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PHYSICAL AND MECHANICAL METALLURGY STUDIES ON DELTA STABILIZED PLUTONIUM-GALLIUM ALLOYS

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PHYSICAL AND MECHANICAL METALLURGY STUDIES ON DELTA STABILIZED PLUTONIUM-GALLIUM ALLOYS

By

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April 1965

BATTELLE-NORTHWEST
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PHYSICAL AND MECHANICAL METALLURGY STUDIES ON DELTA STABILIZED PLUTONIUM-GALLIUM ALLOYS

INTRODUCTION

In unalloyed plutonium, the delta phase region extends from 319 to 451 C. As indicated in the Pu-Ga phase diagram, Figure 1,[1] the addition of appropriate quantities of gallium stabilizes the delta phase over a much wider range of temperatures including ambient and lower temperatures.

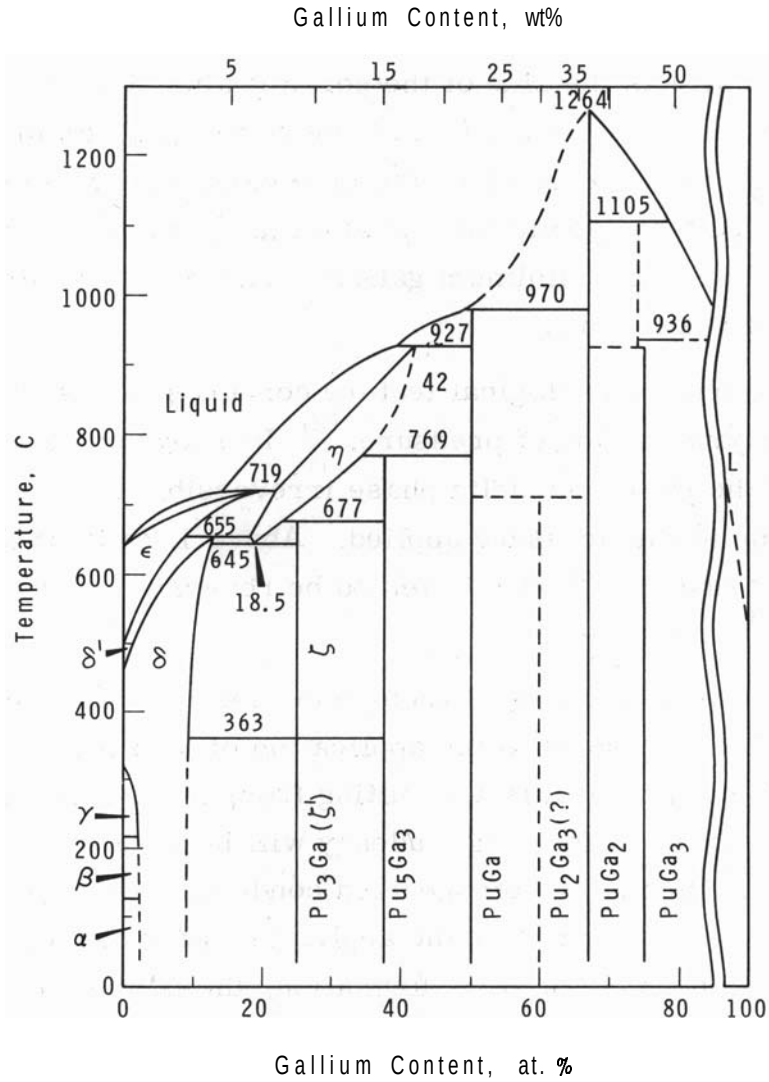


FIGURE 1

Plutonium-Gallium Phase Diagram[1]

The metallurgy of delta stabilized Pu-Ga alloys is complicated by two phenomena. During nonequilibrium cooling through the liquid plus epsilon and epsilon plus delta regions, coring, a type of segregation occurs. This results in a variation in gallium content within each grain, the central portion having a much higher gallium concentration than near the grain boundary. Using microprobe analysis, Johnson[2] has determined that the cored grains in a cast Pu–1.0 wt% Ga alloy range from 0.1 wt% gallium near grain boundaries to 1.5 wt% gallium at the center. Coring can be removed by an appropriate anneal in the 400 to 500 C temperature range. Diffusion occurs and a uniform distribution of gallium results. Since cooling rate during and after solidification affects the size of the cored grains, the rate of diffusion, i.e., homogenization will be dependent on the cored grain diameter. As cored grain diameter decreases, the rate of homogenization should increase. Here it was assumed that despite the variation in cored grain diameter obtained, the maximum and minimum gallium concentration within each grain was independent of grain size.

