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Y-1931

CAST COMPOSITE ELECTRODES FOR ARC MELTING A URANIUM-NIOBIUM-ZIRCONIUM ALLOY

D. W. Hackett

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UNION CARBIDE

OAK RIDGE Y-12 PLANT
OAK RIDGE, TENNESSEE

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ABSTRACT

Composite electrodes have been arc melted to form ingots of the uranium-7 1/2 niobium-2 1/2 zirconium alloy (Mulberry). These electrodes were produced by casting uranium around strips of niobium and zirconium. This alloy has been compared analytically to Mulberry produced by arc melting using elemental strip electrodes. The cast electrode produced alloy well within the required limits and is chemically equal to, and even possibly superior to, alloy using a strip electrode.

CONTENTS

SUMMARY

A new consumable electrode for arc melting was evaluated as a possible replacement for the strip-type electrode currently used in the processing of a uranium-niobium-zirconium alloy (Mulberry). In the new electrode, the uranium is cast around strips of niobium and zirconium. Arc melting of this electrode produces a satisfactory first-melt ingot. A subsequent second arc melt of four first-melt ingots, welded to form an electrode, improves homogeneity. Analytical samples were taken from first-melt and second-melt ingots, and one second-melt ingot was sectioned at various levels for macroexamination. The macro sections did not have any niobium inclusions, segregation, or other defects, and the analytical data were within the limits required for the Mulberry process. Furthermore, the arc-melting characteristics were superior and the process offers other advantages.

INTRODUCTION

A uranium-7.5 weight percent niobium-2.5 weight percent zirconium alloy (referred to as Mulberry) is now produced in the Oak Ridge Y-12 Plant.[a] In the first of two arc-melting operations for processing this alloy, under the present method, the electrode is made up of seven strips of the elemental constituents in their proper proportions. These strips (~ 4" W x 56"L) are then bonded together across the edges by using the gas tungsten-arc welding process.

A new electrode configuration was investigated in an effort to reduce the scrap loss of the relatively expensive niobium and to eliminate the processing necessary to build the strip electrode. Elimination of two pickling operations and the problems associated with the disposal of the solutions was also desirable. In this study, a method has been devised to cast the uranium around strips of niobium and zirconium to form the first-melt consumable electrode.

Studies in the past have been primarily concerned with chemical inhomogeneity in the first-melt and second-melt ingots, and the various methods to improve it such as altering the design of the strip electrode and varying the arc-melting parameters. Effects of segregation on the physical and mechanical properties of the cast Mulberry have also been studied.

This work is the initial attempt to use a vacuum-induction-cast composite electrode for the first arc melt.

(a) Operated by the Union Carbide Corporation's Nuclear Divison for the US Atomic Energy Commission.

CASTING ELECTRODES FOR ARC MELTING

PRELIMINARY DEVELOPMENT

Vacuum-induction casting of uranium around strips of niobium and zirconium was successfully accomplished on several castings made in the laboratory facilities. The electrodes, approximately two inches in diameter by nine inches long, were then subsequently arc melted to produce miniature first-melt Mulberry ingots. Microexamination, density measurements, and chemical analyses of these ingots were sufficiently encouraging to scale up the process to cast and subsequently arc melt production-size electrodes.

PRODUCTION-SIZE ELECTRODE DEVELOPMENT

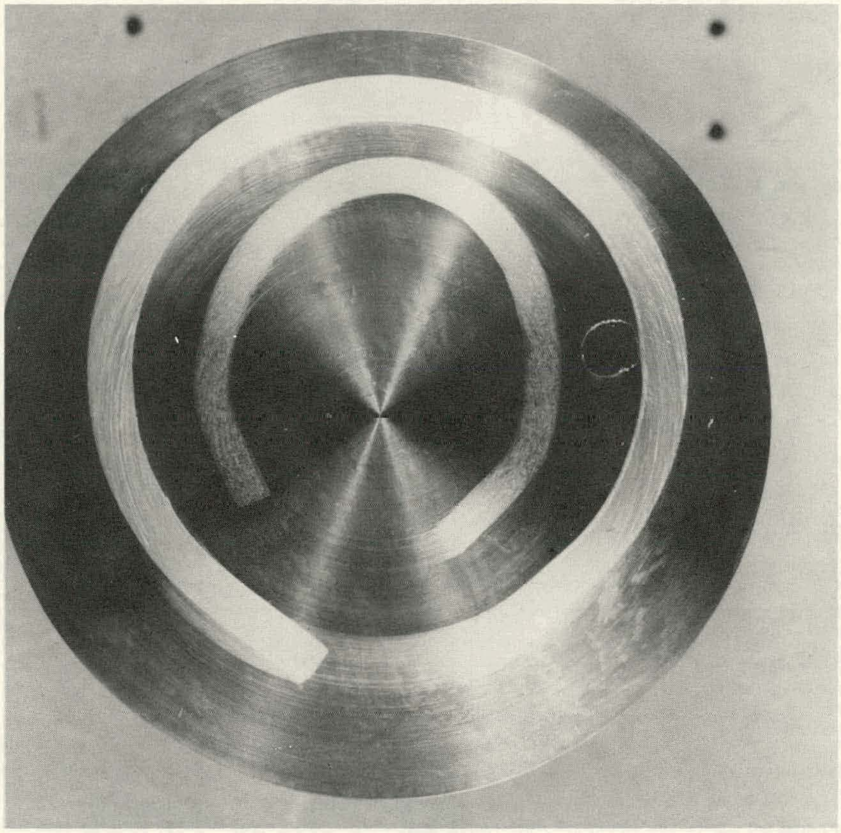
Initially, a mold was designed to "gang cast" four electrodes at one time. A pour was made in a medium-size production furnace to produce three 4-inch-diameter electrodes, 36 inches long. Experience in casting the electrodes in the laboratory furnace had indicated that positioning and maintaining the position of the niobium and zirconium could be a problem. From a few cursory tests it was determined that the niobium and zirconium strips could be readily formed into a circular shape in the as-received condition. Circular strips would not only hold their position better, but would be easier to insert as one piece with less chance of scratching or chipping the mold coating. (If used as flat pieces, the strips would need to be cut into several pieces to fit into the cavity.) A cross section through a typical composite electrode can be seen in Figure 1.

