UNCLASSIFIED

253

C[redacted]AL

NMI-1180

METALLURGY AND CERAMICS (M-3679, 20th Ed.)

UNITED STATES ATOMIC ENERGY COMMISSION

GRAIN REFINEMENT OF CAST URANIUM BY HEAT TREATMENT

By
G. W. Powell
J. L. Klein
D. Krashes

July 5, 1957

Nuclear Metals, Inc.
Cambridge, Massachusetts

RESTRIC[redacted] DATA

This docu[redacted]ins restricted data as defined[redacted]mic Energy Act of 1954. Its tra[redacted] or the disclosure of its contents [redacted]any manner to an unauthorized person is prohibi[redacted]

This documen[redacted]onfidential-Restricted D[redacted] to civilian applica-tion[redacted]tomic energy.

Technical Information Service Extension, Oak Ridge, Tenn.

AEC RESEARCH AND DEVELOPMENT REPORT

C[redacted]AL

UNCLASSIFIED

DECLASSIFIED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

CONFIDENTIAL

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

This report has been reproduced directly from the best available copy.

Printed in USA. Charge 65 cents. Available from the Technical Information Service Extension, P. O. Box 1001, Oak Ridge, Tennessee. Please direct to the same address inquiries covering the procurement of classified AEC reports.

CONFIDENTIAL

AEC Technical Information Service Extension
Oak Ridge, Tennessee

UNCLASSIFIED

CONFIDENTIAL

NMI-1180

Grain Refinement of Cast Uranium by Heat Treatment

G. W. Powell, J. L. Klein, and D. Krashes

July 5, 1957

Nuclear Metals, Inc.
Cambridge, Massachusetts

Contract No. AT(30-1)-1565

A. R. Kaufmann
Technical Director

This document is
PUBLICLY RELEASABLE
B Steele
Authorizing Official
Date: 4-17-17

Classification canceled (or changed to UNCLASSIFIED)
by authority of ... dated 3-16-60
by ... TIE, date 4-29-60

CONFIDENTIAL

UNCLASSIFIED

DECLASSIFIED

ABSTRACT

The thermal cycling, irradiation behavior, and fabrication characteristics of uranium are strongly dependent upon grain size; a smaller grain size yields the better properties. Grain refinement of cast uranium may be accomplished either by hot working in the high alpha temperature range or by heat treatment; refinement by the latter method was the object of this investigation. The variables studied were time, temperature, specimen size, multiple quenching, and alpha-annealing after quenching.

The results of this investigation have led to the following conclusions, within the scope of the experimental conditions:

1.) The grain size of cast uranium may be appreciably refined by beta-quenching.

2.) Time and temperature in the beta phase are not significant in determining the final alpha grain size.

3.) Multiple beta-quenches (two to four times) produce better grain size refinement than a single quench.

4.) Alpha-annealing after beta-quenching causes recystallization, but does not greatly affect the grain size.

5.) Gamma-quenching is not recommended as a practical heat treatment.

UNCLASSIFIED

TABLE OF CONTENTS

I. INTRODUCTION

The grain size and grain orientation of massive uranium have considerable influence on its fabrication qualities and on its behavior during thermal cycling and irradiation. Both thermal cycling and irradiation produce surface roughening in coarse-grained uranium, and either process can cause axial changes of length when the metal exhibits a high degree of preferred orientation.[1] Observation of alpha-extruded uranium disclosed that pieces extruded from coarse-grained billets have rough or striated surfaces, whereas pieces extruded from fine-grained billets are smoother.[2] Since rough-surfaced fuel elements are undesirable, the grain size of cast uranium is refined to enhance the surface smoothness of subsequently extruded forms. Two different methods of grain refinement have been used in preparing extrusion billets: (1) hot working in the high alpha-temperature range (1000°F to 1200°F) by either hammer forging, press forging, or pre-extrusion, and (2) heat treatments resulting in the formation of new grains by heating and cooling through one or more phase changes. Grain refined billets made by any of these processes result in extruded products having about the same surface quality, although the grain size of hot-worked cast uranium is an order of magnitude finer than that of heat treated cast uranium.

Because the hot working of cast uranium is dependent upon the availability of extrusion and forging presses of appropriate size, heat treatment is sometimes the only feasible method of obtaining grain refinement prior to final extrusion. Although it has been known that heat treating can produce grain refinement in cast uranium, the effects of many of the heat treatment variables had not been established, including temperature of treatment, time at temperature, specimen size, and alpha-annealing after heat treatments. The purpose of the work reported here

----

(1) F. G. Foote, Nuclear Metallurgy, Vol.1, AIME, 1955.

(2) D. Krashes and J. M. Fitzpatrick, "Grain Refinement of Cast Uranium", NMI-1153, Ames Information Meeting, 1956.

DECLASSIFIED

was the determination of the effect of these variables on the grain refinement obtainable by heat treating cast uranium.

II. EXPERIMENTAL METHODS AND MATERIALS

A. Materials

The reactor-grade uranium used in this investigation was obtained from the Feed Materials Processing Center of the National Lead Company in the form of casting No. 1539, 3 inches in diameter by 34 inches long. The analysis of this material, given in the table, indicates an impurity content which may be considered normal.

B. Experimental Methods

Specimens of two different sizes were used. The smaller were 1/2 inch thick, 30° sectors of the 3 inch diameter cross section of the ingot. The large specimens were round slugs, 2 inches thick and 3 inches in diameter. Consequently, a slug was heavier than a pie-shaped specimen by a factor of 48.*

All heat treatments were performed in salt pots containing either a carbonate or chloride-base salt. At temperatures above the beta-gamma phase transformation, the carbonate salt caused extensive corrosion of the uranium, while corrosion was considerably less in the chloride salt. Therefore, most heat treatments above 1418°F (770°C) were carried out in a chloride salt and either a chloride or carbonate salt used at lower temperatures.**

Each heat treatment was initiated by heating into either the beta or the gamma-temperature range. The pie-shaped specimens were given a minimum of 16 minutes and a maximum of 128 minutes in the salt prior to

----

*Results of heat treatment of slugs from 4-3/4 inch diameter and 6-3/4 inch diameter ingots are given in the appendix.

**One disadvantage of molten chloride salts is that they will attack stainless steel pots at the air-salt interfaces. However, it is understood that mild steel pots will contain chloride salts without undue attack, although there is no experience of this at NMI.

quenching. In all experiments with the larger slugs, each heat treatment included a constant heating time of 45 minutes. The range of the heating temperature investigated was from 1250°F (677°C) to 1650°F (900°C). In many cases, the quenched specimens were given a 2 hr alpha-anneal at either 1000°F (538°C) or 1175°F (635°C).

C. Metallographic Preparation

Each specimen was sectioned with a wet cutoff wheel to expose an interior surface for examination. Different polishing and etching procedures were used for macro and microstructure. For macroexamination, the sectioned specimens were ground on a belt sander to a No. 500 grit finish. The specimens were then successively etched in concentrated hydrochloric acid, rinsed in water, rinsed in 50:50 nitric acid and finally washed in cold water. The preparation of the sectioned specimens for microexamination started with polishing the specimens on wheels covered with silicon carbide polishing paper, graded from 320 to 600 grit. Following this, the specimens were polished on silk-covered wheels with an abrasive of Linde A in a 10% oxalic acid solution, then electropolished in an electrolyte comprised of orthophosphoric acid, sulphuric acid and water (1:2:2 by volume).

III. RESULTS AND DISCUSSIONS

A. General Effects of Beta and Gamma-Quenching

The grain structure of cast uranium is refined by heating into the beta or gamma-phase temperature range and quenching. The structural changes and evident grain refinement which occur are shown in Figs. 1 and 2. Both the cast and the heat-treated structures contain numerous subgrains, and the grain boundaries are very irregular. In some of the surface grains of the beta-quenched specimens there is a noticeable tendency to grow perpendicular to the surface. In the gamma-quenched specimens, however, oriented growth is quite pronounced, and the over-all

DECLASSIFIED

grain structure, particularly that of the slug, resembles the over-all structure of an entire casting.

The results presented in Sections B and C, below, show the effect of time, temperature, multiple quenching, alpha-annealing and size of specimen on the grain structure of heat-treated cast uranium.

