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Rolling and Forming of Thorium Metal

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THE DOW CHEMICAL COMPANY
ROCKY FLATS PLANT
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ROLLING AND FORMING OF THORIUM METAL

by

A. E. Calabra

L. L. Zodtner, Section Superintendent

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ABSTRACT

One vacuum induction melted and cast nuclear grade thorium ingot was satisfactorily rolled and formed into a hemispherical shape. Attempts to roll and form two vacuum induction melted and cast alloy grade thorium ingots were not successful.

Rolling and forming procedures are described and metallurgical evaluation is reported.

1. INTRODUCTION

The high ductility and high neutron reflection capabilities of thorium make it well suited for weapon case components. Therefore, at the request of the Lawrence Radiation Laboratory, an attempt was made to fabricate hemispherical parts from thorium ingots by rolling and forming techniques. Ingots cast from both nuclear and alloy grade thorium were to be investigated.

2. SUMMARY

A nuclear grade thorium ingot was hot rolled (canned) and cold rolled (uncanned) into plate and successfully drawn at room temperature into a 13-in. diameter hemispherical shape.

Two thorium ingots cast from alloy grade feed exhibited very brittle properties and were not successfully rolled into plate. The relative impurity of these ingots is believed largely responsible for this brittleness.

If additional rolling and forming demonstrations are to be conducted utilizing vacuum induction melted alloy grade thorium, various casting and heat treating procedures should be investigated in an attempt to alter the impurity state and possibly restore metal ductility.

3. EXPERIMENTAL

Three vacuum induction melted and cast thorium ingots were obtained from the Erwin, Tennessee Plant of the Davison Chemical Division, W. R. Grace and Company. Nuclear grade thorium was used as charge metal for Ingot 1 and alloy grade thorium for Ingot 2 and Ingot 3. All three ingots displayed moderate cold shutting and surface pits, however, not to a degree which required removal by machining.

The three ingots were clad in heavy, mild steel cans prior to rolling. The cavities for Ingots 1 and 2 were machined resulting in a snug ingot fit. (Figure 1) However, the cladding and cavity for Ingot 3 was flame cut (Figure 2) allowing a loose ingot fit and considerable air was trapped with the ingot inside the can. Copper sheets 0.010 in. thick were used to separate the top and bottom ingot surfaces from the can since thorium and mild steel display bonding tendencies under heat and pressure. Prior to welding the ingot cladding components, a thin coating of mold wash was applied to all non-welded areas where there would be mild steel to mild steel contact. This precaution was taken to prevent mild steel bonding during rolling since bonding would complicate subsequent can removal.

The rolling practices and results for each thorium ingot are tabulated in Tables I, II, and III. The pressing procedure for Ingot 1 is tabulated in Table IV. Production rolling and forming facilities located in Building 83 were used for this investigation.

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Samples from each rolled plate were analyzed for impurities. Results are shown in Table V.

Specimens for metallographic examination were prepared as described in Appendix "A". Metallographic examination and photography was done with a Vickers Projection Microscope.

Tensile bars from the nuclear grade annealed plate were machined to ASTM specifications for 1 in. gage length, 1/4 in. diameter. Properties of these bars were determined in a Tinius-Olsen testing machine.

Hardness values were determined on the metallographically prepared specimens using a Gries Hardness Tester with a 10-kg load and diamond pyramid indentor.

4. DISCUSSION

4.1 Fabrication

4.1.1 Hot Rolling

Hot rolling was successful for canned Ingots 1 and 2 (nuclear and alloy grades respectively). During hot rolling of canned Ingot 3 (alloy grade) the can ruptured and rolling could not be completed. Ingot 3 can design, which allowed more entrapped air than did the can design used for Ingots 1 and 2, is felt to be the reason for the rupture. After preheating Ingot 3, the can composite displayed a noticeable bulged condition. The entrapped air was squeezed to the end of the slab during each pass through the rolls. A point was reached where the can could not withstand the internal air pressure and several weld areas ruptured. The can was removed from the thorium by flame cutting revealing a badly fractured slab. (Figure 3) Ingot 3 was then scrapped.

Slabs from Ingots 1 and 2 were left in their cans and annealed after hot rolling.

4.1.2 Cold Rolling

Attempts to cold roll canned Ingots 1 and 2 to 50% reduction had to be terminated because of can weld failures before the desired reduction was obtained. The can assemblies were preheated to 400 F before rolling to increase the ductility of the mild steel. During the rolling operation, the composite slabs curled excessively, which made rolling difficult and may have contributed to the weld failures.

At the gages shown in Tables I and II (item: Cold Roll in Can) the cans were removed from the slabs by shearing. However, the top and bottom can layers spread apart when sheared making this method of can removal difficult. Also significant quantities of dust were dispersed into the air during shearing. For these reasons, flame cutting (used on Ingot 3) appears to be the more desirable method of removing the cans.

4.1.3 Etching

Ingot 1 and 2 slabs were immersed in 8 N nitric acid solution to remove any foreign metal particles possibly embedded in their surfaces and to wash off the oxide dust. No embedded metal was noted: and since thorium and thorium oxide are relatively insoluble in concentrated nitric acid, a water wash would have been sufficient to remove the loose oxide.

4.1.4 Final Cold Rolling

After the "etching" operation, attempts were made to cold roll the two slabs to final gage of 0.440 in. The nuclear grade was rolled without difficulty. (Table I)

Before rolling the alloy grade slab, one edge was sheared to remove shallow saw tooth edge cracks. However, the sheared edge was typical of extremely brittle metal and was almost as rough as the original edge. No additional side shearing was done. The brittle properties were again displayed, during subsequent rolling of the alloy plate, (Table II) by its cracking and alligatoring (splitting of flat slab, plate, or sheet in a plane parallel to the rolled surfaces). As a result insufficient metal was left for part forming.

4.1.5 Forming

The cold rolled nuclear grade plate was recanned in 16-gage mild steel, annealed, uncanned, and pressed into a 13-in. diameter hemispherical part without difficulty. (Table IV)

After recanning in 16-gage mild steel, annealing and uncanning, the part was shipped to Building 44 for final machining into a 0.240-in. wall x 12.882-in. inside diameter part per LRL drawing L16A 6834A (Dow Drawing 1PE6931-1-A).

