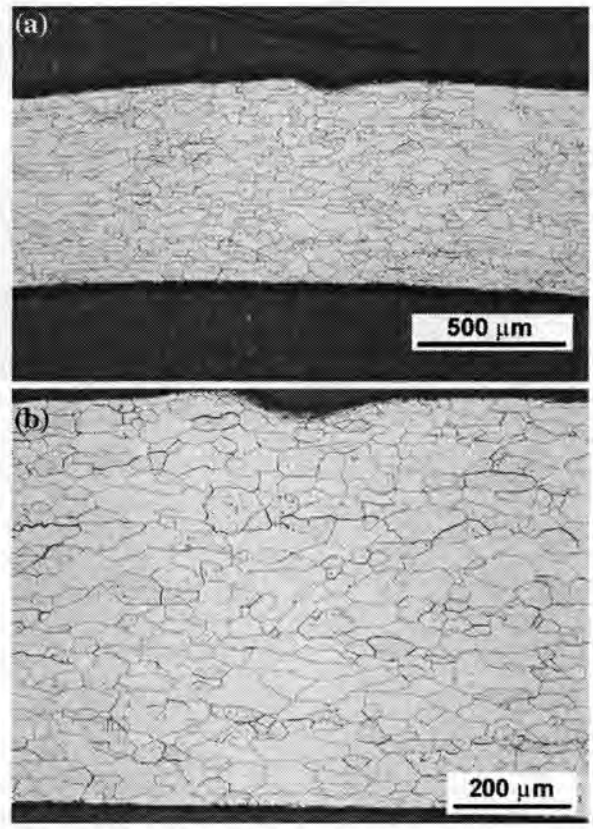


Fig. 22. Optical micrographs at two different magnifications showing the presence of a crease on the outer surface of ORNL-produced cup segment 3625-91-0053 (from cup 3625-05-5022).

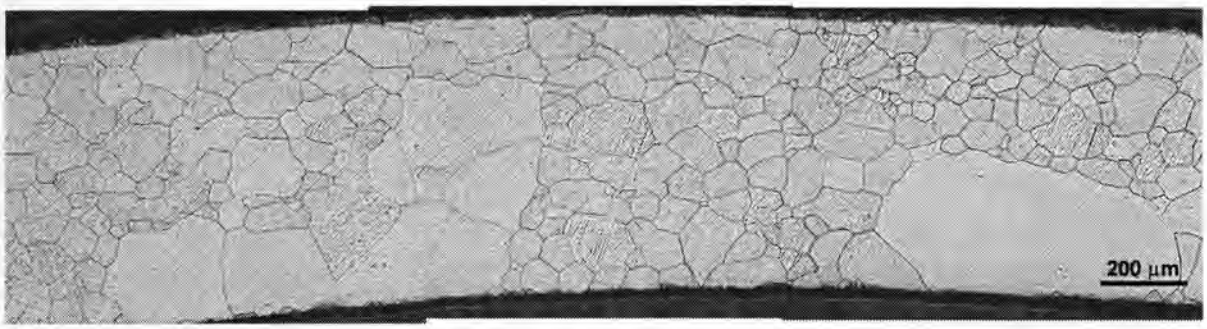


Fig. 23. Optical micrograph showing large grains in the creased region of ORNL production-maintenance cup 3624-05-4013 after annealing for 100 h at 1500°C (area corresponds to #1 in Fig. 4).

Table X. Grain Sizes of Creased CVS Cups Annealed for 100 h at 1500°C

Specimen position in cup[a]	Grain Size[b] (μm) for Cups Identified As		
	3624-05-4013	3625-05-5026	3625-91-0053
1	60 (320)	58 (326)	24 (541)
2	70 (272)	59 (291)	24 (503)
3	70 (267)	62 (274)	--
4	69 (276)	64 (294)	--
5	62 (298)	68 (293)	--
6	59 (313)	69 (275)	--
7	66 (288)	66 (280)	--
8	67 (280)	65 (292)	--

[a]See Fig. 4.
[b]Numbers in parentheses indicate number of intecepts counted for grain size determinations.

SIMULATION OF PILOT PRODUCTION

Cups TC-33 and TC-38 were processed in an attempt to reproduce the fabrication operations involved in the ORNL pilot-production cups. All the pilot-production cups had been deformed (out-of-round) from too much hold-down force during scribing and it was believed that this may have contributed to the non-homogeneous grain growth described above. Because of this deformation, the pilot-production cups had to be re-sized after they were scribed. Cups TC-33 and TC-38 were formed, cleaned, recrystallized, and dimensionally inspected according to standard production procedures. Then they were sized, grit blasted, scribed/deformed, and re-sized. Between each of these operations, the cups were carefully inspected according to standard production procedures. Additionally, cup TC33 was cleaned and given an air burnoff and vacuum outgas heat treatments as described above, while cup TC38 remained in the as-recrystallized condition. The cups were then annealed for 100 h at 1500°C and specimens were cut for grain size measurements in directions parallel and perpendicular to the vent notch.

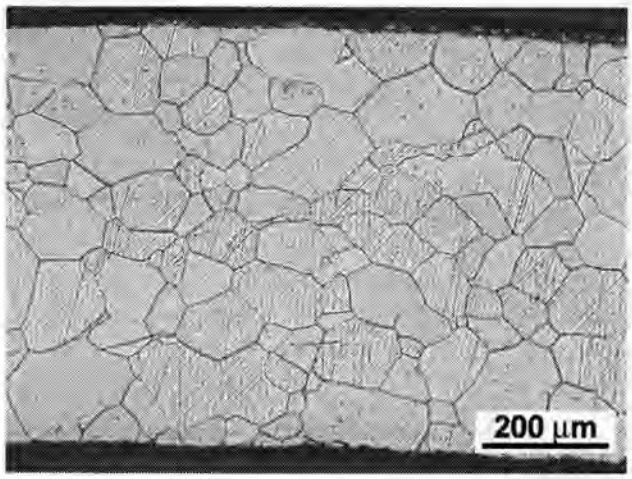


Fig. 24. Optical micrograph showing the general microstructure of cup 3624-05-4013 in an area approximately 3-5 mm away from the crease after annealing for 100 h at 1500°C.

The results are shown in Table IX and in Figs. 28-31 for specimens taken parallel and

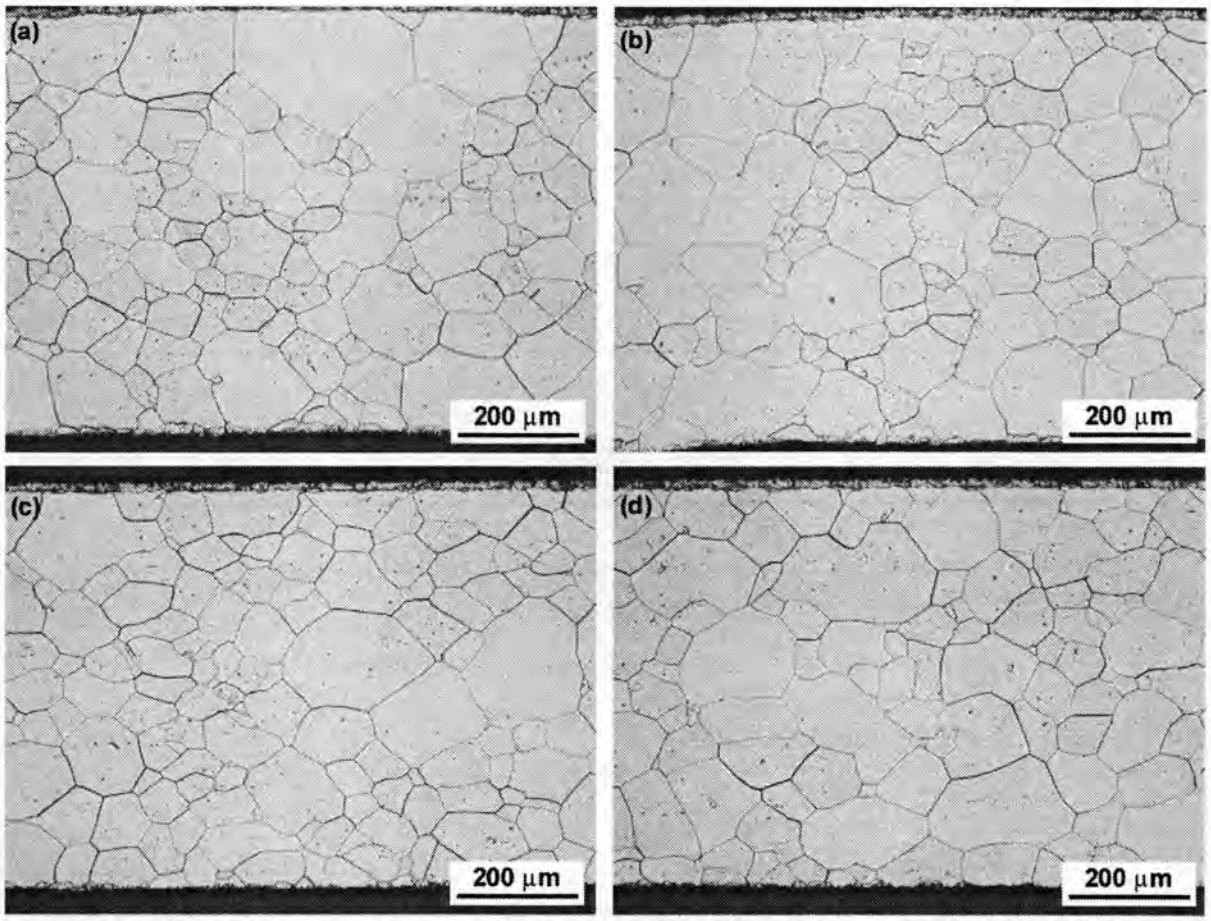


Fig. 25. Optical micrographs showing the grain structure after annealing for 100 h at 1500°C of (a,b) sample #1 and (c,d) sample #4 taken from a region 180° to the crease in cup 3624-05-4013.