B. Heat Treatments Originating in the Beta-Phase Temperature Range

1. Time and Temperature

For the time-temperature studies, some pie-shaped specimens were heated to 1250 (30°F above the alpha-beta transformation), 1300, 1350 and 1400°F (18°F below the beta-gamma transformation) and then held for times of 16, 64 and 128 minutes prior to quenching. Examples of structures obtained by these treatments are illustrated by the macros and photomicrographs of Figs. 3 and 4. Extensive examination of the structures of these specimens has led to the following conclusions for the ranges of times and temperatures in these experiments:

a) For all temperatures within the beta range, the grain size is independent of the time.

b) The grain size is independent of the heating temperature within the beta range.

Two of the possible explanations for these results are as follows:

a) Alpha grains are nucleated primarily at the grain boundaries of the beta phase. Therefore, it must be inferred that it is not possible to heat beta grains to a sufficiently high temperature (because of transformation to gamma-uranium) to exceed the beta-grain size obtained on first transforming the alpha to beta. This may not apply to high purity uranium.

b) Alpha grains are nucleated primarily by impurity particles, such as oxides and carbides. In such a case, the prior beta grain size would have a minor effect on the final alpha grain size.

However, the effect of multiple quenching on grain size, presented below, gives more support to the first explanation.

2. Multiple Quenching

Increased grain refinement may be obtained by heating to and quenching from a given beta temperature several times.[2] To establish the maximum grain refinement obtainable by multiple beta-quenching, several of the pie-shaped specimens were given from one to four quenches, from both 1250°F and 1400°F. The results showed that for either heating temperature the grain structure is definitely refined by a second beta-quench, but that additional quenches do not cause further refinement (see Fig. 5 for series at 1250°F). An incomplete rim of columnar grains which is noticeable in the structure of the specimen given a single quench is broken up by a second quench. A maximum number of beneficial quenches implies an upper limit on the number of alpha nuclei that can form as a result of going through the phase change.

One large slug was given a single quench, and a second slug four successive quenches at 1250°F. Macros of these specimens are shown in Fig. 6,* in which the increased grain refinement obtained by multiple quenching is evident. In a previous unpublished study made by one of the authors, specimens, 2 inches thick with diameters of 4-3/4 inches or 6-3/4 inches, were given as many as six beta-quenches from 1350°F. It was concluded that maximum grain refinement is obtained after three or four quenches. Macros of these specimens are given in the appendix.

----

*The crack in the center of the sample shown in Fig. 6 (b) is often observed in solid massive uranium after multiple beta treatments. Cracks are caused by the tensile stresses at the center of the casting resulting from the combination of two factors: (1) There is a discontinuous linear contraction of about 0.3% when beta-uranium transforms to alpha-uranium, and (2), during quenching, the outer portion of the casting is the first to transform to alpha-uranium, the center last. When cast uranium is drilled to form a billet for a tube extrusion and then given a multiple beta treatment, no cracking is observed. Since the center of a cast uranium billet is probably the least sound and weakest portion, removing this by machining accounts for the lack of cracking in this case.

DECLASSIFIED

3. Alpha-Annealing After Beta-Quenching

Gardner[3] has shown that beta-quenched wrought uranium may be made to recrystallize by an alpha-anneal. The large volume change accompanying the beta to alpha-phase transformation and the high anisotropic thermal expansion of uranium are thought to induce sufficient strain in the lattice during rapid quenching to cause recrystallization during an alpha-anneal.[4] Since this type of heat treatment, beta-quenching followed by alpha-annealing, is a potentially useful method by which to obtain grain refinement, several of the pie-shaped specimens were quenched from either 1250 or 1400°F and then annealed at either 1000 or 1175°F for 2 hr. The microstructures of the specimens quenched from 1250 and annealed at 1175°F are presented in Fig. 7. The following observations were made:

a) Recrystallization did not occur during the 2 hr anneal at 1000°F but did during the anneal at 1175°F.

b) The recrystallized grain size for specimens annealed 2 hr at 1175°F is independent of the number of beta quenches (Fig. 7).

c) The grain size of the recrystallized structure is larger than that of the as-quenched structure after four beta-quenches.

The first observation is in agreement with the data of Wright and Riches for wrought beta-quenched dingot uranium, which indicate incubation periods of roughly 3 minutes at 1175°F and 11 hr at 1000°F.

It was concluded from this series of experiments, that for the times and temperatures employed, the grain size is not profoundly changed by alpha-annealing. The recrystallized structure, however, may become more random, although this was not investigated. To obtain further grain refinement by alpha-annealing of beta-quenched uranium, it would probably be necessary to resort to annealing at lower temperatures for more ex-

----

(3) Gardner, H. R. "Grain Refinement Produced by an Alpha Phase Anneal of Beta Phase Heat Treated and Water Quenched Uranium", January 3, 1955, HW-34368.

(4) Wright, P.D., and Riches J. W., "Contribution to the Study of Recrystallization of Uranium Quenched from Beta Phase", May 18, 1956, HW-4311.

tended periods of time.

Alpha-annealing experiments were also made with the large slugs, and some interesting results were obtained. Figs. 8 and 9 are macros and photomicrographs, respectively, for single beta-quenched slugs in the as-quenched and annealed states. Annealing definitely causes recrystallization in these large sizes, being slightly more pronounced in the rim than in the core. After annealing, there is not much difference between the structures resulting from four beta quenches and that obtained with a single quench.

When beta-treated cast uranium billets are heated to 1100 to 1200°F for extrusion, they are necessarily alpha-annealed. Without doubt, the good surface obtained on tubes extruded from beta-treated, cast billets is partially attributable to the inadvertent alpha-anneal resulting from the billet heating.

C. Heat Treatments Originating in The Gamma-Temperature Range

1. Time and Temperature

The investigation of the time-temperature relationships in the gamma-temperature range was not very extensive. The pie-shaped specimens were held for only two times, 16 and 64 minutes, and two temperatures, 1450 and 1650°F. Photomicrographs of the structures of these specimens are shown in Fig. 10. On the basis of rather limited experimentation, it is concluded that gamma-quenching can produce grain refinement only with short times at low temperatures.

2. Multiple Quenching

Macros and photomicrographs of the core material of specimens given from one to four quenches from 1450°F are shown in Fig. 11. Although the grain size of the core material was not profoundly affected, the columnar grains which persisted through the second quench were broken up by the third.

The large slugs were also given multiple-quenching heat treatments,

UNCLASSIFIED

but regardless of the number of quenches from 1450°F, the over-all structure remained unchanged (Fig. 2). Since the growth of columnar grains is favored by slow cooling rates, the repeated formation of columnar grains in the large slug even after several quenches might be expected.*

3. Alpha-Annealing After Gamma-Quenching

The microstructures of some pie-shaped specimens, quenched either one or four times from 1450°F and then annealed at either 1000 or 1175°F, are shown in Fig. 12. Contrary to the results obtained with the annealed beta-quenched specimens, recrystallization occurred at 1000°F. For some unknown reason, however, the specimens annealed at 1175° F showed only partial recrystallization when the specimen was given a single quench, and no signs of recrystallization when the specimen was given four quenches. When these experiments were carefully rerun, the same results were obtained. Since recrystallization occurred at 1000°F, the absence or incompleteness of recrystallization at 1175°F is unexpected, and without any obvious explanation. The grain size of the specimens annealed at 1000°F is, on the average, coarser than that of the core material after quenching. Annealing, however, does result in a structure which has a uniform grain size, without columnar grains.

Gamma-quenching plus annealing of the large slugs did not cause recrystallization of any portions of the specimens. It will be recalled that alpha-annealing of beta-quenched slugs did cause recrystallization (see Figs. 8 and 9)

4. Practical Usefulness of Gamma-Quenching

Gamma-quenching appears to offer no advantages over beta-quenching, gives less satisfactory results, and is not recommended for practical application.

----

* Columnar grains at the rim have a tendency to have the [001] parallel to the long axis of the grain.

IV. APPENDIX - RESULTS FOR LARGE SLUGS

The macros presented in Figs. 13 through 16 are of large slugs, 4-3/4 inches and 6-3/4 inches in diameter and 2 inches thick, each quenched from one to four times from 1350°F. A selected few of these slugs were subsequently annealed at 1175°F. Macros of the annealed structures are also shown. The additional grain refinement achieved by multiple quenching of these large sizes is evident, three or four quenches producing maximum refinement.