4.2 Metallurgy

4.2.1 Tensile Properties

Tensile specimens from the annealed nuclear grade thorium plate were cut so that properties could be determined parallel, 45°, and normal to final roll direction. The results are tabulated in Table VI. There is little significant variation between properties in the three directions. The values do show the high ductility of thorium, especially when compared to salt-annealed uranium sheet in which the average elongation is approximately 16% normal to the final roll direction vs. 46% average elongation for the thorium in the same test direction.

4.2.2 Inclusions

Figures 4 and 5 are photomicrographs showing the inclusions in the nuclear and alloy grade plates. (Ingots 1 and 2) The ThO_2 and ThC are the predominate inclusions. Each is identified by its respective color; ThO_2 - dark gray, ThC - off-white[1].

Figure 4A shows one of several elongated inclusions observed in the structure of the nuclear grade plate. Samples from one inclusion were extracted for identification by electron diffraction and found to include some thorium oxide. (Figure 6) However, the inclusion was primarily diatomaceous earth which is commonly used in casting as a mold coating. Numerous diatoms were found in the extracted material as shown in Figure 7.

(1) F. L. Cuthbert, Thorium Production Technology, pp. 282, Addison-Wesley, Reading, Mass., 1958.

4.2.3 Microstructures

Figures 8, 9, and 10 are photomicrographs of the nuclear grade thorium plate in the as-rolled state and in the annealed condition.

Figures 11 and 12 are photomicrographs of the alloy grade plate (Ingot 2) in the as-rolled and annealed conditions respectively.

The grains within the nuclear grade ingot and its annealed plate are larger than those found in the alloy grade ingot and its annealed plate. The relative sizes of the cast grains are reflected in the as-rolled plate structures. Compared to the alloy grade material, the nuclear grade material contained fewer impurities (Table V) and impurity inclusions were generally larger and in a less dispersed state. The condition of the impurities probably contributed greatly to the respective cast and recrystallized grain sizes.

Figure 13 is a photomicrograph of the structure of the highly fractured alloy grade slab - Ingot 3. (Figure 3) Although this ingot was rolled, it exhibited an annealed grain structure rather than a rolled structure. The slab may have been above the recrystallization temperature at the time hot rolling was stopped. (Table III)

Figure 13 also shows the numerous inclusions and/or constituents as the grain boundaries. The boundary phase is probably the primary cause of the brittle characteristics exhibited by this alloy grade ingot.

Figure 14 is the as-mechanically-polished surface showing the boundary inclusions and/or constituents and ThO_2 particles.

The lack of success in rolling Ingots 2 and 3 (alloy grade) is largely attributed to the detrimental effects of impurities. With the exception of magnesium, the impurity levels are greater in the alloy grade thorium than in the nuclear grade thorium.

4.2.4 Hardness

Table VII tabulates the hardness values of nuclear and alloy grade plate (Ingots 1 and 2) both parallel and normal to the final rolling direction and in the unannealed and annealed conditions. The relatively high hardness numbers for the unannealed and annealed alloy grade material can be attributed to its fine grain size and the fine and dispersed inclusion particles.

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TABLE I

Rolling Practice for Ingot No. 1 (Nuclear Grade)

Ingot size	3 in. x 12 in. x 12 in. (wt. = 80 kg)
Canned size	4-1/2 in. x 20 in. x 20 in. (Octagon - see Figure 1)
Preheat	2 hr at 1600 F furnace temperature (Lindberg Furnace - He atmosphere)
Hot roll	2-1/4 in. x 38 in. x 20 in.
Cross roll	1.320 in. x 38 in. x 34 in.
Anneal	1-1/2 hr at 1300 F furnace temperature (Lindberg Furnace - He atmosphere)
Preheat	1 hr at 400 F furnace temperature (Lindberg Furnace)
Cold roll in can	1.120 in. x 39 in. x 39 in. several cracks developed around welds
Uncan and etch	Etch in 8 N HNO_3
Uncanned slab dimension	0.750 in. x 24 in. x 24 in.
Cold roll	0.440 in. x 25 in. x random
Recan	16 gage mild steel
Anneal	1-1/2 hr at 1350 F furnace temperature (Lindberg Furnace - He atmosphere)
Remove can	Hand shears
Square shear	24 in. x 24 in.
Circle shear	23 in. diameter

TABLE II

Rolling Practice for Ingot No. 2 (Alloy Grade)

Ingot size	3 in. x 12 in. x 12 in. (wt. = 79 kg)
Canned size	4-1/2 in. x 20 in. x 20 in. (Octagon - see Figure 1)
Preheat	2 hr at 1600 F furnace temperature (Lindberg Furnace - He atmosphere)
Hot roll Cross roll	2-1/4 in. x 38 in. x 20 in. 1.320 in. x 38 in. x 33 in.
Anneal	1-1/2 hr at 1350 F furnace temperature (Lindberg Furnace - He atmosphere)
Preheat	400 F furnace temperature (Lindberg Furnace - air atmosphere)
Cold roll in can	0.870 in. x 39 in. x random
Uncan and etch	Etch in 8 N HNO_3
Slab dimension	0.600 in. x 23 in. x 31 in. (Moderate edge cracks and one edge slightly alligatored)
Side shear	One edge (Brittle-broken sheared edge)
Cold roll	0.580 in. x 22 in. x random (Slab cracked and alligatored severely)
Anneal*	1-1/2 hr at 1350 F furnace temperature (Lindberg Furnace - He atmosphere)

*Note: There was insufficient metal for part forming. Metallurgical specimens were taken and the remainder of the material was scrapped.

TABLE III

Rolling Practice for Ingot No. 3 (Alloy Grade)

Ingot size	3 in. x 12 in. x 12 in. (wt. = 78 kg)
Canned size	4-1/2 in. x 20 in. x 20 in. (square - see Figure 2)
Preheat	1-1/2 hr at 1800 F furnace temperature (Lindberg Furnace - He atmosphere)
Hot roll	2.500 in. x 20 in. x 36 in.
Cross roll	1.625 in.* x 30 in. x 36 in.

*Note: At 1.625 in. gage the can tore open near the weld at one corner. The can was then cut open with a torch revealing a severely broken thorium slab - approximately 1.08-in. thick. (Figure 3)

TABLE IV

Forming Practice for Nuclear Grade Thorium Circle
(Ingot No. 1)

Circle size	0.440 in. x 23 in. diameter
Lubricated	Johnson's Lubricating Wax
Punch size	12.728 in. diameter hemisphere
Press*	75 F
Can	16 gage mild steel
Part anneal	30 min at 1350 F furnace temperature (Lindberg Furnace - He atmosphere)
Can removed	Yes

*Note: Press cycle determined by first drawing mild steel blanks of the same gage as the thorium. The cycle which gave a satisfactory part was used to draw the thorium and it was noted that the thorium drew with less difficulty than did the mild steel.