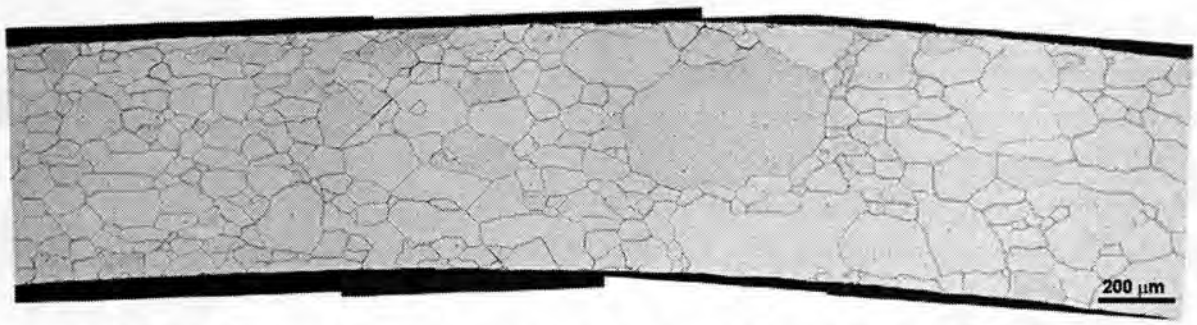


Fig. 26. Optical micrograph showing large grains in the creased region of cup 3625-05-5026 after annealing for 100 h at 1500°C.

perpendicular to the vent notch. There are several large grains of approximately 150-250 μm diameter scattered throughout the microstructures, which is not unusual for DOP-26 iridium alloys. Non-homogeneous grain growth, of the type seen in the ORNL-pilot-production cups (Fig. 16), was not observed in these specimens. Average grain sizes in all three regions of the specimens (top, bend and center) from both cups were comparable to data taken from the cups produced at Y-12 (see Table V and Fig. 8).

Average grain sizes in cup TC-33 were 53-62 μm, while grains sizes in cup TC-38 were 57-69 μm.

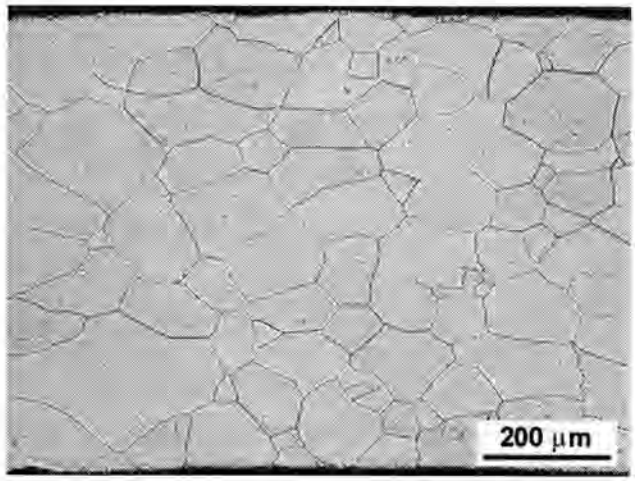


Fig. 27. Optical micrograph showing the grain structure after annealing for 100 h at 1500°C of a sample taken from a region 180° to the crease in cup 3625-05-5026.

STRAIN-INDUCED GRAIN GROWTH IN DOP-26 IRIDIUM

In order to determine if some small amount of strain induced during fabrication could produce non-homogeneous grain growth upon heat treating at high temperatures, specimens from a single DOP-26 blank (CR4-9) were machined and strains of up to 5% were induced in them using tensile, compression, and bend loading as described in the Experimental Procedures section. This blank had been recrystallized for 1 h at 1300°C and in the as-received condition contained a uniform microstructure with an average grain size of approximately 15-20 μm, which is typical for DOP-26 sheet receiving this same heat treatment [11]. Table XI lists the grain sizes, after annealing for 100 h at 1500°C, measured as a function of induced strain in the specimens. Optical micrographs of tensile specimens given strains of 1.75, 2.5 and 5% and then annealed for 100 h at 1500°C are shown in Fig. 32. Only nominal grain growth was noted in the specimen strained to 1.75% (Fig. 32a), where average grain sizes of 40-50 μm were measured. The first isolated large grains were observed in the specimen strained to 2.5% (Fig. 32b), even though the average grain size in this specimen was determined to be 40-50 μm. The large grains were randomly dispersed throughout the length of the specimen and always appeared to be growing inward from the surface. Uniform grain growth and average grain sizes of approximately 300 μm (as shown in Fig. 32c) were observed in those tensile specimens that were strained to 3.75 and 5%.

Rolling to produce compression strains of 1.8 to 5.6% resulted in uniform microstructures and average grain sizes of 126 to 178 μm (see Table XI and Fig. 33). The data show an apparent tendency for slightly smaller grain sizes as the rolling strain is increased.

As would be expected, grain growth in the bent specimens was not uniform along the length of the specimens. Away from the bend, the grain sizes were approximately the same for all four of the specimens and measured between 48 and 57 μm (see Table XI), a grain size which is comparable to Y-12 cups annealed for the same time and temperature. In the region of the bend, however, grain growth was observed even in the specimen strained to 1.3%. Figure 34 shows that large grains in that specimen are beginning to form at both surfaces in a region near the bend, indicating that the anomalous grain growth can be nucleated in both tension and compression regions of bend specimens. In the specimen with 1.7% strain (Fig. 35), only a narrow band of small grains remained in the center of the bend region and the average grain size in the bend region was 183 μm. For the 2.5 and 4.9% specimens (Figs. 36 and 37,

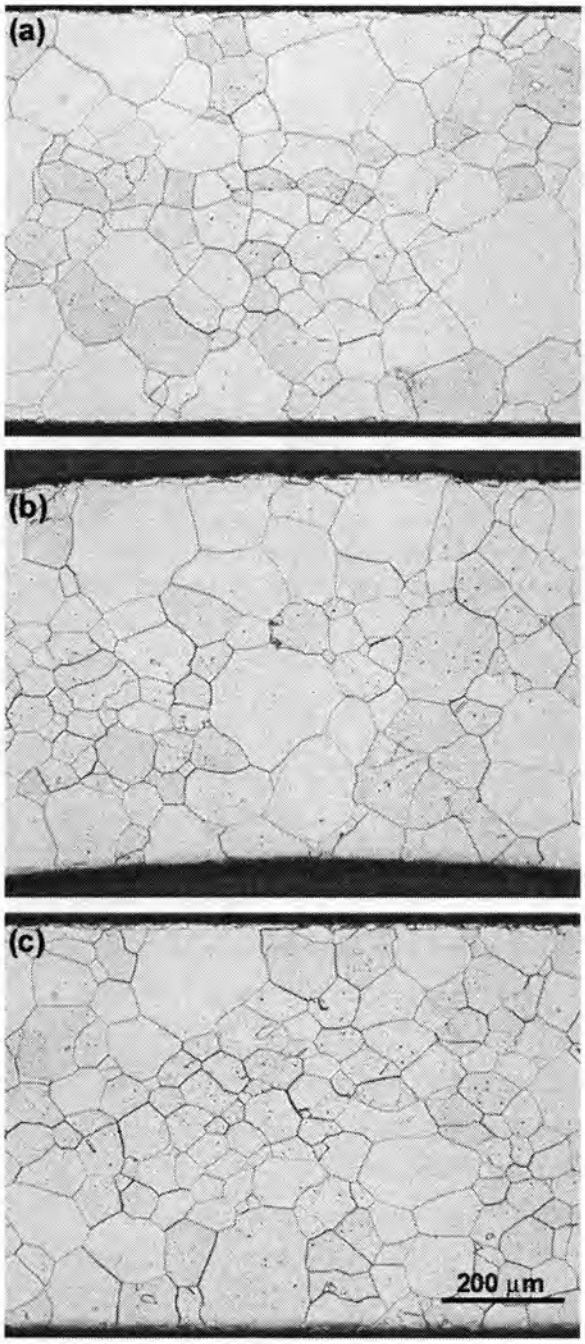


Fig. 28. Optical micrographs showing grain size in areas of cup TC-33, parallel to the vent notch, after annealing for 100 h at 1500°C, (a) top, (b) bend, and (c) center.

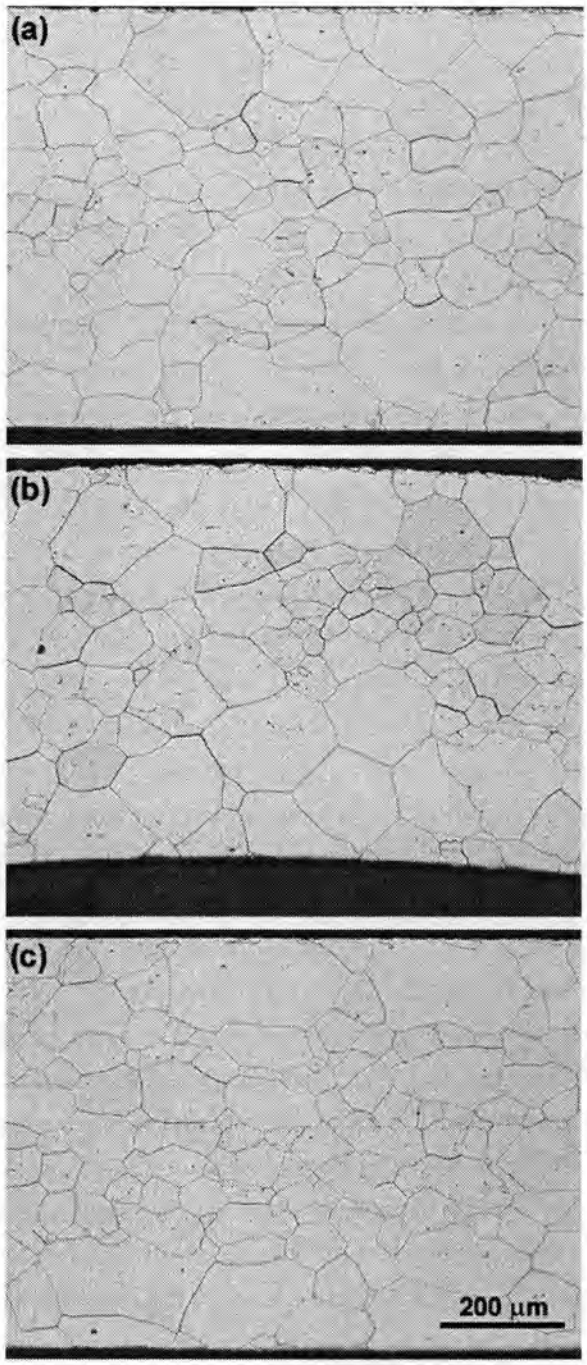


Fig. 29. Optical micrographs showing grain size in areas of cup TC-33, perpendicular to the vent notch, after annealing for 100 h at 1500°C, (a) top, (b) bend, and (c) center.

