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Metallurgy Division

INVESTIGATION OF FACTORS INFLUENCING THE QUALITY OF CAST URANIUM-ALUMINUM ALLOY REACTOR SLUGS

C. D. Smith

Period covered by work - March and April, 1950

Date Issued: JUN 12 1950

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Abstract

A brief study was made of several factors influencing the quality of cast uranium-aluminum alloy reactor slugs. Included are the effects of melt temperature, melt time, pouring rate and mold temperature on segregation, porosity and surface quality of the slugs.

Introduction

In February, 1950, the Laboratory received an urgent request to study the feasibility of producing a total of 6000 uranium-aluminum alloy reactor slugs at a very high production rate. The specifications are listed in Table I below.

TABLE I

Slug Specifications

Material	U-Al alloy
U content	7.50% (approx.)
U enrichment	95% (approx.)
Length	8.0" ± 0.010"
Diameter	1.359" + 0.001"
	- 0.002"

Included in the possible production sites was the Y-12 facility. Since ORNL had experience in slug production the various aspects were discussed in a meeting with Y-12 personnel. Shortly afterward a decision was made to

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have the work done at Y-12. This work was started in the 9212 Area under the direction of J. M. Herndon.

To achieve a high production rate large 60 pound alloy batches yielding from 25 to 30 slugs were used. Considerable difficulty was experienced in obtaining sound castings and uniform distribution of uranium. These difficulties were discussed in several joint meetings and recommendations made based on ORNL experience. In addition it seemed desirable to test systematically with natural uranium alloys the several factors affecting slug quality. This was done at ORNL and the information transmitted verbally to expedite matters.

Previous Work at ORNL

A total of 229 enriched U-Al alloy (4.5% U) slugs, 4" long by 1.359" diameter had been cast and machined in the spring of 1948 for irradiation at Hanford.[1] After irradiation this material was used to test the efficiency of a remote controlled chemical pilot plant.

During this run it had been observed that slug inhomogeneity can occur. According to the phase diagram[2] (Figure I) if an alloy of U and Al containing less than 13% U is melted and cast, the first constituent to freeze should be aluminum. The surfaces of the slugs should therefore be lower in uranium than the center. This condition was found to exist. Of the 229 slugs 144 were from virgin melts, ranging in composition from 4.23 to 4.39% U. The balance was from remelts of shavings and scrap from virgin runs. Composition ranged from a very low value of 4.08 to 4.37% U despite the fact that an additional small amount of uranium was added in some cases.

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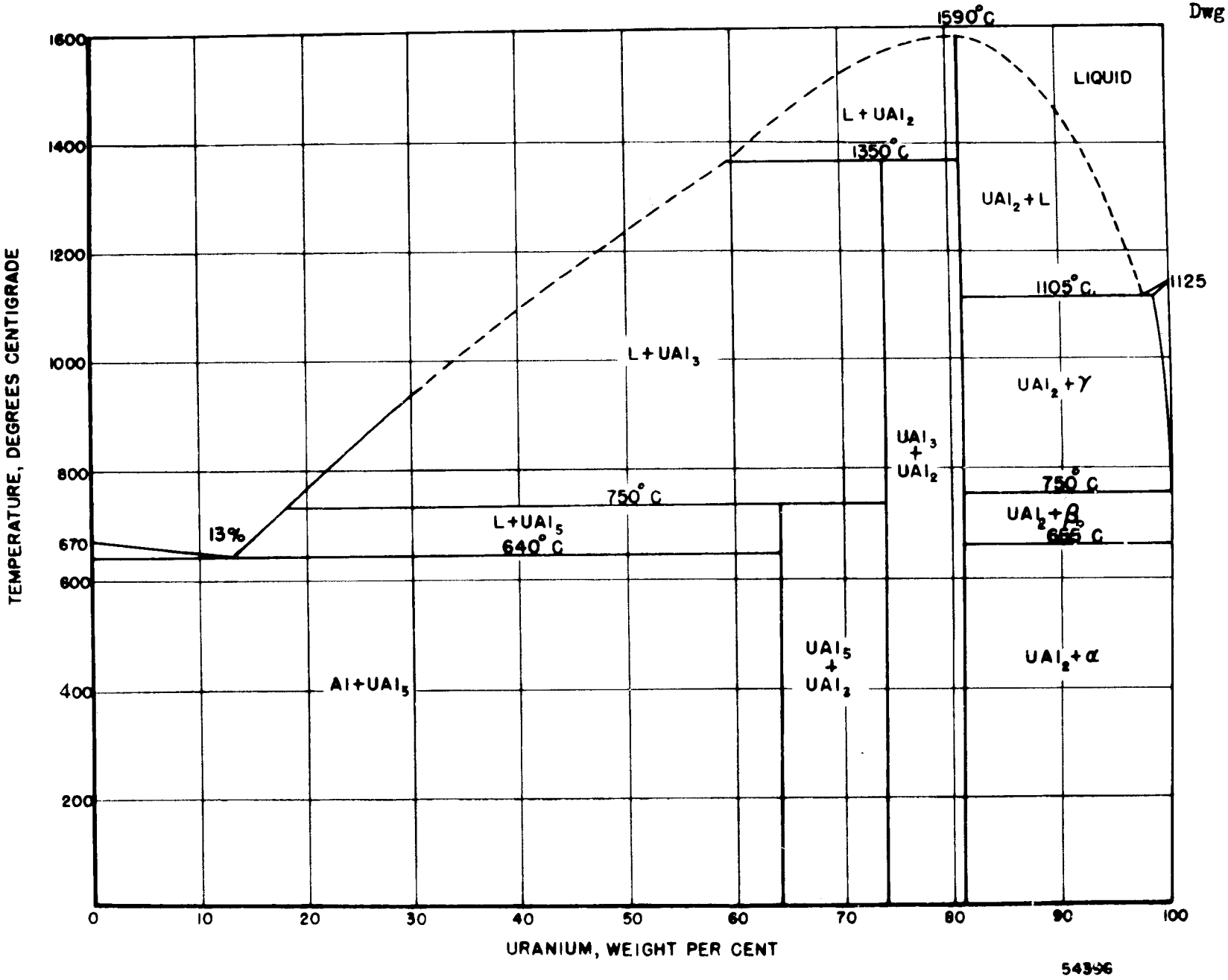

