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STRUCTURAL CHANGES ASSOCIATED WITH
HIGH-TEMPERATURE DEFORMATION OF URANIUM

BY

F. N. RHINES

AEC RESEARCH AND DEVELOPMENT REPORT

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STRUCTURAL CHANGES ASSOCIATED WITH
HIGH-TEMPERATURE DEFORMATION OF URANIUM

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Sponsored by

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PREFACE

The work reported herein was performed at the University of Florida as part of a research project sponsored by the National Lead Company of Ohio.

This topical report is based on the University of Florida's report entitled *Influence of Minor Components Upon the Constitution and High Temperature Deformation Structure of Uranium*, dated June, 1966. Their report was divided into two parts, the first being concerned with structural changes associated with high temperature deformation, the second with the constitution of the system uranium-carbon-iron-silicon. This report covers only the first part of the University of Florida's report. The work is reported verbatim, except for a few minor deletions which have been made with the consent of Dr. Rhines and which do not affect the interpretation of the work performed.

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I. INTRODUCTION

This research has been concerned with structural changes associated with high temperature deformation. It is felt that the findings present in this report provide new and deeper insight into the metallurgical behavior of uranium and some of its alloys when in use as nuclear fuel.

A. Scope

The hot deformation behavior of alpha uranium is of interest with respect to mill practice and control in the production of uranium for fuel elements and with respect to the service characteristics of uranium as a nuclear fuel. As the metal leaves the rolling mill, its structure and associated properties are determined by: (1) the cast microstructure and analysis of the ingot, (2) the rate and extent of deformation in each pass through the mill, (3) the temperature of the metal in each pass, (4) the interval between passes, and (5) the rate of cooling after the final pass. These conditions are subject to individual control, within limits, and their control has much to do with establishing the subsequent pattern of behavior of the uranium in the reactor. While working rates similar to those encountered in mill practice are not approached in hot tensile testing, it has been found that deformation rates thus attained in the laboratory are sufficient to arrest annealing and, therefore, provide a useful index of in-mill conditions.

Alpha uranium, having an orthorhombic crystal structure, is characterized by different properties along its three mutually perpendicular crystal axes. Temperature rise (Table I) is accompanied by expansion upon its $[100]$ and $[001]$ axes, and simultaneous contraction upon its $[010]$ axis, whose room temperature parameters are $2.85\text{\AA}$, $4.96\text{\AA}$, and $5.87\text{\AA}$ respectively. Slip occurs most readily in the $[100]$ direction upon the (010) plane (see Figure 1a). Cross slip has also been observed in the same direction upon other planes, including: (001) , (011) , (013) and (023) . The principal twinning plane is (130) with less certain modes reported as $(\bar{1}72)$, (112) and $(\bar{1}76)$ (see Figures 1b and 2, and Tables II and III). The number of modes of deformation that can operate in a given case is greater, presumably, at high than at low temperature, but specific temperature limits for the several deformation processes are not known.

During its use as a reactor fuel, uranium is subject to constant and fairly violent deformation at temperatures covering much of the thermal range of stability of the alpha phase. Upon the basis of the common observation^{15,16,17} that uranium metal fuel develops gross intergranular cavities in the temperature range between the beginning of recovery (about 300°C) and the onset of recrystallization (about 550°C), it has been postulated that uranium, like other metals, is subject to grain boundary shearing and consequent intergranular cracking during creep at moderately elevated temperature¹⁸. Preliminary tests of this postulate by Angerman¹⁵ and by Shober¹⁹ confirmed a loss of ductility of uranium within the expected temperature interval, but no intergranular cracks were found by either investigator. It was presumed that a detailed study of microstructural evolution during hot deformation would serve to correlate these seemingly contradictory findings.

TABLE I Thermal Expansivity of Uranium (10^{-6} per $^{\circ}\text{C}$)
Parallel to (After Foote¹):

Axis	25 - 125 $^{\circ}\text{C}$	25 - 325 $^{\circ}\text{C}$	25 - 650 $^{\circ}\text{C}$
[100]	21.7	26.5	36.7
[010]	-1.5	-2.4	-9.3
[001]	23.2	23.9	34.2

TABLE II Twinning Modes in Alpha Uranium
(After Lloyd and Chiswick²)

K_1	K_2	U_1	U_2	Shear
(130)	(1 $\bar{1}$ 0)	[$\bar{3}$ 10]	[110]	0.299
(1 $\bar{7}$ 2)	(112)	[$\bar{3}$ 12]	Irrational	0.227
(112)	(1 $\bar{7}$ 2)	Irrational	[$\bar{3}$ 12]	0.227
(1 $\bar{7}$ 6)	(111)	[512]	Irrational	0.216

Experience has demonstrated that small critical quantities of such alloying agents as silicon and aluminum tend to suppress swelling (i.e., gross cavity formation) in uranium fuel elements. Since this effect is clearly structure-dependent, it was anticipated that a comparative study of the evolution of microstructure during the hot deformation of alloys would indicate the mechanism by which swelling is suppressed and thereby serve as a guide in future alloy composition control and mill practice.

An understanding of alloy behavior must be based upon the phase constitution, inasmuch as alloying elements in solid solution and those present in the form of separate phases influence metallurgical processes in quite distinct ways.

B. Nature of the Experimental Approach

Since the subject of study is the evolution of microstructure during hot deformation, it was necessary either to make metallographic observations during deformation at temperature, or to arrest the structural change for subsequent observation. Hot microscopy is of little use in a case of this kind, because the microstructure becomes visible only after elaborate etching which, of course, cannot be performed continuously during deformation.

In the course of a previous study of hot-deformation at the University of Florida, however, it had been found that the microstructure that exists *during* hot working can be preserved for subsequent examination by quenching the specimen while deformation is in progress. This is accomplished by the use of a portable tensile straining device by means of which the specimen under test can be immersed in a high temperature solder bath and then, without interrupting straining, be transferred to cold water in about one second. Using this device, tensile test bars of commercial uranium and eight alloys (Table IV) were strained and quenched according to a program which

provided ranges of temperature (300 to 600°C), strain rate (0.009 to 0.75/minute), and extent of strain (0 to fracture). From these tests a direct measure of the strain to fracture was obtained.

Subsequently, the microstructure of each specimen was analyzed by quantitative metallography. Where the microstructure was homogeneous, these measurements provided precise and geometrically meaningful determinations of grain growth, sub-grain formation, recrystallization, mechanical twin formation, and anisotropy, of grain shape. Inhomogeneous microstructures were also subject to statistically exact analysis except in the matter of anisotropy, which must be treated as a local property.

TABLE III Observed Deformation Mechanisms at Various Temperatures

Experimental Conditions		Observed Deformation Mechanisms					Remarks	Reference
Temp. (°C)	Method of Deformation	Twin Planes	Slip Systems	Cross-Slip Systems	Kink			
					Plane	Axis		
-196	Compression	(130) (172) (176)	{110}[001]	—	—	<001>	{010} slip not observed (172) twinning infrequent	3, 4
-196	Tension	(130) (176) (172) (112)	(110) [100] (110) [001]	—	—	—	(101)[100] slip observed in one crystal	5, 6
R.T.	Not specifi- cally stated, also includes thermal twins	(130) (172) (112) (121)*	{010}[100] {110} —	{011}[100] {013}[100] (001) [100] {023}[100]*	to [100]	—		7, 8
R.T.	Compression	(130) (172) (176)	(010) [100] {001} —	(010) [100] (001) [100]	to [100]	—		2
R.T.	Tension	(112) (130) (172) (176)	(010) [100] (110) [001] (011) [100]	(001) [100]	—	—		9
350	Compression	(130) (176)	{010} —	—	—	—	Kinking observed as was cross-slip, {172} twinning not observed	3
450	Tension	(130) (172)	(010) [100] {110} —	—	—	—	Kinking different than that observed by Ref. 2 and 8 was noted	10, 11, 12, 13, 14
500	Tension	—	{110}<110>	—	—	—	Very little twin- ning observed	10, 11, 12, 13, 14
600	Compression	(130) (172)	(001) [100] (010) [100] {110}<110> {021}<112> {111} —	—	—	—	{021}<112> slip very minor	

*Not well substantiated

FIGURE 1 (a) Slip Systems in Alpha Uranium. Arrows Denote Slip Directions. (b) Twinning Planes in Alpha Uranium

FIGURE 2 Shear Diagrams for All Known Twinning Modes in Alpha Uranium

Initially, the commercial uranium, which was used as a starting material for the manufacture of the alloys, was evaluated in detail. This provided a basis of comparison by which the effects of alloying could be assessed. It thus became possible to ascertain whether a given alloying addition promotes or retards grain growth, subgrain formation, recrystallization and twinning, at specific temperatures and strain rates.

In order to infer the mechanisms of low ductility rupture and cavity formation, the structural evolution was correlated with measurements of elongation at fracture, with known reactor behavior, and with the results of thermal cycling tests. The latter tests were performed by alternately heating and cooling samples of the metal and alloys between temperature limits established by two solder baths. Uranium, thus treated, develops intergranular cracks. Such cracking is suppressed by alloying in much the same way as is swelling under neutron irradiation. Finally, a study of the evolution of microstructure during thermal cycling provided a direct association with the behavior of uranium in hot tensile straining.

These studies indicate that continuous unidirectional tensile strain does not lead to grain boundary shearing in uranium at any temperature, because the stresses that might be expected to induce grain boundary shearing are relieved by other processes including twinning and recrystallization which occur adjacent to grain boundaries at unexpectedly low temperature. Stresses established by temperature change (as contrasted with tensile loading), however, are mainly shears parallel with the grain boundaries and, as such, are most easily relieved by intergranular shearing. Accordingly, thermal fluctuations, whether induced by thermal cycling, or by thermal spikes (nuclear events) are capable of producing intergranular fissures of the kind that appear in other metals during high temperature creep. Silicon seems to suppress this effect by maintaining a grain size so fine that stress levels of sufficient magnitude to initiate cracking at quadruple points are not attained.

It is seen that silicon acts as a scavenger, driving iron (and probably other elements) out of solution in uranium, while it produces particles of U_3Si and perhaps other insoluble phases. The "purging" of the uranium matrix, effected by silicon, is believed to account for a more active recrystallization of uranium containing silicon, and the presence of insoluble silicides is believed to account for arrested grain growth in uranium silicon alloys. Together these factors act to maintain a fine grain size, even in the presence of plastic deformation at high temperature.

C. Scientific Background of the Deformation Study

At this point it is appropriate that the state of the knowledge of the hot deformation of polycrystalline bodies be summarized. The topics introduced here will be considered again in the discussion section of this report where the new findings will be analyzed.

The grain structure of a polycrystalline aggregate is most conveniently described as a three-dimensional network of grain boundaries. It has been shown²⁰ that this network may be described in terms of its crystal edges, which meet in sets of four (quadruple points) to form the crystal corners. Grain interfaces have in common with all surfaces an inherent energy that gives rise to a tension (λ) in the plane of the surface. Where the surfaces meet along triple lines the interfacial forces, seeking balance, require the surfaces to meet at three included angles of 120° each. Where triple lines meet at quadruple points, a balance of surface forces demands six included angles of $109^\circ 28' 16''$ each (i.e., where $\cos \theta = -\frac{1}{3}$). If the surface is curved the vector sum of the surface tension forces lies perpendicular to the surface and at each point upon the surface has a value:

$$F = \frac{\lambda}{2} \left(\frac{1}{r_1} + \frac{1}{r_2} \right)$$

where r_1 and r_2 are the principal radii. This force acts to move the surface toward its center of curvature, thus tending to flatten curved boundaries.

Although the source of grain boundary energy lies within the two dimensional interface, it is valid and somewhat simpler to visualize the grain boundary network as being made up of a system of quadruple points mutually connected by triple lines, which are the edges of the grain surfaces.

Grain growth occurs whenever four quadruple points, that are mutually connected by six triple lines, come together; these four points are annihilated as a grain disappears and one new quadruple point is formed. This kind of event is spontaneous because it is always associated with a net evolution of grain boundary energy. Sets of four quadruple points connected by only six triple lines arise occasionally in the course of the "grain boundary migration" that occurs in response to the imbalance that always exists along triple lines and at quadruple points. This imbalance necessarily prevails in all polycrystalline networks because there exists no space filling stacking of polyhedra wherein the edges and corners have equilibrium angles.²¹

Every grain boundary consists physically of a specific array of dislocations. The angular difference between the conjoint crystals can always be expressed in terms of the relative number of Burger's vectors of two edge dislocations of a different vector in each crystal, plus a rotation in the plane of the boundary.²² The area of boundary is measurable in terms of the total length of dislocation edge. A flat grain boundary has the same dislocation "composition" everywhere; a doubling of its area is accomplished simply by a doubling of the length of each of the four families of edge dislocations. A curved grain boundary, however, is made up of a varying dislocation composition consisting of all of the dislocations that would make up a flat boundary joining the same edges, plus additional dislocations that introduce curvature. The latter appear as pairs of dislocations of opposite sign, which confer displacement of the boundary outward from the straight path and then back, in like amount. Curved grain boundaries can straighten during annealing without requiring any deformation of the metal because all that is required is that pairs of dislocations be eliminated. As curvature subsides, dislocations of the same vector, but of opposite sign, come together and annihilate each other, with evolution of elastic energy, but no shape change of the body as a whole.

Pinning occurs when a grain boundary is interrupted by an interphase boundary, particularly one of high stability. In order to detach a grain boundary from a "pin" it is necessary to supply enough new dislocations so that the boundary can bend around the particle and meet itself at a point on the detaching side of the particle. This can occur during deformation, or it can occur in previously distorted metal. It does not occur, in general, after the first moments of a long anneal.

The effectiveness of pinning is expected to increase with: (1) particle size, (2) number of particles, and (3) stability of phases. Among the alloys studied in this research U_3Si seems to be the most effective pinning agent.

Twinning in alpha uranium occurs when its critically resolved shear stress is reached before that for slip. The state that precedes twinning is one of elastic stress; the dislocations that are involved in twinning shear must originate within the crystal, usually somewhere upon a grain boundary.

The initial twin is in the form of a thin "slab" which usually spans the crystal. Continued deformation by twinning is accomplished by growth in the thickness of the twin "slab." Exploited to its limit, however, twinning can produce only a few percent of extension of the metal; twinned crystal is not usually in an orientation suitable for further extension by secondary twinning, and the critical resolved shear stress for crystal slip is soon reached.