respectively), average grain sizes of 336 and 289 µm, respectively, were measured in the bend region. Since the specimens were between 630 and 680 µm thick, this means that there were sometimes only two grains through the thickness. In both these samples, as one moves away from the bend region toward the straight ends of the specimen, there is a region of large surface grains with smaller grains along the

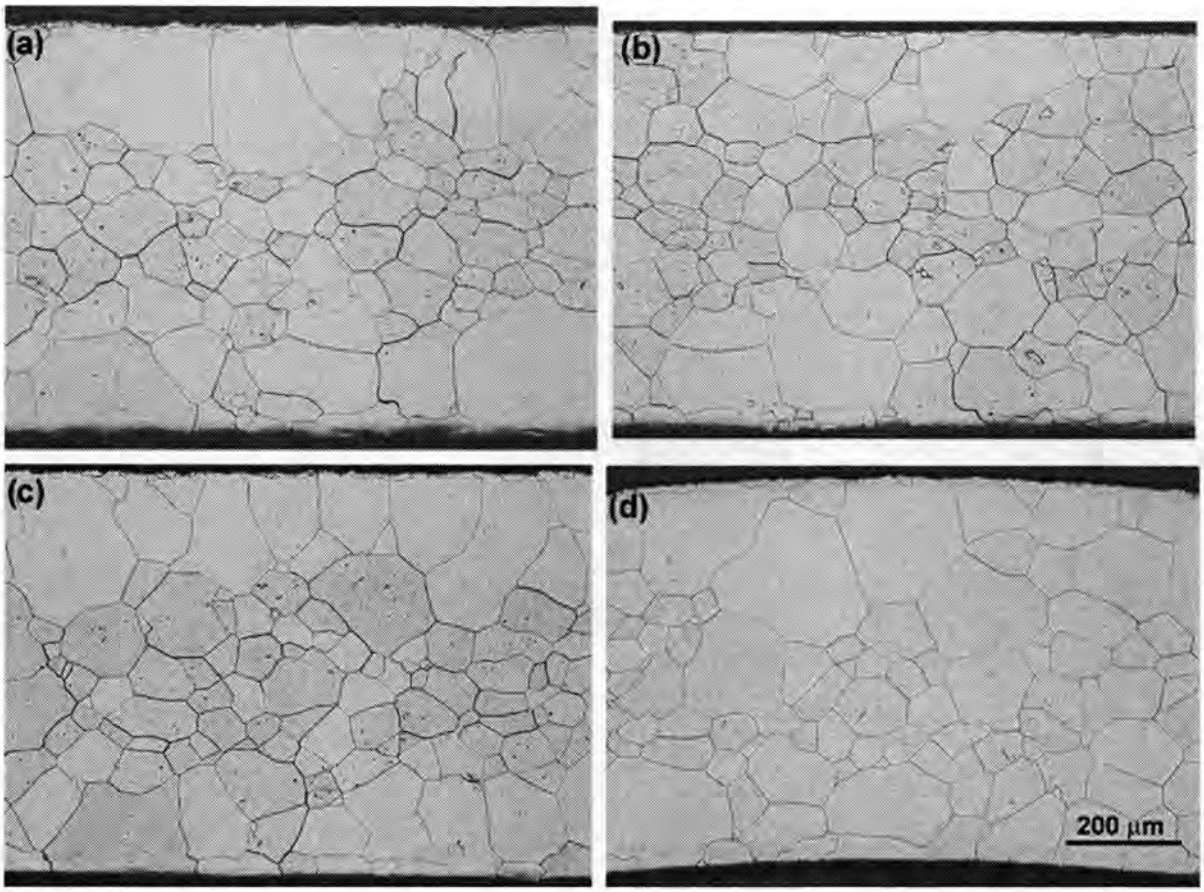


Fig. 30. Optical micrographs showing grain size in areas of cup TC-38, parallel to the vent notch, after annealing for 100 h at 1500°C, (a) top, (b) center, (c) just above the bend, and (d) bend.

centerline (see Fig. 37b), as was observed in the bend region of the specimen with 1.7% strain. All of these observations indicate that grain growth is more rapid and final grains are much larger in the areas of the specimens that received more cold work during bending.

The optical microstructure of a specimen with 2.5% bending strain that was not annealed is shown in Fig. 38. Grains in this sample were slightly elongated along the rolling direction and had average diameters of approximately 16 μm which is comparable to previously determined values for as-received DOP-26 sheet material [11] and to clad vent set cups produced both at ORNL and Y-12 (see Table III). Also for comparison, the grain size range for specimens taken from old- and new-process DOP-26 blanks given the same heat treatment range is shown in Table XI [11].

Additional specimens were cut from blank AR2-9 and bent as noted above to produce approximately 2.5 and 5% strain. These specimens were given the same recrystallization, air-burnoff, and vacuum-outgas thermal treatments as are given to CVS cups during fabrication. They were then heat treated for one month at 950°C to simulate fueled CVS storage life and three months at 1330°C (both in an argon-filled quartz tube) to simulate that length of time in space. Figures 39 and 40 show the optical microstructures of the 2.5 and 5% samples, respectively, after these heat treatments. The specimen that received 2.5% strain had a few large grains on the outer surface of the bend region (Fig. 39a), while the

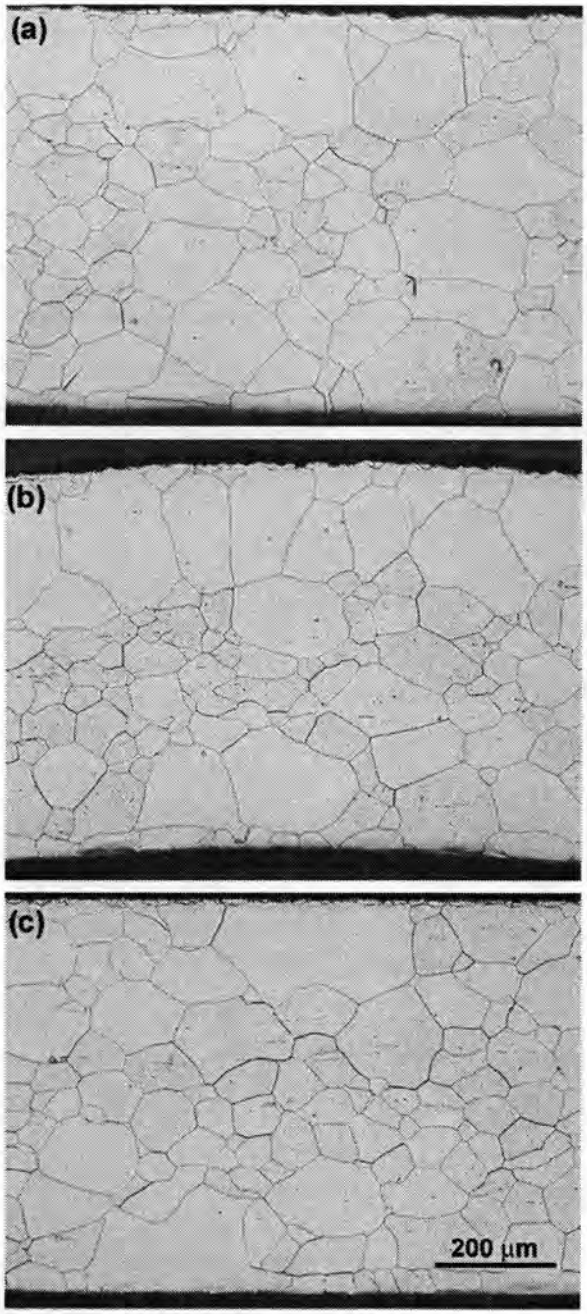


Fig. 31. Optical micrographs showing grain size in areas of cup TC-38, perpendicular to the vent notch, after annealing for 100 h at 1500°C, (a) top, (b) bend, and (c) center.

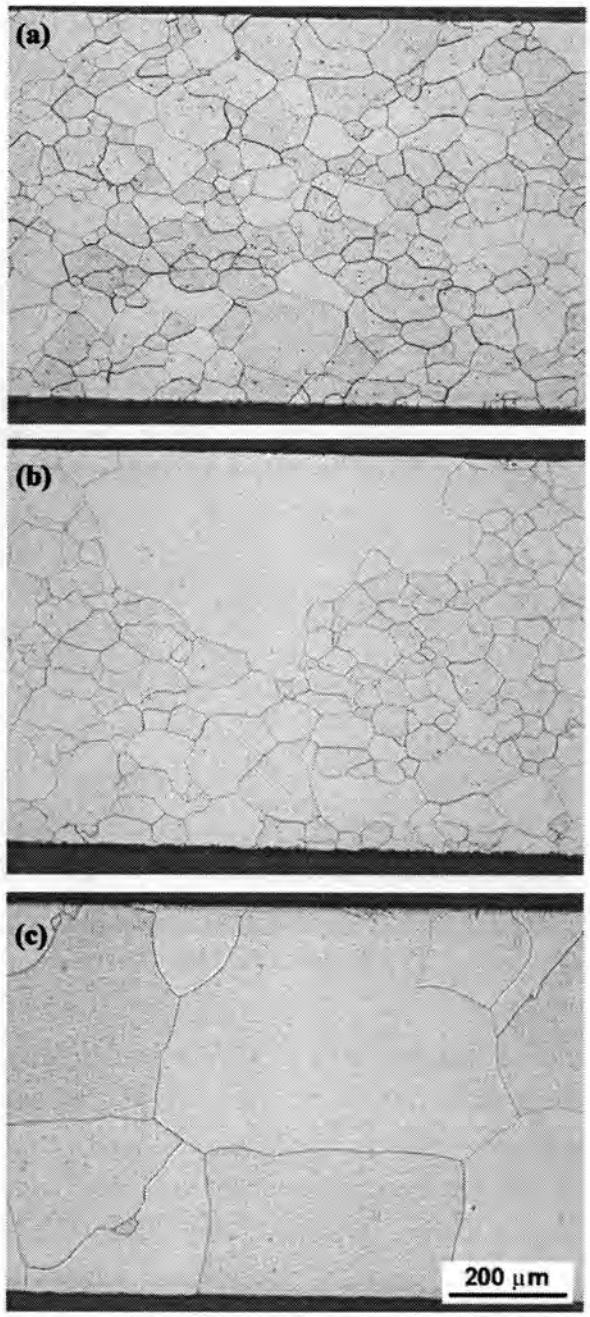


Fig. 32. Optical micrographs showing the grain size of DOP-26 sheet specimens given tensile strains of (a) 1.75, (b) 2.5, and (c) 5% and then annealed for 100 h at 1500°C.

specimen receiving 5% strain contained a band of larger grains along both the inner and outer surface of the bend region (Fig. 40a). None of these large grains appeared to extend more than approximately 100 μm into the cup. Grain sizes were measured in the bend region and in areas away from the bend where no induced strain was present to affect the grain growth. In all these cases, average grain sizes were 23-28 μm.