FIG.1

THE ALUMINUM-URANIUM ALLOY SYSTEM.

Figure II shows the two types of as-cast slugs. The larger slugs were cast with a modified type of riser to permit easier chucking for machining.

Equipment and Materials

1. 20 KW Ajax spark gap high frequency converter
2. Heating coil - 15" length x 7-1/2" ID
3. Insulating brick - Kastellite
4. Graphite items (reactor grade)
   A. Crucibles

	Length, inches	
	13	15
ID Top, inches	4.5	5.5
ID Bottom, inches	3.5	4.5
Wall Thickness, inches	0.5	0.5

   B. Stirring rods
   C. Sample spoons
   D. Molds - 1.5" ID
5. Pouring tongs
6. Aluminum pig, 99.74% pure
7. Uranium metal discs

Thickness, inches	1/4 (approx.)
Diameter, inches	1.2 (approx.)
Weight, gm.	130 (approx.)

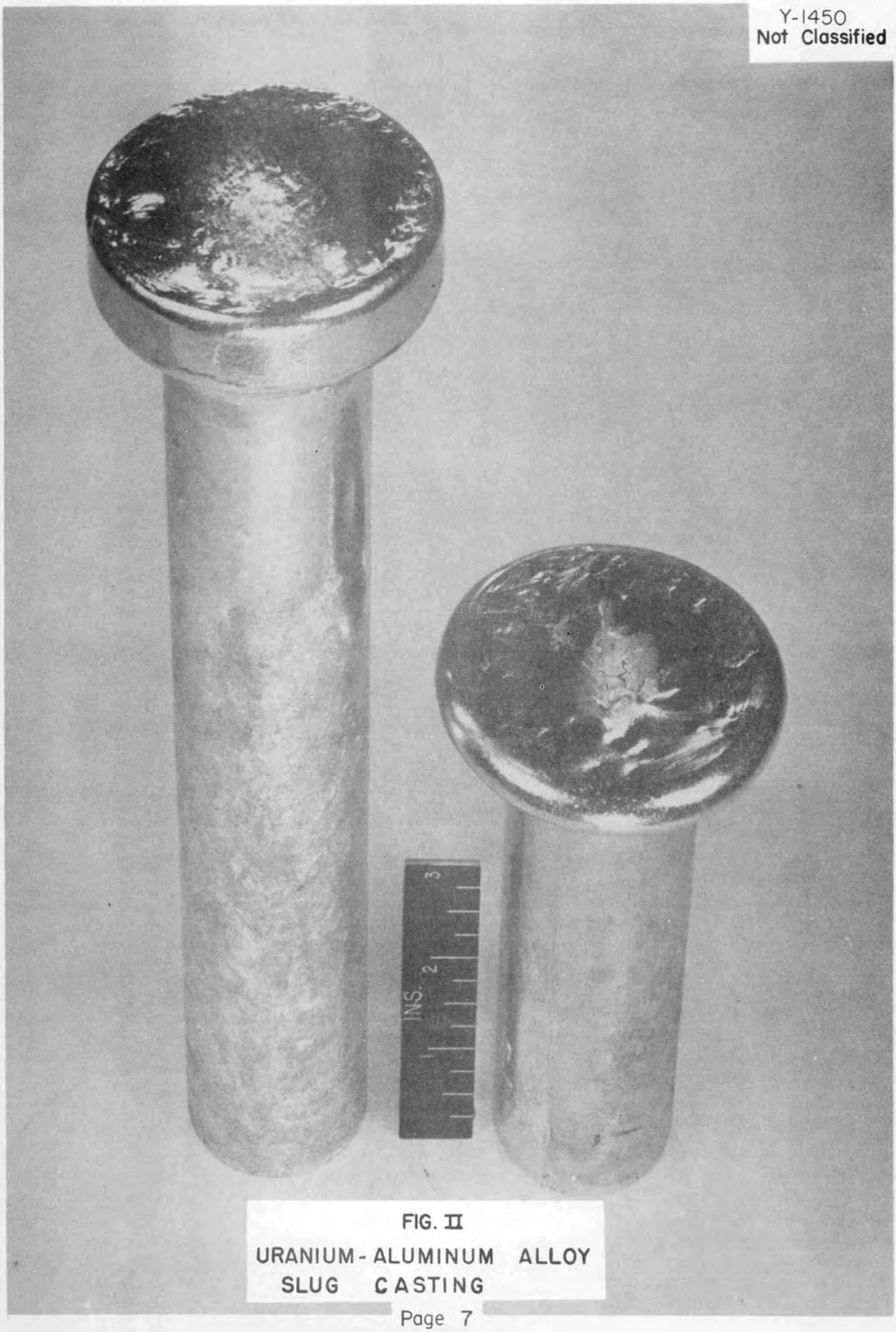

FIG. II
URANIUM - ALUMINUM ALLOY
SLUG CASTING

Melting Procedures

A series of 21 heats was melted and poured under various conditions as shown in Table II. The larger heats, N41 through N48, were melted in the large crucible; all others in the smaller one. For pouring, the crucible was removed from the furnace, inserted in the pouring tongs and tilted. Two samples, weighing about 6 gm. each, were taken from the melt just prior to removal of the crucible from the furnace. All slugs were radiographed after casting to determine porosity.

Dissolution Rate