Once the crystal is distorted by plastic flow, twinning can no longer occur. Neither is twinning observed after subgrains have formed, although crystal matter that has undergone twinning can subsequently become subgrained.

During thermal cycling twinning is at first abundant. With continued thermal cycling the characteristic twin forms disappear from the microstructure. This is not surprising, since twinning might be expected to produce a "totally twinned" structure, which would have the appearance of a normal polycrystalline body. Such a change would yield no preference in crystal orientation that could be detected by ordinary x-ray methods.

Under conditions of neutron exposure alpha uranium seems also to twin profusely. Many of the cavities that form are located upon twin boundaries. There is at present insufficient experience available to be able to say whether twinning disappears with longer exposure in a reactor.

Subgrains, in contrast with twins, develop *after* slip has occurred and are thus a product of both plastic strain and a residual elastic strain. Dislocations that are already present, and that represent the crystal distortion, translate and, at elevated temperature, climb to arrange themselves into two-dimensional arrays that then exist as boundaries between neighboring segments of crystal subgrains. The elastic strain field that surrounded each dislocation is thus diminished and what is left of it becomes the source of the boundary energy. Polygonization, as this process is called²³ begins with the nucleation of a small disc of boundary, which has a dislocation composition corresponding to the orientation difference between the conjugate segments of crystal. This patch of boundary increases in area as more dislocations are added at its edges and until intersection with another patch of boundary occurs. A triple line is thereby formed. When all patches of boundary have met other boundary at all edges, a complete three-dimensional boundary network will have formed. It does not differ, in principle, from any other grain boundary network and is in fact part of the total grain boundary network, but tends to have some characteristic features of its own: (1) the boundaries are mostly of small angle; (2) the crystal segments (subgrains) tend to be small; and (3) the included angles at triple lines are usually not of the equilibrium 120° magnitude, but tend toward 90° and 180° , because the geometric probability of boundary intersection maximizes at 90° . These features of the subgrained structure give to it its typical "masonry" appearance.

Continued annealing causes the subgrain boundary network to tend toward equilibrium angles. This is not usually accompanied by growth except along old grain boundaries where irregularity in the original subgrain size is most probable. Oversized subgrains tend to consume their neighbors and launch into aggressive growth. This constitutes "recrystallization."

"Viscous" flow may occur locally in directions of maximum (absolute) shear stress. The sites upon which such shearing occurs are thin "slabs" that extend across grain boundaries through many grains and "slabs" lying upon suitably orientated grain boundaries. So-called "viscous" flow is not viscous in the classical sense, but rather occurs as a series of relatively large shears with rest periods between, repeating, however, upon exactly the same site. The conditions of its occurrence are: (1) that simultaneous slip upon several slip systems must be in progress and (2) that the temperature must be above that of recovery.

No detailed explanation of "viscous" flow is available at present. It appears that in a crystalline body undergoing complex slip something resembling a turbulent state develops at occasional sites within crystals and at certain types of obstructions, such as grain boundaries. Where this happens, the crystalline material is converted structurally into something more closely resembling a liquid, and for a moment, it yields freely in the plane of the shear stress; i.e., it exhibits no resistance to shearing. Once the immediate stress is relieved the metal quickly recovers its crystalline structure and properties. Presently, a new zone of turbulence is developed, usually upon an old site. Like subgraining, "viscous" flow seems to be an alternate to twinning. Unlike subgraining, the occurrence of "viscous" flow required not merely that slip has occurred, but that *complex slip be concurrent*.

Where "viscous" flow crosses grain boundaries a sharp step is produced. These steps have long been recognized in samples of hot deformed metals and the authors have called them serrations. Their effects upon metal behavior appear to be of two kinds. First, no substantial grain boundary migration is possible while the grain boundaries are serrated. Hence, the occurrence of viscous flow effectively halts grain growth. Second, subgrains tend to assume the planes of "viscous" flow as positions of their boundary systems. Accordingly, there is usually correspondence between the spacing of grain boundary serrations and the subgrain size.

"Viscous" flow upon grain boundaries leads to the formation of intergranular voids, which lower the elongation to fracture in tensile straining. This is the phenomenon of "intermediate temperature embrittlement",¹⁸ which has been found common to many metals and which is believed to be responsible for the formation of large cavities in uranium during service in the reactor.

II. EXPERIMENTAL MATERIALS, APPARATUS, AND PROCEDURES

Research with uranium required that a capability be developed in terms of alloys for study, equipment for the safe handling and processing of specimens, and techniques for properly studying uranium and its alloys.

A. Alloys for High-Temperature Deformation Studies

The materials utilized in high-temperature deformation studies consisted of a commercial uranium (hereafter designated as the "base material") and alloys of this material with silicon, iron, aluminum and carbon. The alloys were prepared by the National Lead Company of Ohio. All materials were received in the form of 3-inch tensile bars. A cylinder of the same material $\frac{1}{2}$ -inch in diameter by $\frac{1}{4}$ -inch long accompanied each tensile bar. Chemical analyses of all materials are included as Table IV; these analyses were made by the National Lead Company of Ohio.

The as-received microstructures of the base material and of alloys "P" (199 ppm Si), "C" (287 ppm Fe), "A" (135 ppm Fe), "L" (122 ppm Si), "V" (399 ppm Si), "E" (203 ppm Al), "H" (750 ppm Al) and "X" (795 ppm C) are illustrated in Figure 3.

The processing history furnished by the National Lead Company of Ohio with the base material indicated that the final, structure-determining stages in processing were: (1) heating 0.625-inch

round rods at 1050°C for 30 minutes, (2) rolling to 0.525-inch in three passes and (3) water quenching. Since all microstructures included in Figure 3 show evidence of deformation, it is presumed that the final rolling was performed at a temperature high in the alpha field and that subsequent cooling was too rapid to allow effective annealing.

All alloys were received as button-headed tensile bars in rough-machined condition. These were heat-treated to develop a reproducible equiaxed grain structure with a uniform grain size. The gage section was then machined to the dimensions specified in Figure 4.

TABLE IV Chemical Analyses of All Alloys Prepared for the Hot-Working Studies (in ppm)

Element	Alloy Designation								
	Base Material	"L" F-33930	"P" F-33948	"V" 20385	"A" F-33918	"C" F-33945	"E" 20389	"H" 20402	"X" 20398
Si	13	122*	199*	399*	<10	22	68	71	32
Fe	60	82	66	67	135*	287*	50	62	70
Al	16	<6	—	<6	8	—	203*	750*	<6
C	81	76	137	65	51	142	48	80	795*
N	22	64	74	50	44	40	32	22	80
Cr	15	14	14	9	10	11	10	13	14
Mg	14	<4	<4	—	<4	<4	—	—	—
Mn	9	9	7	6	8	8	6	4	9
Ni	24	37	37	40	30	33	35	68	42
Pb	1	<1	<1	—	2	<1	—	—	—

*Intentional additions.

(a) BASE MATERIAL

FIGURE 3 The As-Received Structures of the Various Alloys Utilized in the Hot Deformation Experiments. (200X)

(b) "P" ALLOY (199 ppm Si)

(c) "L" ALLOY (122 ppm Si)

FIGURE 3 (Continued)

(d) "C" ALLOY (287 ppm Fe)

(e) "A" ALLOY (135 ppm Fe)

FIGURE 3 (Continued)

(f) "V" ALLOY (399 ppm Si)

(g) "E" ALLOY (203 ppm Al)

FIGURE 3 (Continued)

FIGURE 3 (Continued)

(i) "X" ALLOY (795 ppm C)

(h) "H" ALLOY (750 ppm Al)

FIGURE 4 Tensile Bar for High - Temperature Deformation

B. Apparatus for High - Temperature Deformation Studies

The apparatus required for the high-temperature deformation work included specimen machining and metallographic preparation equipment housed in specially constructed glove boxes in addition to an apparatus for hot-deforming uranium alloy specimens.

1. Glove Boxes

For the performance of dust generating operations involving uranium, three glove boxes equipped with exhaust fans and filters were constructed. The largest contained a South Bend 9 - inch lathe for the finish machining of tensile bars. The second contained a Buehler Metallurgical cut-off wheel. A belt grinder, a Buehler Handi-Met wet grinder and an 8 - inch Buehler polishing wheel occupied the third box, thus providing a capability for preparing metallographic microsections.

2. High - Temperature Straining Apparatus

A device suitable for conducting controlled straining experiments at elevated temperature had been developed in connection with an earlier research. Only slight modification was necessary to permit its use with uranium.

The device, described by the sketch in Figure 5, consisted of a simple, inverted, portable tensile machine which permitted the specimen and grips to be immersed in a constant temperature bath and then quickly removed to a water quenching bath without interrupting the straining. Being made of A.I.S.I. 316 stainless steel and Inconel X, the grips and straining section of this device were suitable for immersion in molten solder. The rate of movement of the upper crosshead could be varied by removing a gear reducer and by changing gears in the gear train. Thus, a step-wise range of strain rates from 0.75 to 0.009 inches/inch/minute was available by the use of various combinations of gears and gear reducers. The pot furnace was of commercial manufacture (Hevi-Duty) and was equipped with $\pm 5^{\circ}\text{C}$ temperature control. Preliminary work indicated the most satisfactory heating medium for temperatures below 600°C was a 60 Sn-40 Pb solder. An alloy pot (35% nickel, 15% chromium, balance iron) proved quite satisfactory. The furnace was also equipped with an exhaust hood.

FIGURE 5 High-Temperature Strain Apparatus.

C. Procedures

1. Preliminary Heat Treatment

The as-received materials exhibited a considerable variation in microstructure, Figure 3. In order to insure that all bars of a particular alloy were structurally the same at the beginning of the working period, and that the structures of the various alloys were as similar as possible (at least in type, i.e., completely annealed), all bars were given a preliminary anneal in the solder bath. The $\frac{1}{2}$ -inch-diameter by $\frac{1}{4}$ -inch-long cylinder supplied with each tensile bar was heat-treated along with its corresponding tensile bar. Half of each cylinder was retained for reference to the initial microstructure. These specimens were designated by an "a" after the specimen number, i.e., P-8a. The other half of the cylinder was subjected to the same subsequent thermal history as its corresponding tensile bar. The annealing conditions utilized for the various alloys are presented in Table V, and the structures produced by these treatments are illustrated by Figure 6. An equiaxed structure was not produced in the "H" alloy (750 ppm Al) by this treatment. Accordingly, the "H" alloy was not included in subsequent investigations.

2. Machining of Tensile Bars

Subsequent to the preliminary heat treatments, the gage section of each tensile bar was machined to a standard diameter. For the base material and the "P", "A", "C", and "X" alloys, this diameter was approximately 0.155 inches. A slight attack of the "V" and "E" alloys by the solder bath necessitated machining the gage section of the "V" bars to a diameter of about 0.135 inch and the gage section of the "E" bars to a diameter of about 0.145 inch. After machining, the gage length of each bar was measured with an optical comparator, the overall length with a vernier caliper, and the gage diameter with a micrometer. The bars were then ready to be hot-worked.

(a) BASE MATERIAL

(b) "P" ALLOY (199 ppm Si)

FIGURE 6 Structures Produced in the Various Alloys by the Heat Treatments Described in Table V. (200X)

(c) "V" ALLOY (399 ppm Si)

(d) "C" ALLOY (287 ppm Fe)

FIGURE 6 (Continued)

(e) "A" ALLOY (135 ppm Fe)

(f) "E" ALLOY (203 ppm Al)

FIGURE 6 (Continued)

(g) "X" ALLOY (795 ppm C)

FIGURE 6 (Continued)

TABLE V Alloys and Preliminary Heat Treatments Used

Alloy Designation	Alloy Addition	Annealing Conditions	
		Temp ($^{\circ}$ C)	Time (hr)
Base Material	none	650	1
P	199 ppm Si	650	1
V	399 ppm Si	650	4
A	135 ppm Fe	650	$\frac{1}{2}$
C	287 ppm Fe	650	$\frac{1}{2}$
E	203 ppm Al	650	1
H	750 ppm Al	*	•
X	795 ppm C	650	1

*No satisfactory set of conditions has yet been devised for this alloy.

3. Hot Working and Annealing After Hot Working

All hot-working experiments utilized a single procedure, as did all experiments which involved annealing after hot working. To perform the actual straining experiments the gear reducer and gear train were set for the desired rate of extension, a specimen was placed in the grips and all slack removed from the system. In all experiments a small piece of uranium of the same chemical analysis and having the same initial structure as the tensile bar was wired to the upper grip of the strain jig, so that it hung adjacent to the gage section of the tensile bar. Although this

piece was not worked, it experienced the same thermal history as the tensile bar. Thus, it afforded a check on the structural changes which occurred during hot working. These pieces are designated throughout by a "T" after the specimen number, i.e., P-8T.

The jig was then lowered into the molten solder and 20 minutes was allowed for temperature equilization. Following this period of time the motor was switched on for a time calculated to give the desired total extension. At the end of this time the jig was quickly removed from the solder pot and water quenched. The motor was turned off as the jig cleared the solder pot. The bars were then wiped free of any excess solder using a propane torch to just melt the solder. The gage length, gage diameter, and total length were then remeasured.

Annealing after deformation was performed simply by stopping the motor and allowing the jig to remain in the solder pot the desired time before quenching.

Tensile bars of each alloy have been extended at several different temperatures, but predominately at 600°C, 500°C, 400°C, and 300°C. Two strain rates, 0.009/minute and 0.75/minute, were used to elongate the tensile bars to various amounts of total strain up to fracture. The conditions of deformation experienced by each bar are given in Table VI. A number of quantitative metallographic measurements were made on the specimens listed.

