NMI-2800
April 18, 1960

PROGRESS REPORT TO THE OAK RIDGE OPERATIONS OFFICE FOR THE PERIOD NOVEMBER 1, 1959 TO APRIL 1, 1960

STUDY OF THE BETA TREATMENT OF URANIUM

R. B. Russell

nuclear metals, inc.
concord, massachusetts

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AEC RESEARCH AND DEVELOPMENT REPORT

Progress Report to the Oak Ridge Operations Office

for the Period November 1, 1959 to April 1, 1960

Study of the Beta Treatment of Uranium

R. B. Russell

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I. INTRODUCTION

The purpose of the beta-treatment study of uranium is to determine the effect of the following variables on the beta-treated product: size, composition, and heat treatment as these variables alter the texture, grain size, and residual stress of final dingot or ingot alpha uranium. It was suggested by a working committee of the Fuel Element Development Committee (FEDC) that the final data should be in such a form that it would be relatively easy for a metallurgist to beta treat uranium satisfactorily within the range of variables covered in this study. The final data could be in the form of empirically derived equations and/or graphs. The following factors in beta treatment will be the first considered:

A. Composition

The metal studied will have a composition which may vary between typical dingot and typical ingot uranium.

B. Sizes

The sizes to be investigated are one-half inch to 2-inch diameter rod, and tubes one-inch to 4-inch OD with 1/4-inch to one-inch wall thicknesses. Certain nominal combinations of these dimensions are listed in Table I.

C. Prior Condition

Prior conditions initially to be investigated include alpha rolled plus recrystallized, and alpha extruded plus recrystallized.

D. Beta Treatment

1. Temperature and Time in the Beta Phase

The temperature and time in the beta phase will be the same for all runs and will conform to the HAPO slug specifications[1] of 715 to 740°C for 11 to 20 minutes. The aim will be 730°C for 15 minutes.

2. Cooling Rates

The extremes in cooling rate provided by a cold water spray quench and a slow cool in still air, together with cooling at intermediate rates, will be investigated. Both continuous and interrupted rates will be studied.

E. Post Beta Treatment Annealing

The effect of post beta treatment annealing will be examined. Annealing temperatures probably will be about 600°C, the temperature ordinarily encountered in some canning operations.

The degree of warping, the grain size (primarily 3X macro), and texture will be determined by conventional methods.

II. PROGRESS OF WORK

The progress to date has been chiefly confined to organizing the program and setting up equipment. A limited amount of experimental data has been obtained.

A. Uranium Required and Present Inventory

The uranium which would satisfy the nominal size requirements[2] of the FEDC working committee are listed in Table I together with the present inventory of metal received from other sites. The partial analysis of three ingots representing the 1.22-inch diameter rod (1-7/32 inches) received from NLO ranged, in ppm:

C	Fe	Ni	Cr	Si
400/460	94/115	20/27	10/14	30/50

A partial analysis of the 3-inch diameter rod from NLO was, in ppm:

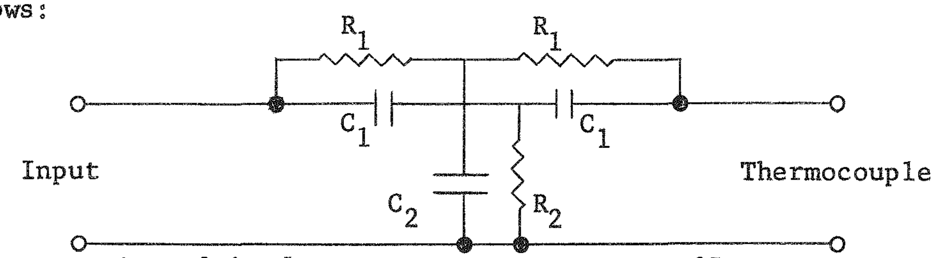
C	Fe	Ni	Cr	Si
470	87	23	14	20

B. Equipment

1. Temperature Recording

Very rapid temperature recording is now possible with a Brush horizontal mounting, rectilinear, thermal writing oscillograph recently acquired. This has a capacity of eight recording channels (Model RD-2684-50). It has at present four channels. It is expected that at least two more channels will be acquired. The frequency response is flat up to 70 cycles per second, which is considerably faster than needed for the present experiment. The maximum sensitivity of the instrument is 0.1 millivolt per millimeter of chart, or, when using a Chromel-Alumel thermocouple, about 2.5°C per millimeter. The width of each channel is 40 mm, so that at maximum sensitivity, a temperature range of 100°C can be recorded on each chart channel with a Chromel-Alumel thermocouple. Since one of the most important inflections on the cooling curve is the beginning of the beta-to-alpha transition which ranges from 666°C downward depending on the cooling rate, a determination of cooling rate in the temperature range 730 to 530°C will be necessary.*

From the start, operation of the oscillograph has been plagued by 60-cycle pickup, the amplitude of which depended on the sensitivity being used. Such sources as internal electronic (instrumental instabilities) and fluorescent lights were eliminated. All thermocouple leads were sheathed in grounded metal mesh, but the pickup reappeared when the surface thermocouples were spot welded to the Jominy sample. Some improvement was obtained by trial reversals of the 110-volt power plugs. The pickup has now been virtually eliminated even at maximum sensitivity by placing across each input terminal of the oscillograph a 60-cycle filter as follows:



$C_1 = 0.1\ \mu f$ $R_1 = 27\ K\Omega$

$C_2 = 0.2\ \mu f$ $R_2 = 13.5\ K\Omega$

---

* According to P. Duwez, J. Appl. Phys. 24 (1953) 152-156, a depression of the transformation to 530°C would correspond to a cooling rate of about 1000°C/sec.

The thermocouples being used are Chromel-Alumel. Surface thermocouples are spot welded to an electropolished uranium surface. Depth thermocouples are Inconel shielded* so that they may reach (unshorted) the bottom of a thermocouple well drilled to a suitable depth in the uranium. Temperature gradients will be measured in both end-quench Jominy-type rods as well as totally-quenched rods and tubes of other sizes.

2. Jominy End-Quench Apparatus

A Jominy end-quench apparatus has been constructed with a 1/2-inch ID water pipe (see Fig. 1). A mixer valve** has been installed so that the temperature of the water fountain can be maintained at constant temperatures in one range of about 4 to $52^{\circ}C$ (40 to $125^{\circ}F$) or in another of 49 to $80^{\circ}C$ (120 to $175^{\circ}F$). Without auxiliary heating or refrigerating units, a range of about 10 to $60^{\circ}C$ (50 to $140^{\circ}F$) is presently obtainable. It is expected that this more limited temperature range will be adequate for some time. To obtain more severe quenches, the height of the water fountain may be increased or a spray may be used.

3. Heat-treating Salt

For the beta treatment of all uranium specimens other than Jominy samples, Nu-Sal (KCl-NaCl) will be used. This salt is specified by HAPO for heat treatment of slugs.

4. X-ray Equipment

A new Philips High-Angle Goniometer-Electronic Circuit Panel has just been installed. Automation equipment of a type designed by Dr. E. F. Sturcken and his associates at SRL has been ordered (March 24, 1960), but delivery cannot be expected until about June 15, 1960. Part of the unit, the Program Controller, will be constructed at NMI.

- - - - - - - - - -

* Ceramo wire, 1/16-inch diameter, Thermo Electric Co., Saddle Brook, New Jersey.

** Photoguard, Powers Regulator Co., Skokie, Illinois.

A rotating specimen holder capable of taking 1-13/16 inch diameter specimens is being constructed at NMI. The design is that of R. N. Thudium at NLO.[3] The holder should be completed by May 1, 1960. Since the preliminary work is entirely on one-inch diameter Jominy specimens, our present one-inch sample holder is entirely adequate.