One of the important variables in determining the dissolution rate is the temperature. To test its effect five heats of 7.5 wt. % U-Al alloy were prepared. Each contained 260 gm. of uranium (two discs) and was used to produce three alloy slugs. The aluminum was heated to the desired temperature, the uranium discs added and the melt stirred until the solid uranium could no longer be felt with the stirring rod. In each case, the melt was held an additional ten minutes at temperature with frequent stirring. The results are listed in Table III and plotted in Figures III-A and III-B on semi-log paper. The straight-line plots indicate that simple diffusion is the controlling mechanism of solution.

TABLE II

Melting Conditions

Heat Number	Type of Charge*	Melting Time, min.	Max. Temp., °C	Pouring Temp., °C	Mold Temp., °C	No. Slugs
N34	V	50	750	750	150	3
N35	V	40	800	800	150	3
N36	V	55	750	750	300	3
N37	V	40	800	800	300	3
N34A	S	30	750	750	150	3
N35A	S	40	700	700	150	3
N36A	S	35	725	725	150	3
N37A	S	40	725	725	150	3
N34B	S	35	725	725	150	3
N35B	S	30	725	725	150	3
N38	V	50	750	725	150	3
N39	V	40	775	725	150	3
N41	V	40	800	750	150	6
N42	S	40	800	750	150	6
N43	V	45	800	725	150	4
N44	V	40	800	725	150	4
N45	S	45	800	700	150	3
N46	S	45	800	725	150	3
N47	S	40	750	725	150	6
N48	S	35	750	725	150	6
N49	V	170	700	700	150	4

* V = Virgin

S = Scrap

TABLE III

Time Required for Dissolution of Uranium in Molten Aluminum

Heat No.	Temp., °C	Total Time, Min.	U Charge, gm.	Av. Rate, gm./min.	Analysis, %U
N37	800	3	285	95.0	7.56
N35	800	3	249	83.0	7.60
N34	750	15	261	17.4	7.36
N36	750	20	256	12.8	7.57
N49	700	140	266.5	1.9	7.54

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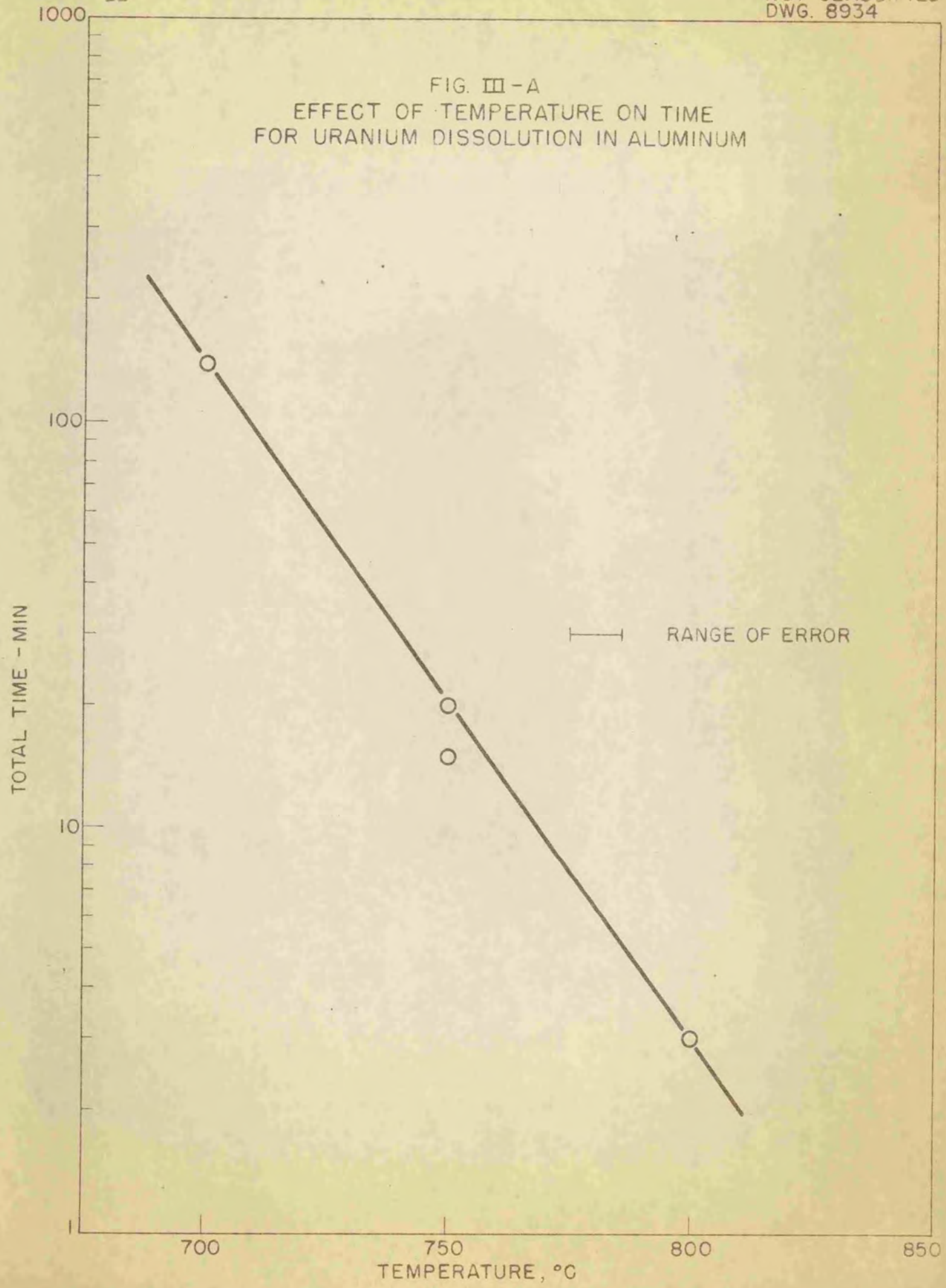