TABLE VI Specimens Extended and Working Conditions

Specimen Number	Temp (°C)	Rate (/min)	Extension (%)	Remarks
83	450	0.75	34	fracture
92	450	0.009	43	fracture
78	400	0.75	34	fracture
84	350	0.75	32	fracture
81	350	0.75	31	fracture
79	350	0.009	33	fracture
73	220	0.75	41	fracture
76	220	0.009	30	fracture
49	255	0.75	30	no anneal
48	255	0.75	36	fracture
50	545	0.75	31	fracture, no anneal
51	545	0.75	48	
82	600	0.009	66	fracture
9	600	0.009	55	
6	600	0.009	42	
2	600	0.009	27	
1	600	0.009	9	
23	600	0.08	56	fracture
22	600	0.08	37	
20	600	0.08	11	
11	600	0.75	63	fracture
12	600	0.75	47	
13	600	0.75	41	jig accidentally ran several seconds after quench
61	600	0.75	38	repeat of 13
15	600	0.75	25	
16	600	0.75	14	
17	600	0.75	38	2 min anneal
67	600	0.75	37	4 min anneal
58	600	0.75	37	8 min anneal
65	600	0.75	38	16 min anneal
66	600	0.75	40	32 min anneal
63	500	0.009	65	fracture
71	500	0.009	54	
36	500	0.009	44	
38	500	0.009	36	
47	500	0.009	26	
29	500	0.009	17	
45	500	0.009	8	
42	500	0.009	8	
85	500	0.75	52	fracture
30	500	0.75	52	fracture
28	500	0.75	47	
27	500	0.75	34	
26	500	0.75	23	
25	500	0.75	11	
5	500	0.75	22	broken removing from jig
53	400	0.009	35	fracture
55	400	0.009	26	

TABLE VI (Continued)

Specimen Number	Temp (°C)	Rate (/min)	Extension (%)	Remarks
52	400	0.009	18	
64	400	0.009	8	
62	400	0.75	42	fracture
60	400	0.75	36	
54	400	0.75	22	
39	400	0.75	12	
93	300	0.009	24	fracture
74	300	0.009	23	fracture
80	300	0.009	16	
77	300	0.009	5.5	
69	300	0.75	43	fracture
72	300	0.75	34	
57	300	0.75	24	
56	300	0.75	11	
21	R.T.	0.08	7	fracture
18	R.T.	0.75	3.5	fracture
P-70	600	0.009	70	fracture
P-65	600	0.009	69	
P-38	600	0.009	57	
P-94	600	0.009	41	
P-63	600	0.009	27	
P-77	600	0.009	11	
P-14	600	0.75	59	fracture
P-23	600	0.75	62	
P-22	600	0.75	51	
P-18	600	0.75	36	
P-13	600	0.75	21	
P-5	600	0.75	11	
P-85	500	0.009	63	
P-93	500	0.009	63	
P-71	500	0.009	55	
P-84	500	0.009	43	
P-83	500	0.009	35	
P-72	500	0.009	26	
P-69	500	0.009	17	
P-68	500	0.009	7	
P-21	500	0.75	48	fracture
P-15	500	0.75	50	
P-24	500	0.75	36	
P-19	500	0.75	23	
P-20	500	0.75	11	
P-39	400	0.009	36	fracture
P-48	400	0.009	35	
P-51	400	0.009	26	
P-33	400	0.009	15	
P-52	400	0.009	4	
P-17	400	0.75	35	fracture
P-8	400	0.75	21	
P-16	400	0.75	10	

TABLE VI (Continued)

Specimen Number	Temp ($^{\circ}\text{C}$)	Rate (/min)	Extension (%)	Remarks
P-43	300	0.009	27	fracture
P-42	300	0.009	26	
P-40	300	0.009	16	
P-53	300	0.009	5	
P-44	300	0.75	45	fracture
P-41	300	0.75	35	
P-36	300	0.75	22	
P-34	300	0.75	9	
P-64	500	0.75	40	fracture
P-98	500	0.75	45	fracture
P-54	450	0.75	30	fracture
P-62	450	0.009	49	fracture
P-55	400	0.75	29	fracture
P-66	350	0.75	32	fracture
P-67	350	0.75	32	fracture
P-74	350	0.009	24	fracture
P-75	220	0.75	41	fracture
P-97	220	0.009	41	fracture
P-30	600	0.75	39	anneal 6 minutes at 600°C
P-76	600	0.75	39	anneal 12 minutes at 600°C
P-46	600	0.75	39	anneal 24 minutes at 600°C
P-79	600	0.75	41	anneal 48 minutes at 600°C
P-89	600	0.75	39	anneal 96 minutes at 600°C
P-95	600	0.75	38	anneal 144 minutes at 600°C
A-3	600	0.75	14	
A-5	600	0.75	29	
A-22	600	0.75	40	
A-2	600	0.75	51	fracture
A-4	600	0.009	15	
A-6	600	0.009	26	
A-7	600	0.009	40	fracture
A-16	500	0.75	12	
A-17	500	0.75	24	
A-21	500	0.75	39	
A-9	500	0.75	48	fracture
A-15	500	0.009	14	
A-18	500	0.009	29	
A-41	500	0.009	40	fracture
A-74	450	0.75	52	fracture
A-54	450	0.009	61	fracture
A-45	450	0.009	41	fracture
A-19	400	0.75	13	
A-11	400	0.75	27	
A-23	400	0.75	38	
A-25	400	0.75	48	fracture
A-12	400	0.009	9	
A-43	400	0.009	19	
A-52	400	0.009	28	
A-66	400	0.009	37	

TABLE VI (Continued)

Specimen Number	Temp (°C)	Rate (/min)	Extension (%)	Remarks
A - 78	400	0.009	40	fracture
A - 77	350	0.75	40	fracture
A - 55	350	0.75	39	fracture
A - 75	350	0.009	36	fracture
A - 8	300	0.75	12	
A - 10	300	0.75	24	
A - 20	300	0.75	37	
A - 14	300	0.75	49	
A - 58	300	0.75	50	fracture
A - 13	300	0.009	9	
A - 56	300	0.009	17	
A - 59	300	0.009	27	
A - 38	300	0.009	37	
A - 57	300	0.009	33	fracture
A - 69	300	0.009	29	fracture
A - 68	220	0.75	43	fracture
A - 67	220	0.009	44	fracture
A - 44	220	0.009	44	fracture
A - 1	600	0.75	39	anneal 24 minutes at 600°C
A - 39	600	0.75	38	anneal 48 minutes at 600°C
A - 46	600	0.75	34	anneal 96 minutes at 600°C
A - 53	600	0.75	41	anneal 96 minutes at 600°C
C - 2	600	0.75	7	
C - 5	600	0.75	24	
C - 6	600	0.75	37	
C - 7	600	0.75	47	
C - 11	600	0.75	47	
C - 1	600	0.75	54	fracture
C - 13	600	0.009	15	
C - 9	600	0.009	30	
C - 8	600	0.009	28	fracture
C - 95	600	0.009	42	
C - 53	600	0.009	43	
C - 3	500	0.75	8	
C - 4	500	0.75	27	
C - 90	500	0.75	40	
C - 94	500	0.75	46	fracture
C - 100	500	0.75	43	fracture
C - 32	500	0.009	14	
C - 10	500	0.009	27	
C - 39	500	0.009	41	
C - 101	500	0.009	42	fracture
C - 45	450	0.75	54	fracture
C - 41	450	0.009	64	fracture
C - 23	450	0.009	47	fracture
C - 96	400	0.75	15	
C - 108	400	0.75	24	
C - 99	400	0.75	37	
C - 107	400	0.75	40	fracture

TABLE VI (Continued)

Specimen Number	Temp ($^{\circ}$ C)	Rate (/min)	Extension (%)	Remarks
C-92	400	0.009	9	
C-97	400	0.009	18	
C-103	400	0.009	27	
C-111	400	0.009	37	
C-49	400	0.009	42	fracture
C-40	350	0.75	34	fracture
C-106	350	0.75	41	fracture
C-26	350	0.009	30	fracture
C-34	300	0.75	12	
C-37	300	0.75	23	
C-43	300	0.75	40	fracture
C-29	300	0.009	8	
C-30	300	0.009	18	
C-44	300	0.009	27	
C-48	300	0.009	38	fracture
C-102	220	0.75	52	fracture
C-57	220	0.75	37	fracture
C-24	220	0.009	39	fracture
C-91	600	0.75	40	anneal 24 minutes at 600 $^{\circ}$ C
C-28	600	0.75	39	anneal 48 minutes at 600 $^{\circ}$ C
C-104	600	0.75	37	anneal 96 minutes at 600 $^{\circ}$ C
E-2	600	0.75	16	
E-1	600	0.75	27	
E-3	600	0.75	39	fracture
E-6	600	0.009	16	
E-7	600	0.009	30	
E-8	600	0.009	43	
E-11*	600	0.009	30	fracture
E-21	600	0.009	39	fracture
E-18*	500	0.75	13	
E-19	500	0.75	14	
E-17*	500	0.75	26	
E-12*	500	0.75	31	fracture
E-44*	500	0.009	14	
E-26	450	0.75	31	fracture
E-20	450	0.009	33	fracture
E-16	400	0.75	14	
E-13	400	0.75	20	fracture
E-49*	400	0.009	7	
E-15	400	0.009	18	
E-10	400	0.009	24	fracture
E-22	350	0.75	28	fracture
E-25	350	0.009	25	fracture
E-29	300	0.75	11	
E-30	300	0.75	15	
E-32	300	0.75	30	fracture
E-14	300	0.009	9	
E-51*	300	0.009	16	

*Unintentionally annealed above the α - β transformation temperature.

TABLE VI (Continued)

Specimen Number	Temp (°C)	Rate (/min)	Extension (%)	Remarks
E-24	300	0.009	16	
E-31	300	0.009	24	fracture
V-24	600	0.75	13	
V-1	600	0.75	22	
V-4	600	0.75	26	
V-31	600	0.75	40	
V-33	600	0.75	37	fracture
V-52	600	0.009	16	
V-32	600	0.009	35	
V-21	600	0.009	41	fracture
V-26	500	0.75	16	
V-11	500	0.75	22	
V-13	500	0.75	30	fracture
V-6	500	0.009	15	
V-15	500	0.009	29	
V-9	500	0.009	33	fracture
V-42	400	0.75	12	
V-56	400	0.75	25	fracture
V-17	400	0.009	8	
V-10	400	0.009	19	
V-8	400	0.009	28	
V-55	400	0.009	30	fracture
V-58	350	0.009	25	fracture
V-38	300	0.75	12	
V-44	300	0.75	19	fracture
V-12	300	0.009	8	
V-45	300	0.009	12	fracture
V-40	300	0.009	18	fracture
X-10	600	0.75	14	
X-12	600	0.75	26	
X-7	600	0.75	41	
X-5	600	0.75	50	fracture
X-6	600	0.009	13	
X-8	600	0.009	29	
X-11	600	0.009	42	fracture
X-1	500	0.75	7	
X-2	500	0.75	29	
X-3	500	0.75	38	
X-4	500	0.75	48	fracture
X-9	500	0.009	13	
X-20	500	0.009	29	
X-17	500	0.009	42	
X-23	500	0.009	47	fracture
X-35	450	0.75	36	fracture
X-33	450	0.009	45	fracture
X-24	400	0.75	12	
X-16	400	0.75	23	
X-18	400	0.75	36	
X-22	400	0.75	40	fracture

TABLE VI (Continued)

Specimen Number	Temp (°C)	Rate (/min)	Extension (%)	Remarks
X-19	400	0.009	9	
X-13	400	0.009	19	
X-30	350	0.75	25	fracture
X-27	350	0.009	34	fracture
X-29	300	0.75	4	
X-26	300	0.75	21	
X-32	300	0.75	30	fracture
X-15	300	0.009	9	
X-21	300	0.009	18	
X-31	300	0.009	22	fracture

4. Metallography

No single metallographic procedure proved satisfactory for all specimens. Through the penultimate step, however, all of the procedures are identical. These steps consist of: (1) mounting a ½ to 1-inch section of the gage length in Nu-weld, a cold-setting dental plastic, (2) wet grinding on a 240-grit silicon carbide belt, (3) wet grinding on 240, 320, 400, and 600 silicon carbide metallographic papers, (4) mechanical polishing, first with 6 micron diamond paste on a nylon cloth and then with 1 micron diamond paste on Microcloth, and finally (5) mechanical polishing on a Snytron vibratory polisher using Linde "A" on Microcloth. The final electropolishing procedures, however, varied from alloy to alloy.

Electropolishing was performed in two steps. The first was the major polishing step; the second consisted of a short time at a relatively low voltage (in the polishing solution) in order to remove the surface film formed during the first step. Conditions for both steps are outlined in Table VII. All polishing was performed at room temperature and utilized a stainless steel cathode.

After electropolishing, the specimens were anodized in a 40% aqueous solution of sodium citrate.* The best anodizing conditions for all materials were found to be 13 volts for 2 to 4 seconds. This procedure also utilized a stainless steel cathode and was performed at room temperature.

In recent work with the "T" and "α" specimens of the "V" alloy it was found that the electropolishing techniques described above were not workable. After many experiments, the following procedure was established:

- Alloy specimens are oxidized in air for 24 hours to form a uniform yellow oxide after Snytron polishing with Linde alumina.
- Electropolish in single ortho at 15 volts for 1 minute.
- Adjust voltage to maintain a 15-ma cell current for 3 to 8 minutes until the specimen surface is bright.
- Reduce current to 5 ma for 1 minute.

* In later work, particularly with "V" alloy specimens, a saturated aqueous solution of sodium citrate was found to result in better anodized coatings.

TABLE VII Electropolishing Solutions and Operating Conditions Used in Polishing the Various Materials Utilized in the Hot-Working Studies

Alloy	Solution	Time (min)	Voltage (volts)	Time (sec)	Voltage (volts)
Base Material	single ortho*	20	14	30	6
"P" alloy	double ortho**	15	3.5 - 4.0	30	1
"V" alloy	single ortho*	10	10	30	1
"A" alloy	double ortho**	15	8	30	1
"C" alloy	double ortho**	15	8	30	1
"E" alloy	single ortho*	10	10	30	2
"H" alloy	single ortho*	10	10	30	1
"X" alloy	double ortho**	20	8	30	1

*5 parts H_3PO_4 , 5 parts ethylene glycol, 8 parts C_2H_5OH

**10 parts H_3PO_4 , 5 parts ethylene glycol, 8 parts C_2H_5OH

A properly prepared specimen examined under polarized light will show excellent contrast between grains and between mechanical twins and the matrix grain. It is also possible to detect lattice bending and, to some degree, subgrain formation. Metallographic examination should be carried out at magnifications in excess of 500X.

5. Quantitative Metallography

All quantitative metallographic data resulted from two types of measurements: (1) point counting and (2) intercept counting. All intercept counts and some of the point counts were performed at a magnification of 525X; the remainder of the point counts were performed at a magnification of 340X.