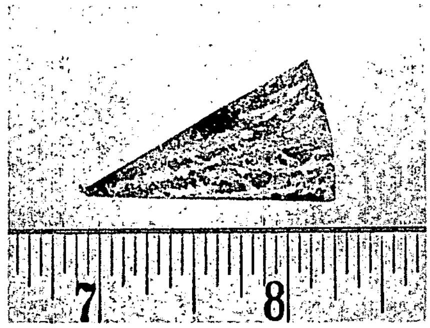
UNCLASSIFIED

V. TABLES AND FIGURES

Table

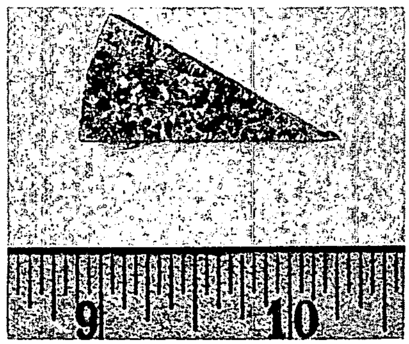
Chemical Composition of Ingot No. 1539

Chemical Analyses in ppm			Spectrographic Analyses in ppm							
Cl	C	N	B	Cd	Cr	Fe	Mg	Mn	Ni	SiO_2
1	430	76	0.2	0.2	6	25	6	4	45	20



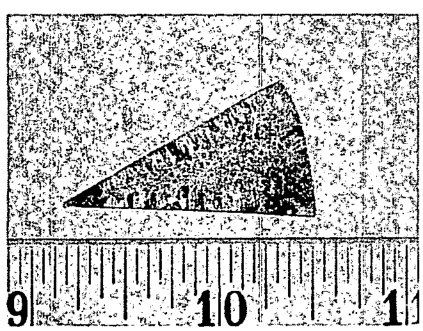
1X RF 3515

(a) As-cast.



1X RF 3435

(b) Water-quenched from 1250^{o}F.

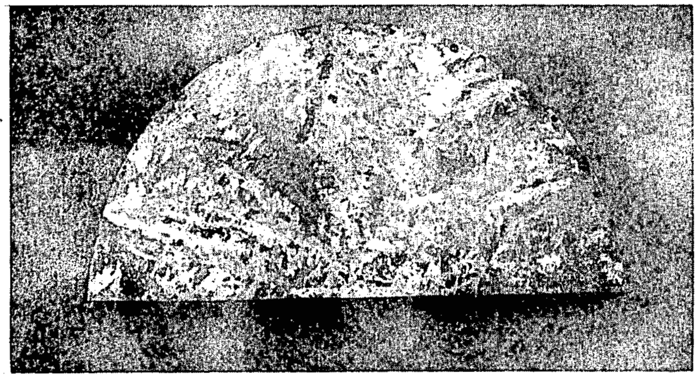


1X RF 3852

(c) Water-quenched from 1450^{o}F.

<u>Fig. 1</u> - Small specimens of cast uranium, showing grain refinement obtained by water-quenching from (b) beta phase and (c) gamma phase temperatures.

UNCLASSIFIED

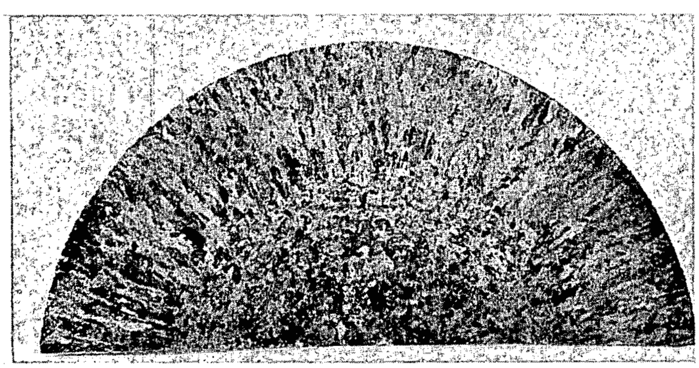


1X RF 4437

(a) As-cast.

1X RF 4327

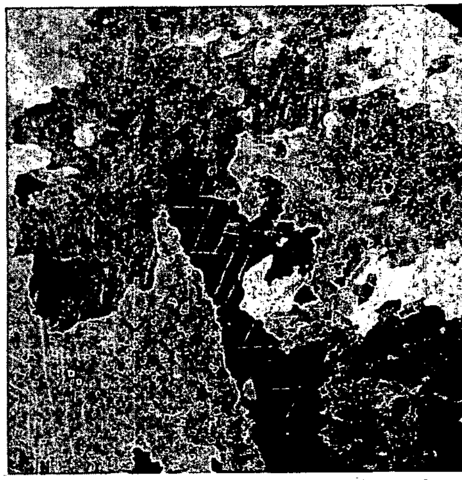
(b) Water-quenched from 1250°F (β).



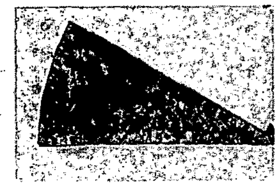
1X RF 4326

(c) Water-quenched from 1450°F (γ).

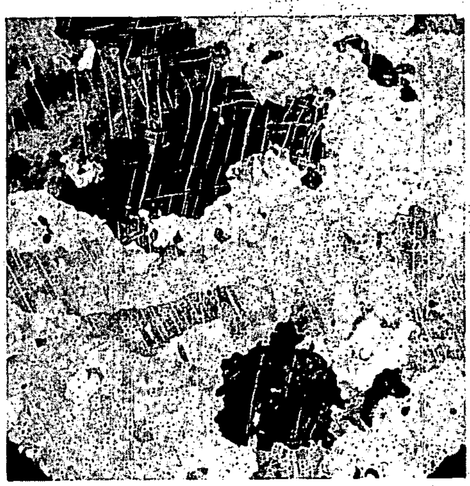
<u>Fig. 2</u> - Large slugs (3 inches diameter x 2 inches thick) of cast uranium, showing grain refinement obtained by water-quenching from (b) beta phase and (c) gamma phase temperatures.



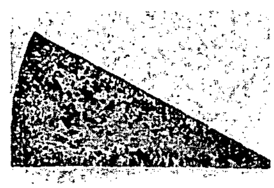
16 min



50X U-269-4 1X RF 3440

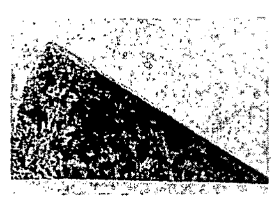


64 min



50X U-269-6 1X RF 3444

128 min



50X U-269-5 1X RF 3442

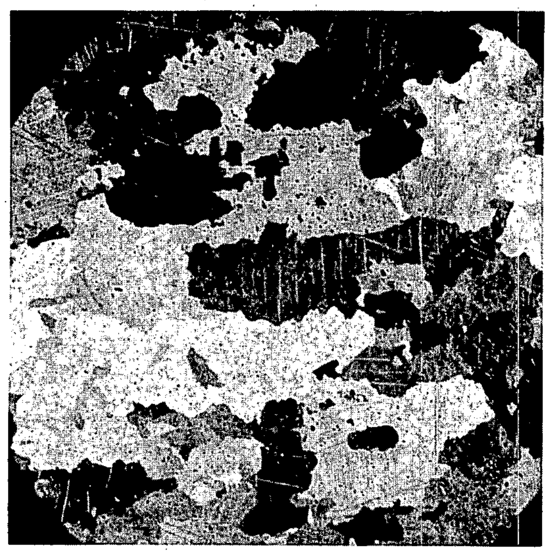
Fig. 3 - Effect of holding time at 1300°F on the grain structure of beta-quenched cast uranium.