FIG. III-A
EFFECT OF TEMPERATURE ON TIME
FOR URANIUM DISSOLUTION IN ALUMINUM

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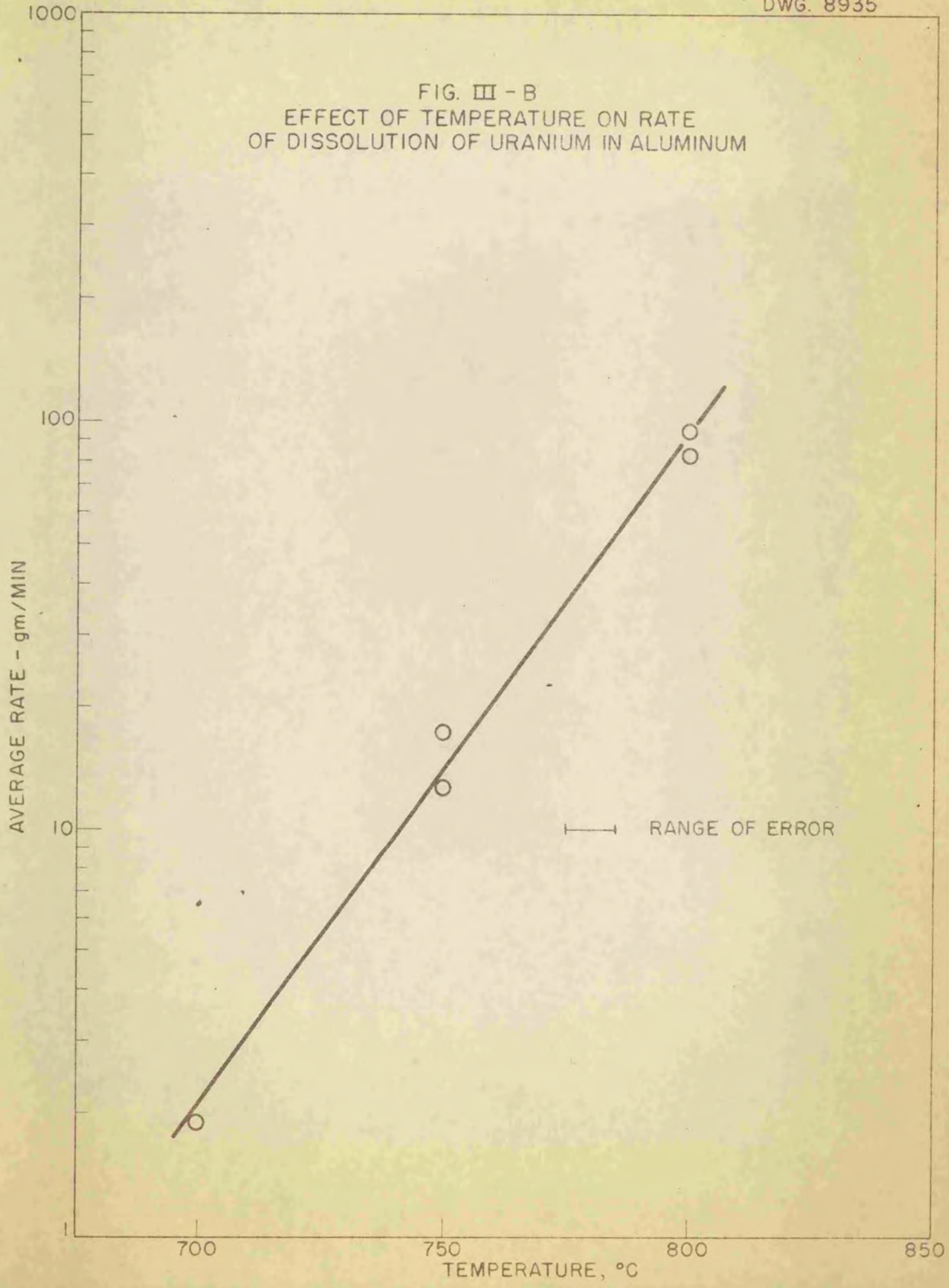
FIG. III - B
EFFECT OF TEMPERATURE ON RATE
OF DISSOLUTION OF URANIUM IN ALUMINUM