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TABLE V

Chemical Analysis
(ppm unless noted)

	Ingot 1 (Nuclear Grade) Sample*		Ingot 2 (Alloy Grade) Sample*			Ingot 3 (Alloy Grade) Sample*	
	A	B	A	B	C	A	B
Al	<20	<20	1500	1500	500	60	60
B	<1	<1	40	40	15	6	4
Be	100	150	200	200	150	200	200
C	185	345	490	885	460	1250	1250
Ca	50	50	15	15	35	50	50
Co	--	--	--	--	--	<2	--
Cr	100	80	80	80	300	>80	>80
Cu	5	5	15	15	20	50	40
Fe	820	680	1100	1100	1300	1145	1175
Mg	1500	500	20	20	200	200	>200
Mn	--	--	--	--	--	30	--
Mo	--	--	--	--	--	150	--
N	65	70	250	230	300	125	205
Ni	8	3	200	200	200	206	207
Si	50	40	120	120	120	250	200
U	600	600	400	400	400	800	750
V	--	--	--	--	--	<1	--
Zn	<20	<20	<20	<20	<20	<20	<1
Zr	<25	<25	200	200	200	<25	<25
ZrO_2	--	--	--	--	--	<35	205
Total	3549	2589	4650	5025	4220	4685	4652
ThO_2	1.5%	1.1%	1.2%	1.0%	1.2%	1.0%	0.96%

*Samples taken at random from different locations on the rolled plates.

TABLE VI

Properties of Rolled and Annealed Nuclear Grade Thorium

Ingot No. 1

(1) Roll Direction	(2) No. of Samples	Ultimate Strength p.s.i.			Yield Strength p.s.i. 0.2% offset			% Elongation			(3) Hardness DPH	% Area Reduction
		Low	Avg	High	Low	Avg	High	Low	Avg	High		
N	3	38,600	40,800	43,700	27,800	28,900	30,900	43	46	49	92	58
45°	2	38,100	38,100	38,100	30,100	30,100	30,100	40	41	42	---	48
P	3	38,400	39,700	42,000	30,300	31,900	35,000	38	43	46	91	53

(1) N - Specimens cut so tests would be normal to the final rolling direction.
45° - Specimens cut so tests would be 45° to the final rolling direction.
P - Specimens cut so tests would be parallel to the final rolling direction.

(2) Samples machined to ASTM standard 1-in. gage round bars.

Note: All above were pulled at a strain rate of 0.080 in/in/min. One 45° sample was tested at a strain rate of 0.800 in/in/min with the following results:

Ultimate Strength - 41,500
Yield Strength - 33,600
% Elongation - 35%

An increase is shown in both ultimate and yield strengths, and a lower elongation, compared to those 45° samples pulled at a slower strain rate.

(3) Diamond pyramid hardness.

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TABLE VII

Hardness of Rolled Thorium Plate

Ingot No.	Condition	Direction	Diamond Pyramid Hardness Number
1	Unannealed	Normal	120
		Parallel	121
1	Annealed	Normal	92
		Parallel	91
2.	Unannealed	Normal	180
		Parallel	177
2	Annealed	Normal	146
		Parallel	146