Casting parameters for this and subsequent runs in the production furnace were:

Charge:	350 kilograms of high-purity depleted uranium (Derby metal)
Coating:	Zirconite Mold Wash A (for the mold); Mold Wash B (for the crucible) - brushed
Pressure:	600 micrometers (approximately)
Pour Temperature:	1,285° C (by optical pyrometer)
Mold Temperatures:	500° C (bottom); 1,100° C (top) (by thermocouple)

The furnace was held at 1,285 ± 10° C for 20 minutes prior to pouring and back filled with argon 20 minutes after pouring was completed. Figure 2 provides a schematic view of the mold and crucible setup for these castings.

At the bottom of the electrode, the niobium extended one inch below the end of the uranium and zirconium to establish equilibrium melting conditions more rapidly during the initial arc melting. For these and all subsequent electrodes, the alloying elements were sheared to widths calculated from nominal thicknesses, namely: 0.230 inch (niobium) and 0.175 inch (zirconium), to yield weight percentages of 7.5 and 2.5 percent, respectively. The strips were not trimmed to an exact weight, but the weights of the strips were recorded for information purposes.



MS-71-0461-2
Figure 1. CROSS-SECTION VIEW OF A COMPOSITE ELECTRODE WITH THE URANIUM CAST AROUND THE FORMED ZIRCONIUM AND NIOBIUM STRIPS. (Full Size)

The cast surfaces of the electrodes were cleaned by grit blasting, and a two-inch-diameter uranium adapter was fusion welded to the top. No machining or cutting of the electrode was performed prior to the welding.

During arc melting of the four-inch electrodes, arcing to the side walls of the five-inch cup occurred. Figure 3 is a view of an electrode that arced to the side. Approximately one third of the niobium had melted, but more of the uranium had melted due to the arcing. One of the three electrodes was melted into a six-inch cup (see Figure 4). Prior to completion, the melting was stopped. As can be seen, the uranium was melted at an even rate about one inch ahead of the niobium and 1/4 inch ahead of the zirconium. This even "burn off" was characteristic of all but a few electrodes in which the clearance between the electrode and the cup wall was 1/2 inch or less, and caused arcing.

Three additional 4-inch-diameter electrodes were cast using the same mold. Prior to arc melting, extra care was taken to ensure that the electrodes were quite straight and centered in the cups. The melting amperage was also increased. The arc still tended to jump to the side wall and was not stable or well defined on the television screen. The remaining four-inch-diameter electrodes were melted into six-inch-diameter cups with one being a melt in which one and a half electrodes were welded together and sucessfully arc melted.

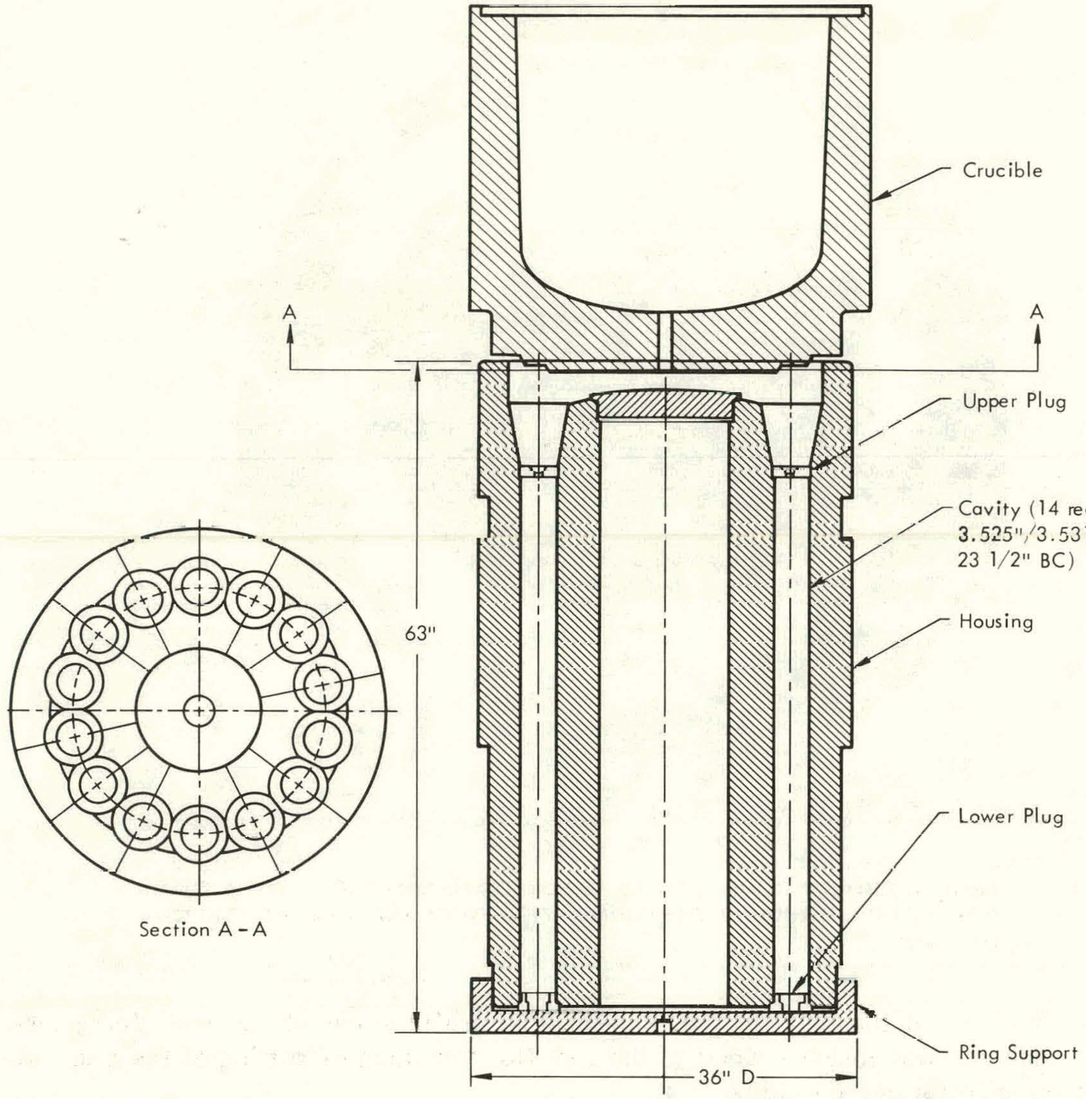


Figure 2. COMPOSITE ELECTRODE MOLD AND CRUCIBLE. (14 Cavities; Material - Graphite)