Surface Quality of Slugs

In general, surface quality is dependent on pouring temperature, mold temperature and rate of pouring. Varying the mold temperature between 150°C and 300°C (Heats N34 through N37) had little effect on surface quality. As a matter of safety, to insure against presence of moisture, all later heats were poured in molds heated to 150°C. In heats N34 through N37A, the pouring temperature was varied from 700°C to 800°C. Visual examination of slugs from these runs showed that the higher temperatures produced a cleaner surface. Differences were slight, however, indicating that within these limits, pouring temperature is relatively unimportant.

Tilting of the mold at start of pour with a gradual return to a vertical position during pouring was tried in the case of the first two slugs of Heats N34A and N35A. Although the surface of the tilted slugs was superior, differences were too slight to justify further consideration.

The rate of pouring of any metal has a very marked effect on surface quality of the resulting ingot. If the initial rate is too rapid, the metal splashes and freezes, producing scabs on the sides of the ingot which are sometimes detrimental. In the case of too slow pouring, solidification of the ingot proceeds too rapidly, resulting in contraction of the top circumference and subsequent bleeding of new molten metal between mold wall and the contracted portion. This type of defect is commonly called a cold shut. The ideal situation, of course, is to start pouring at a very slow rate and then increase to a rate which is too fast for cold shutting to occur. This optimum condition can only be ascertained by a trained operator by trial and error. To determine its effect on surface, pouring rate was varied from very fast (ca. 6 sec./slug) in Heat N43

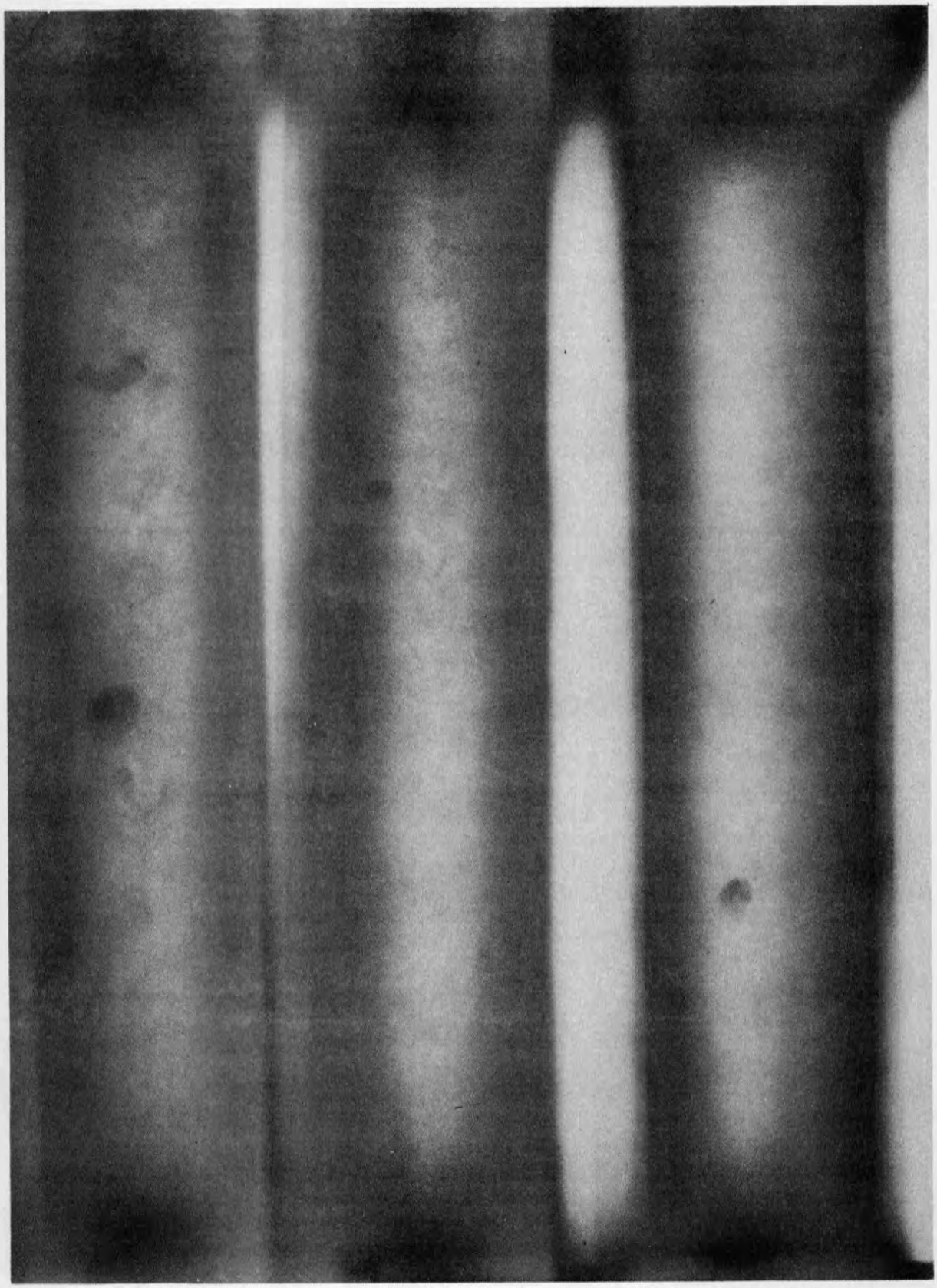
to very slow (ca. 12 sec./slug) in Heat N44. In the first case, considerable splashing occurred and resulted in defects which were not removed by machining. The slow rate of pour, of course, resulted in slugs having very deep cold shuts which also would not machine out.