Table XI. Effect of Induced Strain on Grain Size in DOP-26 Sheet Material (Blank CR4-9)
(all specimens annealed for 100 h at 1500°C)

Type of Strain	Strain (%)	Grain Size[a] (μm)	
Unstrained	0	71-82 (280)[b]	
Tensile	1.75	41 (598)	
	2.5	48 (531)	
	3.75	290 (108)	
	5.0	309 (94)	
Rolling	1.8	178 (115)	
	2.7	172 (117)	
	3.85	139 (142)	
	5.6	126 (151)	
		Away from bend	In bend region
Bending	1.3	50 (537)	66 (225)
	1.7	57 (117)	183 (43)
	2.5	48 (271)	336 (23)
	4.9	51 (129)	289 (27)
Bending (no anneal)	2.5	16 (369)	

[a]Numbers in parentheses are the number of intercepts counted to determine average grain size.
[b]Data for unstrained material taken from Ref. 11.

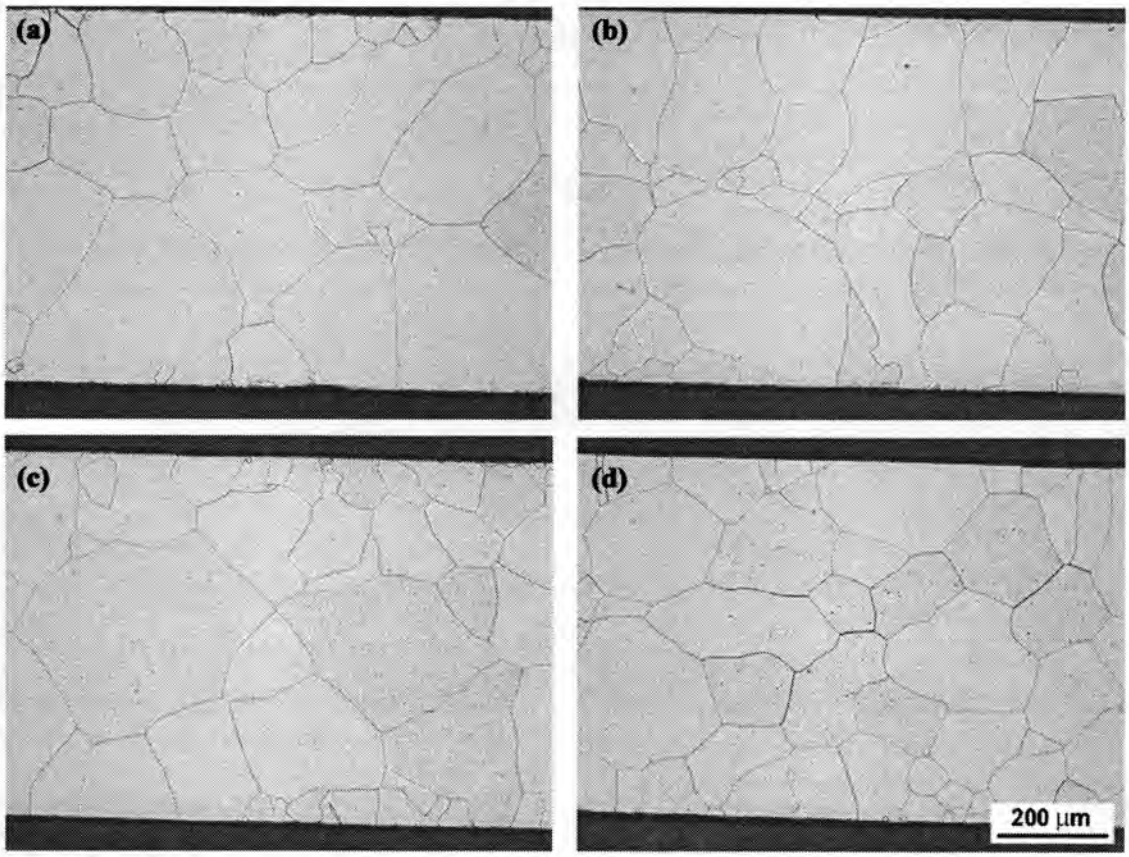


Fig. 33. Optical micrographs showing the grain size of DOP-26 sheet specimens that were rolled to a strain of (a) 1.8, (b) 2.7, (c) 3.85, and (d) 5.6% and then annealed for 100 h at 1500°C.

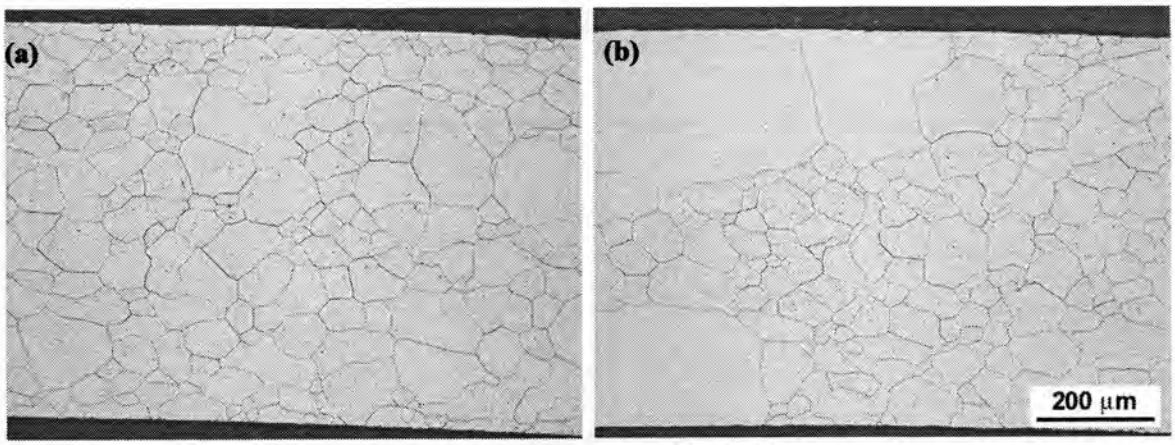


Fig. 34. Optical micrographs showing the grain size of DOP-26 sheet specimens that have been bent to a strain of 1.3% and then annealed for 100 h at 1500°C: (a) away from and (b) near center of bend region.

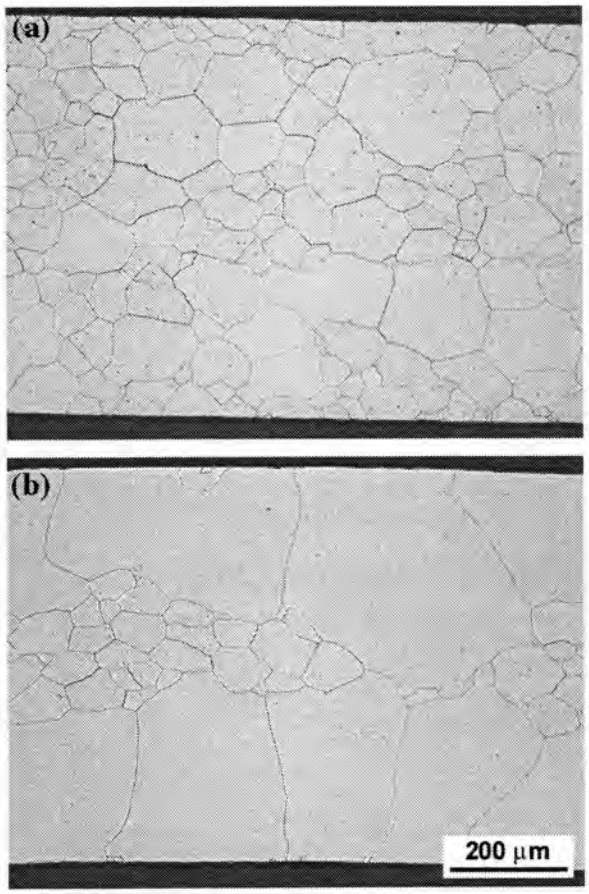


Fig. 35. Optical micrographs showing the grain size of DOP-26 sheet specimens that have been bent to a strain of 1.7% and then annealed for 100 h at 1500°C: (a) away from and (b) near center of bend region.

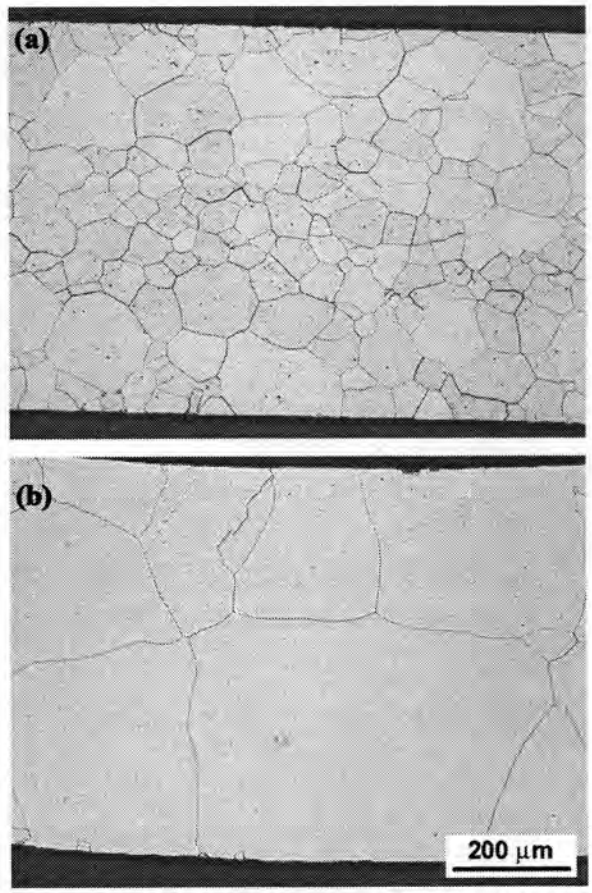


Fig. 36. Optical micrographs showing the grain size of DOP-26 sheet specimens that have been bent to a strain of 2.5% and then annealed for 100 h at 1500°C: (a) away from and (b) near center of bend region.