Graphite sleeves were inserted into the four-inch-diameter mold to reduce the diameter to 3 1/2 inches, and five electrodes were successfully cast into a five-inch-diameter cup. Four of these first-melt ingots were welded to make up a single second-melt electrode. Table 1 lists the sampling and analytical data for the ingot that was arc melted from this second-melt electrode.

The two arc-melting operations were accomplished in an air atmosphere at somewhat less than 100 micrometers pressure using water-cooled copper cups and back filling with argon during the cool down. The parameters were nominally as follows:

Five-Inch-Diameter Ingots

Amperage at Start	2,500 - 2,800 amperes
Amperage during Melting	4,200, then reduced to 3,200 amperes
Voltage	23 - 25 volts
Electrode	3 1/2 inches in diameter; 121 kilograms in weight
Ingot	5 inches in diameter; 110 kilograms in weight
Time to Melt	28 minutes

Seven-Inch-Diameter Second-Melt Ingots

Amperage	7,800 then reduced to 3,000 amperes
Voltage	24 - 32 volts
Electrode[(b)]	5 inches in diameter; 360 kilograms in weight
Ingot	7 inches in diameter; 300 kilograms in weight
Time to Melt	35 minutes

A new mold was designed to cast 14 electrodes (3 1/2" D x 48" L; see Figure 2) using one of the large furnaces in the uranium foundry. Two sets of seven electrodes were to be cast; one set with flat strips of niobium and zirconium and the other with the strips formed into semicircles, as indicated in Figure 1. To obtain the proper proportions with flat strips, four niobium strips were placed to form a square, with two zirconium strips across the opposite corners.

The surface area of the flat strips was the same as that of the formed strips, excluding the edges. The 14 electrodes were to be used to statistically evaluate the electrode design.[(c)] The chemistries of the two types were to be compared to each other and to the production Mulberry stream in the first-melt and second-melt ingot stages.

Casting parameters were the same except that the uranium charge was 1,700 kilograms, the average pressure 450 micrometers, and the mold temperature somewhat higher. The actual mold temperature was not measured. Due to a short coil in this large furnace, the temperature at the top of the mold was sufficiently high to produce a reaction between the molten uranium and coated graphite. This problem had not occurred in any of the previous runs.

(b) Electrode was made up of five 5-inch-diameter ingots that were cropped and welded.

(c) Memorandum from C. W. Holland to D. W. Hackett; August 30, 1971.

There was a small amount of niobium and zirconium pickup in the chip samples machined from the bottom of the risers above the electrodes. This action could be controlled in several ways, including keeping the mold temperature lower.

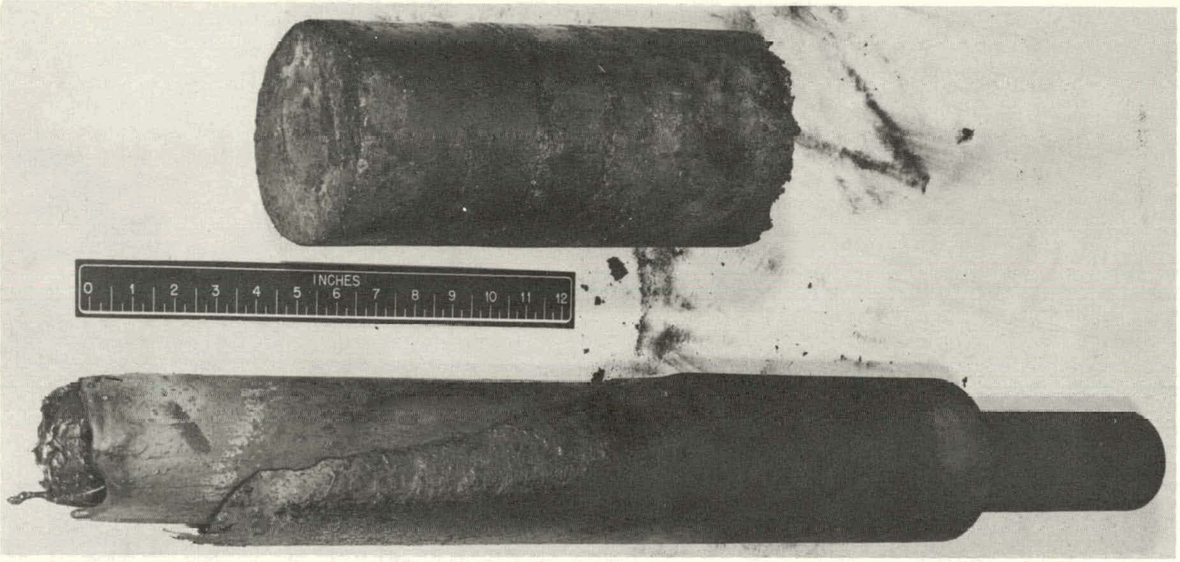


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Figure 3. A FOUR-INCH-DIAMETER COMPOSITE ELECTRODE WHICH ARCED TO THE SIDE OF A FIVE-INCH-DIAMETER CUP, AND THE RESULTANT CASTING. (Note the Unusual "Burn Off" of the Uranium)

Variations in the uranium, niobium, and zirconium levels are shown graphically in Figure 5 for the 7-inch-diameter second-melt ingots and the four 5-inch-diameter ingots making up each of them. In Figure 6, the variation of each component along the length of the larger ingots is presented. The sampling plan and analytical values for both size ingots are included in the Appendix (Figure A-1). Mulberry specification SG-225510 gives the limits as follows: niobium, 6.8 - 8.2%; zirconium, 2.25 - 2.5%; and uranium, 89.0% (minimum).