DECLASSIFIED

50X 1250°F U-265-5

50X 1300°F U-269-5

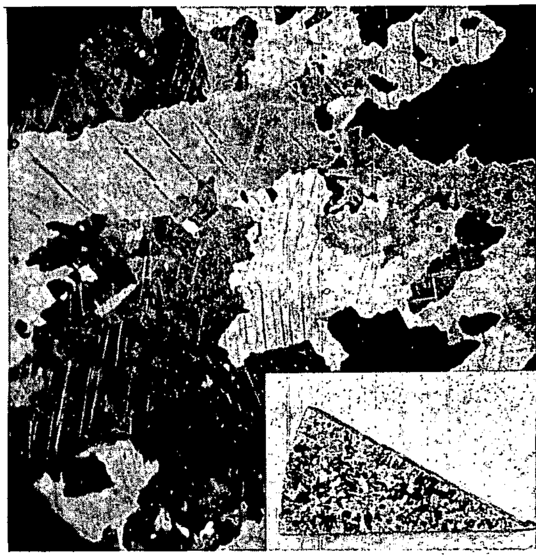
50X 1350°F U-269-3



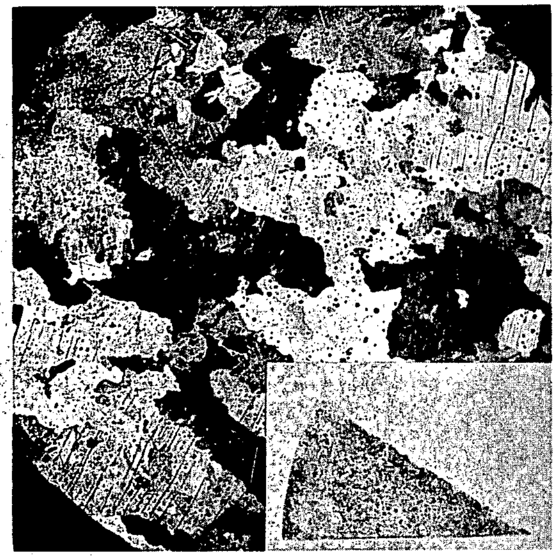
50X 1400°F U-265-3

Fig. 4

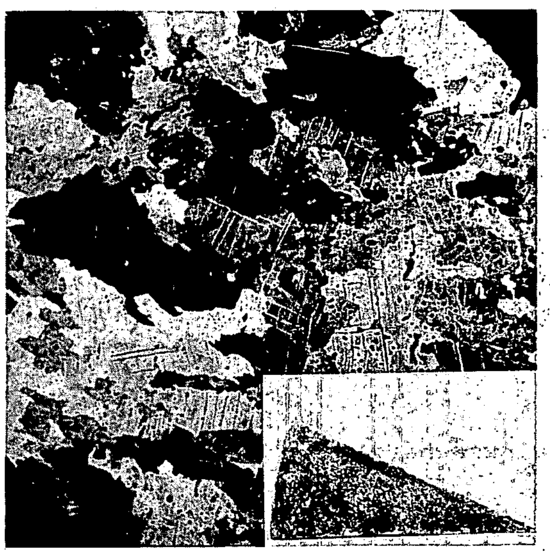
Effect of heating temperature in beta range on grain structure of beta-quenched cast uranium held for 128 min at temperature.



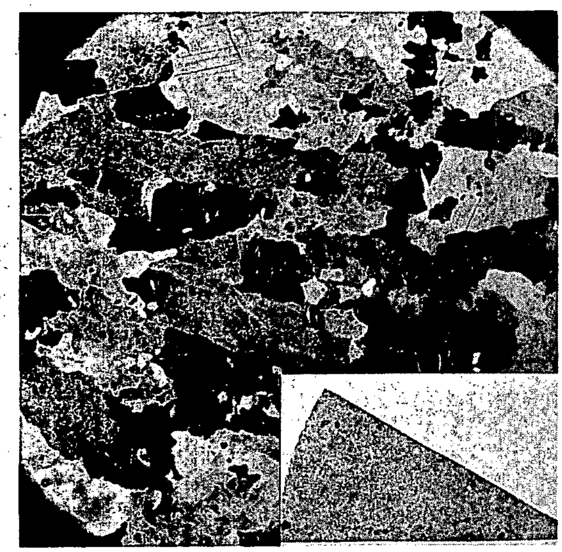
U-254-8 RF 3435
50X 1X
One beta quench.



U-270-4 RF 4027
50X 1X
Two beta quenches.



U-270-5 RF 4028
50X 1X
Three beta quenches.

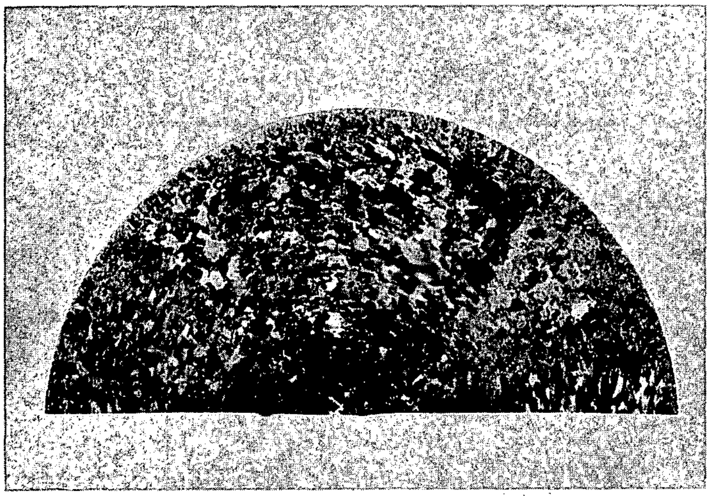


U-254-3 RF 4058
50X 1X
Four beta quenches.

Fig. 5

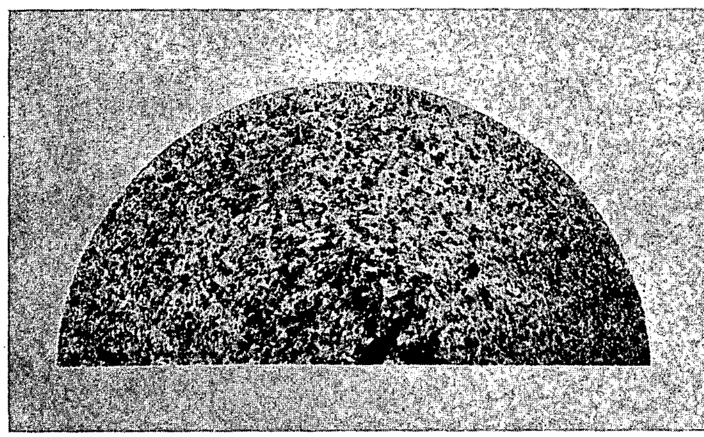
Effect of repeated beta quench treatments at 1250°F on structure of small samples of cast uranium.

UNCLASSIFIED



1X RF 4327

(a) One beta quench.



1X RF 4329

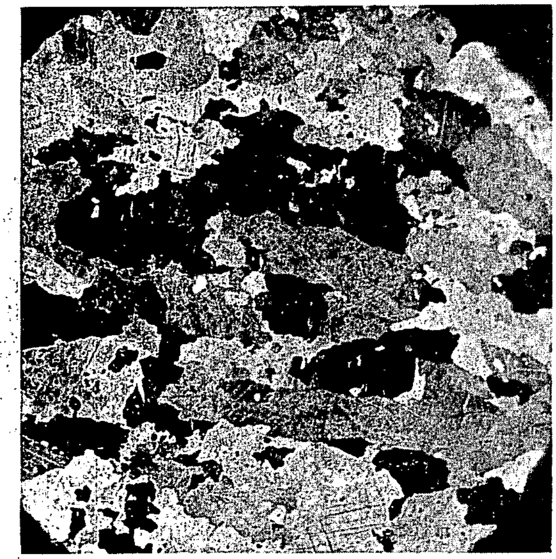
(b) Four beta quenches.

Fig. 6

Effect of repeated beta-quench treatments at 1250°F on structure of 2 inch thick, 3 inch diameter slugs of cast uranium.

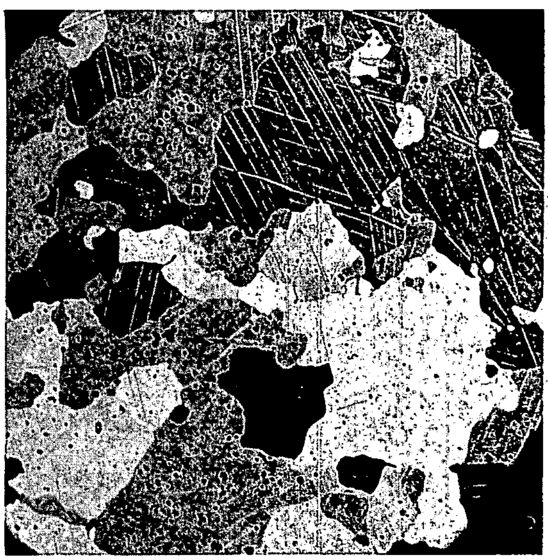
50X U-254-8

One quench.



50X U-254-3

Four quenches.