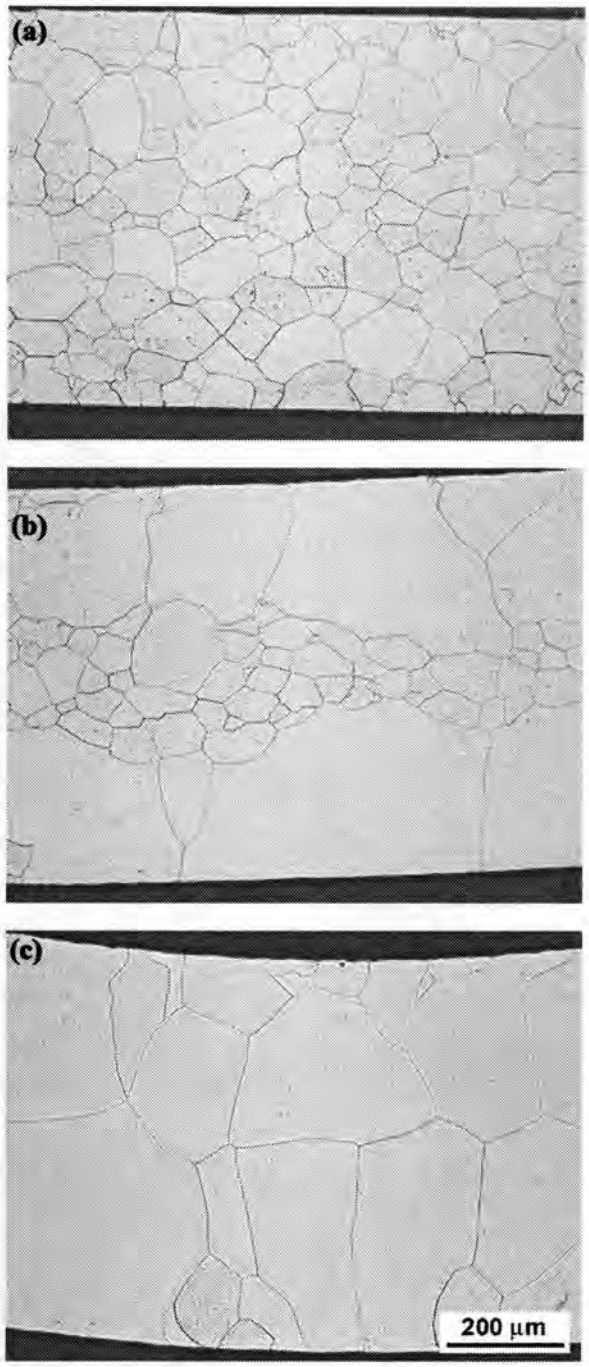


Fig. 37. Optical micrographs showing the grain size of DOP-26 sheet specimens that have been bent to a strain of 4.9% and then annealed for 100 h at 1500°C: (a) away from, (b) at start of, and (c) near center of bend region.

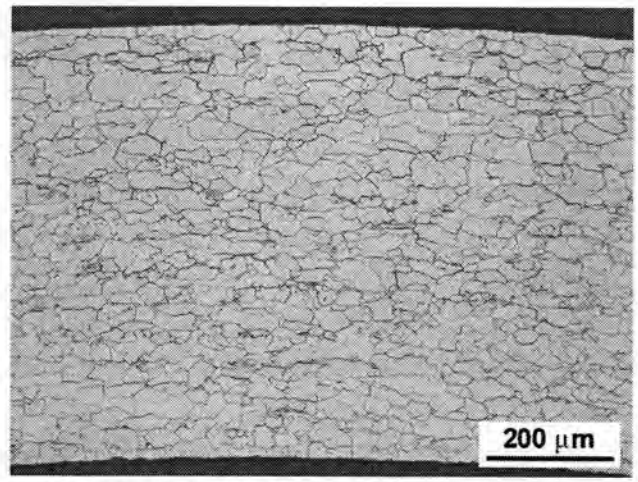


Fig. 38. Optical micrograph showing the grain size of a DOP-26 sheet specimen that has been bent to a strain of 2.5% with no subsequent anneal.

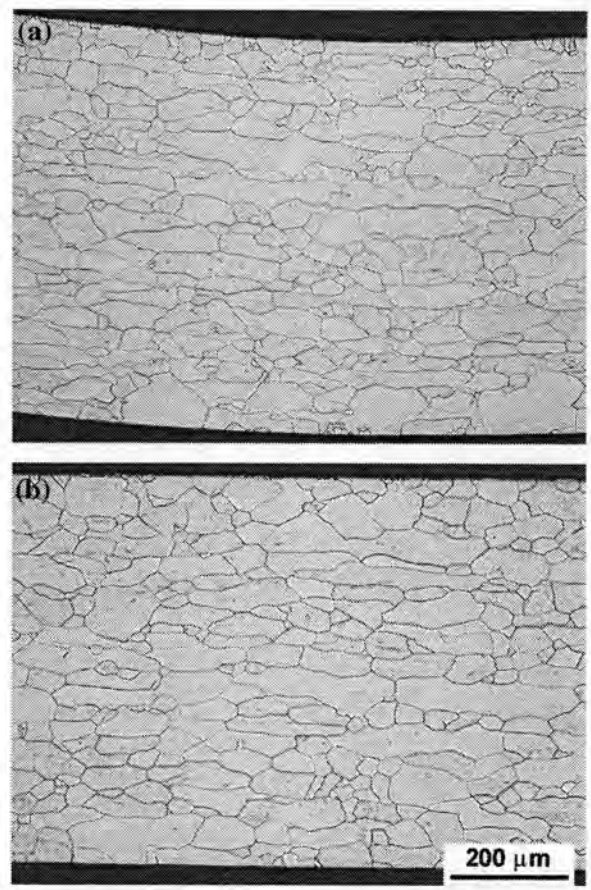


Fig. 39. Optical micrographs showing the grain size of DOP-26 sheet specimens that have been bent to a strain of 2.5% and then exposed to heat treatments to simulate the life of a CVS cup: (a) near center of bend region, (b) away from bend region.

This same trend was observed for those specimens that received an additional heat treatment of 3 min at 1580°C to simulate reentry into the earth's atmosphere. No large grains were observed in any region of the specimen that had been bent to 2.5% strain (see Fig. 41) and the average grain size was 28 and 31 μm in areas away from the bend and in the bend, respectively. The specimen bent to 5% strain (Fig. 42) had a band of larger grains along both the inner and outer surface of the bend region that appeared to extend approximately 125-150 μm into the cup. The grains were uniform in size outside of this strained region with an average grain size of approximately 26 μm.

CALCULATION OF GRAIN SIZE OF DOP-26 CLADS IN THE F-5 GENERATOR

Recently, DOE has begun to assess options for providing power systems for NASA's Ice/Fire missions. One option is the use of the F-5 generator which has been in storage at Mound Laboratories with full fuel complement for almost 15 years. It was therefore necessary to approximate the effect of this

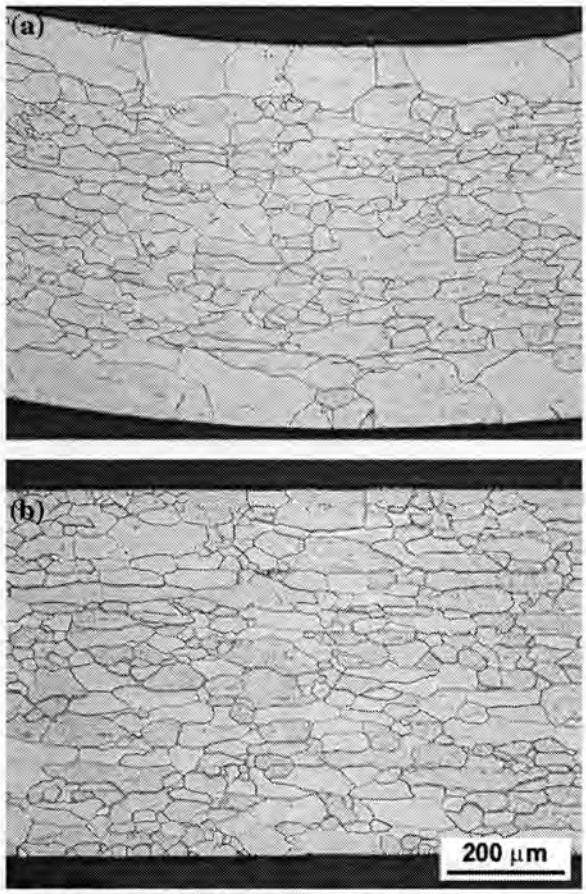


Fig. 40. Optical micrographs showing the grain size of DOP-26 sheet specimens that have been bent to a strain of approximately 5% and then exposed to heat treatments to simulate the life of a CVS cup: (a) near center of bend region, (b) away from bend region.