It was known that the niobium and zirconium weight percentanges would be on the high side because the weights of the strips were greater than calculated from the nominal thicknesses. Also, the uranium shrinkage was approximately three percent by weight greater than anticipated. Since the main concern was the variation in composition within an ingot and from ingot to ingot and not attaining an exact composition, no efforts were made to change the weights of the strips which had been cut and formed. Also, there was little doubt that the charge weight needed to yield mean percentages of 7.5 for the niobium and 2.5 for the zirconium could be more readily determined with the results obtained from this run. This conclusion was verified in a final run. Also, it was proven that if approximately one inch, more or less, had been removed from either end, the variability in the 5-inch ingot and, hence, the 7-inch second-melt ingots would have been appreciably reduced. Chemical data for these final two ingots are presented in Table 2. The top three inches of these ingots did not have any niobium and the top two inches contained no zirconium. The change was made so that, in normal operation, if any of the stub end of the electrode remained after arc melting, only uranium would be scrapped.

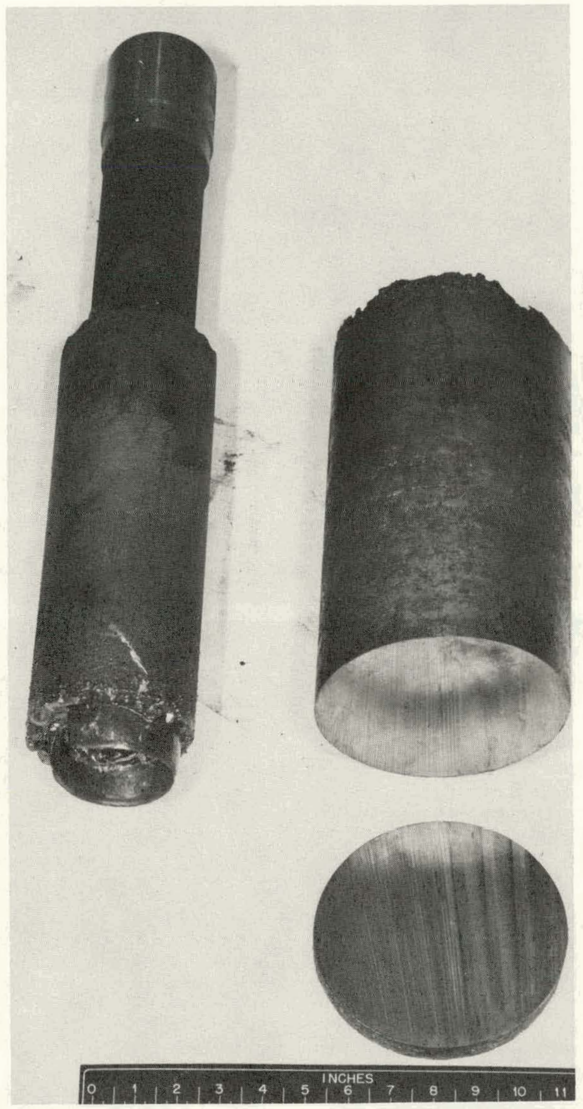

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Figure 4. MELTING THE FOUR-INCH-DIAMETER COMPOSITE ELECTRODE INTO A SIX-INCH-DIAMETER CUP. (This Operation was Accomplished without any Side-Wall Arcing; and Melting the Uranium, Zirconium, and Niobium was very Uniform)