50X U-265-7

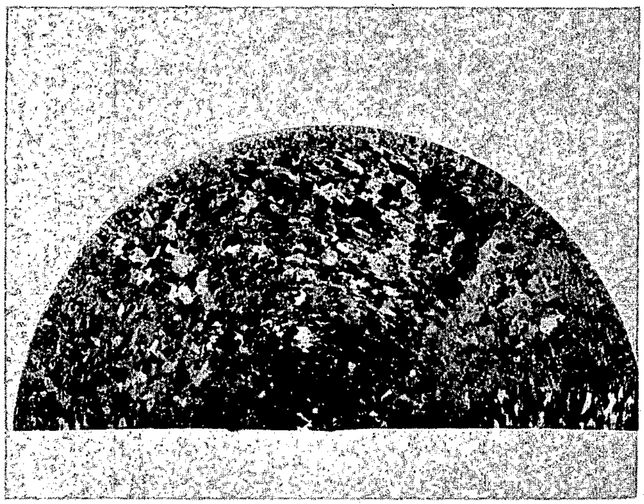
One quench plus 2 hr anneal at 1175°F.

50X U-254-1

Four quenches plus 2 hr anneal at 1175°F.

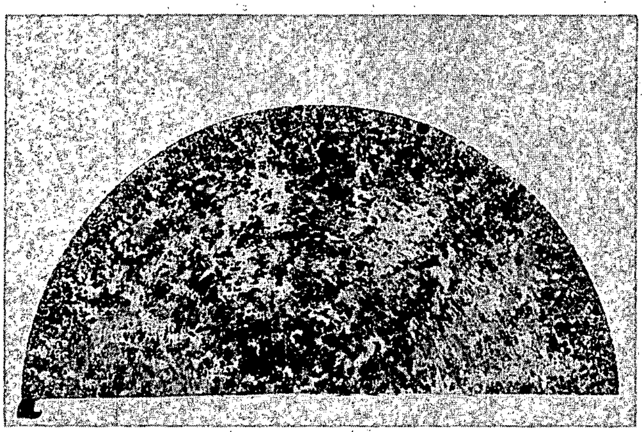
Fig. 7 - Effect of alpha-annealing on beta-treated cast uranium.

DECLASSIFIED



1X RF 4327

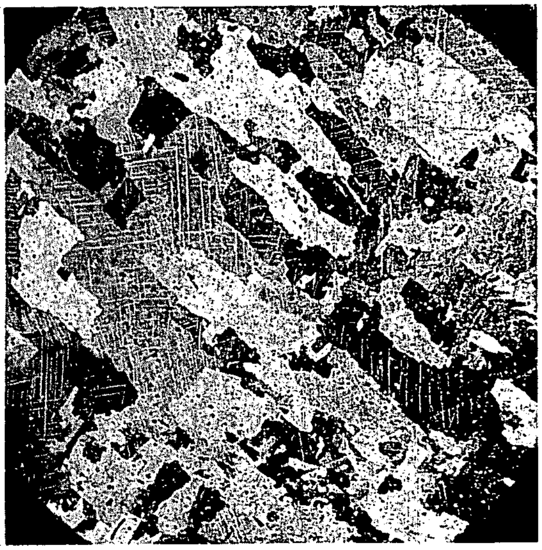
One quench.



1X RF 4328

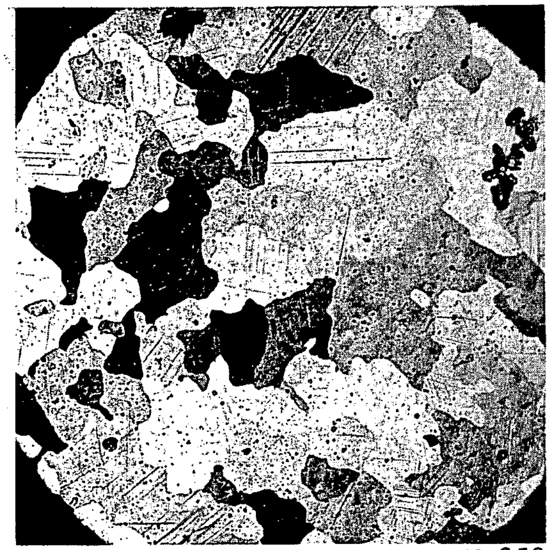
One quench plus 2 hr anneal at 1175°F.

Fig. 8 - Effect of alpha-annealing on beta-quenched cast uranium slugs, 2 inches thick x 3 inches diameter.



50X U-259-8

(a) As-quenched, rim.



50X U-259-6

(b) Annealed, rim.

50X U-259-7

(c) As-quenched, core.

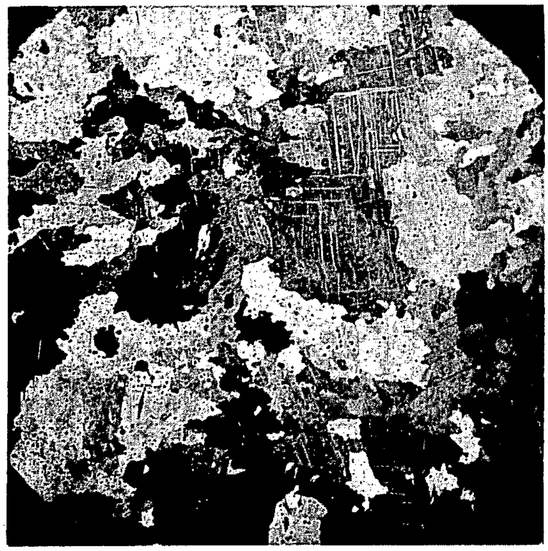
50X U-259-5

(d) Annealed, core.

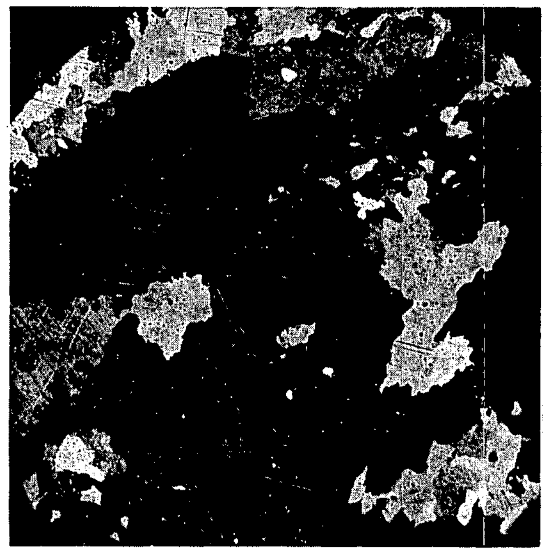
Fig. 9

Photomicrographs of sections shown in Fig. 8.

DECLASSIFIED

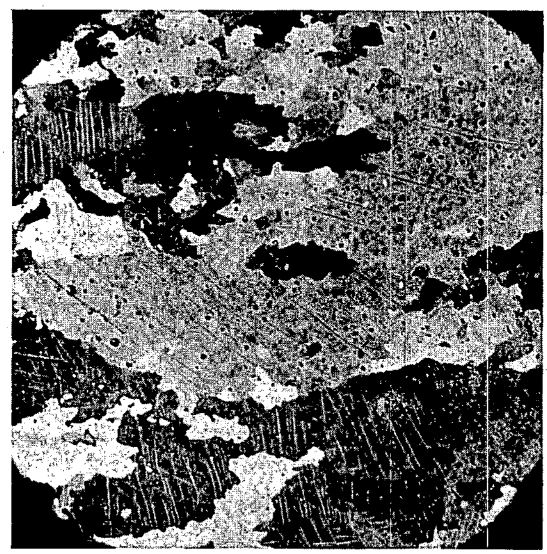


50X U-254-10
16 min at 1450°F.



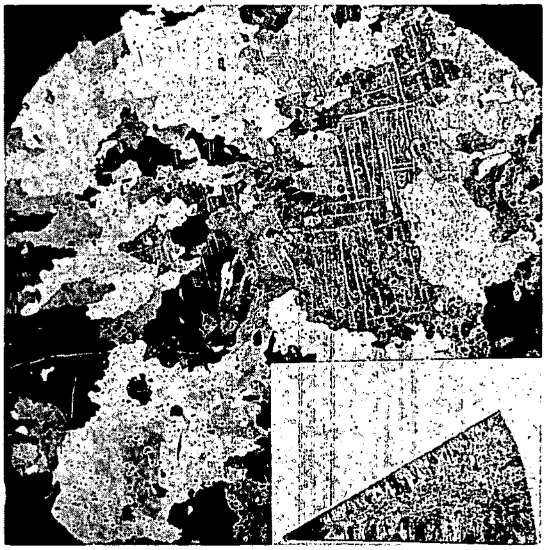
50X U-255-2
16 min at 1650°F.

50X U-255-15
64 min at 1450°F.