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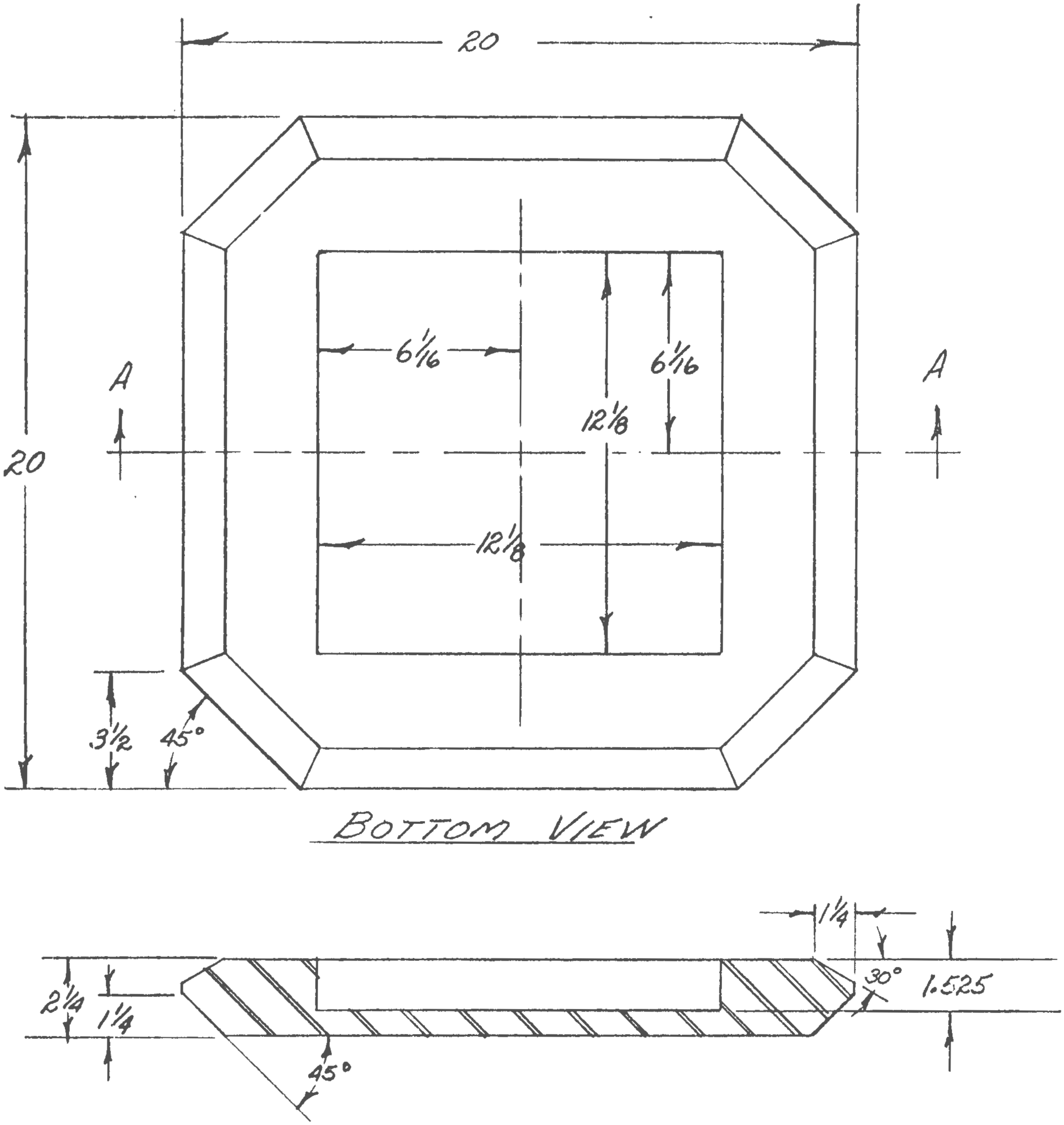
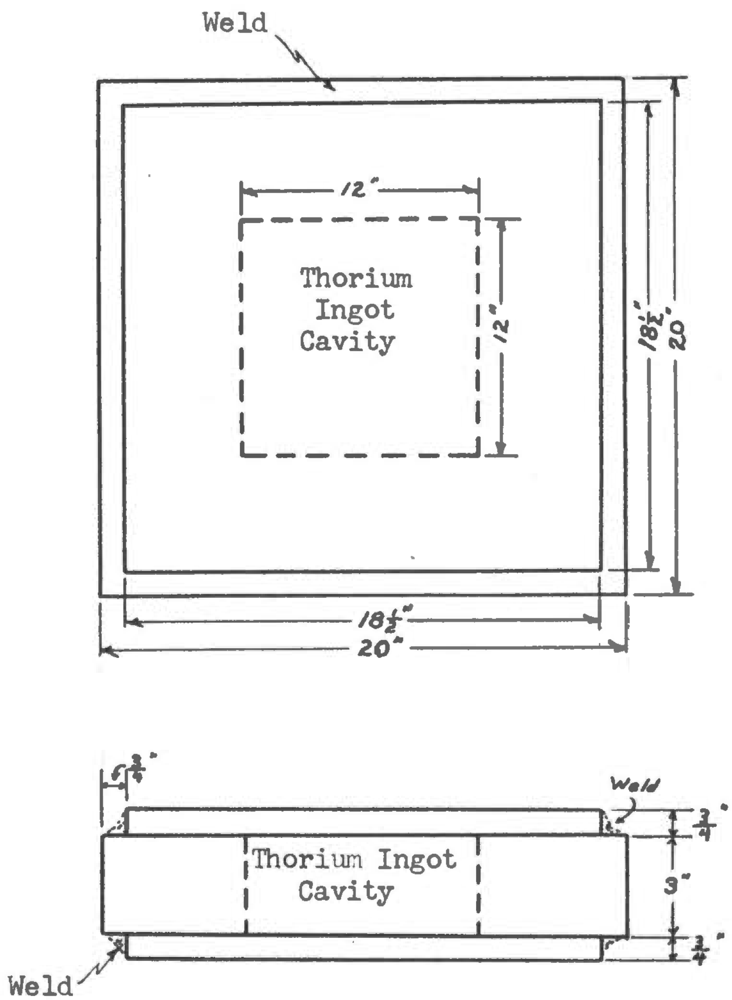