The second phenomenological feature concerns metastability with with respect to the application of pressure.[3] In alloys containing less than 1.2 wt% gallium, the stabilized delta phase irreversibly transforms to alpha phase in proportion to the pressure applied. Above 1.2 wt% gallium, the formation of alpha phase under pressure is felt to be reversible and is undoubtedly controlled by reaction kinetics.

The Pu–1.0 wt% Ga alloy, therefore even when completely homogenized will be metastable with respect to the application of pressure. Because of the 0.1 to 1.5 wt% gallium gradient resulting from coring, the degree of pressure metastability or alpha forming tendency will be greater. A test for proximity to an equilibrium or homogenized condition would then be the determination of density before and after the application of pressure. The greater the density increase because of alpha formation, the farther the alloy is from the equilibrium state. As the equilibrium or homogenized condition is approached, the density change should decrease and level off at a constant value.

The objective of this study was to obtain a fundamental understanding of the influence of the indicated metallurgical phenomena on the physical and mechanical metallurgy of delta stabilized Pu-Ga alloys. Studies initiated include: resolution of the relative contribution of liquid plus epsilon and epsilon plus delta coring to the resultant gallium concentration gradient as indicated by response to heat treatment, cooling rate and homogenization anneals; the effect of pressure and/or mechanical deformation and heat treatment on the delta ⇄ alpha transformation; the effect of gallium content on selected physical and mechanical properties; and determination of the recrystallization kinetics of cold rolled alloy.

SUMMARY AND CONCLUSIONS

Studies of coring and homogenization in the Pu–1.0 wt% Ga delta stabilized alloy indicated that: (a) the coring which occurs in the epsilon plus delta region is controlling for homogenization of the alloy and (b) the rate of homogenization is not only temperature dependent but an inverse function of cored grain size which in turn is controlled by cooling rate through the epsilon plus delta region. X-ray line broadening and density-compression-density techniques were developed to permit estimation of state of homogenization.

In a study of the metastability of the alloy with respect to pressure and subsequent annealing, it was found that (a) alpha plus delta phase mixtures in cored alloy were unstable during room temperature storage after annealing at temperatures below 280 C, while (b) alpha plus delta phase mixtures in homogenized alloy were stable under the same conditions.

For gallium compositions ranging from 0, 37 to 1. 26 wt% in homogenized alloy, lattice parameter, density, and pressure metastability decreased while hardness increased as gallium content increased.

On 94% cold-rolled alloy, recrystallization occurred at 250 C. Possible stress relief and age hardening effects were noted. In addition, during extended anneals grain coarsening and grain boundary void formation were observed.

During tensile and compressive testing at temperatures from -32 to 500 C it was determined that strength decreased uniformly with temperature

while ductility in general increased for low iron alloy. As iron was increased, a hot short behavior was noted leading to a loss of strength and ductility. Testing speed up to 1.0 in./min at temperatures from -32 to 300 C had virtually no effect on strength or ductility.

Strain hardening occurred in homogenized, cold-rolled alloy while the alpha phase formation in cored, cold-rolled alloy gave strengthening over and above that from strain hardening. The alpha phase formation also increased the modulus of elasticity.

Several metallurgical conditions of alloy sheet were fatigue tested in cyclic flexure at room temperature. As the 0.2% yield strength increased from 11,000 to 32,000 psi, fatigue strength increased from 10,000 to 15,000 psi. The cored, as-cast, and homogenized, 94% wrought, 300 C annealed conditions had the lowest fatigue strength, while the cored, 86% wrought sheet had the highest fatigue strength.

EXPERIMENTAL PROCEDURE

Material

The Pu-Ga alloy used in this work was prepared from arc-melted Pu–11.1 wt% Ga master alloy. Dilution with unalloyed plutonium to the desired composition was done in either induction or resistance heated vacuum furnaces where it was also cast to shape. This procedure minimized if not prevented the formation of Pu-Ga compounds which tie up gallium thus lowering the effective gallium available for stabilization while not altering the total gallium content. The average and range of impurity content of the alloy is presented in Table I.