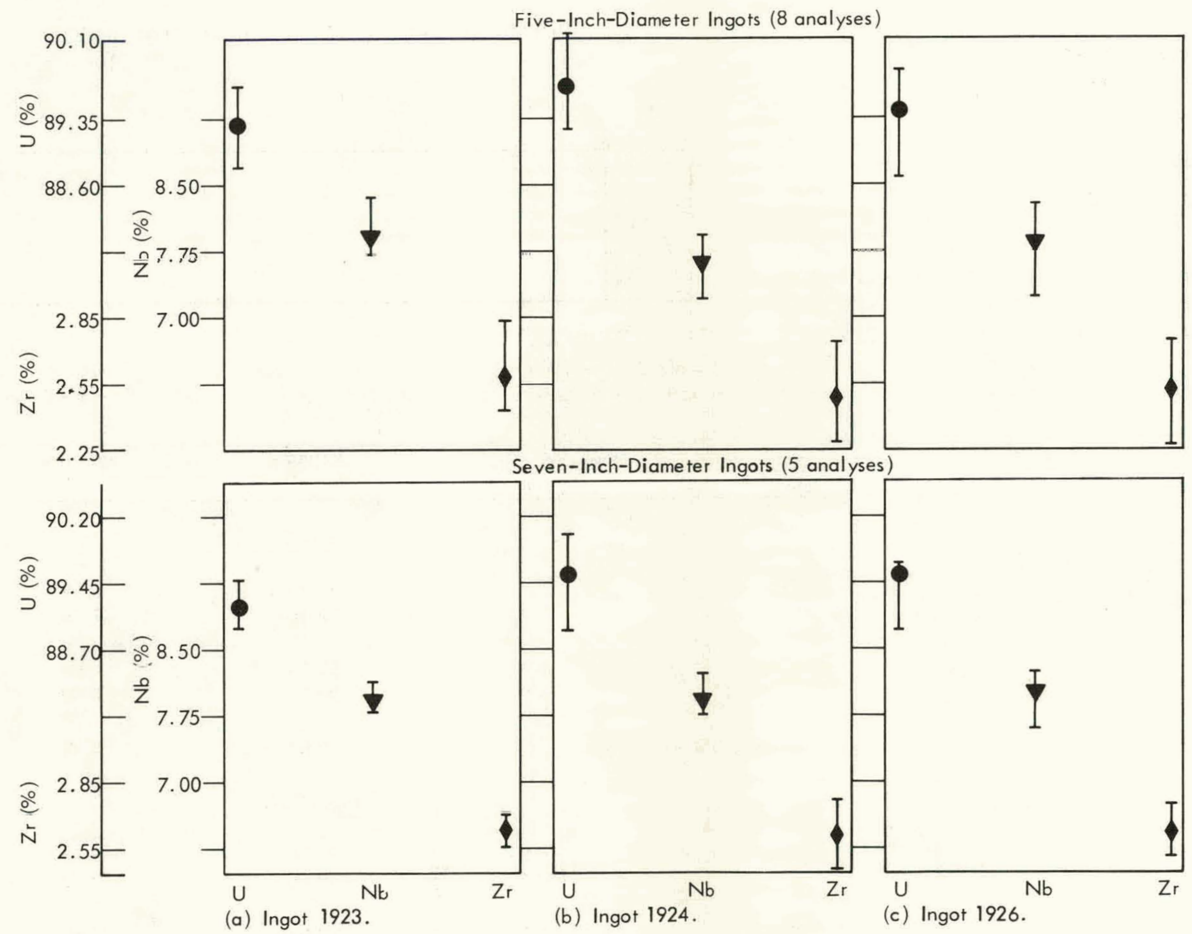


Figure 5. AVERAGE AND RANGE OF COMPOSITION OF TWO SIZES OF SECOND-MELT INGOTS.

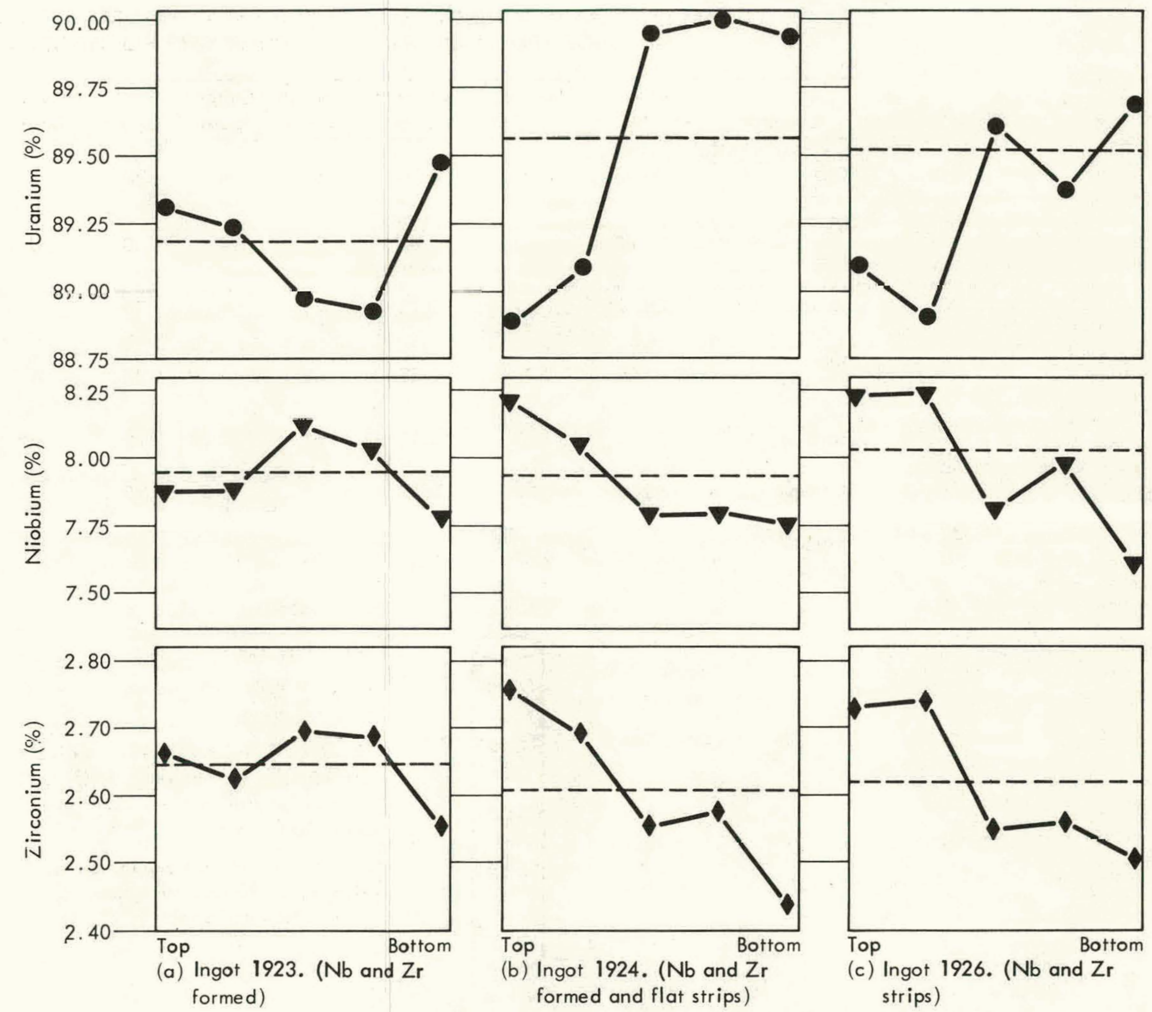
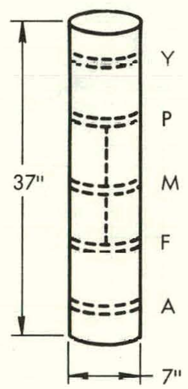


Figure 6. VARIATION IN COMPOSITION FROM TOP TO BOTTOM. (Seven-Inch Second-Melt Ingots)