Porosity

Porosity in aluminum alloy castings can be classified into two types; that caused by shrinkage and that caused by release of some gas from solution due to lowered solubility as the metal freezes. The gas which gives most trouble in the latter case is hydrogen.[3] Primary piping (large void at top of casting open to the air) can be controlled by the use of "hot tops" or a sufficiently large riser of metal at the top of the casting. Secondary piping (usually occurring in the center of the casting) can be minimized within the limitations of mold size by pouring slowly enough so that the ingot is fed continuously.

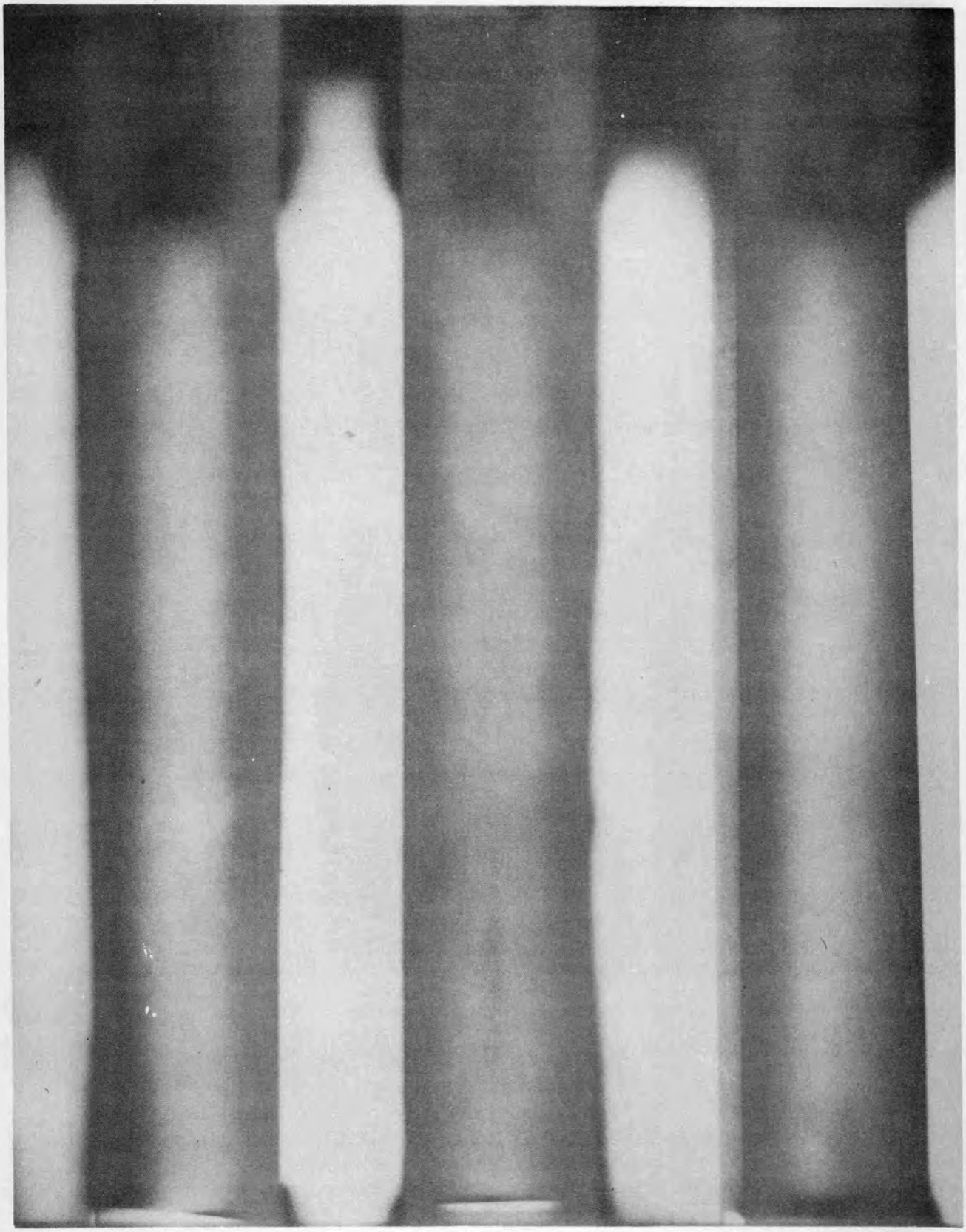
Figure IV-A is a typical radiograph showing porosity in a group of three slugs poured at 800°C from a virgin melt. Figure IV-B is a typical radiograph of sound slugs poured at 725°C from a scrap melt. The small amount of secondary piping shown is not considered to be detrimental. A study of the radiographs and the machined slugs indicates the following:

1. Remelt slugs are superior to virgin slugs cast under the same conditions.
2. Low melting and casting temperatures (725°C) are better than higher temperatures (ca. 800°C).