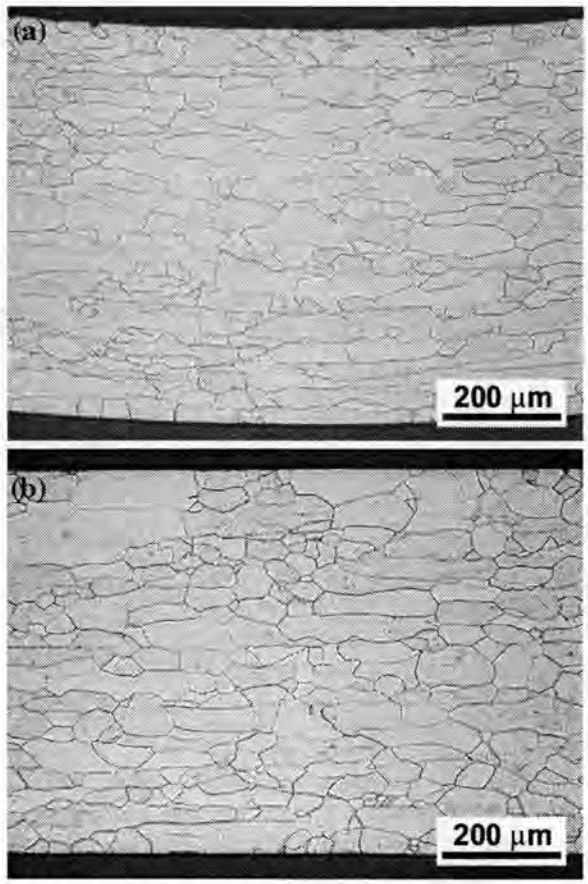


Fig. 41. Optical micrographs showing the grain size of DOP-26 sheet specimens that have been bent to a strain of 2.5% and then exposed to heat treatments to simulate the life of a CVS cup and reentry into earth's atmosphere: (a) near center of bend region, (b) away from bend region.

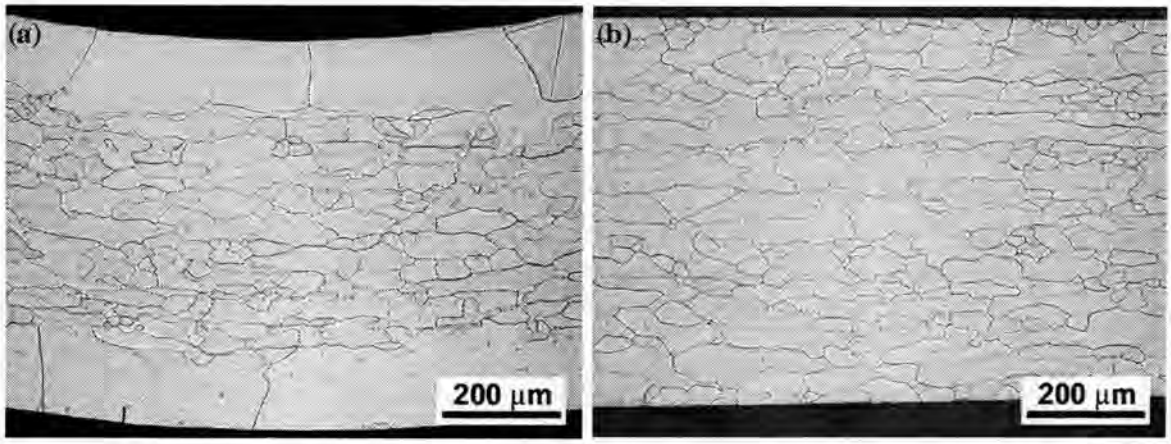


Fig. 42. Optical micrographs showing the grain size of DOP-26 sheet specimens that have been bent to a strain of 5% and then exposed to heat treatments to simulate the life of a CVS cup and reentry into earth's atmosphere: (a) near center of bend region, (b) away from bend region.

length of storage on the grain growth of the DOP-26 material used in that unit to encapsulate the PuO_2 fuel. In order to do that, an equation was developed to describe grain growth in DOP-26 iridium. This equation was then used to estimate the grain size of the DOP-26 material in the F-5 generator.

Since the F-5 generator has been sitting with a full load of PuO_2 fuel, any grain growth that it has experienced has occurred in a low-pressure oxygen environment. Grain growth in oxygen occurs by the dissolution of the pinning particles in DOP-26 (the Ir_5Th intermetallic precipitates), followed by the outward diffusion of thorium along the grain boundaries, and eventual reaction with oxygen to form ThO_2 particles on the outside surfaces [6,11]. This process results in the near-surface grains growing at a faster rate than those in the interior, unlike in vacuum where grain growth is uniform through the sheet thickness. Over the last several years, grain growth studies have been conducted at ORNL on DOP-26 iridium sheet in low-pressure oxygen (1.3 and 13.3 mPa), at temperatures in the range 1230-1330°C and for times up to 3000 h [11]. From these studies, the *average* grain sizes transverse to the sheet thickness were used to develop the grain growth equation.

In addition to the shorter-term (< 3000 h) ORNL data, grain sizes were available [16] for two plutonia-fueled clads that were shelf-aged at Los Alamos National Laboratory (LANL) for 50,000 h at an estimated temperature of ~1287°C [17]. The temperature dependence of the equilibrium oxygen pressure over plutonia ($PuO_{1.98}$) can be described by the following expressions [18]:

$$\log P(O_2)\ (\text{atm}) = 3.933 \times 10^{-4} - 52897/T\ (\text{K})\ ,$$
$$\log P(O)\ (\text{atm}) = 13.852 - 39715/T\ (\text{K})\ .$$

The O_2 and O pressures in the fueled clads during shelf aging can be calculated from these equations to be 1.1×10^{-29} Pa and 2.5×10^{-7} Pa, respectively.

Theory of Grain Growth

If one considers the balance of surface tension forces at a triple point (Fig. 43), mechanical stability requires that the included angle $\theta = 120°$. In general, only hexagonal grains have average internal angles of 120° (Fig. 43); grains with more than 6 sides have $\theta > 120°$, and grains with fewer than 6 sides have $\theta < 120°$. To more closely approach the equilibrium angle of 120°, grain boundaries in non-six-sided grains tend to become curved (inwards or outwards depending on whether they have more or less than 6 sides). As a result, grains with fewer than 6 sides (which tend to be the smaller grains), have boundaries that are concave towards the grain interior, whereas the larger grains with more than 6 sides tend to be convex (hexagonal grains tend to have flat boundaries).

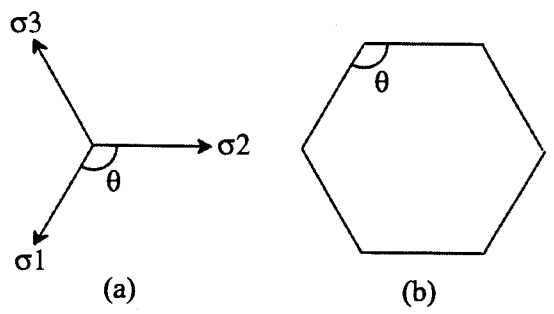


Fig. 43. Schematic 2-D diagrams showing (a) surface tension forces at a triple point and (b) a hexagonal grain with internal angle $\theta = 120°C$.

Grain boundary motion is the result of atomic diffusion across a curved boundary from the concave to the convex side. In other words, boundaries move towards their centers of curvature. This process results in the bigger grains (which have convex boundaries) getting bigger still and the smaller grains (which have concave boundaries) getting smaller and eventually disappearing. The average grain size thus increases. In normal grain growth, the rate at which grain size increases is proportional to boundary curvature, which in turn is inversely proportional to grain size (smaller grains have fewer sides so their edges have to curve more to approach the equilibrium angle of 120°). This dependence can be expressed by the following equation [19]:

$$dD/dt = K/D \ , \tag{1}$$

where D is the grain size at time t and K is a constant. Integrating between D_o, the grain size at time $t = 0$, and D, the grain size at time t, yields the following expression [19]:

$$D^2 - D_o^2 = Kt \ . \tag{2}$$

Experimentally, this square root dependence of grain size on time ($D \propto t^{0.5}$) is not often observed; usually it is seen only in pure metals where the grain boundaries are not pinned by impurities, alloying elements, or second-phase particles. More commonly, it is found that grain growth follows the following equation [19,20]:

$$D - D_o = K t^n \ , \tag{3}$$

where n is the growth law exponent. Usually $n < 0.5$ because grain growth can be hindered by a number of factors. In particular, impurities can slow down diffusion, and dispersed phases can pin the grain boundaries, leading to decreased grain growth [19]. Measured values for n include 0.4 for grain growth in zone-refined lead [21-23] and 0.1-0.3 for aluminum and brass at temperatures above 400°C [24]. Generally, the exponent n tends to increase with temperature and approach the value 0.5 [19].

The temperature dependence of grain growth can be expressed in the above equation through the K term which can be replaced by

$$K = K_o \, e^{-Q/RT} \quad , \tag{4}$$

where Q is an empirical heat of activation for grain growth, T is absolute temperature, and R the gas constant [19].

Grain Growth Projection

The grain growth data of new-process DOP-26 obtained at ORNL are shown in Tables XII and XIII [11]. Grain size measurements were also made at LANL on cups FC0220 and FC0060 (Table XIV) which had been shelf-aged for 50,000 h with plutonium oxide fuel inside [16]. It was estimated that during this shelf-aging, the iridium was exposed to a temperature of approximately 1287°C, produced by the decay of the plutonium fuel [17].

Our earlier study [11] had concluded that the grain growth rates of DOP-26 sheet material were not dependent on oxygen pressure, at least for the two pressures that we investigated (1.3 and 13.3 mPa). However, as mentioned above, the oxygen pressure inside the fueled clads is expected to be on the order of 10^{-7} Pa [18], which is four orders of magnitude lower than the lowest oxygen pressure at which the DOP-26 sheets were annealed at ORNL (it is extremely difficult to obtain base pressures lower than 10^{-4} Pa and, therefore, oxygen pressures lower than 10^{-3} Pa, in laboratory furnaces operating at temperatures around 1300°C). In general, the driving

Table XII. Grain Size of DOP-26 Sheet Material (D2 ingot) at Temperatures of 1230, 1280, and 1330°C in an Oxygen Partial Pressure of 1.3 mPa [11]

Exposure time (h)	Grain Size[a] (μm) at		
	1230°C	1280°C	1330°C
0	22 (641)	23 (687)	19 (330)
144	--	26 (598)	--
167	--	--	26 (238)
335	--	--	30 (215)
380	--	32 (495)	--
756	--	--	38 (165)
764	--	35 (462)	--
768	27 (535)	--	--
1508	--	36 (445)	--
1512	29 (497)	--	--
1522	--	--	38 (172)
2250	--	35 (424)	--
2252	28 (521)	--	--
2282	--	--	56 (238)
3000	31 (460)	40 (376)	52 (133)

[a]Numbers in parentheses are the number of intercepts counted to determine average grain size.

force for Th diffusion from the grain interiors to the outside surfaces is expected to decrease with decreasing oxygen pressure. This would, in turn, slow down the grain growth rates, especially at sufficiently low oxygen pressures where the availability of oxygen atoms is the rate limiting step. It is not known how low the oxygen pressure would have to be before it had a measurable effect on grain growth rates. For the present analysis it is assumed that changes in the oxygen pressure, in the range 10^{-7} to 10^{-3} Pa, do not significantly affect grain growth rates. Therefore, the ORNL data that was used for comparison with the LANL data are those obtained at 1280°C (which is close to the estimated temperature of the fueled clads, 1287°C) and an oxygen pressure of 1.3 mPa (which was assumed to be close enough to the estimated pressure inside the fueled clads, 10^{-7} Pa).