TABLE I

AVERAGE CHEMICAL ANALYSIS OF Pu-Ga ALLOY

Element	Analysis, ppm	Element	Analysis, ppm
Aluminum	41	Magnesium	85
Calcium	4	Manganese	34
Chromium	75	Nickel	120
Copper	9	Silicon	41
Iron	106	Carbon	255
		Total	770

Techniques

Metallography - Specimens are mounted in polyester resin in 3/4 in. ID mounts. The mounted specimen is faced flat on a small lathe in increments of 0.010, 0.005, 0.002, 0.002, 0.001, and 0.001 in. in that order to minimize the introduction of surface strain. Rough polishing is accomplished on the 180, 3260, 600 grit cloths on rotating laps. A Syntron vibratory polisher equipped with silk cloth and Linde A is used for 2 hr to produce an intermediate polish. The final polish for as-polished viewing is obtained during a 1 hr period on a Syntron equipped with microcloth and Linde A. Carbon tetrachloride is used as a lubricant in the above operation

X-Ray Line Broadening - The specimen is taken after the intermediate polishing step and electropolished for 100 sec at 20 v and 0.8 - 1.0 amp in a bath of 20 parts tetraphosphoric acid, 30 parts water and 50 parts 2-ethoxyethanol. A Norelco diffractometer using CuK_a radiation and a scanning speed of 0.25°/min is used to obtain the (331) peak on a rotating specimen. After determination of the half-height width, the specimen is electropolished for an additional 20 sec. Again the (331) peak is obtained, half-height width determined, etc. The cycle is repeated until the total electropolishing time reaches 180 to 200 sec. At this point, the half-height width has generally passed through a minimum value and is starting to increase with electropolishing time. The minimum half-height width is taken as the true value of broadening for comparison to other specimens.

CORING AND HOMOGENIZATION

Since coring can take place during cooling through both the liquid plus and epsilon plus delta regions, the question of the relative contribution of each region is of interest. From the phase diagram, Figure 1, the maximum concentration gradient of gallium that can occur in the liquid plus epsilon region is approximately 35% less than that occurring in the epsilon plus delta region. This, coupled with the very rapid homogenization normally taking place in the region of incipient solidification in cored alloys and during subsequent cooling to 640 C, in the Pu–1.0 wt% Ga alloy tends to minimize the gallium concentration gradient. Therefore, it would be expected that the coring taking place in the epsilon plus delta region would have the major contribution to gallium gradient. Nevertheless, to resolve this question,

various combinations of heat treatment and cooling condition can be used to produce cored structures largely characteristic of the individual two-phase regions. The rate of homogenization response of these cored structures to annealing between 400 and 500 C then can be used to determine the contribution of the coring in each region to the resultant structure. It must be kept in mind, however, that cored grain size is itself a variable that will affect rate of homogenization determinations.

Preliminary Studies

State of Homogenization Determinations - Two methods which can be used to indicate state of homogenization are alpha⇄delta pressure metastability or tendency to form alpha phase during compression, and decrease in X-ray line breadth. To establish a base point for estimation of state of homogenization in Pu–1.0 wt% Ga alloy using alpha phase formation tendency, 1/8 x 3/4 x 7 3/4 in. sheet was cast. The melt was poured at 885 C into a mold at 390 C followed by furnace cooling to room temperature, heat 23-1-7, Figure 2a. An indication of the alpha forming tendency was obtained by both compressing portions of the sheet with 150,000 psi and rolling to 94% reduction in thickness. During compression the density increased from 15.74 to 16.10 g/cc indicating that approximately 10% alpha phase had formed. The effect of percent reduction during rolling on density is illustrated in Figure 3. It is apparent that appreciable alpha formation does not occur until unit roll pressures reach a certain level, roll pressure increasing as sheet thickness decreases.[4] King[5] has also studied the effect of compression and rolling on alpha forming tendency in the Pu–1.0 wt% Ga alloy, Figure 3. An almost linear relationship between reduction in thickness during compression and density increase was obtained. The relationship between reduction during rolling and density was similar to the author's except that the alpha forming tendency of King's material was higher, possibly because of the larger cored grain size which apparently promotes alpha phase formation under pressure.