Intercept counts yield directly N_L , the number of intercepts made with a certain microstructural feature by unit length of test line. N_L is related to the surface area per unit volume possessed by the feature of interest through the expression:

$$S_V = 2N_L \quad (1)$$

Point counts were used to determine the volume fraction of the structure which is occupied by the feature of interest. The ratio P_P of the total number of points falling on the feature of interest to the total number of points applied to the structure gives the volume fraction V_V of the structure occupied by the feature of interest:

$$P_P = V_V \quad (2)$$

Both types of measurements were performed with the aid of a 7×7 grid inserted into a 10X eyepiece. New areas were brought into the field of view by moving the microscope stage a predetermined amount. When measurements of N_L and $(N_L)_{total}$ were performed, the eyepiece (and hence the grid) was rotated 9° between areas. This procedure eliminated any orientation dependence in the results due to the position of the test line with respect to the tensile axis of the specimen. A total of 20 areas was counted as a group. In most cases counts from two groups were used in calculating the reported values for the various N_L 's.

The following measurements were made, where possible and applicable, on the extended specimens:

- a. $(N_L)_L$ — the number of grain boundary intersections per unit length of line parallel with the tensile specimen axis.
- b. $(N_L)_T$ — the number of grain boundary intersections per unit length of line perpendicular to the axis.
- c. $(N_L)_{L(total)}$ — the total number of grain boundary and subgrain boundary intersections per unit length of line parallel to the specimen axis.
- d. $(N_L)_{T(total)}$ — the total number of grain boundary and subgrain boundary intersections per unit length of line perpendicular to the specimen axis.
- e. N_L — the number of grain boundary intersections per unit length of line independent of specimen orientation.
- f. $(N_L)_{(total)}$ — the total number of grain boundary and subgrain boundary intersections per unit length of line independent of specimen orientation.
- g. $(V_V)_{N-T}$ — the volume fraction of non-thermal twins, i.e., those twins formed during working at the working temperature as opposed to thermal twins formed during rapid cooling.
- h. V_V — the volume fraction of strain-free (recrystallized) material; measured only in specimens annealed after hot working.

Measurements of $(N_L)_L$, $(N_L)_T$ and N_L were also performed on a number of the unstrained "T" specimens and on the "a" specimens of the "V" alloy. The measurements of the "a" specimens provided data which were used to define the starting microstructures, and those of the "T" specimens were used to define the microstructure which had not been subjected to deformation, but had the same thermal history as the corresponding deformed tensile specimens. Throughout this report, values of N_L are in units per millimeter.

6. Thermal Cycling

Two different sets of preliminary thermal cycling experiments were conducted, each with a different apparatus.

The first apparatus was a cam-actuated machine designed to cycle mechanically a small piece of uranium between a solder pot and a blast of room temperature air to produce a thermal cycle between approximately 600°C and 60°C. A sketch of this apparatus is included as Figure 7. The cam, utilized in all experiments, resulted in the thermal cycle outlined below.

<u>Time (sec)</u>	<u>Temp (°C)</u>	<u>Position of Specimen</u>
0	60	just in
25	605	in
48	605	just out
66	150	out
122	64	out-end of cycle

The temperature cycle was measured with the aid of a chromel-alumel thermocouple embedded in a 10-gram cylinder of uranium (base material).

FIGURE 7 First Thermal Cycling Apparatus

After a predetermined number of cycles the specimens were prepared for metallographic examination by the procedures outlined above. Quantitative metallography was employed to monitor the structure changes that resulted from thermal cycling.

A second apparatus was designed to cycle a uranium specimen between two controlled temperature limits by alternate immersion in solder baths held at two different temperatures. A schematic drawing of this apparatus is given in Figure 8. Transfer time between the pots was about 5 seconds, and the holding time in each pot was 15 seconds. Pot 1 was set at a temperature of 230°C and pot 2 at 511°C. The thermal cycle for this apparatus is outlined below.

<u>Time (sec)</u>	<u>Temp (°C)</u>	<u>Position of Specimen</u>
0	230	just out of pot 1
5		in pot 2
20	508	just out of pot 2
25		in pot 1
40	233	just out of pot 1
45		in pot 2
60	509	just out of pot 2
65		in pot 1
80	232	just out of pot 1
85		in pot 2
100	509	just out of pot 2
105		in pot 1
120	231	just out of pot 1

FIGURE 8 Second Thermal Cycling Apparatus

The temperature cycle was measured in the same manner as with the first apparatus. Specimen preparation and evaluation were performed in essentially the same manner as previously described.

III. EXPERIMENTAL WORK AND RESULTS

The metallographic study has dealt with the effect of hot deformation upon the uranium grains, their mutual grain boundaries, and the second phase particles. Features of the grain boundary that vary during hot deformation are: (1) the total area of the grain boundary, (2) the contour of the grain boundary, whether smooth or irregular, (3) the anisotropy of orientation exhibited by grain boundaries, and (4) the appearance and area of subgrain boundaries. In addition to their external form, the grains themselves may vary with respect to: (1) the size of the subgrains, (2) the presence, or absence, and degree of lattice bending, and (3) the amount and form of mechanical twinning.

As an aid in following the metallographic presentation, a brief description of the physical appearance of some of these features may prove useful. Perhaps the most striking microstructural feature of hot deformed materials is the grain boundary serrations, which appear during deformation above the recovery temperature. These appear as almost straight-sided zigzags of the grain boundary (see Figure 9a). Both the size and spacing of the serrations depend upon conditions of deformation.

Another prominent microstructural feature of uranium, deformed at elevated temperature, is the large, well defined subgrains (also illustrated in Figure 9a). These are delineated by relatively sharp changes in "shading" within the large grains. The tone contrast (orientation difference) and quantity of subgraining are also strong functions of the deformation conditions. With increasing deformation the subgrains become more distinct. Faster straining and lower temperature produce more and smaller subgrains.

Two other microstructural features that become prominent as the temperature of deformation is decreased are: (1) lattice bending and (2) deformation twinning. Both of these are illustrated in Figure 9b. Lattice bending appears as a continuous change in shading of the individual grains and is readily apparent in several of the grains in the middle of the photomicrograph. Deformation twins formed at the deformation temperature appear as lenticular stripes extending from boundary to boundary of a grain. These are characteristically of much greater width than are the more numerous thin thermal twins. A number of deformation twins appear in the extensively elongated, grey grain which is slightly below center at the far right of Figure 9b. Nearly all grains contain a few of the thin thermal twins which are formed by thermal stresses during cooling from the deformation temperature.

(a) SPECIMEN NO. 15

(b) SPECIMEN NO. 62

FIGURE 9 Microstructural Features of Hot Deformed Uranium. (a) Grain Boundary Serrations and Subgrains, (b) Lattice Bending and Deformation Twins. (200X)

A. Metallography of the Initial Structures

As a reference state, the microstructure of each alloy (Base, "P", "V", "C", "A", "E", and "X") is given in Figure 6a through g. These samples had been annealed (see Table V) subsequent to their receipt. In general the microstructures were characterized by equiaxed straight-sided grains. Few if any subgrains were present. The large number of very thin mechanical twins which appear throughout the structures are thermal twins, formed during the quench from the annealing treatment; these were disregarded when considering the high temperature deformation process.

Since some differences in grain size exist among the several alloys, quantitative metallographic measurements were made to establish a starting point for each material. These measurements are recorded in Table VIII. In three of the four types of metal which were studied extensively by quantitative metallography, namely; base material and "P" and "C" alloys the initial grain sizes among different specimens of each alloy were observed to be uniform. The initial structures of the "V" alloy specimens, however, exhibited a wide range of grain sizes from specimen to specimen.

Microstructural parameters of each individual "a" specimen of the "V" alloy are given in Table IX. This starting state for each "V" alloy specimen was used to make adjustments in the calculated grain boundary anisotropy (e_g), grain boundary area (S_V), and subgrain boundary area ($S_{V(SB)}$) for the corresponding hot worked specimens, so as to compare among alloys on a fixed basis. (These terms are defined below.) A derivation of these adjustments is given in the Appendix.

TABLE VIII Quantitative Values of Annealed As-Received Alloys*

Alloy	$(N_L)_L$ (/mm)	$(N_L)_T$ (/mm)	N_L (/mm)
Base Material	27	29	28
P	48	52	49
V	67	75	72
A	41	45	45
C	40	42	38
E	32	29	30
X	44	48	43

*See Table V for standard annealing treatments.

TABLE IX Quantitative Values for "V" Alloy "a" Specimens
(Not Subjected to Hot Deformation)

Specimen Number	$(N_L)_L$ (/mm)	$(N_L)_T$ (/mm)	(N_L) (/mm)	$S_V = 2N_L$ (/mm)
V - 24a	70	88	81	162
V - 1a	50	55	53	106
V - 4a	43	54	51	102
V - 31a	67	79	75	150
V - 33a	89	102	103	206
V - 52a	70	83	73	146
V - 32a	89	97	92	184
V - 21a	73	85	82	164
V - 26a	74	88	80	160
V - 11a	43	53	46	92
V - 13a	66	67	71	142
V - 6a	48	54	49	98
V - 15a	44	50	47	94
V - 9a	39	46	43	86
V - 42a	63	69	61	122
V - 56a	75	84	75	150
V - 17a	67	77	75	150
V - 10a	42	74	45	90
V - 8a	34	42	39	78
V - 55a	—	—	—	—
V - 38a	60	68	68	136
V - 44a	62	81	67	134
V - 12a	94	103	101	202
V - 45a	45	52	52	104
V - 40a	53	65	62	124

Note: All specimens pre-annealed at 650°C for 4 hours.

B. Quantitative Metallographic Measurements Obtained from Hot-Worked Specimens

Quantitative metallographic measurements were obtained from specimens of the base material and "P", "C", and "V" alloys extended at 600°C, 500°C, 400°C, and 300°C. They are presented in Tables X, XI, XII, and XIII, respectively. These measurements were used to characterize the hot-worked structures through the calculation of the following quantities:

a. e_g , a measure of grain boundary anisotropy calculated from the equation:

$$e_g = \left(\frac{(N_L)_T}{(N_L)_L} \right)^{2/3} - 1 \quad (3)$$

b. S_V , the total grain boundary area per unit volume, calculated from the expression:

$$S_V = 2N_L \quad (1)$$

c. $S_{V(SB)}$, the total subgrain boundary area calculated from the expression:

$$S_{V(SB)} = 2 (N_L)_{total} - N_L \quad (4)$$

TABLE X Quantitative Values for Percent Extension of Base Material

Specimen Number	Extension (%)	$(N_L)_L$ (/mm)	$(N_L)_L(\text{total})$ (/mm)	$(N_L)_T$ (/mm)	$(N_L)_T(\text{total})$ (/mm)	N_L (/mm)	$(N_L)_{\text{total}}$ (/mm)	$(V_V)_{N-T}$ (%)
600°C, rate of extension = 0.75/min								
16	14	30	34	35	40	32	38	—
15	25	36	43	38	46	36	47	—
61	38	38	52	43	54	36	49	—
12	47	38	55	42	60	42	58	—
11	63	41	58	44	60	40	60	—
600°C, rate of extension = 0.009/min								
1	9	28	30	30	32	28	29	—
2	27	31	38	36	41	31	40	—
6	42	33	39	37	44	34	41	—
9	55	30	38	33	44	33	45	—
10	59	38	44	40	46	34	42	—
82	66	34	42	36	42	38	43	—
500°C, rate of extension = 0.75/min								
25	11	25	—	29	—	28	—	0.003
26	23	25	—	31	—	27	—	0.009
27	34	23	—	33	—	26	—	0.006
28	47	22	—	36	—	30	—	0.000
30	52	20	—	35	—	29	—	0.006
500°C, rate of extension = 0.009/min								
45	8	29	31	33	34	32	33	0.000
29	17	25	29	32	35	26	34	0.000
47	26	20	38	30	45	28	42	0.000
38	36	27	40	34	46	30	43	—
36	44	26	40	34	49	30	41	—
71	54	32	46	37	52	34	50	—
63	65	35	49	43	59	35	52	—
400°C, rate of extension = 0.75/min								
39	12	24	—	29	—	28	—	0.022
54	22	21	—	28	—	27	—	0.022
60	36	20	—	32	—	27	—	0.022
62	42	19	—	31	—	25	—	0.033
400°C, rate of extension = 0.009/min								
64	8	26	—	29	—	27	—	0.006
52	18	22	—	30	—	25	—	0.012
55	26	22	—	29	—	25	—	0.011
53	35	21	—	30	—	26	—	0.000
300°C, rate of extension = 0.75/min								
56	11	25	—	29	—	28	—	0.080
57	24	22	—	29	—	25	—	0.122
72	34	20	—	30	—	26	—	0.108
69	43	22	—	36	—	27	—	0.164
300°C, rate of extension = 0.009/min								
74	5.5	24	—	27	—	26	—	0.014
80	16	25	—	30	—	27	—	0.028
77	23	24	—	33	—	27	—	0.044

TABLE XI Quantitative Values for Percent Extension of "P" Alloy

Specimen Number	Extension (%)	(N _L) _L (/mm)	(N _L) _L (total) (/mm)	(N _L) _T (/mm)	(N _L) _T (total) (/mm)	N _L (/mm)	(N _L) _{total} (/mm)	(V _V) _{N-T} (%)
600°C, rate of extension = 0.75/min								
P-5	11	41	41	52	55	44	47	—
P-13	21	49	—	60	—	—	—	—
P-18	36	49	59	59	71	51	64	0.000
P-22	51	54	—	68	—	—	—	—
P-23	62	49	67	66	79	57	70	—
600°C, rate of extension = 0.009/min								
P-77	11	45	48	52	54	47	49	0.000
P-94	41	41	51	53	62	50	58	—
P-70	70	45	52	60	69	53	64	0.000
500°C, rate of extension = 0.75/min								
P-20	11	42	—	53	—	44	—	0.000
P-24	36	34	—	53	—	46	—	0.000
P-21	48	34	—	58	—	47	—	0.000
500°C, rate of extension = 0.009/min								
P-69	17	36	43	44	51	40	46	0.000
P-83	35	39	51	52	64	45	56	0.000
P-71	55	41	59	57	75	52	70	—
400°C, rate of extension = 0.75/min								
P-16	10	41	—	55	—	47	—	0.008
P-8	21	39	—	53	—	45	—	0.022
P-17	35	34	—	50	—	43	—	0.020
400°C, rate of extension = 0.009/min								
P-52	4	43	—	45	—	42	—	0.006
P-33	15	—	—	—	—	44	—	—
P-51	26	33	—	43	—	38	—	0.003
P-48	35	—	—	—	—	42	—	—
P-39	36	33	—	46	—	39	—	0.000
300°C, rate of extension = 0.75/min								
P-34	9	41	—	48	—	44	—	0.090
P-36	22	33	—	46	—	42	—	0.130
P-44	45	27	—	44	—	37	—	0.110
300°C, rate of extension = 0.009/min								
P-53	5	42	—	47	—	44	—	0.030
P-40	16	39	—	49	—	45	—	0.060
P-43	27	35	—	48	—	41	—	0.110