Table XIII. Grain Size of DOP-26 Sheet Material (D2 ingot) at Temperatures of 1230, 1280, and 1330°C in an Oxygen Partial Pressure of 13.3 mPa [11]

Exposure time (h)	Grain Size[a] (μm) at		
	1230°C	1280°C	1330°C
0	--	--	24 (597)
166	--	--	--
170	--	--	35 (408)
332	--	34 (423)	--
338	28 (499)	--	--
674	27 (511)	--	--
696	--	37 (386)	--
720	--	--	40 (362)
1370	27 (546)	--	--
1494	--	33 (429)	--
2186	--	--	--
2285	--	--	--
3000	32 (444)	35 (395)	50 (592)

[a]Numbers in parentheses are the number of intercepts counted to determine average grain size.

Table XIV. Grain Size of Fueled Clads Shelf-Annealed at ~1287°C for 50,000 h [16,17]

Cup Identification	Grain Size (μm)
FC0060 vent	56
FC0060 shield	50
FC0220 vent	47
FC0220 shield	43

Table XV. Grain Growth Constants Determined by Linear Fit of Data to Equation (3)

n	D_o	K	R[a]
0.5	29.5	0.1	0.893
0.4	28.1	0.3	0.910
0.3	25.7	0.9	0.929
0.25	23.9	1.7	0.937
0.2	21.7	3.1	0.940
0.15	19.1	5.7	0.925
0.1	16.8	10.0	0.877

[a]Correlation coefficient for fit of data to equation.

The ORNL grain growth data (Table XII, 1280°C) and the LANL fueled clad data (Table XIV) were fitted to Eq. (3) using various values of the grain growth exponent n. The results are shown in Table XV. The best fit to the data was obtained for $n = 0.2$ which had a correlation coefficient of 0.94 (Table XV and Fig. 44). A similar plot (Fig. 45) of data taken from specimens annealed in 13 mPa of oxygen at 1280°C (Table XIII) showed that both the 1.3 and 13 mPa data fit well with the long-term LANL data.

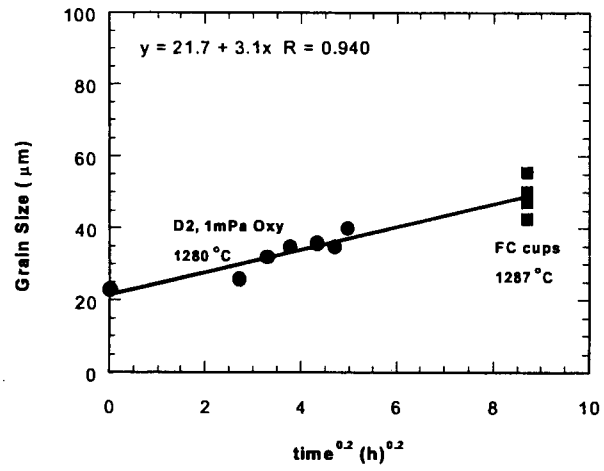


Fig. 44. Line fit for grain size data using n = 0.2.

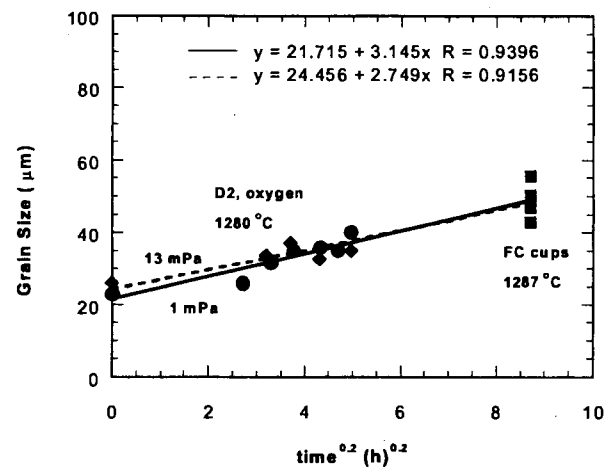


Fig. 45. Comparison of line fits of grain size data for 1 and 13 mPa oxygen (n = 0.2 was used for both line fits).

The ORNL data can be used to determine the temperature dependence of the grain growth exponent, *n*. Using Eq. (3) and various values for *n*, the grain growth data listed in Table XII for new-process DOP-26 annealed in an oxygen pressure of 1.3 mPa at temperatures of 1230, 1280, and 1330°C were plotted. The *n* values that resulted in the best fits (highest correlation coefficients) are listed in Table XVI along with the corresponding D_o and *K* values. As expected [6], *n* is seen to increase with temperature.

In summary, data generated over the last several years for the grain growth of new-process DOP-26 annealed in low-pressure oxygen at temperatures ranging from 1230 to 1330°C were evaluated in terms of the grain growth law expressed in Eq. (3) above. Included in this analysis were grain size measurements taken from optical micrographs of fuel cups FC0220 and FC0060 which had been shelf-aged at Los Alamos for 50,000 h with plutonium oxide fuel inside. The ORNL and LANL grain size data (at 1280 and 1287°C, respectively) when plotted together fit the following equation:

$$D = 21.7 + 3.1t^{0.2},$$

where D is the grain size (in mm) at time t (in h).

Table XVI. Grain Growth Constants for Line Fits of Data at 1.3 mPa Oxygen Pressure

Temperature (°C)	n	D_o	K	R[a]
1230	0.35	22	0.5	0.967
1280	0.35	23	1.0	0.953
1330	0.5	18	0.7	0.960

[a]Correlation coefficient for fit of data to equation.

Grain Growth in the F-5 Generator

The F-5 generator has been in storage at Mound Laboratories for almost 15 years (5426 days = 130,224 h) at a temperature of approximately 920°C. For most of that time it has been exposed to inert gas (argon) with only short times at vacuum or other atmospheres. Gas taps have revealed little exposure to oxygen over time because any oxygen present reacts with the molybdenum foils and graphitics.

Table XVII shows grain growth data for old-process DOP-26 (ZR ingot) heat treated in vacuum. This data is shown plotted in Figs. 46 and 47 according to Eq. (3) using n values of 0.5 and 0.2, respectively. This data is from sheet material, but grain size studies of the Y-12 produced clad vent set cups confirm that grain growth in the cup material is comparable to that of sheet material, especially for temperatures

Table XVII. Effect of Annealing Time on Average Grain Size (ST Direction) of ZR (old process) and D2 (new process) DOP-26 Alloys [11]

Temperature (°C)	Annealing Time (h)	Grain Size[a] (μm)	
		ZR	D2
1300	1	15 (268)	24 (322)
	2	17 (232)	--
	4	19 (94)	25 (166)
	18	21 (128)	28 (289)
	70	24 (329)	31 (340)
	250	34 (223)	30 (404)
	480	32 (282)	36 (274)
	1000	34 (146)	42 (138)
1400	1	23 (340)	29 (397)
	2	26 (208)	30 (134)
	4	24 (317)	34 (300)
	16	31 (310)	39 (283)
	100	52 (218)	50 (210)
	250	63 (40)	56 (153)
	520	74 (59)	63 (73)
	1000	99 (109)	72 (159)
1500	0.2	22 (367)	30 (336)
	0.5	25 (331)	32 (302)
	1	30 (225)	33 (80)
	2	36 (310)	39 (123)
	4	39 (233)	42 (192)
	18	59 (165)	52 (194)
	100	82 (127)	71 (153)
1600	0.2	30 (352)	37 (292)
	0.5	41 (169)	43 (134)
	1	45 (182)	48 (74)
	4	90 (70)	77 (161)

[a]Numbers in parentheses indicate number of intercepts counted for grain size determinations.

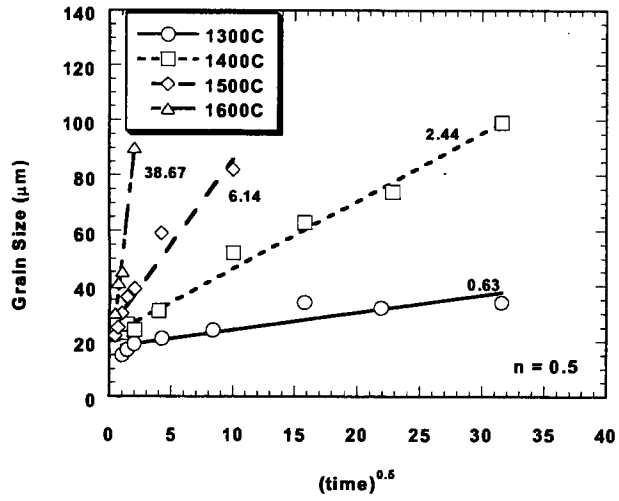


Fig. 46. Line fit of grain size data using Eq. (3) and n = 0.5.

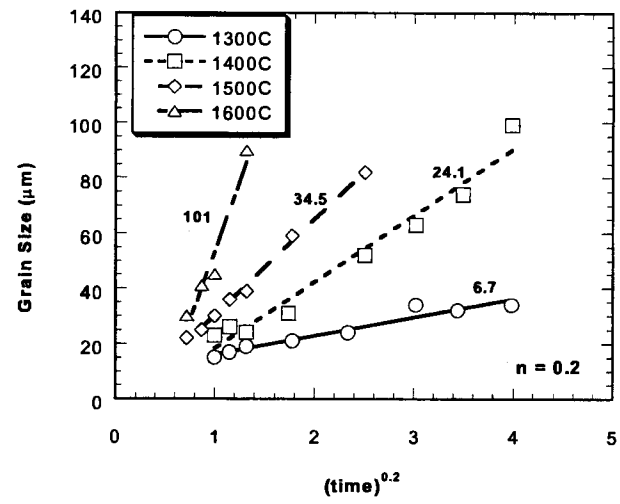


Fig. 47. Line fit of grain size data using Eq. (3) and n = 0.2.

below 1500°C [11]. According to Eq. (3), the slopes of the line fits in Figs. 46 and 47 are the values K for each temperature. The values determined for K at each temperature are noted in the figures. The temperature dependence of grain growth can then be determined using Eq. (4). The results are shown in Table XVIII and Fig. 48.