C. Plant Visits

A. Boltax[4] visited SRL on December 14, 1959 to discuss beta treatment of uranium with C. E. Paetschke and W. McDonell. The details of the beta treatment of triple dip canned Mark VII-A slugs as well as hot-press bonded Mark VII-A slugs were examined. The group at SRL found that oil-quenching from the beta phase produced the best combination of fine grain size with the least texture. The opinion at SRL is that the most significant cooling rate is that through the beta-to-alpha transformation. X-ray techniques and automation were discussed with J. Legeros, H. Minton, and E. F. Sturcken.

R. Russell visited HAPO on January 4 and 5, MCW on January 6, and NLO on January 7 and 8, 1960.[5] Opinions and facts about beta treatment, measurement of warp, texture and grain size, quenching stresses, hydrogen pickup, etc., were gathered.

D. Bibliography

A literature survey on beta treatment has not been started, but a survey is planned and is expected to be completed by June 1, 1960. The bibliography will not be included in a report but the information will, of course, be used on this program.

E. Experimental

1. Jominy End-quench Experiments

While awaiting the arrival of the desired starting uranium, preliminary Jominy end-quench experiments on as-cast metal were run to demonstrate any trends or peculiarities of the total Jominy-oscillograph-thermocouple system. The cast metal, a 1-1/4 inch uranium rod cast into a ZrO_2-washed graphite mold, was machined to one-inch diameter by 6-inch length Jominy rods and end-quenched. Since it is generally known that different temperature gradients induce differences in texture,(6) it would be desirable to obtain temperature gradients which are all perpendicular to the water-quenched plane surface of the Jominy bar, because x-ray diffraction analysis covers the entire cross section at different distances from the water-quenched end. For this reason, the first Jominy bar, J1, was wrapped with three or four layers of asbestos tape so that radial temperature gradients would be lessened. The tape also holds the thermocouple leads in place and, to some extent, limits oxidation during the half-hour heating to the beta temperature in the air furnace. Air oxidation was so severe that the two spot-welded surface thermocouples flaked off and the water-quenched end was insulated by the heavy oxide layer. The results of texture and temperature measurements are summarized in Table II. The 3X photomacrographs of the as-cast and Jominy sections are shown in Fig. 2. The second Jominy bar, J2, is to be end-quenched from an argon atmosphere in order to prevent excessive oxidation. If argon is successful, all future Jominy bars will be heat treated in argon.

The results in Table II show that very little, if any, growth index (G_3) change was induced by water quenching, although there is some change in 3X grain size (see also Fig. 2). However, a study of crystallographic (inverse) pole figures, representing the different distances shown in Table II, reveals no systemic nor extreme texture changes except in the case of the 001 pole density which rises rather abruptly from values of 1.0 - 1.8 at distances of 0 - 2.7 inches to a pole density of 7.2 at 3.7 inches from the water-quenched end. It is possible that this high 001 pole density was inherited from the as-cast texture which showed, not surprisingly, large texture variations from one position to another. Accordingly, we will assume for the present that this anomaly was inherited from the prior condition of the bar. The low cooling rate is probably due to the heavy oxide layer on the water-quenched end of the sample. In this first experiment, the gain on the oscillograph was not set high enough to give much precision to the cooling rates cited.