TABLE XII Quantitative Values for Percent Extension of "C" Alloy

Specimen Number	Extension (%)	$(N_L)_L$ (/mm)	$(N_L)_L(\text{total})$ (/mm)	$(N_L)_T$ (/mm)	$(N_L)_T(\text{total})$ (/mm)	N_L (/mm)	$(N_L)_{\text{total}}$ (/mm)	$(V_V)_{N-T}$ (%)
600°C, rate of extension = 0.75/min								
C-2	7	38	39	42	43	40	43	0.000
C-5	24	42	—	54	—	47	—	—
C-6	37	40	51	49	60	44	54	—
C-1	54	51	62	67	79	58	69	0.000
600°C, rate of extension = 0.009/min								
C-13	15	40	41	50	52	45	48	0.000
C-9	30	41	51	48	54	44	52	0.000
C-53	43	39	48	42	50	39	47	—
500°C, rate of extension = 0.75/min								
C-3	8	40	—	51	—	44	—	0.000
C-4	27	28	—	38	—	35	—	0.000
C-94	46	27	—	47	—	35	—	0.006
500°C, rate of extension = 0.009/min								
C-32	14	34	41	40	46	40	46	0.006
C-10	27	37	46	47	58	38	48	0.000
C-101	42	35	47	45	59	41	55	0.000
400°C, rate of extension = 0.75/min								
C-96	15	32	—	43	—	38	—	0.028
C-108	24	38	—	56	—	46	—	0.011
C-107	40	30	—	51	—	41	—	0.008
400°C, rate of extension = 0.009/min								
C-92	9	31	—	36	—	39	—	0.012
C-103	27	27	—	40	—	37	—	0.022
C-49	42	28	—	48	—	40	—	0.008
300°C, rate of extension = 0.75/min								
C-34	12	32	—	40	—	36	—	0.122
C-37	23	26	—	40	—	36	—	0.178
C-43	40	34	—	53	—	43	—	0.170
300°C, rate of extension = 0.009/min								
C-29	8	33	—	40	—	38	—	0.022
C-44	27	29	—	45	—	38	—	0.058
C-48	38	30	—	46	—	40	—	0.044

TABLE XIII Quantitative Values for Percent Extension of "V" Alloy

Specimen Number	Extension (%)	(N _L) _L (/mm)	(N _L) _L (total) (/mm)	(N _L) _T (/mm)	(N _L) _T (total) (/mm)	N _L (/mm)	(N _L) _{total} (/mm)	(V _V) _{N-T} (%)
600°C, rate of extension = 0.75/min								
V-24s	13	68	78	80	89	72	85	—
V-1s	22	47	59	62	71	47	63	—
V-4s	26	44	54	61	74	50	66	—
V-31s	40	71	94	99	119	84	106	—
V-33s	37	65	79	79	99	72	91	—
600°C, rate of extension = 0.009/min								
V-52s	16	60	69	74	90	68	85	0.556
V-32s	35	67	80	91	107	74	90	—
V-21s	41	74	85	97	119	91	108	—
500°C, rate of extension = 0.75/min								
V-26s	16	71	81	91	102	82	95	0.556
V-11s	22	47	55	72	84	59	72	—
V-13s	30	61	72	88	100	78	95	—
500°C, rate of extension = 0.009/min								
V-6s	15	56	63	68	74	64	71	1.667
V-15s	29	55	72	69	85	64	82	—
V-9s	33	46	59	58	72	57	69	1.112
400°C, rate of extension = 0.75/min								
V-42s	12	53	64	69	78	60	71	0.556
V-56s	25	65	75	92	105	79	89	1.112
400°C, rate of extension = 0.009/min								
V-17s	8	46	51	47	54	43	48	1.112
V-10s	19	40	50	49	57	45	52	0.556
V-8s	28	37	46	59	70	45	55	1.667
V-55s	30	63	75	91	105	83	92	—
300°C, rate of extension = 0.75/min								
V-38s	12	53	59	68	76	62	67	6.67
V-44s	19	50	56	77	85	63	72	5.55
300°C, rate of extension = 0.009/min								
V-12s	8	67	75	90	100	79	90	0.556
V-45s	12	45	52	60	67	51	57	5.00
V-40s	18	60	67	78	86	73	79	—

In addition, information on the effects of composition and working conditions (temperature, rate and extension) upon the amount of non-thermal twinning was obtained from the measurement of $(V_V)_N - T$.

1. *Total grain boundary area, S_V* , (a measure of grain size) is plotted as a function of the total extension for the various working temperatures in Figures 10 and 11 (base material), Figures 12 and 13 ("P" alloy), Figures 14 and 15 ("C" alloy) and Figures 16 and 17 ("V" alloy). Also plotted on all of the above graphs at zero extension are the values of total grain boundary area obtained from the appropriate "T" specimens; these specimens had had the same thermal history as had the extended specimens, but were not deformed. In the single case of the "V" alloy, because of the necessity of adjusting for a non-uniform initial grain size, the percentage change in the grain boundary area over that present in the initial state has been plotted in Figures 18 and 19. The following interpretations are drawn from these results:

- a. Normally (i.e., typically in the base material) the grain boundary area remains almost unchanged through-out the working cycle, except at the highest temperature of deformation.
- b. An increase in grain boundary area occurs with high temperature of working and with decrease in the extension rate. An increase in grain boundary area implies the appearance of new grains.
- c. A decrease in grain boundary area (corresponding to an increase in the average grain size) is marked chiefly in the presence of alloying agents, notably "V" alloy. This condition implies a definite growth of the original grains.
 - (1) The decrease in grain boundary area as observed in the "V" alloy occurs at all working temperatures at the outset of deformation. This trend tends to become reversed as deformation continues, ultimately becoming an increase in grain boundary area.
 - (2) The grain boundary area decrease is at least as marked at the lowest working temperature (300°C), and possibly more so, than at higher temperatures.
 - (3) The ultimate increase in grain boundary area is greatest at the higher temperatures and appears to maximize around 500°C.

2. *The ratio e_g^{24}* (a measure of anisotropy) is plotted as a function of total extension in Figures 20 and 21 (base material), Figures 22 and 23 ("P" alloy, 199 ppm Si), Figures 24 and 25 ("C" alloy, 287 ppm Fe), and Figures 26 and 27 ("V" alloy, 399 ppm Si). Adjusted values of grain boundary anisotropy (e_g) for "V" alloy are given in Figures 28 and 29 and in Table XIV (see also Appendix). A slope of e_g versus total extension equal to 45° corresponds to grain deformation by plastic effects alone; grain growth toward an equiaxed shape decreases the slope; change toward a more elongated grain increases the slope. It is apparent from these plots that the degree of grain boundary anisotropy is a function of the composition of the material as well as of its rate and degree of extension. These effects may be summarized as follows:

- a. Normally (i.e., typically in the base material) the slope is 45°, except at high temperature and at slow strain rate, where grain growth begins to be prominent.
- b. Deviation toward smaller slope occurs at lower temperatures and lower strain rates.
- c. Positive deviation tends to occur at the lower temperatures and during the early part of extension; it is accentuated by the presence of alloying elements, especially silicon.
 - (1) The temperature of maximum positive deviation is higher at faster strain rates.
 - (2) The elongated grains that give rise to the positive deviation are considerably longer than could be produced by the test bar extensions (Figure 30).

- (3) Elongated grains are bordered along their long edges by new grains (Figure 30). From this it is inferred that elongated grains owe their proportions, in part at least, to having been made narrow by the encroachment of small grains along two sides.

3. *Subgrain boundary area* could be measured only for those specimens which were worked at 600°C at both rates and at 500°C at a strain rate of 0.009/minute. These results are plotted versus total extension in Figures 31, 32, and 33 for the base material, "P" alloy, and "C" alloy, respectively. Plots for "P" and "C" alloys are quite similar. The subgrain size is consistently smaller for those specimens of both alloys worked at 600°C and 0.75/minute than for those worked at 600°C and 0.009/minute. The plots indicate, however, that the subgrains formed during working at 500°C at 0.009/minute are smaller than those formed at either rate at 600°C, at least at high extensions. On the other hand, comparison with plots obtained for the base material, Figure 31, indicates that alloying decreases the average size of the subgrains formed during working at 500°C and 0.009/minute (compare the plot for 500°C and 0.009/minute with that for 600°C and 0.75/minute in each graph). In addition, a large extension at 600°C and 0.009/minute for the base material apparently results in an increase in the subgrain size. Neither alloy shows this effect.

C. Extension to Fracture in the Temperature Range from 220°C to 600°C

All values of extension to fracture for the various materials have been collected into Table XV. These data have also been plotted in Figures 34 (base material), 35 ("P" alloy), 36 ("C" alloy), 37 ("V" alloy), 38 ("A" alloy), 39 ("E" alloy), and 40 ("X" alloy). All alloys except "V" exhibit a minimum in extension at an intermediate temperature. This minimum is best characterized by the temperature at which it occurs (300 to 400°C) and the total extension at this temperature. Appropriate values of these two parameters appear in Table XVI.

Both the rate of working and the alloy composition affect the high-temperature ductility of uranium. These effects may be summarized as follows:

1. All alloys except the "P" alloy (199 ppm Si) have considerably less ductility in the range from 400°C to 600°C than does the base material. For the iron alloys the ductility in this region is sensitive to the rate of working (compare the plots in Figures 36 and 38). This effect was not noted in any of the other materials.
2. *Additions of silicon have little effect on the temperature of the minimum; however, above 200 ppm they do decrease the ductility at the temperature of the minimum. On the other hand, iron additions do not change the temperature of the minimum, but they noticeably increase the ductility at that temperature. Aluminum seems to increase the temperature of the minimum, but decreases the ductility at this temperature. Carbon has little effect on either the temperature of the minimum, or upon the ductility at this temperature.*

D. Photomicrographs and Quantitative Metallographic Measurements of Volume-Fraction Strain-Free Material Obtained from Worked and Annealed Specimens

Specimens of the base material, the "P" alloy, the "A" alloy, and the "C" alloy were extended approximately 39% at 600°C and then annealed various times at 600°C. Structures obtained are included as Figures 41 (base material), 42 ("P" alloy), 43 ("C" alloy), and 44 ("A" alloy).

Quantitative metallographic measurements of the volume fraction of strain-free (recrystallized) material were obtained for each specimen. These results are included in Table XVII. They also have been plotted as Figures 44 (base material), 45 ("P" alloy), 46 ("C" alloy), and 47 ("A" alloy).

Both the quantitative measurements and the photomicrographs illustrate the very great retardation effect of alloying on the evolution of structure during annealing after hot working. Only 32 minutes were required for a specimen of the base material to become essentially strain-free, Figure 44. On the other hand, only 18% of a "P" alloy specimen was strain-free at the end of a 144-minute anneal. This retardation of the structural evolution was also great in the "A" and "C" alloys in which only 14% and 9%, respectively, of the structures were strain-free at the end of a 96-minute anneal. As further interpretation of these observations it may be noted that 14% of recrystallization occurs just before the rapid stage of recrystallization; i.e., it is on the foot of the S curve. No sign of an incipient rapid evolution was noted in the specimens of "P" and "C" alloys annealed 144 and 96 minutes, respectively.

Figures 44, 45, 46 and 47 are also characterized by a small percentage (~6%) of strain-free structure at zero annealing time. Thus, some strain-free grains must have originated and grown during the working period. This phenomenon has also been noted in a study of nickel 200.

E. Experimental Results of Thermal Cycling

Some preliminary data on the structural changes which occur during thermal cycling were obtained using the first apparatus and three specimens of the base material cycled approximately 100, 190, and 320 times between 605°C and 65°C. Structures obtained in each of the three specimens are illustrated by Figures 48, 49, and 50. One photomicrograph of each set was taken from the as-mechanically polished surface under bright field illumination. These illustrate the development of porosity during cycling. The second photomicrograph in each set was obtained from an electropolished and anodized surface. These illustrate the structural changes which resulted from each treatment and should be compared with Figure 6a, which is typical of the starting structure.

Quantitative metallographic measurements of grain boundary area (S_V), twin boundary area ($S_{V_{total}} - S_V$) and total boundary area ($S_{V_{total}}$) (exclusive of twin boundaries, but including subgrain boundaries) were obtained from each specimen. These measurements are included as Table XVIII. They have been plotted versus number of cycles in Figure 51. The data plotted at zero cycles were obtained from a structure similar to that which appears as Figure 6a.

The qualitative microstructural changes which are apparent from a comparison of Figures 48, 49, and 50 are confirmed by the quantitative data plotted in Figure 51. The most important of these structural changes are:

1. Grain boundaries become ragged early in the cycling process. However, the amount of grain boundary area, including irregularities, at any number of cycles up to 320 is no greater than the amount at zero cycles.
2. Subgrains begin to form at a low number of cycles and become smaller and better defined as the number of cycles increase (compare Figures 48 and 50). The decrease in subgrain size is apparent from the marked increase in total boundary area with an increasing number of cycles (Figure 51).

3. The amount of thermal twinning decreases with an increasing number of cycles (compare Figures 6a and 50b). This decrease is especially marked between zero and 100 cycles.

The second thermal cycling apparatus was used to obtain data on the effects of thermal cycling on structural evaluation in the base, "P" (199 ppm Si), and "C" (287 ppm Fe) alloys. Quantitative metallographic measurements of specimens thermally cycled 100, 300, 500, and 1000 times were made to determine grain boundary area (S_V), total grain boundary area ($S_{V_{total}}$) (exclusive of twin boundaries, but including subgrain boundaries), and twin boundary area (S_{V_M}). These measurements are plotted as boundary area versus number of cycles in Figures 52, 53, and 54.

In general, thermal cycling increases the total grain boundary area, decreases the twin boundary area, and has practically no effect on the grain boundary area. The increase in total grain boundary, therefore, reflects an increase in subgrain boundary area. With respect to changes in twin boundary area and total grain boundary area, the "P" (199 ppm Si) alloy is much more stable than either the base material or the "C" (287 ppm Fe) alloy. In regard to the greater stability of the "P" alloy another significant result is that after 1000 cycles no cavities were observed in specimens of the "P" alloy while after 500 cycles cavities were found in the "C" alloy. Cavities were found to lie along grain boundaries and connect quadruple points in the microstructure. This observation supports the assumed mechanism that cavities form at quadruple points and grow along grain boundaries as the result of grain boundary shearing.