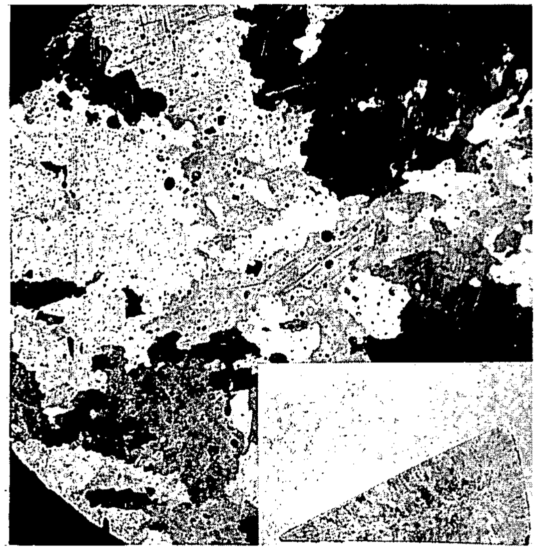


50X U-265-9
64 min at 1650°F.

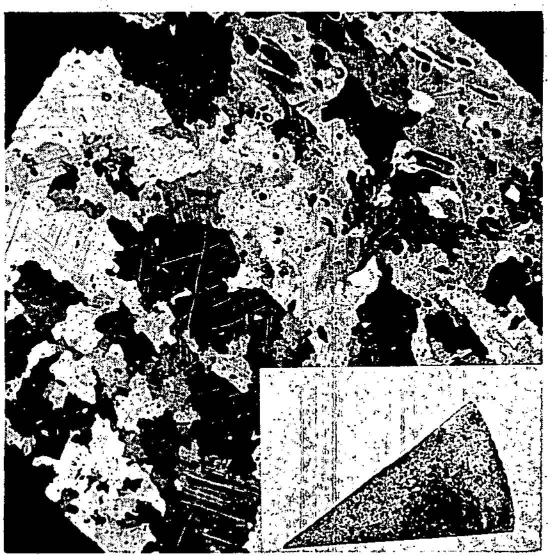
Fig. 10 - Effect of time and temperature in structure of gamma-quenched cast uranium.



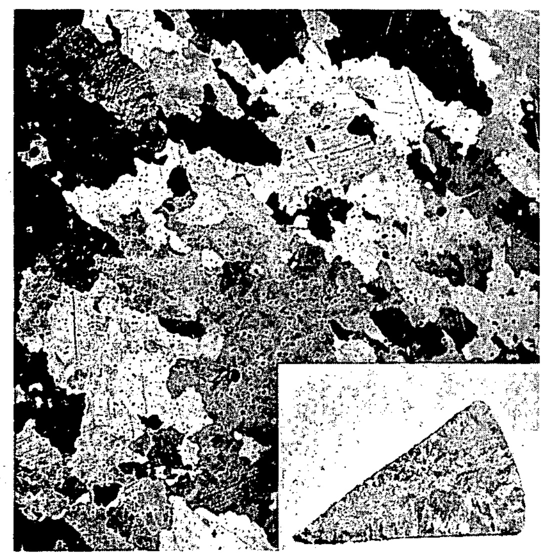
U-254-10 RF 3852A
50X One quench. 1X



U-270-2 RF 3850
50X Two quenches. 1X



U-270-1 RF 3851
50X Three quenches. 1X



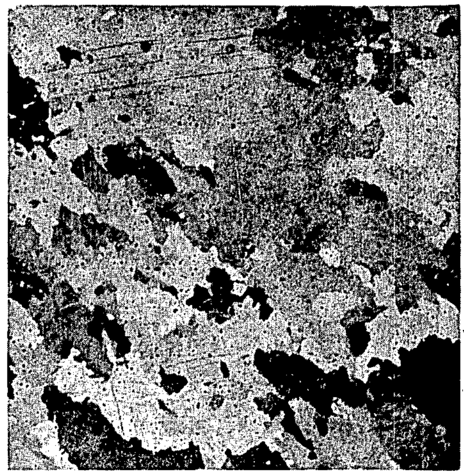
U-254-5 RF 3852B
50X Four quenches. 1X

Fig. 11

Effect of repeated gamma-quenching on structure of cast uranium (all holding times in gamma were 16 min).

DECLASSIFIED

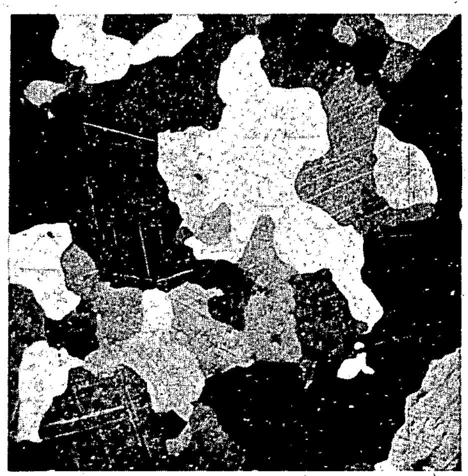
50X U-254-10



50X U-254-5

As-quenched.

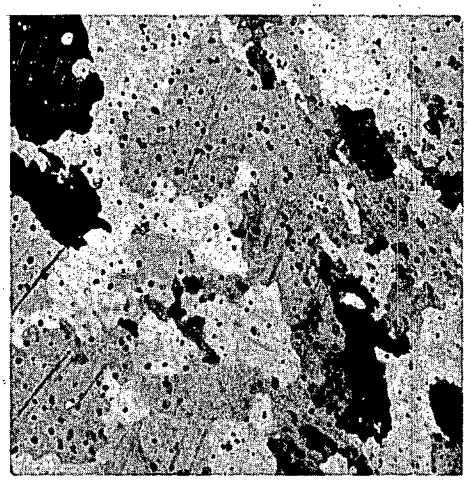
50X U-254-4



50X U-254-6

Annealed 2 hr at 1000°F.

50X U-265-6



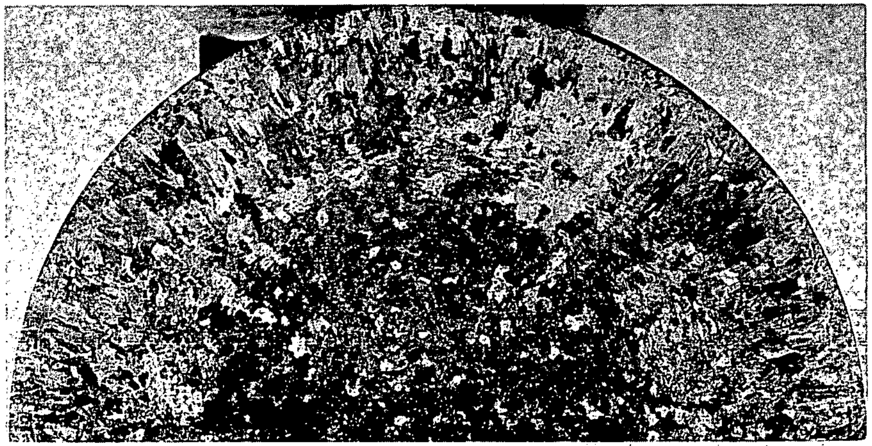
50X U-265-4

Annealed 2 hr at 1175°F.

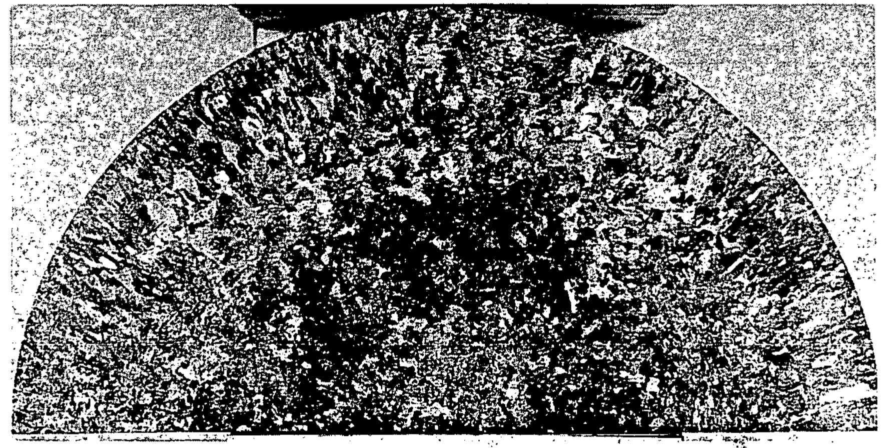
One quench. Four quenches.