37-1 37-2 37-3

FIG. IV-A
RADIOGRAPH OF U-AL SLUGS CAST
FROM A VIRGIN MELT (N37)
AT 800°C



34-B 3 34-B 2 34-B 1

FIG. IV-B

RADIOGRAPH OF U-AL SLUGS CAST FROM A SCRAP MELT (N34B) AT 725°C

Item 1, above, may be an indication that one or both of the metals contained considerable quantities of hydrogen.

As mentioned previously, high melting temperatures (ca. 800°C) are necessary to achieve rapid dissolution of uranium in aluminum. It appears, then, that it is desirable to melt at a high temperature, cast and then remelt at a lower temperature to achieve homogeneous and sound slugs.

Segregation

From both an accountability and an engineering standpoint, it is desirable that the slugs be uniform in composition. Since the alloy is a mixture of two phases, one of which is much more dense than the other, it is to be expected that some gravity segregation will take place. To study the magnitude of this inhomogeneity, four heats each producing three slugs (N34B, N35B, N38, N39) were melted and cast as shown in Table II. The riser was removed and a 6 gm. sample representing the entire cross-section machined from the top and bottom of each slug. Results of analyses of these samples are listed in Table IV. In every case, the bottom sample had a higher uranium content than the top. Variation in individual slugs averaged about 2% of the uranium content.

As mentioned earlier, work with 4.5% U-Al slugs had indicated that the turnings and scrap had a slightly lower composition than the melt sample. It was desired to determine whether or not the analysis of the original melt accurately represented uranium content of the machined slugs and the scrap and turnings. Two heats (N41, N42) each producing six slugs, were melted and cast, as shown in Table II, and then machined to size. Then, in each case, the

slugs were remelted in one batch and the shavings in another. Because of the small size of the turnings there was considerable oxidation during melting resulting in a large volume of dross. Duplicate samples were taken just prior to pouring, both of which were submitted for analysis. Results are shown in Table V. In both cases, the metal resulting from the scrap melt had a slightly higher U content than that from the machined slug remelt. This may be due to the fact that some of the uranium oxidized during melting was reduced by aluminum. However, all analyses were within the range of accuracy of sampling and analytical procedures.

Two additional heats (N47, N48) were treated as N41 and N42 with the exception that the slugs and scrap were remelted with cryolite as a flux. It has been shown (4) that under these conditions any uranium present as oxide is reduced by aluminum with very high efficiency (ca. 99%). Therefore, a complete material balance should disclose any large variations in homogeneity. Analytical results are listed in Table V. Here, the slug remelt analyses were about the same as the original melt. The scrap remelt analyses were considerably higher. A complete material balance is shown in Table VI. To compute the weight of uranium in each charge it was assumed that the alloy was homogeneous and therefore the analyses of the original melt sample represented the uranium content of both the machined slugs and the scrap. In both cases, slightly lower yields were obtained with the scrap remelts than with the machined slug remelts. This is an indication that the scrap actually had a slightly lower uranium concentration than the slugs. Differences are however within the range of accuracy of the sampling and analytical variables.

TABLE IV

Segregation in 7.5% U-Al Alloy Slugs

Heat No.	Type	Composition, Wt. % Uranium				
		Melt	Slug #1	Slug #2	Slug #3	
N34B	Scrap	7.39	7.44	7.37	7.43	Top
			7.53	7.50	7.47	Bottom
N35B	Scrap	7.52	7.51	7.48	7.49	Top
			7.59	7.71	7.65	Bottom
N38	Virgin	7.53	7.49	7.35	7.51	Top
			7.66	7.60	7.58	Bottom
N39	Virgin	7.53	7.49	7.45	7.53	Top
			7.60	7.62	7.60	Bottom

TABLE V

Segregation - Machined Slugs vs. Scrap

Heat	Type	Flux	Melt Composition, Weight % U		
			Original	Slugs	Scrap
N41	Virgin	None	7.49	7.43	7.49
				7.46	7.51
N42	Scrap	None	7.47	7.45	7.50
				7.48	7.56
N47	Scrap	Cryolite	7.52	7.54	7.71
				7.56	7.71
N48	Scrap	Cryolite	7.51	7.54	7.68
				7.54	7.70

TABLE VI

Uranium Distribution in Turnings and Slugs

Cast Slugs	Heat N47		Heat N48	
Alloy				
Weight, gm.	4967.0		4999.5	
U content, %	7.52		7.51	
U content, gm.	373.52		375.47	
Machined Slugs	**Charge**	**Product**	**Charge**	**Product**
Alloy				
Weight, gm.	3246.0	3215.5	3274.0	3222.5
U content, %	7.52	7.55	7.51	7.54
U content, gm.	244.10	242.77	245.88	242.98
Slag				
U content, gm.		1.33		2.90
U content, % *		0.54		1.17
Scrap				
Alloy				
Weight, gm.	1721.0	1653.0	1725.5	1651.0
U content, %	7.52	7.71	7.51	7.69
U content, gm.	129.42	127.45	129.59	126.96
Slag				
U content, gm.		1.97		2.63
U content, % *		1.52		2.04

* Percent of uranium charged.