TABLE XIV Adjusted Values of Grain Boundary Anisotropy for
"V" Alloy, Extended

Specimen Number	Total Ext. (%)	$N_L(a)$ (/mm)	k	$e_g(s)$	$e_g(a)$	Δe_g	e_g (Adjusted)*
600°C, rate of extension = 0.75/min							
V-24	13	81	2.08	11.3	16.5	-5.2	14.0
V-1	22	53	1.36	18.7	7.0	11.7	15.7
V-4	26	51	1.31	14.8	16.0	-1.2	15.1
V-31	40	75	1.92	23.5	11.2	12.3	17.8
V-33	37	103	2.65	13.8	9.5	4.3	11.1
600°C, rate of extension = 0.009/min							
V-52	16	73	1.87	14.7	12.3	2.4	13.6
V-32	35	92	2.36	22.8	5.3	17.5	12.7
V-21	41	82	2.10	20.0	10.0	10.0	14.8
500°C, rate of extension = 0.75/min							
V-26	16	80	2.05	17.0	11.8	5.2	14.3
V-11	22	46	1.18	33.0	15.7	17.3	30.3
V-13	30	71	1.82	27.7	0.8	26.9	15.6
500°C, rate of extension = 0.009/min							
V-6	15	49	1.26	13.6	8.0	5.6	12.4
V-15	29	47	1.20	16.1	8.8	7.3	14.9
V-9	33	43	1.10	16.5	11.2	5.3	16.0
400°C, rate of extension = 0.75/min							
V-42	12	61	1.57	19.0	6.8	12.2	14.6
V-56	25	75	1.93	28.5	8.0	20.5	18.6
400°C, rate of extension = 0.009/min							
V-17	8	75	1.93	1.3	10.1	-8.8	5.5
V-10	19	40	1.17	14.3	7.2	7.1	13.8
V-8	28	39	1.00	36.3	17.1	19.2	36.3
V-55	30	—	—	27.5	—	—	—
300°C, rate of extension = 0.75/min							
V-38	12	68	1.75	17.9	7.8	10.1	13.6
V-44	19	67	1.72	33.1	19.0	14.1	27.3
300°C, rate of extension = 0.009/min							
V-12	8	101	2.59	21.9	8.6	13.3	13.7
V-45	12	52	1.33	21.0	9.9	11.1	18.3
V-40	18	62	1.59	19.1	14.4	4.7	17.4

$$*e_g \text{ (Adjusted)} = \frac{\Delta e_g}{k} + e_g(a); \Delta e_g = e_g(s) - e_g(a)$$

TABLE XV Percent of Extension to Fracture for All Alloys

Temp (°C)	Base Material		"P" Alloy (199 ppm Si)		"C" Alloy (287 ppm Fe)		"V" Alloy (399 ppm Si)		"A" Alloy (135 ppm Fe)		"E" Alloy (203 ppm Al)		"X" Alloy (795 ppm C)	
			0.75	0.009	0.75	0.009	0.75	0.009	0.75	0.009	0.75	0.009	0.75	0.009
	/min	/min	/min	/min	/min	/min	/min	/min	/min	/min	/min	/min	/min	/min
600	63	59 66	59	70	54	43	37	41	51	40	39	39	50	42
500	52 52	65	48 40 45	63	46 43	42	30	33	48	40	—	—	48	47
450	34	43	30	49	54	47	—	—	52	41	31	33	36	45
400	42 34	35	35 29	36	40	42	25	30	48	40	20	24	40	—
350	32 31	33	32 32	24	41 34	30	—	25	40 39	36	28	25	25	34
300	43	22 24	45	27	40	38	19	12 18	49 50	29 33	30	24	30	22
220	41	30	41	41	52 37	39	—	—	43	44 44	—	—	—	—
R.T.	3.5													

TABLE XVI Parameters of Extension to Fracture

Material	Rate (/minute)	Position of Minima	
		Temp (°C)	Total Extension (%)
Base	0.75	350	31
	0.009	300	23
"P" alloy (199 Si)	0.75	400	29
	0.009	350	24
"C" alloy (287 Fe)	0.75	350	37
	0.009	350	30
"V" alloy (399 Si)	0.75	300*	19*
	0.009	300*	15*
"A" alloy (135 Fe)	0.75	350	40
	0.009	300	31
"E" alloy (203 Al)	0.75	400	20
	0.009	350*	23*
"X" alloy (750 C)	0.75	350	25
	0.009	300*	22*

*Tentative; i.e., lowest value obtained, but not clearly a minimum.

TABLE XVII Volume-Fraction (V_V) of Strain-Free (Recrystallized)
Material Extended Approximately 39% at 600°C and 0.75/minute

Specimen Number	Alloy Addition (ppm)	Annealing Time (min)	V_V
61	Base Material	0	0.05
17		2	0.11
67		4	0.27
58		8	0.59
65		16	0.88
66		32	0.94
P-18	199 Si	0	0.04
P-30		6	0.08
P-76		12	0.10
P-46		24	0.09
P-79		48	0.11
P-89		96	—
P-95		144	0.18
C-6	287 Fe	0	0.06
C-91		24	0.08
C-28		48	0.07
C-104		96	0.10
A-22	135 Fe	0	0.07
A-1		24	0.08
A-39		48	0.09
A-46		96	0.14
A-53		96	0.13

TABLE XVIII Quantitative Values for Thermally Cycled Specimens

Specimen Number	No. of Cycles	S_V (/mm)	$(S_V)_M$ (/mm)	$(S_V)_{total}$ (/mm)
TC-0	0	58	240	58
TC-1	96	61	138	77
TC-2	192	64	109	85
TC-3	320	63	102	88

FIGURE 10 Total Grain Boundary Area Versus Total Extension, Base Material Extended at 0.75/minute

FIGURE 11 Total Grain Boundary Area Versus Total Extension, Base Material Extended at 0.009/minute

FIGURE 12 Total Grain Boundary Area Versus Total Extension, "P" Alloy Extended at 0.75/minute

FIGURE 13 Total Grain Boundary Area Versus Total Extension, "P" Alloy Extended at 0.009/minute

FIGURE 14 Total Grain Boundary Area Versus Total Extension, "C" Alloy Extended at 0.75/minute

FIGURE 15 Total Grain Boundary Area Versus Total Extension, "C" Alloy Extended at 0.009/minute

FIGURE 16 Total Grain Boundary Area Versus Total Extension, "V" Alloy Extended at 0.75/minute

FIGURE 17 Total Grain Boundary Area Versus Total Extension, "V" Alloy Extended at 0.009/minute

FIGURE 18 Percent Difference in Grain Boundary Area Versus Total Extension, "V" Alloy Extended at 0.75/minute

FIGURE 19 Percent Difference in Grain Boundary Area Versus Total Extension, "V" Alloy Extended at 0.009/minute

FIGURE 20 Grain Boundary Anisotropy, e_g , Versus Total Extension, Base Material Extended at 0.75/minute

FIGURE 21 Grain Boundary Anisotropy, e_g , Versus Total Extension, Base Material Extended at 0.009/minute

FIGURE 22 Grain Boundary Anisotropy, e_g , Versus Total Extension, "P" Alloy Extended at 0.75/minute

FIGURE 23 Grain Boundary Anisotropy, e_g , Versus Total Extension, "P" Alloy Extended at 0.009/minute

FIGURE 24 Grain Boundary Anisotropy, e_g , Versus Total Extension, "C" Alloy Extended at 0.75/minute

FIGURE 25 Grain Boundary Anisotropy, e_g , Versus Total Extension, "C" Alloy Extended at 0.009/minute

FIGURE 26 Grain Boundary Anisotropy, e_g , Versus Total Extension, "V" Alloy Extended at 0.75/minute

FIGURE 27 Grain Boundary Anisotropy, e_g , Versus Total Extension, "V" Alloy Extended at 0.009/minute

FIGURE 28 Adjusted Grain Boundary Anisotropy, e_g , Versus Total Extension, "V" Alloy Extended at 0.75/minute

FIGURE 29 Adjusted Grain Boundary Anisotropy, e_g , Versus Total Extension, "V" Alloy Extended at 0.009/minute

(a) C-6 ANNEALED 0 MINUTES

(b) C-104 ANNEALED 96 MINUTES

FIGURE 30 Structures of the "C" Alloy Extended Approximately 39% at 600°C and at 0.75/minute; Then Annealed at 600°C. (200X)

FIGURE 31 Subgrain Boundary Area Calculated from $2[(N_L)_{tot} - N_L]$ Versus Total Extension, Base Material

FIGURE 32 Subgrain Boundary Area Calculated from $2[(N_L)_{tot} - N_L]$ Versus Total Extension, "P" Alloy

FIGURE 33 Subgrain Boundary Area Calculated from $2[(N_L)_{tot} - N_L]$ Versus Total Extension, "C" Alloy

FIGURE 34 Extension to Fracture, Base Material

FIGURE 35 Extension to Fracture, "P" Alloy

FIGURE 36 Extension to Fracture, "C" Alloy

FIGURE 37 Extension to Fracture, "V" Alloy

FIGURE 38 Extension to Fracture, "A" Alloy

FIGURE 39 Extension to Fracture, "E" Alloy

FIGURE 40 Extension to Fracture, "X" Alloy

(a) 17, ANNEALED 2 MINUTES

(b) 66, ANNEALED 32 MINUTES

FIGURE 41 Structures of the Base Material Extended 39% at 600°C and at 0.75/minute; Then Annealed at 600°C. (200X)

(a) P - 18, ANNEALED 0 MINUTES

(b) P - 95a, ANNEALED 144 MINUTES

FIGURE 42 Structures of the "P" Alloy Extended Approximately 39% at 600°C and at 0.009/minute; Then Annealed at 600°C. (200X)

(a) A-22, ANNEALED 0 MINUTES

(b) A-53, ANNEALED 96 MINUTES

FIGURE 43 Structures of the "A" Alloy Extended Approximately 39% at 600°C and at 0.75/minute; Then Annealed at 600°C. (200X)

FIGURE 44 Volume Fraction (V_V) of Strain-Free (Recrystallized) Grains Versus Annealing Time, Base Material Extended 39% at 600°C and at 0.75/minute; Annealed at 600°C

FIGURE 45 Volume Fraction (V_V) of Strain-Free (Recrystallized) Grains Versus Annealing Time, "P" Alloy Extended 39% at 600°C and at 0.75/minute; Annealed at 600°C

FIGURE 46 Volume Fraction (V_V) of Strain-Free (Recrystallized) Grains Versus Annealing Time, "C" Alloy Extended 39% at 600°C and at 0.75/minute; Annealed at 600°C

FIGURE 47 Volume Fraction (V_V) of Strain-Free (Recrystallized) Grains Versus Annealing Time, "A" Alloy Extended 39% at 600°C and at 0.75/minute; Annealed at 600°C

(a) BRIGHT FIELD

(b) POLARIZED LIGHT

FIGURE 48 Structure of a Specimen of the Base Material Cycled 98 Times Between 605°C and 65°C. (200X)

(a) BRIGHT FIELD

(b) POLARIZED LIGHT

FIGURE 49 Structure of a Specimen of the Base Material Cycled 193 Times Between 605°C and 65°C. (200X)

(a) BRIGHT FIELD

(b) POLARIZED LIGHT

FIGURE 50 Structure of a Specimen of the Base Material Cycles 320 Times Between 605°C and 65°C. (200X)

FIGURE 51 Boundary Areas Versus Number of Cycles for Thermally Cycled Specimens of Base Material, Preliminary Data on Structural Changes

FIGURE 52 Boundary Areas Versus Number of Cycles for Thermally Cycled Specimens of Base Material (510°C to 220°C)

FIGURE 53 Boundary Areas Versus Number of Cycles for Thermally Cycled Specimens of "P" Alloy (510°C to 220°C)

FIGURE 54 Boundary Areas Versus Number of Cycles for Thermally Cycled Specimens of "C" Alloy (510°C to 220°C)

IV. DISCUSSION

It should be recognized that alloys of uranium are at least two-phased (i.e., $\alpha\text{U} + \text{UC}$) and that all except the "high-purity" uranums are three phased, containing also U_6Fe or U_3Si . All highly alloyed compositions are doubtlessly four-phased, at least at the lower temperatures. Variation in the composition of the alpha uranium matrix is possible in the three-phased alloys, but not in the four-phased compositions.

A number of questions were proposed, the answers to which should help clarify the behavior of uranium and its alloys.

1. What conditions of temperature, deformation rate, and alloying promote grain growth, and what conditions deter it? Can grain growth be anisotropic?
2. What conditions of temperature, deformation rate, and alloying promote new grain formation (recrystallization)?
3. How does annealing after cold working differ from annealing after hot working?
4. What part does twinning play in the deformation and internal cracking of uranium?
5. How can uranium exhibit intermediate temperature brittleness without developing internal cracks? Why doesn't it crack as other metals do?
6. Why do cracks develop during thermal cycling and why are they prevented by the presence of silicon?

These are mostly complex questions. Partial answers will be found in a perusal of the metallographic results. It will be useful, therefore, to consider what each kind of metallographic measurement has to contribute.

A. Grain Boundary Area (S_V)

In principal, grain boundary area can be *diminished* only by thermal migration of the grain boundaries, as in recrystallization and grain growth. Grain boundary area can be *increased* by plastic deformation alone, and also by polygonization, since subgrain boundaries must be included in the total boundary network of the system. Accordingly, an initial downward slope of the curves of Figures 10 through 19 may be interpreted as grain growth. Were there no grain growth, the grain boundary area would increase during deformation, in direct proportion to the increase in external specimen surface area experienced by the tensile specimen. This does happen initially in some of the alloys of uranium, those shown in Figures 12 and 13 for example.

It is important to note that an initial downward slope of the S_V versus extension curves is perceptible at all temperatures in the base material and that it is more distinct, persisting to greater total extension at 300 and 400°C, than at the higher temperatures. Clearly, *there is pronounced thermal migration of grain boundaries (grain growth) during deformation, even at 300°C*. Moreover, the amount of grain growth must be substantial, because it is susceptible to detection only to the extent that it exceeds the area increase associated with plastic extension of the metal.