Table 1

SAMPLING PLAN AND CHEMICAL DATA FOR THE INITIAL SECOND-MELT INGOT MADE FROM INDUCTION-CAST COMPOSITE ELECTRODES

Location		Uranium (%)	Niobium (%)	Zirconium (%)	Carbon (ppm)	Copper (ppm)	Iron (ppm)	Hydrogen (ppm)	Nitrogen (ppm)	Oxygen (ppm)
Top	Y	89.43	8.03	2.70	80	4	20			
Mid Top	P	89.40	7.98	2.64	75	4	20	6	2	31
Middle	M	89.13	7.92	2.70	71	6	20			
Mid Bottom	F	88.98	7.90	2.62	82	4	20	4	3	31
Bottom	A(1)	90.02	7.65	2.48	61	4	20			
Average		89.39	7.90	2.63	74	4.5	20			
Process Certification(2)		89.87	7.66	2.44	48	11	22			



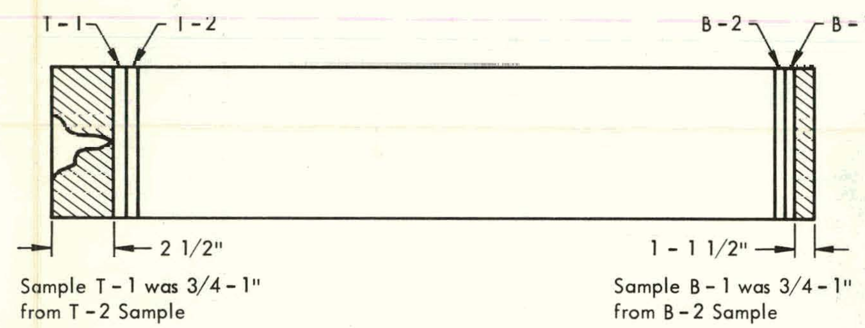
Sampling Procedure:

1. Crop off ends; take normal cut used on production.
2. Cut 3/4± 1/4-inch slices as indicated and identify (5 required). Mill 0.005 inch chips from surface for the analytical sample.
3. Cut center two sections in half and identify as MP and MF. Grind surface for macroexamination.

(1) Sketch showing sample location.
(2) Current Mulberry process averages by T. R. Harvey, August 10, 1971.

Table 2

CHEMICAL ANALYSIS OF THE FINAL TWO INGOTS



Location	Sample	Uranium (wt %)	Niobium (wt %)	Zirconium (wt %)
		Ingot 2012		
Top	T-1	89.86	7.44	2.42
	T-2	89.60	7.55	2.45
Bottom	B-2	89.63	7.62	2.62
	B-1	89.31	7.68	2.57
Average B-2 and T-2		89.62	7.58	2.54
		Ingot 2013		
Top(1)	T-1	91.74	5.72	2.13
	T-2	90.15	7.22	2.38
Bottom	B-2	89.65	7.48	2.53
	B-1	89.37	7.65	2.66

(1) Did not cut a sufficient amount from the top of Ingot 2013.

Electrode 2012 had a stub end of about seven inches remaining, while its companion, Electrode 2013, was melted to the weld attaching it to the adapter. It is readily apparent that, with this modification, more than the normal 2 1/2 inches would have to be cut off if the electrode is melted to the top.

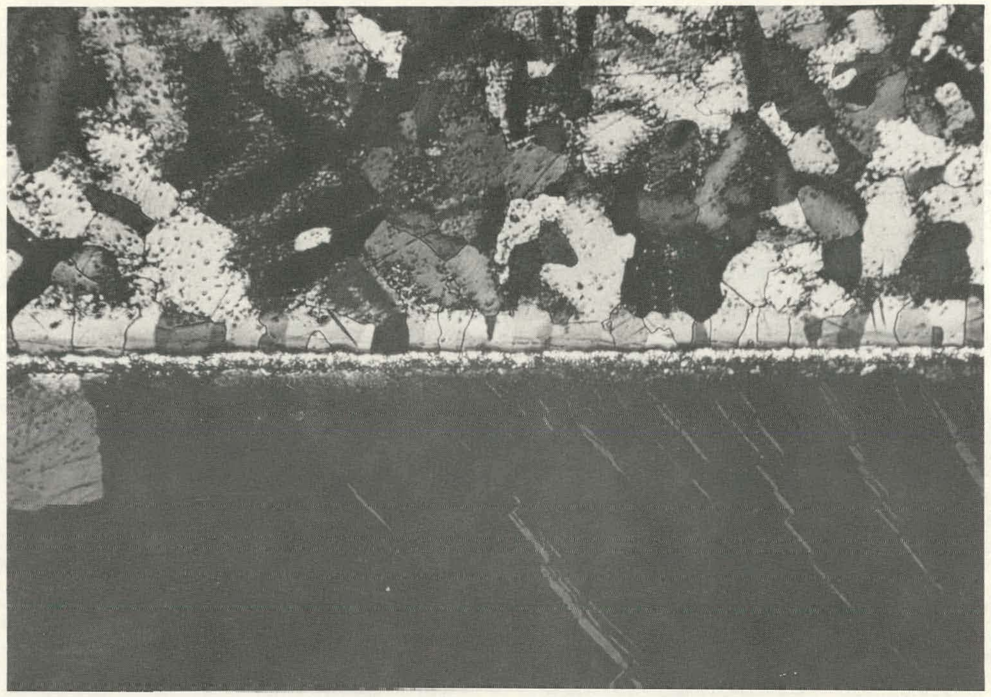
CONCLUSIONS AND RECOMMENDATIONS

This work has demonstrated that a composite electrode, in which the low-melting constituent (in this instance, uranium) is cast to enclose the remaining two constituents (niobium and zirconium), can be readily cast and consumably arc melted to produce Mulberry alloy. One of the advantages over the strip electrode would be a more intimate contact between the metals and, consequently, a more stable arc. Variations in the amperage and voltage, as noted from the meters and strip charts, were less when visually compared to the variations normally occurring with the present electrode design. Also, the television screen showed a more stable arc, particularly with the cast composite electrode having the formed strips. Figure 7 shows the uranium/niobium and uranium/zirconium interfaces, etched to reveal the uranium structure, on a cross section through an early electrode. The intimate metal-to-metal contact of the surfaces, which was a uniform and almost a metallurgical bond in some areas, would account for a more stable arc.