MATERIAL - MILD STEEL - 2 REQ'D PER ASSEMBLY

Figure 1

Figure 2

Can Dimensions For Alloy Grade Thorium Ingot - No. 3



Material - Mild Steel
All flame cut

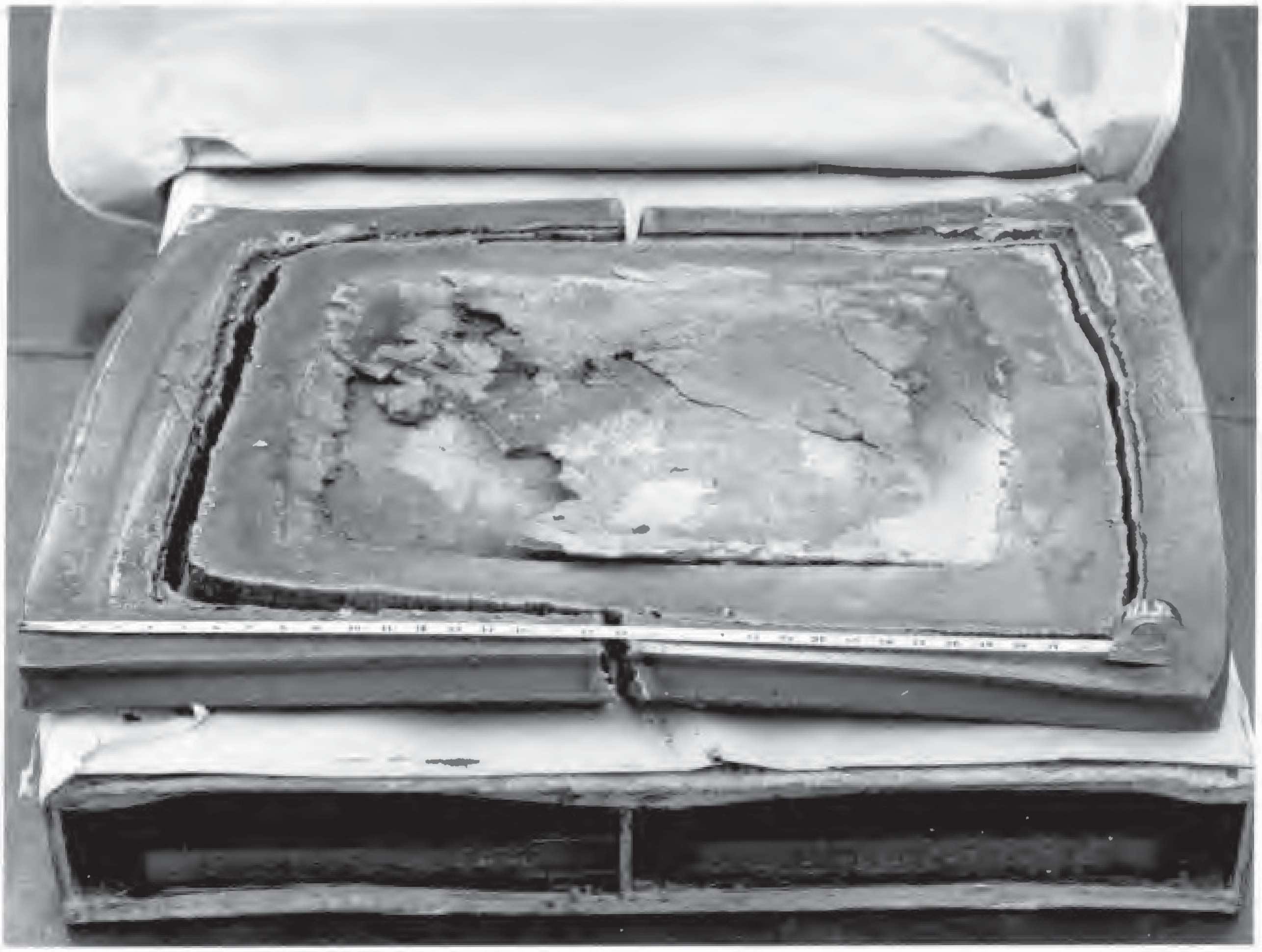


Figure 3

Alloy grade thorium - Ingot 3

Ingot 3 was hot rolled until the can ruptured; at this time the fabrication was stopped. The can components were flame cut for removal revealing the highly fractured slab.

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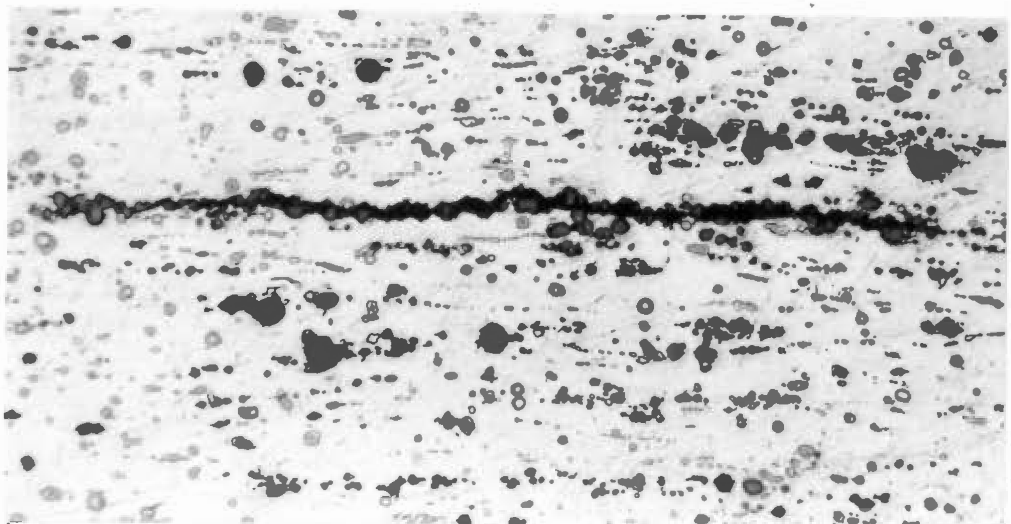
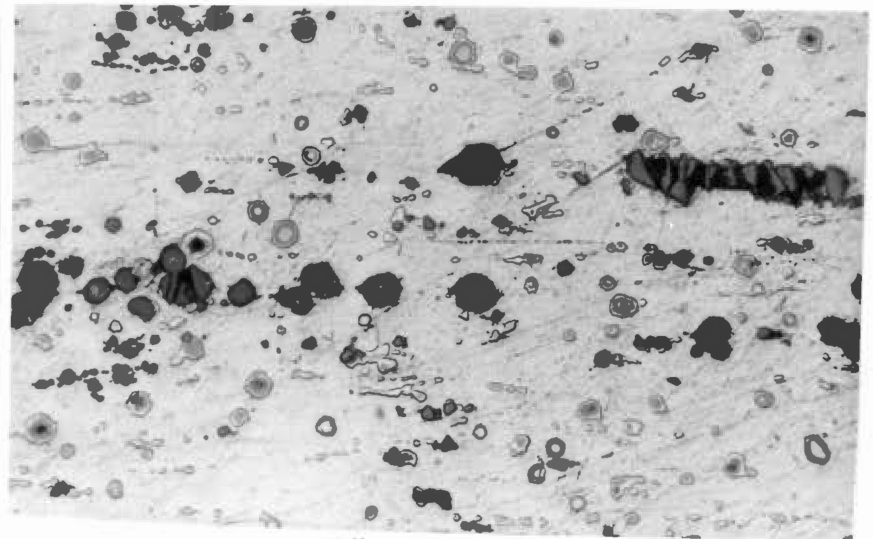
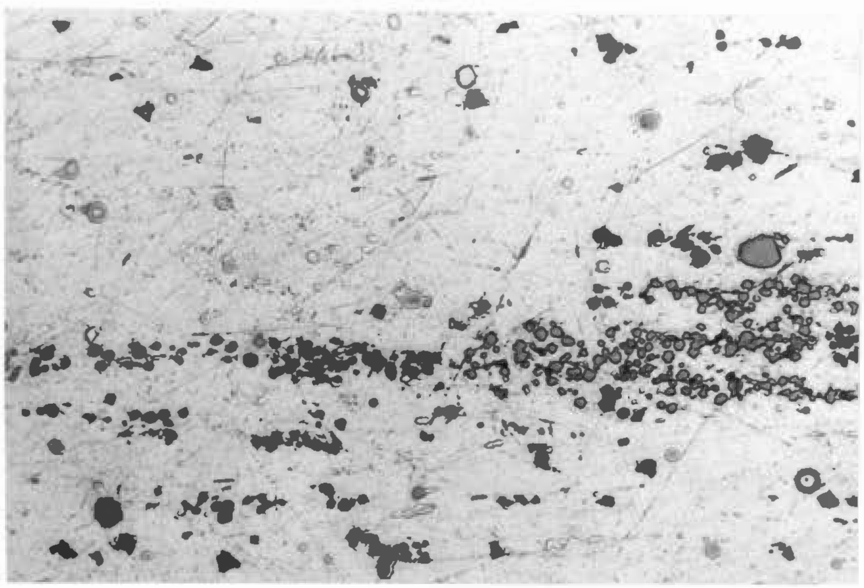


"A"



"B"



"C"

Figure 4 - 200X

Nuclear Grade Plate

The inclusions appear to be ThO_2 (dark gray) and ThC (off-white). The stringer like inclusion ("A") contained some oxide (Figure 6), but primarily diatomaceous earth (Figure 7).