Fig. 12 - Effect of alpha-annealing on gamma-treated cast uranium.

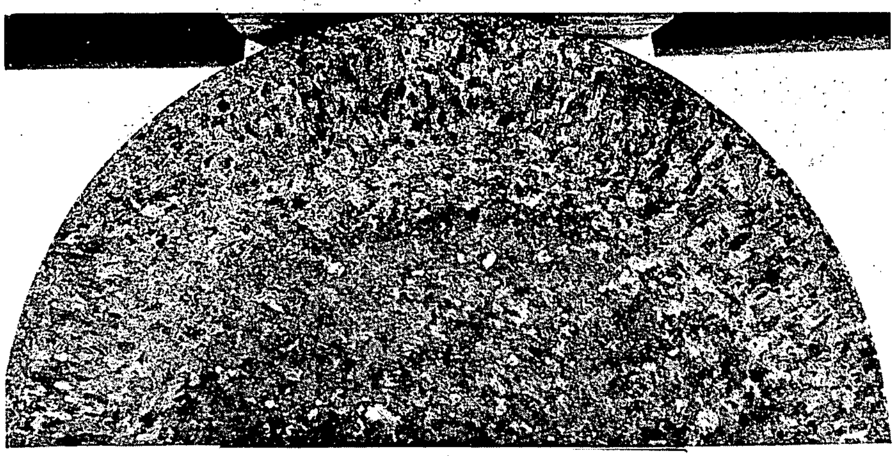
CONFIDENTIAL



Approx. 1X One quench. RF 3052



Approx. 1X Two quenches. RF 3054

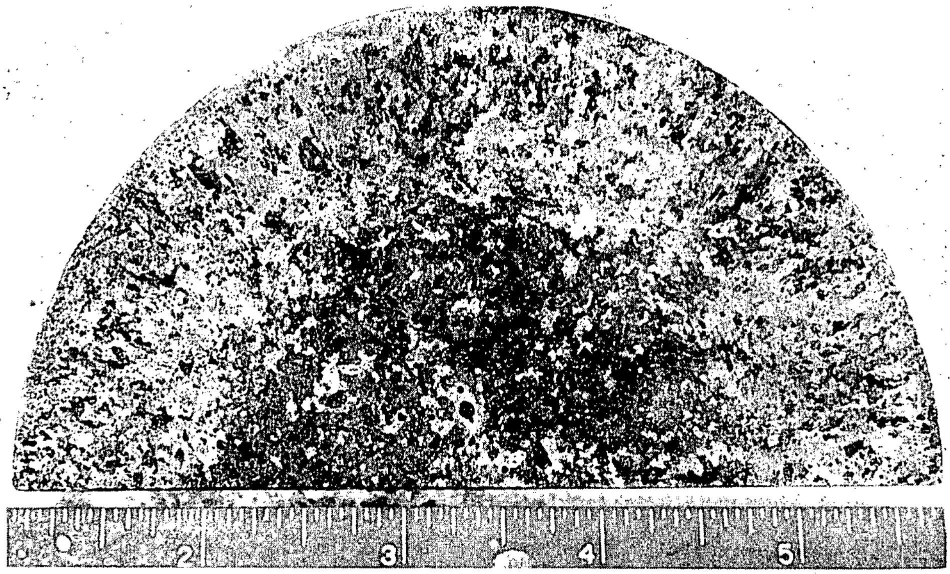


Approx. 1X Three quenches. RF 3056

Approx. 1X Four quenches. RF 3058

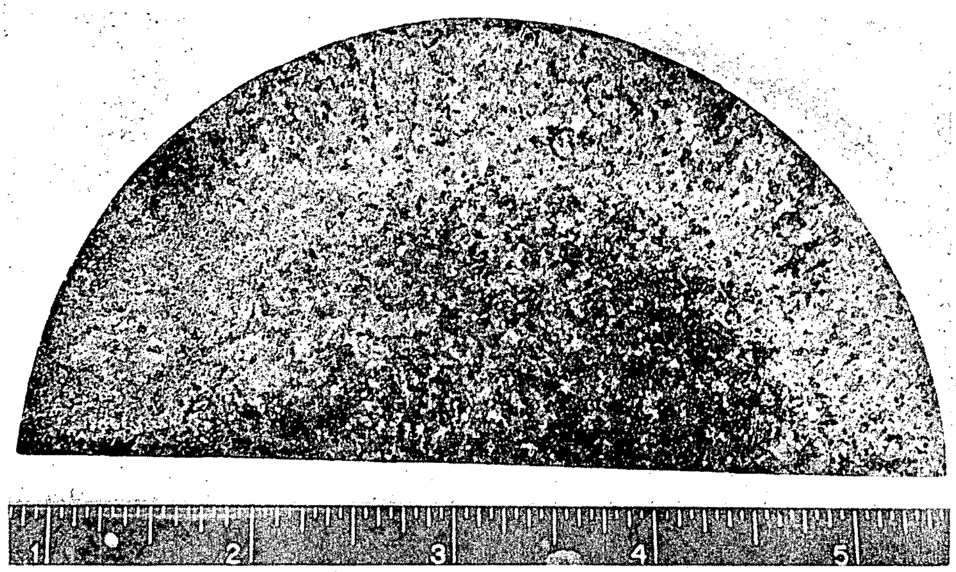
Fig. 13 - Effect of repeated beta treatments at 1350°F on the structure of 2 inch thick, 4-3/4 inch diameter slugs of cast uranium.

CONFIDENTIAL



Approx. 1X RF 3407

One quench plus 2 hr anneal at 1175°F.



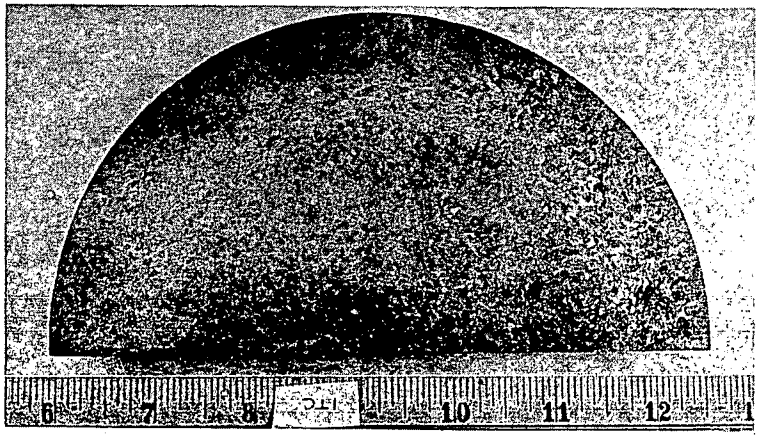
Approx. 1X RF 3408

Four quenches plus 2 hr anneal at 1175°F.

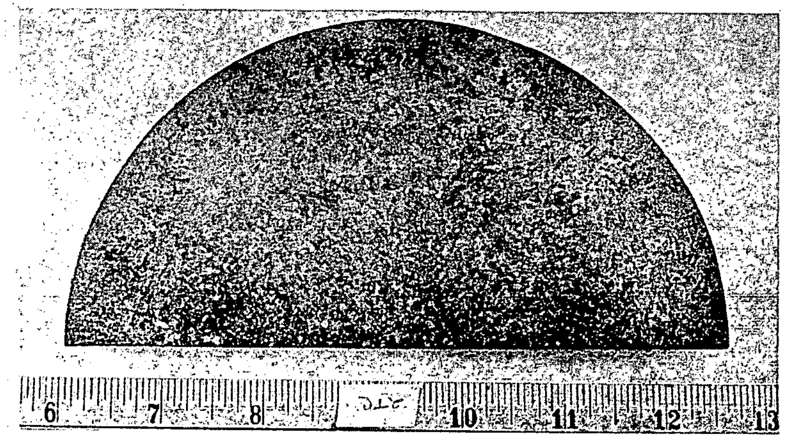
Fig. 14

Effect of alpha-annealing on the structure of 2 inch thick, 4-3/4 inch diameter slugs of cast uranium beta-quenched from 1350°F.