The higher alloy content in the center of the ingots (see Appendix, Table A-1, drilled samples) agrees with the existing data. However, the tendency toward a higher alloy content at the bottom of the ingot is unique and could be the result of earlier and more rapid melting of the niobium. The variation between the top and the bottom was carried over into the second-melt ingots. In Table 2 it can be noted that the variation would be reduced and the top and bottom chemistries would become closer to a mean if another inch had been removed from most of the first-melt ingots. It is probable, also, that extending the niobium something less than one inch would reduce the variations in this design. The chemistry was, however, statistically comparable to the current production stream variations. Microexaminations and macroexaminations of the second-melt ingot did not reveal any niobium-rich inclusions or visible porosity that is characteristic of the ingots from strip electrodes (see Figure 8).

A cost analysis[d] that compares the induction-cast electrode with the welded-strip electrode was made for the two current production programs. It is apparent that the material and machining costs of the graphite mold for induction casting the 14 electrodes would need to be spread over several runs to have an economical advantage. There is no question that the mold could be reused, but the average number of times would be conjectural. However, ten or more runs is reasonable, particularly if inserts are used; and, based on this usage, there is a significant cost advantage to the cast electrode.

(d) Memorandum from J. R. Gray to J. L. Cadden; December 3, 1971.



Zirconium

Interface

Uranium

(a) Uranium-to-Zirconium Interface. MS-71-0267-3

Niobium

Interface

Uranium

(b) Uranium-to-Niobium Interface. MS-71-0267-2

Figure 7. URANIUM-TO-ZIRCONIUM AND URANIUM-TO-NIOBIUM INTERFACES FROM A CROSS SECTION THROUGH A COMPOSITE ELECTRODE. (75X)

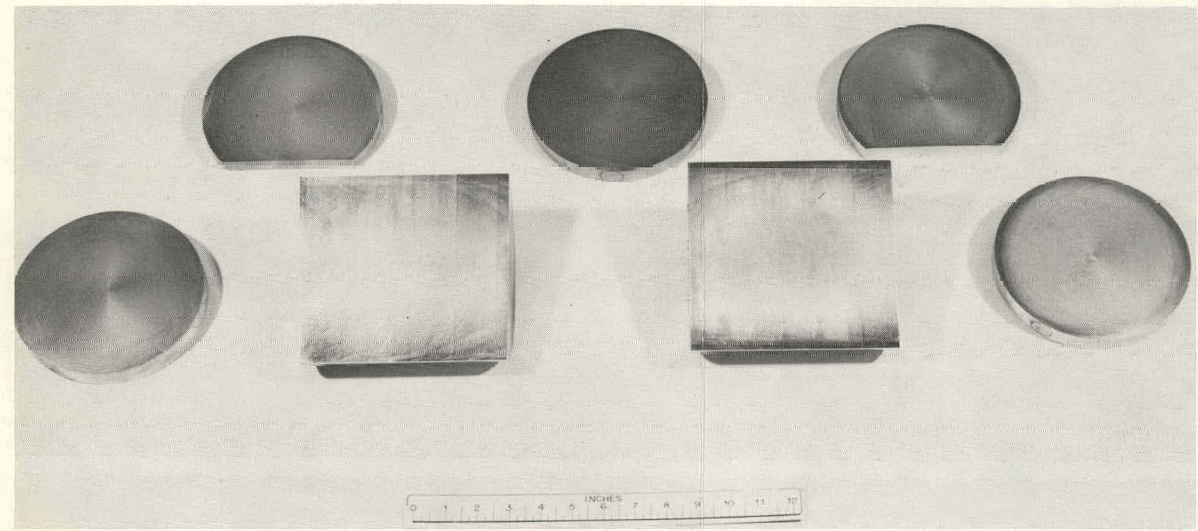


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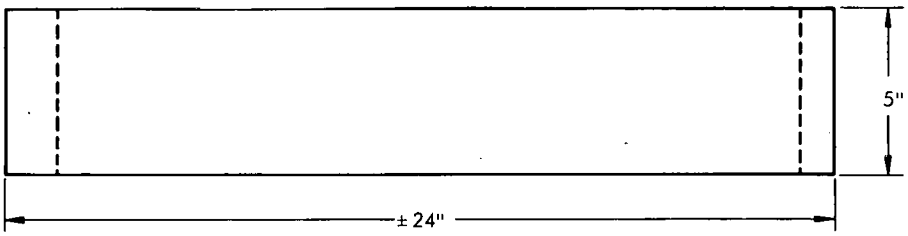
Figure 8. MACROETCHED CROSS SECTIONS OF SEVEN-INCH-DIAMETER SECOND ARC-MELT INGOTS. (No Niobium-Rich Inclusions were Found in these Sections)

ACKNOWLEDGMENTS

The author wishes to thank M. S. Dill of the Plant Laboratory Department for his interest and assistance in processing the relatively large number of analytical samples.

APPENDIX

SAMPLING AND ANALYTICAL DATA



Steps:

1. Cut 1 1/2 inches from the bottom of each ingot and scrap.
2. Cut a 3/4 to 1-inch slice and identify with the ingot number (Number 7095-45-XXXX). Mark the surface nearest the center of the ingot with a "B".
3. Cut sufficient metal from the top of the ingot to give a smooth surface, with no indication of pipe or porosity, and scrap. Minimum of 2 1/2 inches.
4. Cut a 3/4 to 1-inch slice and identify it with the ingot number. Mark the surface nearest the center with a "T".
5. Mill 0.005 inch from the surface of "B" and "T" for analytical samples.
6. Machine a density sample (~ 3/4-inch cube) from the edge and center of both samples from Ingots 1932, -35, -36, -39, -40, and -43. Machine a gas sample (1/8" D x 1" L) from the center of the ingots.
7. Weld the remaining sections of the ingots (4) to make up three second-melt electrodes approximately 72 inches long.
8. Record the location and from which electrode each five-inch ingot has been taken.

Sampling of the second-melt ingots was as outlined in Table A-1.