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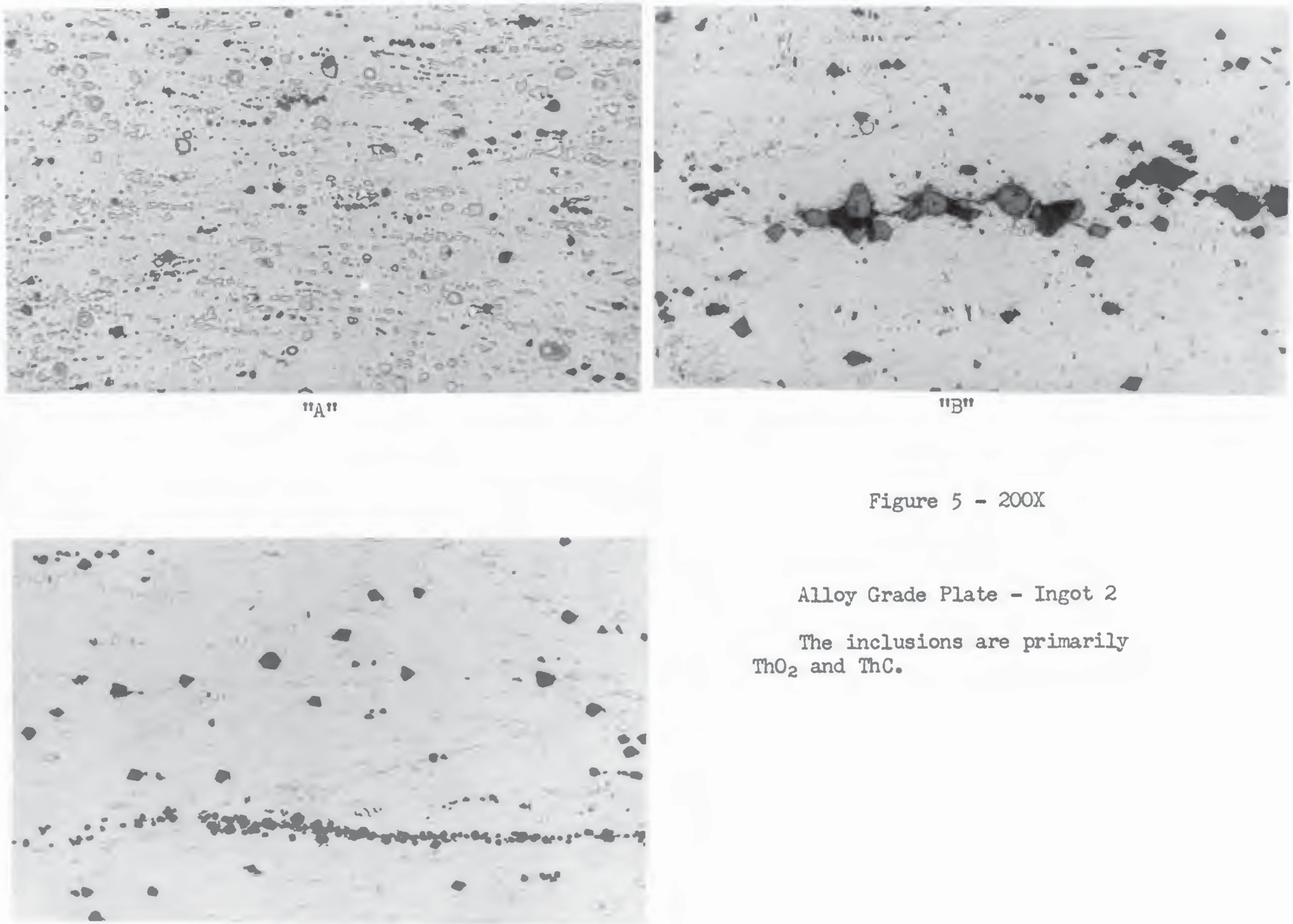

"A"

"B"

"C"

Figure 5 - 200X

Alloy Grade Plate - Ingot 2

The inclusions are primarily ThO_2 and ThC.

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Figure 6 - 7500X

Nuclear Grade Plate

ThO_2 particles from the elongated inclusion (Figure 4-A).

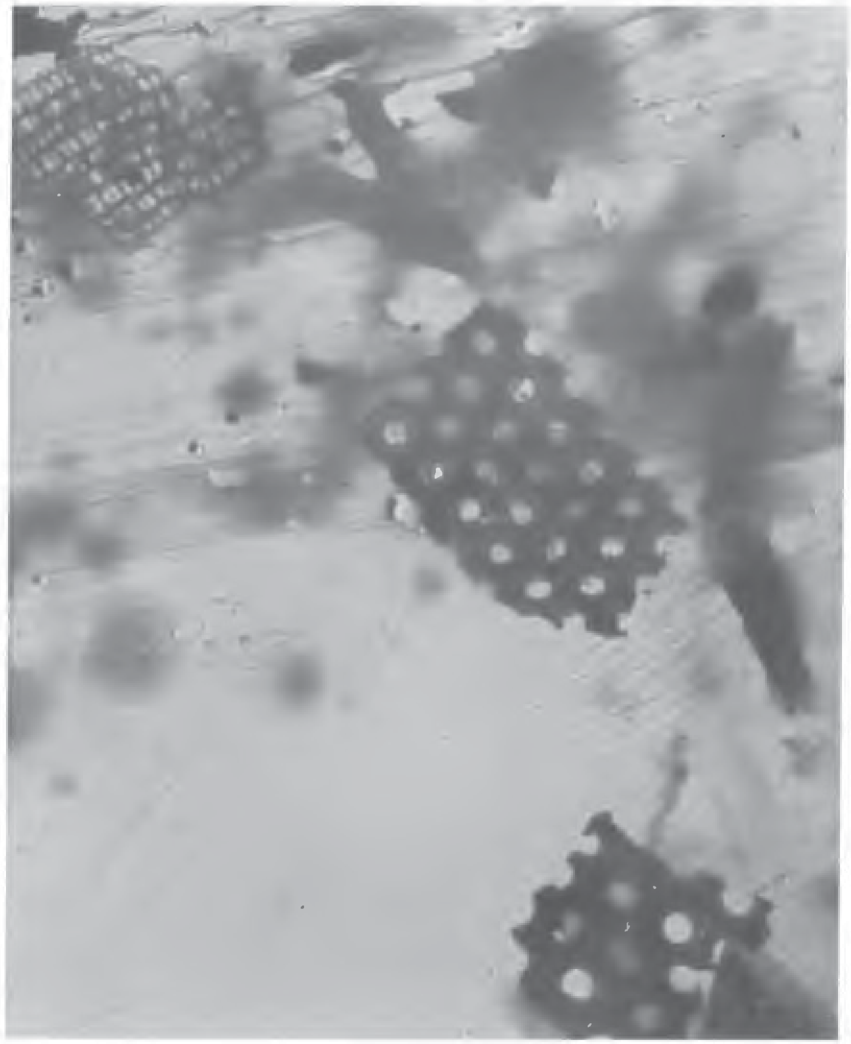

Figure 7 - 8000X

Nuclear Grade Plate

Diatom particles from the elongated inclusion (Figure 4-A)