The effect of isostatic pressure on alpha forming tendency in cored, Pu–1.0 wt% Ga alloy has been studied by King[5], Figure 4. It was determined that no measurable amount of alpha formation occurred until the pressure exceeded 25,000 psi. Above this value, density increases in a linear manner

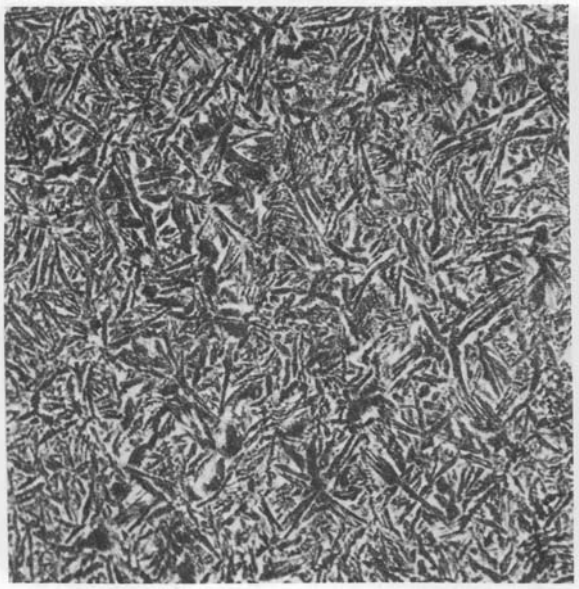


FIGURE 2a

Heat 23-1-7. As-Cast, no Homogenization. Average width of Acicular Cored Grains 0.0068 mm As-Polished. 150 X

FIGURE 2b

Heat 23-1-7. Typical Area After 24 hr Anneal at 425 C and Furnace Cool. As-Polished. 150 X