2. Growth Index

The characterization of a beta-treated sample will be made by a determination of a growth index and a texture. There have been four principal growth indices in use: GI,[7] K,[8] G_2,[9] and G_3.[10] Of these four, K has been used only by NMI in connection with textured uranium.[8] The remaining indices can be determined for either textured or untextured metal. All the indices use the concept of weighting crystallographic pole densities by the factor $C = \cos^2\beta - \cos^2\alpha$ (where α and β are the angles between a given crystallographic pole $hk\ell$ and the 010 and 100 poles, respectively). The GI and K indices are based on a simple point weighting by C, whereas both G_2 and G_3 are further weighted by the angular areas associated with each pole density. In the case of G_3, the crystallographic poles projected onto the surface of a sphere occupy an area which can be divided among spherical polygons, each polygon containing the pole. Each polygon is then assumed to represent a pole and is given the pole density of the pole lying within it. The weight assigned to each area is proportional to the relative area of the polygon. This G_3 method is known as the "Area Weight Method." This method has been further refined by E. F. Sturcken in his G_2 method to a weighting by continuous pole density function, which is essentially the mathematical expression of a pole figure. The characteristics[11] of all these indices are listed in Table III. For the present, G_3 will be computed for this program, but the pole densities will be computed differently from that recommended by Morris in order to be able to plot conventional crystallographic (inverse) pole figures These pole densities are called p_{hm} (original Harris-Morris), while the Morris pole densities, p_m, will not be computed. A sample computation sheet is shown as Table IV in which column (6) gives the value of p_{hm}.

III. FUTURE WORK

A. Equipment

The number of channels on the Brush oscillograph will be increased from four to six. The extra channels can be obtained in about one month.

B. Experimental

Jominy end-quenching of one-inch diameter as-rolled or extruded uranium rods will continue. Some investigation will be made of the prevention of oxidation during beta treatment by plating the Jominy bars with nickel. A wide range of cooling rates will be tried. It is believed that very useful information can be obtained from end-quenches ranging in severity from that obtained by a vigorous water spray to that from a moderate air blast.

Totally quenched rods and tubes will be studied for evidence of radial texture gradients, and these will be correlated with texture data from the Jominy tests.

IV. TABLES AND FIGURES Table I

Nominal Requirements and Present Inventory of Uranium Metal

Nominal Requirements					Present Inventory						
Dimensions (in.)		Type	Weight (lb)	Length (in.)	Dimensions (in.)			Type	Weight (lb)	Source	Date Rec'd
OD	Wall				OD	Wall	Length				
4	1	tube	3115	480							
3	1	tube	2077	480	3	1.5	36	rod	176	NLO	3/21/60
2	1	rod	519	240							
4	1/2	tube	1817	480							
3	1/2	tube	1298	480							
2	1/2	tube	779	480	1.5	0.55	500	tube	553	BB(A)	3/25/60
1	1/2	rod	130	240	1.22	0.61	360	rod	290	NLO	3/21/60
4	1/4	tube	973	480							
3	1/4	tube	714	480							
2	1/4	tube	454	480							
1	1/4	tube	195	480							
1/2	1/4	rod	33	240							

Composition: The partial analysis of three ingots representing the 1.22-inch diameter rod (1-7/32 inches) received from NLO ranged in ppm:

C	Fe	Ni	Cr	Si
400/460	94/115	20/27	10/14	30/50

A partial analysis of the 3-inch diameter rod from NLO was, in ppm:

C	Fe	Ni	Cr	Si
470	87	23	14	20

Table II

G_3, Texture, Thermal Gradients and 3X Grain Size in As-Cast Jominy Bar, J1, End Quenched from 735°C - 15 Minutes, Water Temperature 11°C (52°F)

Distance (in.) from Quenched End	G_3	Texture Type	"Grain" Size (3X)**	Temp. Gradient at Far Center, °C/sec at Transformation		
				Start***	During	End
0	0.00	random	<300 μ	-	-	-
0.25	-	-	-	7.3	1.3	1.8
0.80	+0.04	random	1500	-	-	-
1.77	−0.02	random	2000	-	-	-
2.71	−0.06	random	coarse striped	-	-	-
3.00	-	-	-	6.9	1.5	4.2†
3.68	+0.01	001	coarse striped	-	-	-
0*	−0.17	011-114,221	massive	as-cast condition		
5*	−0.14	102,011	massive	as-cast condition		

* as-cast condition.

** estimated from 3X photomacrographs.

*** approximately 650°C.

† This anomaly may be due to inability to read chart from instrument set at relatively low sensitivity of 1 mv (mm, chart speed 4 mm/sec).