Figure 8 - 200X

Nuclear Grade Thorium - As-rolled (Ingot No. 1)

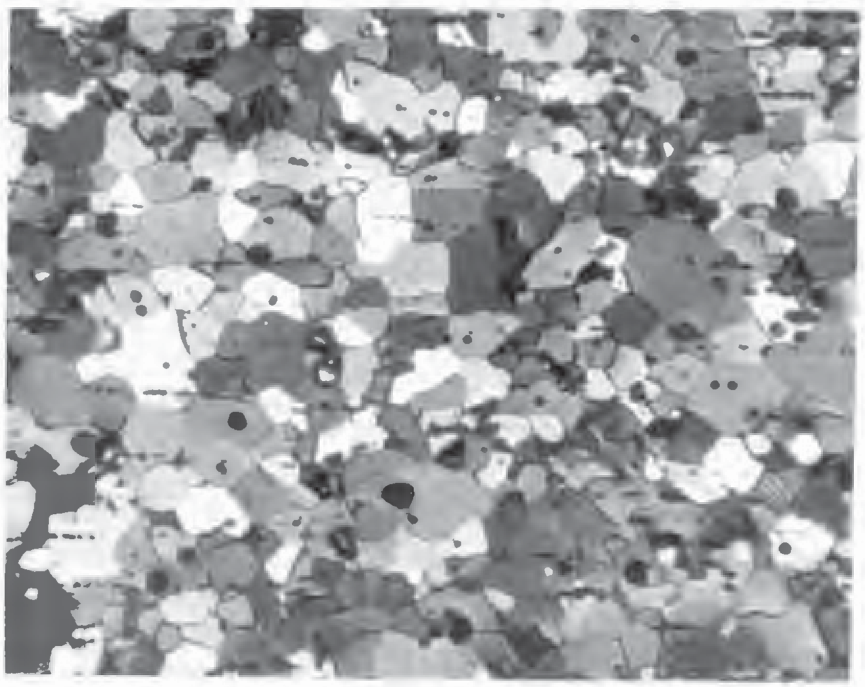

Figure 9 - 200X

Nuclear Grade Thorium - Annealed (Ingot No. 1)

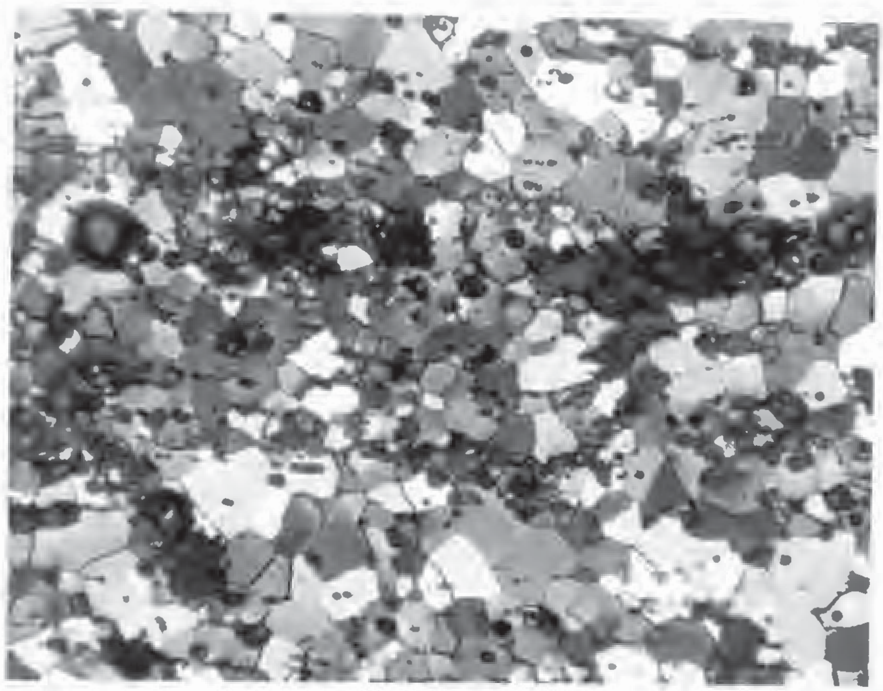

Figure 10 - 200X

Nuclear Grade Thorium - Annealed (Ingot No. 1)

An area of extremely high inclusion materials content.

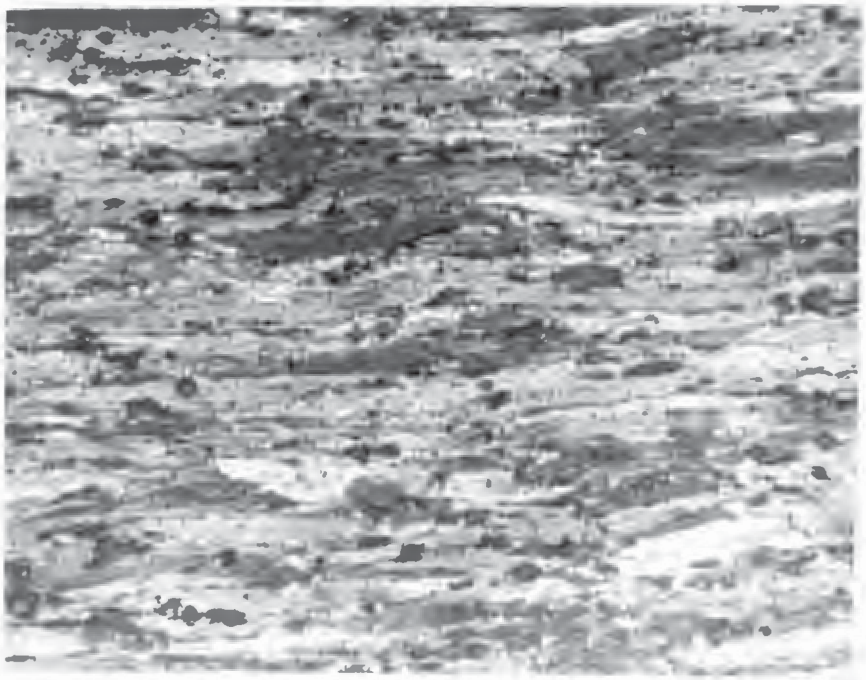

Figure 11 - 200X

Alloy Grade Thorium - As-rolled (Ingot No. 2)