In Fig. 48, the straight-line fits of the data for n-values of 0.5 and 0.2 have been extrapolated down through the temperature of interest for the F-5 generator (920°C). Using this line, the y-values of the points at 920°C were determined to be

$$\ell n\, k\,(0.5) \approx -8.25\,,$$
$$\ell n\, k\,(0.2) \approx -2.97\,.$$

Table XVIII. Parameters for Determination of Temperature Dependence of K Using Eq. (4)

n	T (°C)	K	ln K	1/RT (10^{-5} mole/J)	Q (kJ/mole K)
0.5	1300	0.63	-0.462	7.64	324
	1400	2.44	0.892	7.19	
	1500	6.14	1.815	6.78	
	1600	38.67	3.655	6.42	
0.2	1300	6.71	1.904	7.64	208
	1400	24.14	3.184	7.19	
	1500	34.46	3.540	6.78	
	1600	100.70	4.612	6.42	

Using these values and the starting grain size of 16 μm in Eq. 3 above, the grain sizes at 920 °C were determined to be

$$D\,(n{=}0.5) = 16.1\ \mu\text{m},$$
$$D\,(n{=}0.2) = 16.5\ \mu\text{m}.$$

Using the available data and Eqs. (3) and (4), the temperature at which grains will grow to 17 μm if left on the shelf for 5426 days was calculated to be 961 °C (for n=0.2) and 1022 °C (for n=0.5). So if the F-5 generator has stayed below 960 °C for most of its life, then little or no grain growth should have occurred.

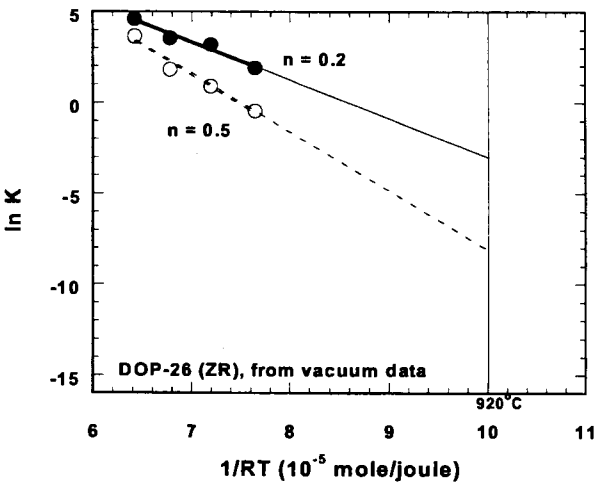


Fig. 48. Determination of temperature dependence of K using Eq. (4) for n values of 0.5 and 0.2.

SUMMARY AND CONCLUSIONS

Because grain size is such an important factor in determining the high-temperature tensile impact ductility of DOP-26 iridium, studies have been conducted to determine the grain growth characteristics of clad vent set cups not only as a function of the heat treatment conditions but also as a function of the position in the cup with respect to both the rolling direction and the cup geometry. These studies were conducted on cups produced at both the Y-12 Plant and ORNL. For those cups produced at ORNL, pilot-production, qualification-production, and maintenance-production cups were included in our studies.

Based on heat treatments of 1 h at temperatures of 1200 to 1800°C and for various times at 1400 and 1500°C, the grain growth characteristics of specimens taken from clad vent set cups produced at the Y-12 Plant (fabricated from new-process DOP-26 blanks) appears to be the same as those of specimens taken from the blanks from which the cups were fabricated. As a function of temperature for one-hour anneals, significant increases in the average grain size occurred only for temperatures at and above 1600°C. As a function of annealing time, the grain diameter increased gradually from an initial size of approximately 20 μm to a diameter of 40-50 μm after 520 h at 1400°C and to 125-150 μm after 240 h at 1500°C. There also does not appear to be any difference in grain growth between specimens taken from different locations in the cup relative to the rolling direction or cup geometry.

In 1996 production of clad vent set cups was moved to ORNL and pilot-production, qualification-production, and maintenance-production cups were subsequently produced. The grain growth behavior of these cups (fabricated from new-process DOP-26 blanks) was studied and the results were compared to the results from cups produced at Y-12. Based on heat treatments of one hour at temperatures between 1200 and 1800°C, the grain growth characteristics of ORNL qualification-production cups are similar to those of Y-12-produced cups and to the blanks from which the cups are fabricated. There does not appear

to be any significant difference in grain growth between specimens taken from different orientations with respect to the rolling direction or from different locations in the cup.

However, in the grain growth study as a function of annealing time, non-homogeneous grain growth was observed in pilot-production cups for the 515 h anneal at 1400°C and for all anneals longer than 1.5 h at 1500°C. For these anneals, some regions of the specimens contained small grains (even much smaller than expected for the heat treating conditions) while other regions had much larger grains. There did not appear to be any correlation between the presence of non-homogeneous grain growth and different regions of the cup or in the orientation of the specimen with respect to the rolling direction. The reason for the non-homogeneous grain growth observed in ORNL pilot-production cups is not fully understood. None of the DOP-26 sheet material that has been examined has shown this type of grain-growth behavior, nor was it observed in the cups produced at Y-12 or the two ORNL qualification cups examined in this study. These results suggest that the non-homogeneous grain growth is related either to inhomogeneities in the DOP-26 sheet material from which the cups were formed (the blanks were not flight quality) or to strain introduced as a result of some cold-working operation performed on the ORNL pilot-production cups during fabrication.

Because it was believed possible that the use of too much hold-down force for the post-grit-blast scribing operation during pilot production could have resulted in residual stress that led to the non-homogeneous grain growth, two cups were formed from blank assemblies and processed in an attempt to duplicate the cold work imparted during pilot production. One cup was studied after a recrystallization anneal of 1 h at 1375°C followed by the cold working operations, while the other cup was recrystallized, cold worked, and then given air burnoff (4 h at 635°C) and vacuum outgas (1 h at 1250°C) heat treatments. Although a few large grains were observed scattered throughout the specimens after a heat treatment of 100 h at 1500°C, the average grain sizes of both cups were 53-69 μm, a range which is comparable to the grain size of the cups produced at the Y-12 Plant. In addition, two ORNL maintenance production cups containing a small crease down the side (caused by the presence of dried and hardened lubricant on the deep-drawing die) had only a few large grains in the area of the crease. The average grain sizes of all four of these ORNL cups were comparable to cups produced at Y-12 receiving the same heat treatment.

In order to determine if induced strain during fabrication could have produced the non-homogeneous grain growth, three avenues were investigated, each using 100 h at 1500°C as a standard heat treatment to reveal the presence or absence of non-homogeneous grain growth. A study was conducted to determine if strains of up to 5% induced in DOP-26 blank material by bending, rolling, or tensile straining (to simulate events in the fabrication of the cups from blanks) could produce this type of non-homogeneous grain growth. The results showed that tensile strains of more than approximately 2.5% or bending strains of more than approximately 1% could produce non-uniform microstructures containing regions of much larger than average grain size. Compression strains (here simulated by rolling of the sheet) could also result in grain growth for very low strains (<1%). However, in this study all specimens with compression strains displayed uniform grain growth throughout the specimen.

Additional specimens were cut and bent to produce approximately 2.5 and 5% strain. These specimens were given the same recrystallization, air-burnoff, and vacuum-outgas thermal treatments as are given to CVS cups during fabrication, plus heat treatments to simulate one month of fueled CVS storage life, three months in space, and reentry into the earth's atmosphere. After the simulation of cup fabrication and short-term storage and space life, average grain sizes were still near the grain size of the as-recrystallized cup. However, a few large grains were present on the outer and inner surfaces of the bend that produced the 5% strain. After the simulation of reentry into earth's atmosphere, the large grains in the bend of the specimen with 5% strain had grown to a thickness of approximately 125-150 μm on each surface, while elsewhere the average grain size was approximately 26 μm. The specimen with 2.5% strain contained no unusually large grains and had an average grain size of 28-31 μm.

In summary, after studying many ORNL-produced clad vent set cups (from pilot-production, qualification-production, and production-maintenance) and attempting to simulate cup fabrication and short-term use, the explanation for the non-homogeneous grain growth in the pilot-production cups is still not completely understood. Studies of induced strain showed that excessive grain growth could result from cold work that might have been introduced in the cups during fabrication and that was not adequately relieved by subsequent stress relief heat treatments. However, grain growth would only be of concern if the cup is then exposed for long times (much longer than the 3 min expected for reentry into earth's atmosphere) to temperatures at or above 1500°C. One possible explanation that was not addressed in this study is that the non-homogeneous grain growth in the pilot-production cups could have been a result of inhomogeneities in the DOP-26 sheet material. During pilot production, non-flight-quality sheet material was used for forming the cups. Inhomogeneities in the base composition or in the distribution of the Ir_5Th precipitates could affect grain growth considerably.

The non-homogeneous grain growth was observed only in two ORNL pilot-production cups. Subsequent investigations of the ORNL qualification-production cups and the production-maintenance cups showed no evidence for non-homogeneous grain growth. Similarly, it was not observed in any of the cups produced at the Y-12 Plant or the DOP-26 blanks that have been examined to date. It is therefore believed that this was a one-time event that should not occur in future flight-quality cups.

Grain size data generated over the last several years for new-process DOP-26 annealed in low-pressure oxygen, along with measured grain sizes for fuel cups FC0220 and FC0060 (shelf aged at Los Alamos for 50,000 h with plutonium oxide fuel inside), were used to determine an equation for grain growth. When plotted together, the data were found to fit best to the following equation:

$$D = 21.7 + 3.1t^{0.2},$$

where D is the grain size (in mm) at time t (in h). These results and data from vacuum-annealed DOP-26 were used to predict the grain size of the F-5 generator which has been in storage at Mound Laboratories for approximately 15 years. It was found that if the F-5 generator has stayed below 960°C for most of its life, then little or no grain growth should have occurred.

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