As hot deformation continues, the slope of the S_V curve tends to reverse, corresponding to a net gain in grain boundary area. This occurs at the smaller deformations, at higher working tempera-

tures and at slower rates of extension. It is intuitively unacceptable that this kind of reversal should result from a decrease in thermal migration of the grain boundaries, particularly since the reversal is more pronounced at higher working temperatures and slower deformation rates. This leaves only polygonization and new grain formation to account for the upward trend of grain boundary area versus extension curves. That this interpretation is tenable is made apparent by consideration of subgrain boundary area versus extension, which approaches zero near 10% of extension (Figures 31 and 32). The appearance of subgrains and the rise in grain boundary area are always sequential events.

A second reversal of slope of the grain boundary area curve, this time in the direction of decreasing area, is observed at the highest temperature and greatest extension (Figures 10, 14 and 18). This clearly corresponds with thermal migration of the newly formed boundaries.

The above discussion may be summarized by saying that *grain growth of the original grains accompanies hot deformation, being more vigorous at higher temperatures and lower strain rates, while new boundary formation begins after a certain degree of extension, which is smaller at higher temperatures and low strain rates, yielding a large increase in boundary area which then succumbs to general grain growth.* In other words, the ideal boundary area versus extension curve has the form of an "S" (actually ∞ when plotted).

The effects of alloying upon the amount of grain boundary may be evaluated both in terms of total area (i.e., relative grain size) and distortion of the "lazy S" curve. A comparison among Figures 10 through 17 makes it clear that alloying increases the grain boundary area (i.e., decreases grain size). The base material exhibits a mean grain boundary area in the neighborhood of 60/mm, while 287 ppm iron (alloy "C") elevates the mean to 75/mm, 199 ppm silicon (alloy "P") to 85/mm, and 399 ppm silicon (alloy "V") to 130/mm. This marked tendency toward grain refinement is also accompanied by a somewhat less clear, but apparently general, delay in the initiation of grain growth; that is, there is usually a small initial rise in grain boundary area preceding the first decline, Figures 12 through 15. Both conditions argue for alloying imposing a restraint upon grain boundary migration. The strength of the restraint appears to increase in the order: iron, silicon and to increase with alloy content. Further evidence of this effect resides in the observation, discussed elsewhere, that alloying suppresses recrystallization during annealing.

B. Grain Boundary Anisotropy (e_g)

The property of microstructure defined as grain boundary anisotropy (e_g) presumes homogeneity of grain shape. In the present studies this condition was sometimes realized and sometimes not. Accordingly, it is necessary to compare the plots of e_g , Figures 20 through 29, with their corresponding microstructures, in order to understand the message that each has to convey. Where the grain shapes appear homogeneous, it is reasonable to distinguish between shapes generated by plastic deformation alone (slope of 45°) and shapes that have resulted from grain boundary migration, with or without recrystallization (slope less than 45°).

Mixed grain shapes would ordinarily be expected to give slopes between 0 and 45° , and this condition is met in many cases. There are some circumstances, however, under which the slope is

found to exceed 45° , particularly during the early stages of deformation; this condition is most pronounced with the "V" alloy (Figures 26 through 29) but is perceptible in most of the alloys studied.

Taken at face value, a slope greater than 45° means that the grains are, on the average, longer, in comparison with their width, than could result from plastic deformation alone (consider Figures 55 and 56). This could result from an accelerated growth in the direction of plastic extension, from a narrowing of the grains perpendicular to the direction of extension, or both. In the absence of subgrain (or new grain) formation, an increase in grain boundary anisotropy beyond that produced by plastic extension can mean only directional grain growth, parallel with the axis of straining. This condition is seen at low temperature and during an early stage of deformation, before polygonization has begun. Although the effect is never large, it seems to be sufficiently distinct to warrant acceptance (see Figures 21 through 27, points lying above the 45° slope lines at first stages of deformation). In the absence of any better explanation, it seems reasonable to conclude that grain boundary pinning is disrupted by plastic deformation in proportion to the shear across the grain boundary. If this is the case, grain boundaries lying normal to the direction of strain should be the most mobile. Preservation of equilibrium angles at grain boundary junctions as the end boundaries move away from the grain center should also shorten the grain boundaries. This would produce pointed grains, such as are to be seen in photomicrographs where the length-to-breadth ratio of the grains is large; consider, for example, Figures 56 and 57. Such pointed grains, which are observed even at 300°C after considerable extension (Figure 58) may be taken as proof of directional growth.

Absolute narrowing of the grains also seems to occur after extensive deformation. This is invariably associated with new grain formation. New grain growth (recrystallization, if you prefer) consistently emanates from old grain boundaries. When the grain boundaries have a dominating orientation parallel to the strain axis, new grain formation tends to narrow the grains, thus further accentuating their axial anisotropy (see Figure 55). At the height of its development, this structural effect appears as an envelope of small grains embedding each large grain (Figure 59, for example).

The new-grain envelopes are sometimes several new grains in width and the new grain tends to be uniform in any one specimen, being coarser at the higher working temperatures and the slower strain rates. Thus, the size of the new grains appears to be established by a dynamic equilibrium between the rate of thermal grain boundary migration and the rate of appearance of subgraining of the new grains, resulting in more new grains being "nucleated" at the interfaces between the envelope and the large old grains. The net anisotropy of grain boundary (e_g) rises rapidly as the original grains are made narrower by the envelope of new grains and then diminishes as the number of new grains becomes so large that their boundaries dominate the average. Thus, the development of an ultimate decrease in slope of the e_g curve appears to be general for those cases in which any part of the slope exceeds 45° ; this decrease develops to a change of sign of the slope at a maximum in the e_g curve of the "V" alloy.

115

FIGURE 55 Structure Near Fracture of a Base Material Specimen Extended 63% at 600°C and at 0.75/minute. (200X)

625

FIGURE 56 Structure of a Base Material Specimen Extended 42% at 400°C and at 0.75/minute. (200X)

30S

FIGURE 57 Structure of a Base Material Specimen Extended 52% at 500°C and at 0.75/minute.
(200X)

69S

FIGURE 58 Structure of a Base Material Specimen Extended 43% at 300°C and at 0.75/minute.
(200X)

125

FIGURE 59 Structure of a Base Material Specimen Extended 47% at 600°C and at 0.75/minute. (200X)

C. Subgrains

Upon first examination of photomicrographs of hot deformed and quenched uranium, the reader may have difficulty in distinguishing between small new grains and subgrains, for the two are often not greatly different in size. The distinction is basically one of origin: subgrains arise in situ by polygonization; whereas new grains are formed by the invasion of a growing crystallite into a pre-existing grain.

The distinguishing characteristics of subgrains are: (1) similarity of orientation among the subgrains making up one old grain, as shown by the passage of thermal twins entirely through the related group of subgrains and as shown by similarity of shade, (2) faintness of their boundaries (owing to orientation similarity) as distinguished from the boundaries of most new grains, and (3) a tendency toward nonequilibrium grain shapes (i.e., grain boundary junction angles nearer 90° than the stable 120°). It is impossible in any one photograph to display the subgrains of all, or even of a majority of the old grains, because the polarizing microscope must be differently adjusted for different orientations of the parent grain. In viewing the photomicrographs, it is most informative to consider the subgrains in any individual grain in which they happen to be clearly resolved and then to assume that the subgrain structure is similar in all other original grains of the same sample.

Grain boundaries between subgrained grains are also to be distinguished from those separating subgrained grains from the shells of new grains. Both kinds of boundaries may be ragged, but the irregularities are different in contour. Parallel sided "teeth," previously referred to as serra-

tions, characterize the mutual boundaries between subgrained grains (for example see Figure 60). (These are not nearly as large or profuse as those observed in alloys of nickel and of aluminum.) Boundaries between old grains and colonies of new grains are made up of outlines of small grains of near equilibrium shape and, thus, are composed of many slightly curved segments joined at angles approaching 120° (see Figure 61).

The formation of subgrain boundaries may be thought of as a "precipitation" of dislocations to form an interface between segments of crystal having slightly different orientation. At high temperature the precipitation probably occurs almost instantly. Subgrains cannot be said to exist, however, until the bits of precipitated boundary have grown laterally to mutual intersection to form a continuous three-dimensional network, enclosing completely and isolating each subgrain. The degree of plastic deformation that must occur before a subgrain boundary network can grow to completion is determined in part by the area of subgrain boundary required to form the network. This, in turn, depends upon the distance between the segments of precipitated boundary that went into the building of the network. Higher temperature and slower deformation might be expected to generate lower degrees of "super-saturation" with respect to dislocations and, therefore, to give a lower nucleation rate and produce more widely spaced subgrain boundaries. In conformity with this expectation, *high temperatures and slow deformations resulted in: (1) coarser subgrains and (2) the appearance of resolved subgrains at smaller total deformation* (see Figures 31, 32, and 33).

Growth of the original grains is most active prior to the completion of the subgrain boundary network. Upon its completion, this network becomes a part of the grain boundary network of the system as a whole, and the original grain boundaries can move only within the restriction that the subgrain boundaries must be carried with them. Accordingly, *subgrains tend to limit grain growth. When new grains appear they are at first innocent of subgraining and their boundaries are free to migrate. Continued plastic deformation causes distortion of each new crystal, however, and subgraining of the new crystal puts a restriction upon its further growth. Accordingly, the size of new grains is established by those conditions that control the evolution of subgraining, namely: temperature, rate of deformation, and alloying.*

The importance of subgrain formation in the present research is, of course, that it is prerequisite to new grain formation and that, correspondingly, new grains make their appearance almost as soon as a subgrain boundary arises from the growth of a subgrain across the grain boundary into one of the three adjoining boundary networks. Adjacent new grains usually have greatly different orientation because few pairs of neighboring new grains arise from the same parent crystal. The new-grain size, of course, reflects the subgrain size as well as its rate of growth into surrounding crystal. Coarse new grains are associated with coarse subgrains (i.e., high temperature, low strain rate, and alloying that promotes sub-boundary formation) and maximum opportunity for growth (i.e., again, high temperature, low strain rate, and also early nucleation).

Alloying may be expected to modify subgrain formation chiefly through the anchoring of dislocations by solute atoms. To overcome such interference, dislocation climb must occur. This is a diffusion process and, as such, is time consuming, although its rate must increase sharply with rising temperature. Thus, *the "precipitation" of subgrain boundaries may be expected to be delayed approximately in proportion to the concentration of the solute element.*

This prediction appears to account for much of the special alloy behavior that is encountered. Aluminum, according to Petzow and Tank,²⁵ having a solubility near 1% at 600°C, promotes vigorous grain growth during hot deformation at temperatures up to nearly 600°C where new grains appear and growth is suppressed. Iron, with its solubility in alpha uranium near 0.1% at 600°C, promotes grain growth at temperatures up to nearly 500°C where, again, "recrystallization" begins. *Silicon not only provides very low solubility, but also seems to drive other elements out of solution in uranium. In doing so, it actually lowers the temperature of the beginning of "recrystallization" and thereby deters grain growth.* In the "V" alloy (399 ppm Si) recrystallization is retarded by new grain formation at temperatures as low as 300°C.

D. Twinning

Although the evidence at hand is too scanty to provide a clear case, it does strongly suggest that mechanical twinning and subgrain network formation are mutually exclusive processes. Mechanical twins are always absent or, at most, confined to a very few grains, when subgrains are in evidence. Correspondingly, mechanical twins are found at high as well as at low temperatures when extensive deformation has not generated new grains (Figure 61). As this photograph shows, however, subgrains may eventually appear in both mechanical twins and matrix, presumably subsequent to twin formation.

Mechanical twins were observed in all alloys tested at 300°C. At this temperature they were only a little broader than the thermally generated twins, but they were often very profuse (Figure 62). With a rise in the testing temperature, the mechanical twins became coarser and fewer (Figure 56). They also occur in a smaller fraction of the grains as temperature increases (Tables X to XIII). The temperature at which twinning disappears seems to be elevated somewhat by alloying, especially by aluminum.

It is concluded, accordingly, that whereas lattice strain is commonly relieved at high temperature by the "precipitation" of subgrain boundary, any factor that prevents or slows this process may permit a higher stress to accumulate in the crystal and that this higher stress may relieve itself by generating and growing mechanical twins. Thus, mechanical twins are typical of structures produced by deformation at lower temperature, at a faster strain rate, and in alloys.

The initial appearance (Figure 48) and subsequent disappearance (Figure 50) of mechanical twins, as a consequence of thermal cycling may perhaps be explained by a total conversion of twinable grains to their twin orientation, so that they become stabilized. Each cycle of cooling would, of course, still produce "thermal twins."

E. Elongation to Fracture

With the possible exception of alloy "V" (399 ppm Si), which is known to display a maximum resistance to swelling during irradiation, each of the alloys made for this research exhibited a ductility minimum at a temperature at or above 300°C (see Figures 34 through 40). A metallographic search has failed to reveal any significant number of intergranular cavities in fractured samples, even close to the rupture itself. In other metals such cavities have always been found

to be associated with a ductility minimum and vice versa.¹⁸ Furthermore, materials that display the intermediate temperature ductility minimum and that are subject to intergranular cavity formation, rupture with almost no necking. The uranium alloys neck at fracture even in the temperature region of low ductility (see Figures 34 and 63). *It appears necessary to conclude that the ductility minimum in uranium is associated with something other than grain boundary shearing, something that occurs just above the recovery temperature and that promotes localized yielding to fracture.* Thus, the range of lower ductility merely reflects a range in which the interrelation between grain growth and recrystallization results in a coarse grain size. This conclusion does not argue against uranium being susceptible to intergranular cavitation under influences other than simple hot tensile extension.

The failure of uranium to develop intergranular cavities during hot extension, as other metals do, may be due to stress relief adjacent to grain boundaries by mechanical twinning. Mechanical twinning is observable in the temperature range within which grain growth is prevalent. Furthermore, it has been found that intergranular cavities develop during thermal cycling only after twinning has ceased to be evident.

E - 35

FIGURE 60 Structure of an "E" Alloy Specimen Extended 39% at 600°C and at 0.75/minute.
(200X)

E - 185

FIGURE 61 Structure of an "E" Alloy Specimen Extended 13% at 500°C and at 0.75/minute.
(200X)

725

FIGURE 62 Structure of a Base Material Specimen Extended 34% at 300°C and at 0.75/minute.
(200X)

815

FIGURE 63 Tensile Specimen Extended at 350°C and at 0.75/minute.