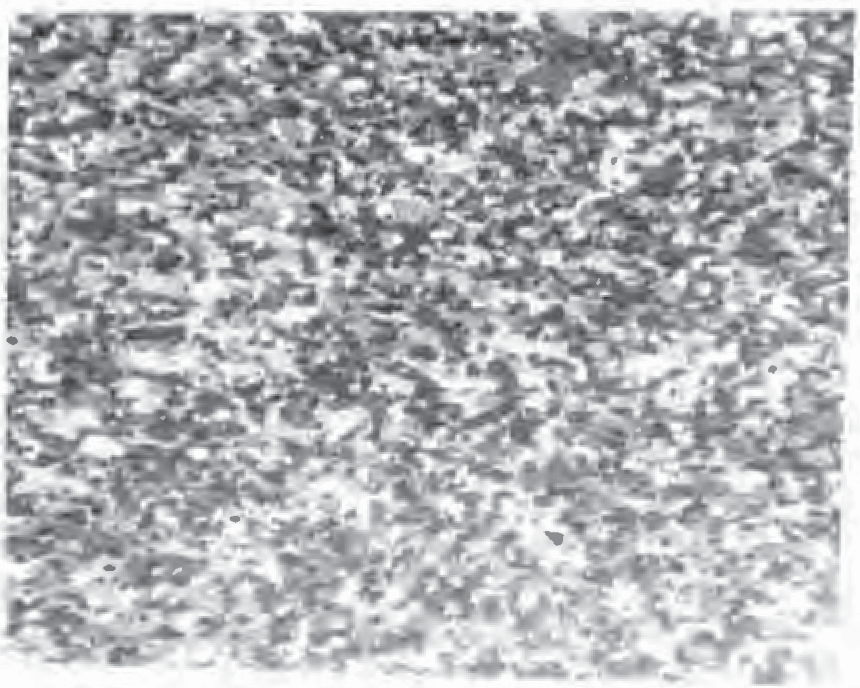

Figure 12 - 200X

Alloy Grade Thorium - Annealed (Ingot No. 2)

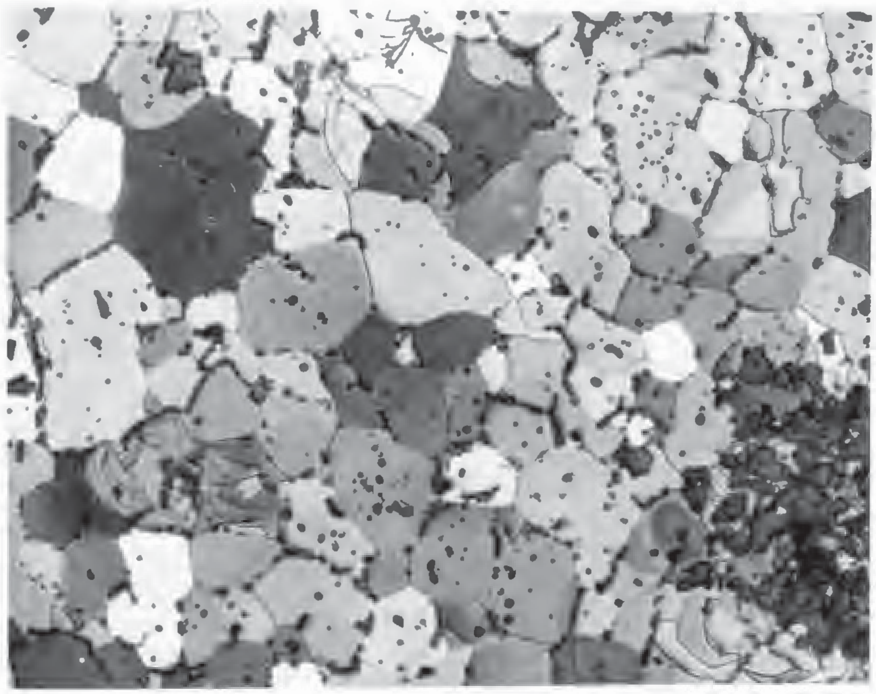

Figure 13 - 200X

Alloy Grade Slab - Ingot 3

The above photomicrograph shows the abundant boundary inclusions and/or constituents. The large clump of inclusions, lower right, appears to be the oxide.

The grain structure is in the recrystallized state.

Figure 14 - 200X

Alloy Grade - (Ingot No. 3) - as-mechanically-polished

ThO_2 inclusions as well as the stringer like boundary inclusions are shown. The large dark inclusions are the oxide.

APPENDIX

Procedure Used In Preparing Thorium Specimens
For Metallographic Examination

1. Mounting: Specimens mounted in Bakelite to facilitate handling.

2. Mechanical grinding: Wet grinding on 180 through 600 grit emery paper.

3. Mechanical polishing: Wet grinding on 0 grit paper lubricated lightly with Aquet solution.* Further mechanical polishing was accomplished on a Syntron Vibratory Polisher using Kitten-ear cloth and a slurry* of Alumina 1 followed by Alumina 2.

   * A 1:20 Aquet to water solution used for polishing on the 0 grit paper and to make up Alumina slurries.

4. Electro polish: Etch

   The procedure used basically followed that of M. H. Cornett.[1]

   Reagent: One part basic solution;

   118 g CrO_3
   100 cc distilled H_2O

   Four parts Glacial Acetic Acid

   Voltage: 25 V (used for the alloy grade thorium)
   35 V (used for the nuclear grade thorium)

   Time: 15 to 20 sec intervals until desired resolution attained.

   Cathode: Stainless steel

(1) F. L. Cuthbert, Thorium Production Technology, pp. 272, Addison-Wesley, Reading, Mass., 1958.