Microstructure

The U-Al phase diagram (Figure I) indicates that the structure of a cast 7.5% U-Al alloy should consist of primary Al plus a eutectic of UAl_5 and Al. Recent work by B. Borie (5) indicates that the compound is actually UAl_4. To study the microstructure of this alloy, one slug from heat N46 was cut longitudinally, polished and etched with hot 10% NaOH. Samples were then cut from the top, middle and bottom of the slug at the edges and center. Microscopic examination showed the primary aluminum surrounded by a eutectic. The grain size increased progressively, from edge to center as shown in Figure V. There was no observable difference in the ratio of amounts of UAl_4 to aluminum. Some microporosity was found in the center of the slug which did not show up in the radiograph of the as-cast slug.

Summary

It appears from previous work and the results of this investigation that reasonably sound and homogeneous slugs of U-Al alloy can be produced by direct alloying of the two metals as follows:

1. Melt aluminum to 800°C
2. Add uranium metal
3. Stir vigorously until U metal is dissolved
4. Stir for an additional 10 minutes
5. Cast and remelt at 725°C
6. Stir vigorously for 10 minutes
7. Cast in warm (150°C) graphite molds at a rate of about 9 seconds per slug (8 inch length)

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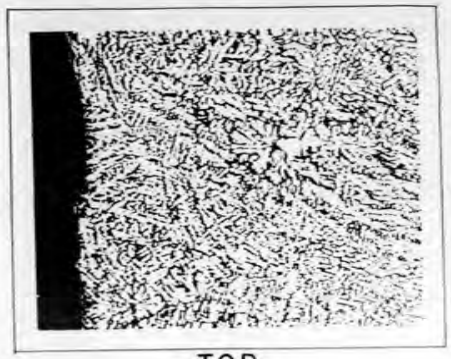

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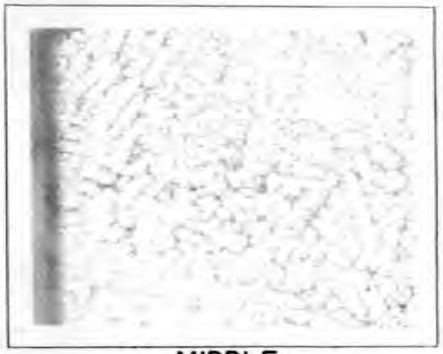

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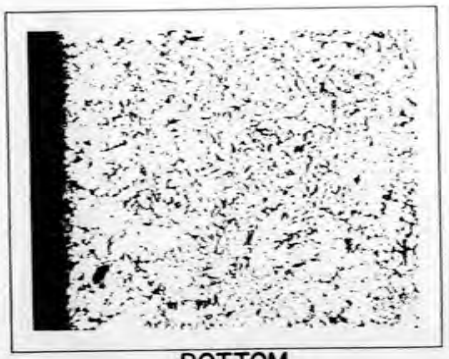

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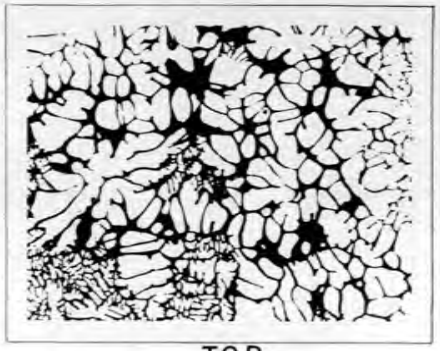

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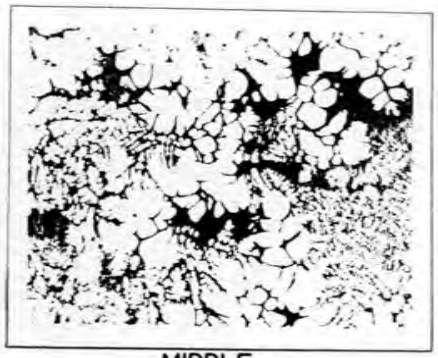

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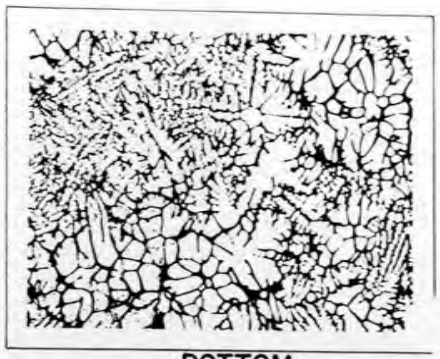

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FIG. V

MICROSTRUCTURE OF 7 1/2 % URANIUM–ALUMINUM ALLOY

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The data presented indicate that some segregation (ca. 2% of contained uranium) is found from top to bottom but that there is no significant difference between the uranium analyses of the machined slugs and the turnings.

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Metallography	R. S. Crouse
	R. J. Gray
Aid in preparation of report	F. Kerze, Jr. (Supervisor)
	G. M. Adamson, Jr.

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