F. Recrystallization After Hot Working

Uranium which has been hot deformed beyond a certain minimum and is then held at an elevated temperature undergoes recrystallization in the conventional sense. New grains grow to impingement, their shapes become equiaxed, and their grain boundaries become smooth (compare Figures 3a and 6a). It is felt that new grains already present along old grain boundaries provide the "nuclei" of recrystallization, because grain growth during annealing starts near old grain boundaries. Presumably, only the newest of the new grains are thus endowed with growth potential because they have not yet developed internal subgrains. Since plastic deformation is not in progress during annealing, the new grains can grow to impingement, in contrast with the growth limitation imposed during hot working.

Both iron and silicon, when added to uranium in impurity amounts, restrain recrystallization during annealing. The time to achieve a certain fraction of consumption of old grains by new subgrain free grains is increased by a factor of 40 by adding either 199 ppm of Si or 287 ppm of Fe. Lesser or greater quantities produce correspondingly lesser or greater effects. It is believed that this restraint is a consequence of grain boundary pinning by small particles of impurity phases.

G. Thermal Cycling

Angerman and co-workers¹⁵ were first to observe that the thermal cycling of uranium leads to intergranular cavity formation. Their findings have been confirmed by the present study, and it has been shown, in addition, that iron confers some resistance to cavitation, while silicon gives

much more protection. This is all consistent with known reactor behavior of uranium alloys. Accordingly, it is of interest to identify those factors that have become operative in thermal cycling and which were not present during hot tensile deformation.

Evident differences in the imposed conditions are: (1) absence of a sustained unidirectional homogeneous stress, (2) cyclic variation in temperature over a broad range, and (3) cyclic stressing in opposite directions between adjacent grains. The latter condition clearly supplies the specific shear stress parallel with certain grain boundaries that would be expected, in the cases of most metals, to induce intergranular shearing and after considerable displacement in one direction, lead to intergranular cavity formation. Continued shearing in one direction is conceivable through the operation of Burke's²⁶ thermal ratcheting mechanism. The temperature interval through which the samples were cycled included essentially the whole range of expected sensitivity to intergranular cracking, as well as that of observed swelling of uranium under irradiation. Thus far, the only factor that appears drastically different from tensile stressing is the local magnitude of the strain in thermal cycling.

A clue to the essential difference between hot tensile straining and thermal cycling may reside in the manner in which mechanical twins appear and disappear. It may be coincidence that cavities appear in unalloyed uranium only after the incidence of mechanical twinning has declined to a low level, and that cavities did not form and twins did not disappear during a like cycling of "P" alloy, containing silicon, while "C" alloy, containing iron, was intermediate in its behavior. It is remarkable also that the base material and the iron alloy, which are prone to twin during tensile straining, become nearly twin-free during thermal cycling, whereas the silicon alloy, which develops few twins during hot tensile straining, becomes heavily twinned during thermal cycling.

Apparently the crystallographic characteristics of the unalloyed uranium are modified progressively by thermal cycling, and this change is resisted by the uranium containing silicon. One possible kind of change is a reorientation of the grains into some kind of preferred orientation. There is a strong tendency for the thermal twins in the base material to gather about one direction and many of these twins seem to run through several grains as though these had a common axis in the habit plane of twinning. These effects are largely absent from the silicon alloy.

Upon this basis it is proposed that thermal cycling induces new grain formation and directional growth of such nature that certain orientation relationships tend to dominate. When this has happened, the local strain among neighboring grains may have been magnified by placing end to end a number of grains that are undergoing the same dimensional change during thermal cycling. Thus, the magnitude of the local strain is increased as cycling continues until local rupture occurs. The silicon alloy is expected to resist this type of structural evolution because its proclivity to develop new grains is greater and these would tend to randomize the orientations present.

H. Alloying Effects

Thus far, the various effects of alloying have been considered piecemeal. It will assist understanding at this point to summarize these effects alloy by alloy.

Silicon appears to occupy the role of a scavenger, converting other impurities to silicides and removing them from solution in uranium as particles of a separate phase. For this reason, silicon provides a maximum resistance to grain growth, wherever this may be accomplished by pinning. And for the same reason, silicon promotes easy polygonization and recrystallization. In hot tensile straining, these characteristics of silicon alloys serve to maintain a homogeneous fine grain size that eliminates the range of lowered ductility. In thermal cycling, silicon is thought also to maintain a fine grain size wherein new grains frequently replace old grains and keep the grain orientation random. Thus, silicon appears as a stabilizer of structure in the presence of deforming influences. Lest it be argued that high-purity uranium would also recrystallize readily, it must be emphasized that the combination of pinning with free recrystallization is the property claimed for the silicon alloy and that pure uranium would provide no pinning constituent.

Iron seems to differ from silicon in that it forms an extensive solid solution in addition to providing second phase particles. This is seen as detracting from its effectiveness in providing pinning of grain boundaries and at the same time establishing a restraint to polygonization. Accordingly, the iron alloys differ from the unalloyed uranium chiefly in that they offer only a modest restraint of grain growth.

Some effects of carbon may be inferred from the known characteristics of uranium containing various concentrations of carbon. Ordinary uranium, which carries an excess above the solubility limit of carbon, resists recrystallization at 300°C, whereas the addition of silicon to the same material lowers the "recrystallization temperature" as observed during deformation. Accordingly, carbon is here regarded as being much like iron in its influence upon the high temperature deformation characteristics of uranium.

Having viewed the effects of aluminum only in a limited degree and at low aluminum concentration, no real appraisal is possible at this time. As of now it appears that aluminum should be quite the opposite of silicon in its effect upon hot deformation behavior. Yet, experience with uranium fuels containing aluminum point toward a behavior similar to that of silicon. Only further research can resolve this paradox.

I. Concerning the Swelling of Uranium During Irradiation

The foregoing studies have perhaps done more to narrow the range of possible explanations of gross swelling of uranium than to pin point its mechanisms. The same is true of the effects of alloying elements upon swelling.

It now appears highly improbable that swelling results from any continuous in-pile deformation of the metals, such as longitudinal growth, because few cavities have been produced in this way. The association of swelling with some unique type of pinning also appears untenable upon the grounds of differences in the structural behavior of U-Si and U-Al alloys. Silicon seems to maintain a fine grain size during deformation, whereas aluminum promotes exaggerated grain growth.

Rather, a reciprocating load many times repeated and stressing the metal by differential dilatation of adjacent grains appears to be requisite to cavity formation. The mechanism of rupture is

most probably by grain boundary shearing, cavity nucleation by hydrostatic tension developed at grain boundary quadruple points, and cavity growth by continued grain boundary shearing.

It is deduced that failure to rupture during simple stressing is the result of the existence of several stress release mechanisms, such as twinning and grain boundary migration, which tend to prevent the localization of stress along grain boundaries. Only when these have been exhausted by repeated functioning can grain boundary shearing be caused to occur.

Aluminum and silicon are alike in the fact that during deformation they promote active grain boundary migration, the one with respect to existing grains and the other with respect mainly to new grain boundaries. Grain boundary mobility has been found to curtail grain boundary shear and to restore good ductility in other metals. This may, therefore, explain in part the beneficial effects of silicon and aluminum. As has been said before, however, the effect of silicon in maintaining a fine grain size provides a most attractive explanation of its role in delaying the swelling of uranium.

V. SUMMARY OF OBSERVATIONS AND CONCLUSIONS

1. *Uranium resembles other metals in most of its high-temperature deformation characteristics.* Grain growth accompanies initial deformation at all temperatures from 300°C up. New grain formation is restrained so long as deformation is in progress and is limited to the maintenance of small grains adjacent to old grain boundaries. There is a ductility minimum between about 300 and 500°C.
2. *The high temperature deformation character of uranium is unique* in the occurrence of mechanical twinning only at lower temperatures and in the failure of uranium to develop intergranular cracks coincident with its intermediate temperature ductility minimum.
3. *Grain growth accompanies deformation* because the dislocations needed to detach the grain boundaries from second phase pins are being supplied and because the grain boundary network is being distorted out of its equilibrium shape. The higher the temperature and the longer the time (slower the strain rate) the more the grain growth. Boundary migration is restrained by increasing the number of "pins" through alloying and by the removal of the driving force that occurs when the development of subgrains diminishes the lattice bending strain and ties down the network at more quadruple points.
4. *Anisotropic grain growth is possible during deformation* because "unpinning" occurs more readily upon boundaries that lie normal to the elongation of the metal.
5. *New grains develop adjacent to old grain boundaries only after subgrains have formed.* They are believed to result from the growth advantage possessed by those subgrains which, being next to grain boundaries, may be larger than certain of their neighbors in adjacent old grains.
6. *New grains maintain a steady size which is larger at higher temperatures and slower deformation rates.* The size limitation is imposed by continued deformation, which presently distorts the new grains, causing them, in their turn, to develop a new generation of subgrains, thereby destroying their growth advantage with respect to their neighbors.

7. *New grain formation during deformation is promoted by alloying which lowers the recovery temperature of alpha uranium (silicon, for example). This effect is associated with the formation of a subgrain boundary network at a lower temperature. This formation is more rapid when dislocation climb is made easier.*

8. *Subgrains, defined by a complete subgrain boundary network, form after the introduction of a certain minimum of deformation needed to supply the dislocations requisite to subgrain boundary formation. Their size is greater at higher temperatures and slower strain rates. Often their size is defined by the distance between adjacent planes of viscous shear that produce grain boundary serrations. When newly formed, their boundaries often meet at 90° and 180° angles.*

9. *Annealing after hot working results in total recrystallization just as does annealing after cold working. The newest generation of new grains grow to mutual impingement, because, in the absence of deformation, they retain their growth advantage over their neighbors. Second phase pinning produces a finer recrystallized grain size by slowing grain growth and thus permits more of the new grains to compete in growth.*

10. *Twinning occurs at a critical resolved shear stress that is so low in the temperature range 300 to 500°C that cross slip and the resulting grain boundary shearing may fail to develop. Thus the occurrence of twinning is associated with the nonformation of intergranular cracks during hot tensile straining. Alloying which hardens the alpha uranium tends to promote twinning with increasing temperatures.*

11. *Twinning and subgrain boundary formation appear to be mutually exclusive processes. No twins are found in metal which has previously developed subgrains. Twins themselves, during subsequent deformation, may become subgrained.*

12. *Thermal cycling produces twins which disappear as cycling continues. Possibly the grains become so reoriented by twinning that further twin-forming stresses cannot occur.*

13. *The tensile ductility minimum at intermediate temperatures occurs without the usual intergranular cracking. Instead of a "brittle fracture," failure is associated with a highly ductile type of fracture. Apparently, the low ductility is to be associated with the coarse grain size that is produced by deformation in the 300 to 500°C temperature range. Alloying with silicon maintains a fine grain size and removes the ductility minimum.*

14. *Intergranular cracks develop during thermal cycling after twins have disappeared and when the grain size is coarse. Twinning is presumed to relieve stress before cross slip and grain boundary shearing can occur, thus preventing intergranular cracking. A fine grain size is always associated with a low cracking tendency, because little shearing stress can build up between quadruple points where these are close together.*

15. *Silicon maintains a fine grain size during tensile straining and thermal cycling, because it promotes new grain formation by "purifying" the alpha-uranium phase and lowering the recovery temperature; it simultaneously restrains grain growth by supplying U_3Si pins.*

16. *The swelling of uranium during neutron irradiation is believed to be associated with the type of intergranular crack formation that is developed by thermal cycling. Cavity formation during irradiation is much more severe than during thermal cycling because of the more frequent and more intense local straining associated with thermal spikes.*

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APPENDIX – ADJUSTMENT OF THE QUANTITATIVE METALLOGRAPHIC PARAMETERS

A. Grain Boundary Area (S_V)

The observed wide variation in grain size among specimens of the "V" alloy in the as-annealed condition ("a" specimens) made it impossible to show any reasonable trends in the structural evaluation during hot deformation by plotting grain boundary area (S_V) versus total extension. Thus, in order to obtain a more reliable relation between grain boundary area and total extension, the percent difference in grain boundary area between the hot deformed condition ("s" specimen), and the as-annealed condition (corresponding "a" specimen) was plotted versus total extension. The percent difference in grain boundary area was calculated from the following relation:

$$\text{Percent difference in } S_V = \frac{S_{V(s)} - S_{V(a)}}{S_{V(a)}} \times 100 \quad (\text{A-1})$$

where,

$S_{V(s)}$ = Grain boundary area of "s" specimen.

$S_{V(a)}$ = Grain boundary area of corresponding "a" specimen.

B. Grain Boundary Anisotropy (e_g)

A qualitative examination was made of the relation between grain boundary anisotropy obtained from "s" specimens and the grain sizes of the corresponding "a" specimens. From this examination it seemed that the larger grained specimens produced abnormally large values for grain boundary anisotropy. An adjustment of the grain boundary anisotropy values for the "s" specimens was made assuming that there existed a linear relation between grain size and grain boundary anisotropy. In order to adjust the value of grain boundary anisotropy (e_g), as determined from the measurements of hot deformed specimens ("s" specimens), it was necessary to obtain an adjustment factor, k , from the N_L measurements of the "a" specimens as follows:

$$k = \frac{N_L(a)}{N_L(a)_r} \quad (\text{A-2})$$

where,

k = Adjustment factor.

$N_L(a)$ = N_L value for any specimen "a" specimen.

$N_L(a)_r$ = Reference value of N_L arbitrarily chosen from one "a" specimen. In this work

$N_L(a)_r = 39$.

The adjusted value of $e_g(\%)$ is given by the equation:

$$e_g(\text{adjusted}) = \frac{\Delta e_g}{k} + e_g(a) \quad (\text{A}-3)$$

where,

$$\Delta e_g = e_g(s) - e_g(a)$$

$e_g(s)$ = Grain boundary anisotropy for "s" specimen.

$e_g(a)$ = Grain boundary anisotropy for corresponding "a" specimen.

k = Adjustment factor corresponding to particular "a" and "s" specimens.

C. Subgrain Boundary Area ($S_{V(SB)}$)

An apparent grain size effect was observed in the values of subgrain boundary area of the "s" specimens. The subgrain boundary area values seemed to be low for the large grained specimens. As in the case for grain boundary anisotropy, a linear relation between grain size and subgrain boundary area was assumed such that an adjusted subgrain boundary area was determined as follows:

$$S_{V(SB)} (\text{adjusted}) = k S_{V(SB)} \quad (\text{A}-4)$$

where,

k = The adjustment factor defined in Eq. A-2.

$S_{V(SB)}$ = Subgrain boundary area determined from "s" specimens.