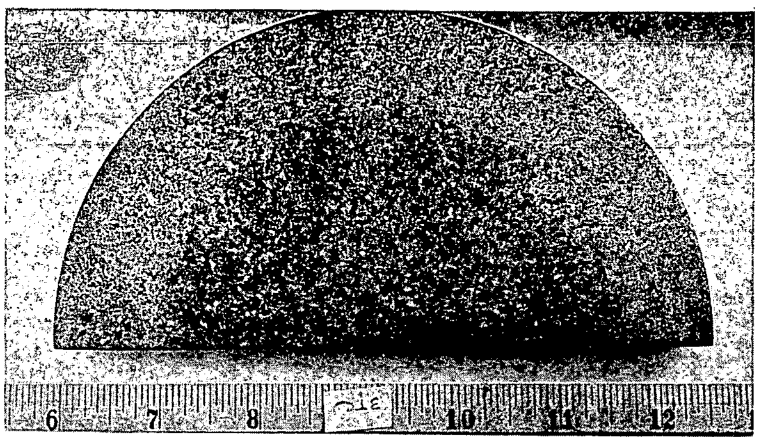
CONFIDENTIAL



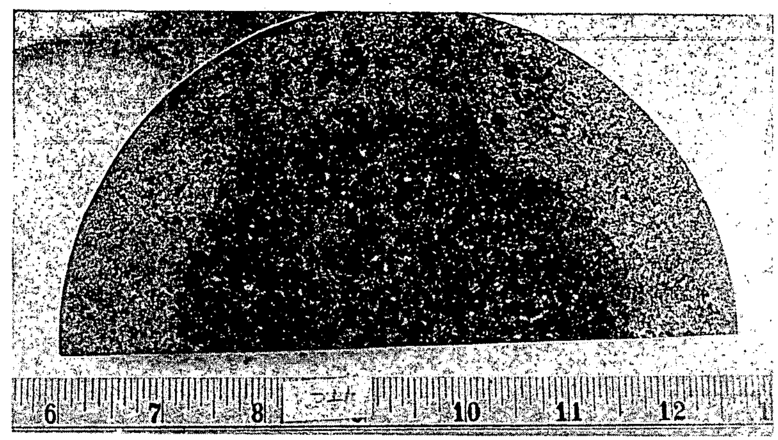
1/2X One quench. RF 3063



1/2X Two quenches. RF 3065



1/2X Three quenches. RF 3067



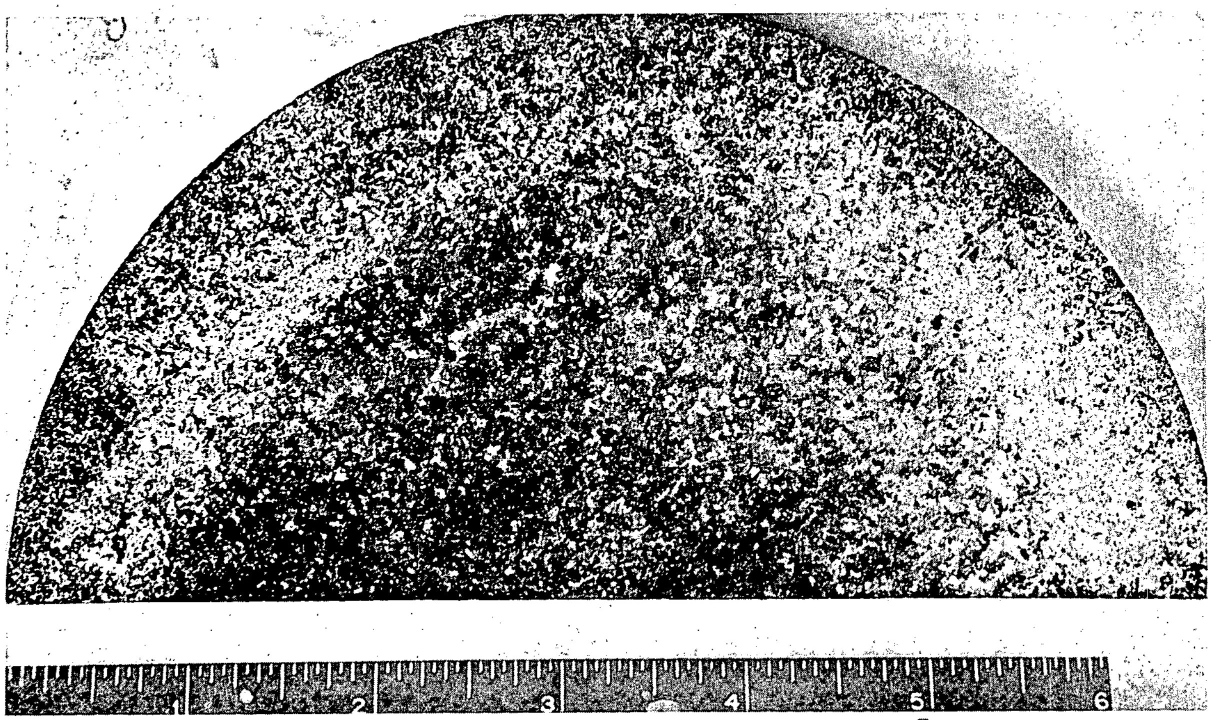
1/2X Four quenches. RF 3069

Fig. 15 - Effect of repeated beta treatments at 1350°F on the structure of 2 inch thick, 6-3/4 inch diameter slugs of cast uranium.

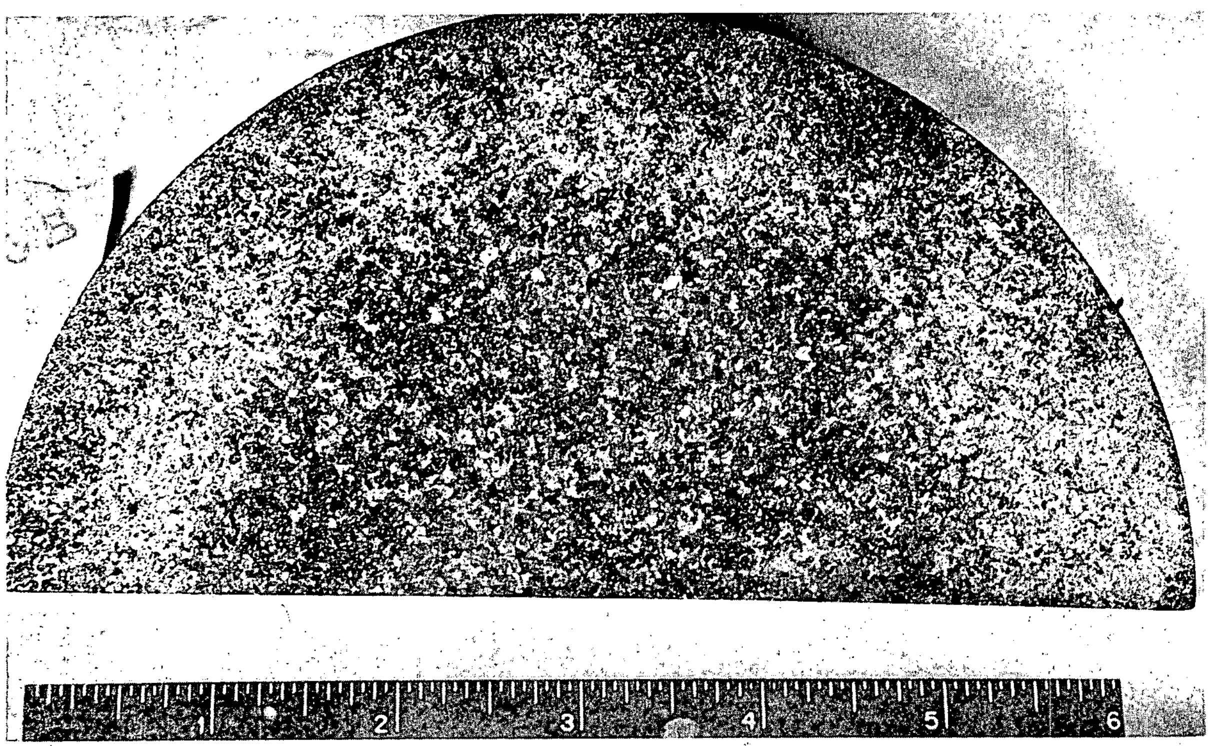
CONFIDENTIAL

UNCLASSIFIED

CONFIDENTIAL



Approx. 1X Two quenches plus 2 hr anneal at 1175°F. RF 3409



Approx. 1X Three quenches plus 2 hr anneal at 1175°F. RF 3410

Fig. 16 - Effect of alpha-annealing on the structure of 2 inch thick, 6-3/4 inch diameter slugs of cast uranium beta-quenched from 1350°F.

CONFIDENTIAL

UNCLASSIFIED