TABLE III

Comparison of Characteristics Among Different Growth Indices

Method	Harris-Morris	GI (Sturcken)	K_{29} (Russell)	G_3 (Morris)	G_2 (Sturcken)
n	---	14	29	10, 14	10, 15, 18, 20
Pole Density	$p_{hm} = n \frac{I}{I_o} \frac{1}{\sum_n (I/I_o)}$	$TC \equiv p_{hm}$	$p_{hkl} \equiv p_{hm}$	$p_m = \frac{I}{I_o} \frac{1}{\sum_n (A_w I/I_o)}$	Legendre polynomial (See Ref. 8b, Pt. I, eq. 7)
random	1	1	1	1	
perfect	n	n	n	$1/A_w \sim n$	
Growth Index	---	$GI = \sum_n (p_{hm} C^*)$ * modified	$K_{29} = \frac{420}{n} \sum_n (p_{hm} C)$	$G_3 = \sum_n (p_m A_w C)$	$\int_0^1 \int_0^{\pi/2} p(u,\phi)(1-u^2)\cos^2\phi \, du \, d\phi$
random	---	$\sum C^* \sim 0$	$\sum C \sim 0$	$\sum A_w C \sim 0$	0
perfect	---	$\pm 14C^*$ (± 14 max)	$\pm 14.5C$ (± 14.5 max)	$\pm C$ (± 1 max)	± 1 max.
Severity of Texture	---	---	$T = \frac{\sum_n (I \lvert \log p \rvert)}{\sum_n I}$ let $\lvert \log (p < .005) \rvert = 2.3$ T = 0 (random)	$J' = \frac{\pi}{2} \sum_n (p_m^2 A_w)$ J = 1 (random)	$J = \int_0^1 \int_0^{\pi/2} p^2(u,\phi) \, du \, d\phi$ J = 1 (random)
$\sum p$	n	n = 14	n = 29	---	---
$\sum p A_w$, $\sum A_w$	---	---	---	1	1
$\sum C$	---	+1.25 (n = 13)	+0.0132 (n = 29)	+0.4947 (n = 18) -0.2764 (n = 10)	
$\sum A_w C$	---	---	---	+0.003 (n = 18) +0.004 (n = 10)	---
Ref.	1, 2	3	10	5, 6, 7	8b

[redacted]

[redacted]

Table IV

Sample X-ray Data Sheet for Determination of Texture and G_3

hkl	1 s.f.	2 Area	3=I (1) x (2)	4 I_o	5=I/I_o 3/4	6=P_{hm} $\frac{5}{\text{av. }5}$	7 A_w x100	8=$A_w I/I_o$ 5 x 7	9 C	10=$CA_w I/I_o$ 8 x 9
020				6.40	*		3.18		+1	+
110				72.87			6.34		−.6184	−
021				100.00			6.08		+.7393	+
002				51.44			3.04		0	0
111				58.04			7.38		−.4876	−
022				3.55						
112				47.74			5.22		−.2968	−
130				3.34			3.57		+.3616	+
131				39.34			6.84		+.3255	+
040				6.81						
023				16.52			6.59		+.2395	+
200				8.69			2.34		−1	−
041				4.74						
113				11.31			4.77		−.1804	−
132				3.56			5.29		+.2525	+
221				19.29						
004				5.20						
202				10.34						
133				14.37			7.95		+.1844	+
114				9.50			7.49		−.1169	−
043				2.04						
150				6.89			3.72		+.7096	+
223				10.91			4.87		−.3843	−
152				11.49			6.89		+.5961	+
310				3.57						
025				5.15						
204				5.55						
312				7.72			8.45		−.8294	−

Sum(8): +

−

Sum(10):

Filter factor =
Sum(5)/23 = Av(5) =
1/Av(5)
Sum(6) = 25 ?

$$G_3 = \frac{\text{Sum(10)}}{\text{Sum(8)}} = \boxed{}$$

Ref.:
Prior:
Location:
Calc. by:

Sample No.:
Quench:
3X Grain Size:
Date:

Pole Fig.:
Remarks:

- - - - - - - - - - -

* Av of 020 and 040.