Figure A-1. CHEMICAL SAMPLING PLAN FOR THE 14 FIVE-INCH-DIAMETER ARC-MELTED INGOTS MADE FROM INDUCTION-CAST COMPOSITE ELECTRODES.

Table A-1

CHEMICAL ANALYSES OF FIRST AND SECOND-MELT INGOTS

Ingot Number	Location	Uranium (%)	Niobium (%)	Zirconium (%)	Copper (ppm)	Iron (ppm)	Carbon (ppm)	Hydrogen (ppm)	Nitrogen (ppm)	Oxygen (ppm)	Density (3/4" cube) Center (g/cc)	Density (3/4" cube) Edge (g/cc)
						Five-Inch Diameter (first melt)						
1944	Top	89.27	7.84	2.48	10	20	46					
	Bottom	89.05	8.06	2.57	10	20	85					
1940	Top	89.72	7.73	2.43	6	7	33	4	6	47	16.5408	16.4372
	Bottom	89.37	7.33	2.63	7	33	49	3	4	43	16.3123	16.3692
1934	Top	89.51	7.30	2.58	10	20	43	5	5	70		
	Bottom	89.13	7.31	2.62	8	20		6	15	76	16.2884	16.3865
1936	Top	89.44	7.76	2.57	5	28	50	3	11	71	16.4531	16.4238
	Bottom	88.79	8.36	2.83	10	26	99	3	13	71	16.2624	16.3438
1932	Top	89.23	7.85	2.75	10	16	45	7	15	59	16.9417	16.3723
	Bottom	89.44	7.94	2.60	< 10	41	31	7	1	48	16.2707	16.3675
1937	Top	90.11	7.54	2.39	7	18	44	6	10	70	16.3067	16.5250
	Bottom	90.24	7.34	2.49	8	18	35	6	17	41	16.4039	16.6285
1942	Top	89.33	7.94	2.55	10	20	46					
	Bottom	89.43	7.79	2.48	10	20	270					
1939	Top	90.30	7.20	2.28	< 10	15	21	8	24	112	17.1271	16.5982
	Bottom	89.85	7.53	2.49	143	19	39				16.4750	16.5096

Table A-1 (Continued)

Ingot Number	Location	Uranium (%)	Niobium (%)	Zirconium (%)	Copper (ppm)	Iron (ppm)	Carbon (ppm)	Hydrogen (ppm)	Nitrogen (ppm)	Oxygen (ppm)	Density (3/4" cube) Center (g/cc)	Edge (g/cc)
Five-Inch Diameter (first melt)												
1941	Top	89.82	7.78	2.50	10	25	74					
	Bottom	89.18	8.28	2.54	10	30	74					
1933	Top	89.86	7.47	2.36	5	25	34					
	Bottom	88.67	8.23	2.75	6	35	44					
1943	Top	89.72	7.54	2.27	6	30	24	5	2	38	17.0478	16.5352
	Bottom	88.99	8.28	2.67	47	20	58	8	18	72	16.2737	16.3075
1935	Top	89.88	7.24	2.49	8	30	88	3	10	62	16.4276	16.8052
	Bottom	89.12	8.09	2.54	5	26	64	3	5	48	16.3060	16.3768
1931	Top[3]	89.93	7.54	2.35	5	38	31					
	Bottom	89.74	7.79	2.48	7	45	43					
1938	Top[3]	89.71	7.46	2.42	7	18	30	7[1]	5[1]	55[1]	16.6644	16.4866
	Bottom	90.06	7.61	2.50	7	20	211	6[2]	12[2]	70[2]	16.3941	16.4125
Seven-Inch Diameter (second melt)												
7095-46-0923												
Top	Y	89.33	7.88	2.67	2	25	36	6[1]	20[1]	76[1]	16.2695	16.4428
	P	89.25	7.90	2.63	2	25	32	6[2]	12[2]	73[2]		
	M	88.98	8.14	2.70	2	25	46	7[1]	15[1]	61[1]	16.1063	16.4392
	F	88.93	8.04	2.68	25	30	62	7[2]	24[2]	74[2]		
Bottom	A	89.47	7.78	2.56	2	30	54	4[1]	13[1]	46[1]	16.3617	16.4613
								6[2]	12[2]	52[2]		
Center Drilled	M	88.04	8.77	2.83	10	25	92					

Table A-1 (Continued)

Ingot Number	Location	Uranium (%)	Niobium (%)	Zirconium (%)	Copper (ppm)	Iron (ppm)	Carbon (ppm)	Hydrogen (ppm)	Nitrogen (ppm)	Oxygen (ppm)	Density (3/4" cube) Center (g/cc)	Edge (g/cc)
Seven-Inch Diameter (second melt)												
7095-46-0924												
Top	Y	88.90	8.22	2.76	6	40	88	2[1]	3[1]	58[1]	16.12	16.29
	P	89.09	8.06	2.69				2[2]	2[2]	53[2]		
	M	89.86	7.79	2.56	5	25	74	3[1]	3[1]	37[1]	16.16	16.39
	F	90.00	7.30	2.58	10	25	64	2[2]	8[2]	48[2]		
Bottom	A	89.94	7.76	2.44	5	25	50	2[1]	2[1]	55[1]	16.42	16.36
								2[2]	5[2]	25[2]		
Center Drilled	M	88.52	8.50	2.71	5	22	107					
7095-46-0926												
Top	Y	89.10	8.23	2.73	6	20	42	2[1]	7[1]	47[1]	16.11	16.41
	P	88.91	8.23	2.74	8	16	48	3[2]	8[2]	18[2]		
	M	89.62	7.81	2.55	11	19	44	0.94[1]	7[1]	36[1]	16.19	16.42
	F	89.33	7.97	2.56	8	10	40	3[2]	4[2]	23[2]		
Bottom	A	89.69	7.60	2.51	9	14	32	7[1]	11[1]	62[1]	16.34	16.36
								4[2]	7[2]	27[2]		
Center Drilled	M	88.69	8.56	2.72	9	20	58					

(1) Edge.
(2) Center.
(3) On hold—not used to make up second-melt ingot.

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