32-2-3/60-150

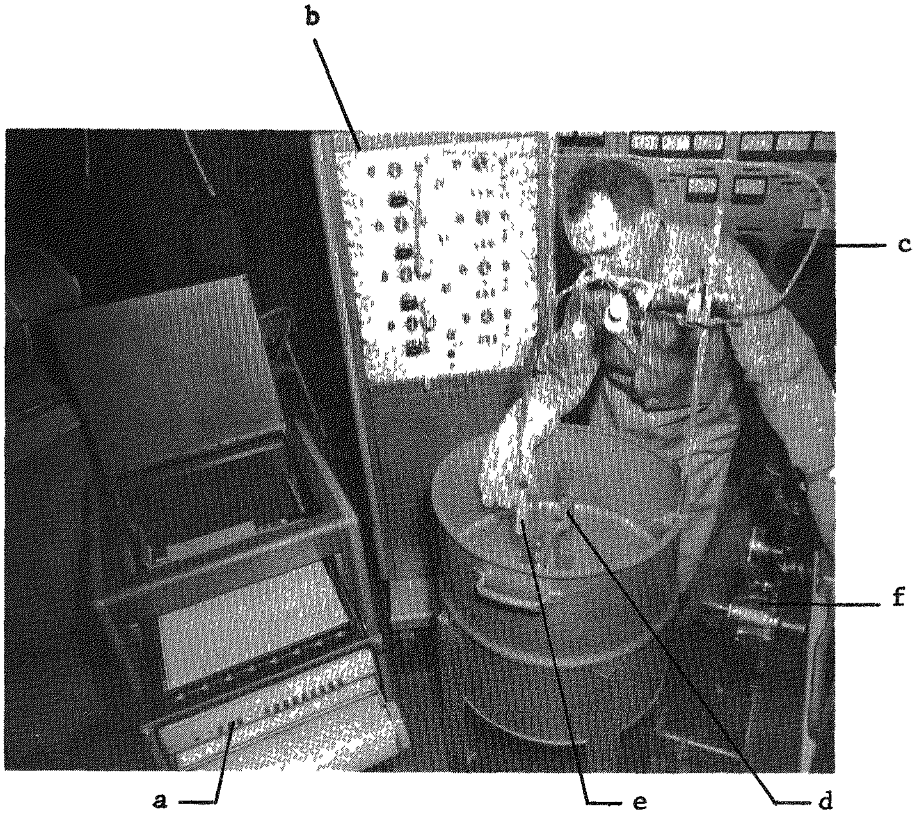


Fig. 1 - Jominy end-quench setup.

(a) Brush oscillograph for recording temperature-time data.

(b) Brush amplifier console.

(c) thermocouple leads.

(d) jig for holding sample.

(e) sample wrapped in asbestos tape.

(f) water mixing valve to control temperature of water fountain.

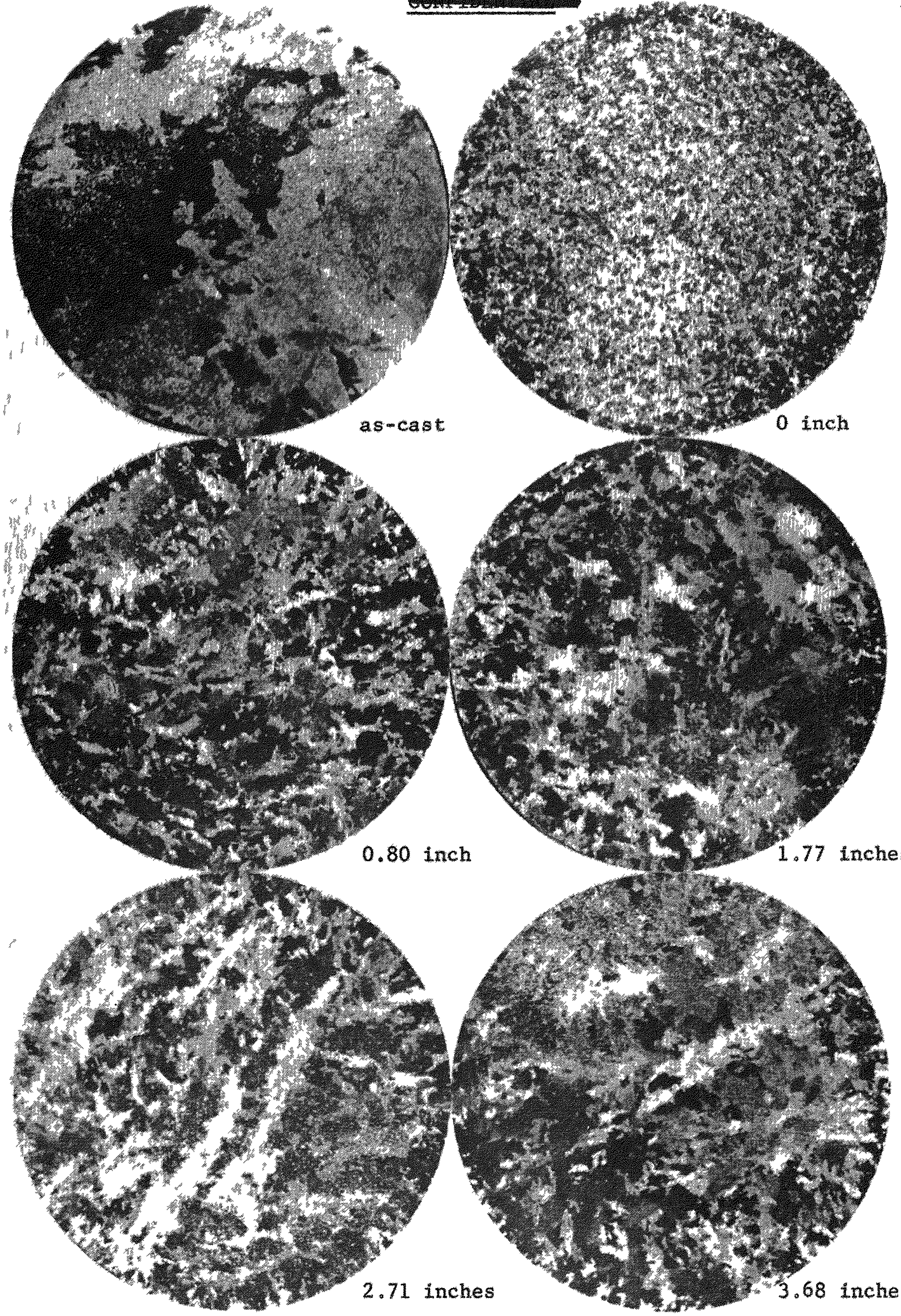


Fig. 2 - Photomacrograph showing relative "grain" or patch size of an as-cast uranium rod and sections of end-quenched Jominy bar J1 shown at various distances from the water-quenched end.

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8. R. B. Russell, Balanced Textures in Uranium, NMI-1230, December 8, 1959, SECRET.

9. E. F. Sturcken, "A Generalized Growth Index Formalism (G_2)", Minutes of Meeting at NLO, Cincinnati, Ohio, November 9-10, 1959, P. R. Morris, Chairman (to be published).

10. P. R. Morris, Ref. 9.

11. Discussed in a memorandum from R. B. Russell, NMI-RBR-27 (March 21, 1960), Addendum (March 31, 1960), distributed to the x-ray committee which last met on November 10, 1959 at NLO with P. R. Morris, Chairman.

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