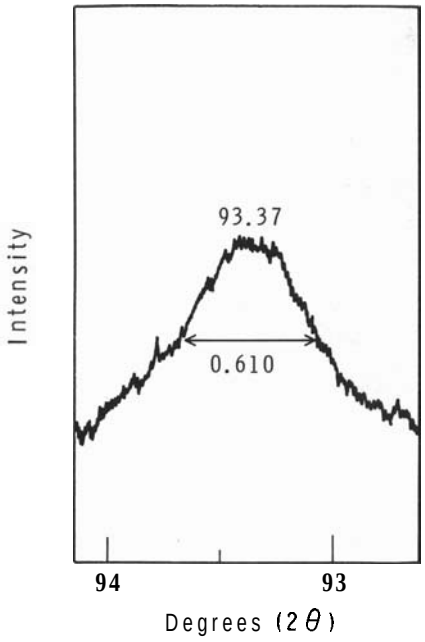


FIGURE 2c

X-Ray Diffraction Peak, (331) Plane, for Above Structure

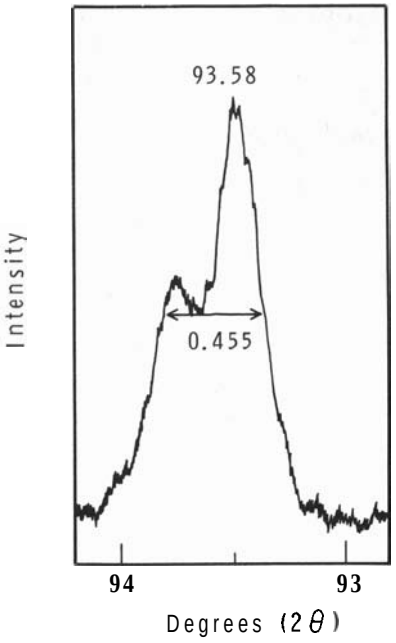


FIGURE 2d

X-Ray Diffraction Peak, (331) Plane for Above Structure

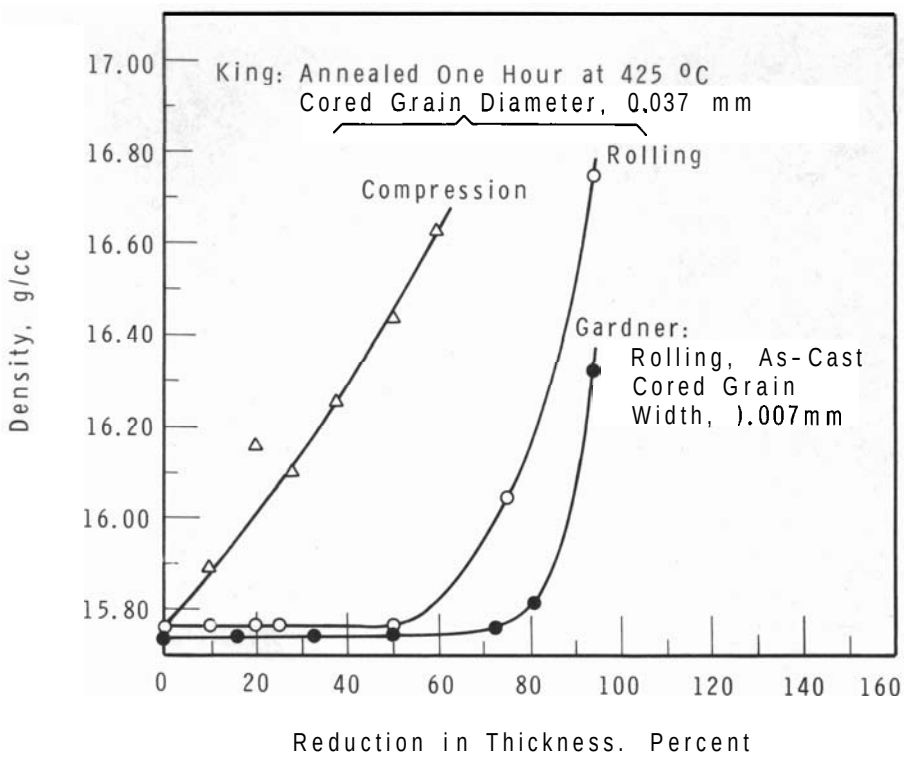


FIGURE 3

Effect of Compression and Rolling on Density of a Cored Pu-1.0 wt% Ga Alloy
Initial Sheet Thickness: 0.130 in.

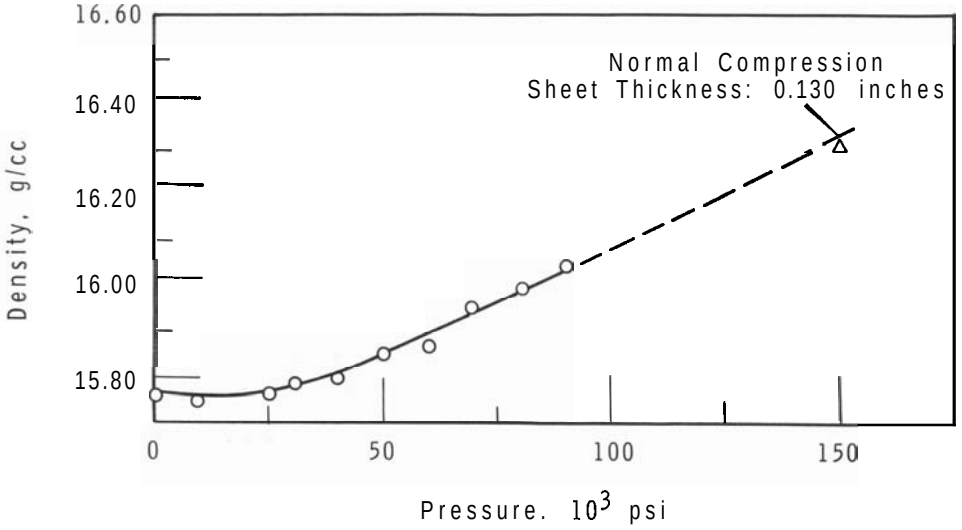


FIGURE 4

Effect of Isostatic Pressure and Compression on Density of a Cored Pu-1.0 wt% Ga Alloy Heat Treated for 1 hr at 425 C
Original Cored Grain Diameter: 0.037 mm

with increasing pressure. It is interesting to note that the application of 150, 000 psi in uniaxial compression produces essentially the same density increase as would be expected from a 150,000 psi isostatic pressure based on linear extrapolation from the 90, 000 psi isostatic result.

After consideration of the above data, it was decided to use the 150, 000 psi compression rather than a particular reduction during rolling to obtain an estimate of state of homogenization. The greater the density increase after the compression, the greater the departure from a homogenized or equilibrium condition.

Use of X-ray line breadth for rate of homogenization studies is based on the fact that line breadth decreases as homogenization occurs. In an experimental demonstration, portions of Heat 23-1-7 were annealed at 425 C for times up to 975 hr and the line breadth determined, Table II.

TABLE II

EFFECT OF ANNEAL TIME AT 425 C ON X-RAY LINE BROADENING OF (331) PEAK, HEAT 23-1-7

Anneal Time, hr	Half-Height Line Breadth (2θ)	Resolution of K_{α_1}, K_{α_2} Doublet
0	0.610	No
1	0.580	No
2	0.565	No
6.67	0.530	No
24	0.480	Yes
48	0.490	Yes
200	0.480	Yes
975	0.490	Yes

As expected, line breadth decreases with increasing annealing time. A typical broadened X-ray peak for the as-cast material is illustrated in Figure 2c. After the 24 hr anneal, the K_{α_1}, K_{α_2} doublet is resolved indicating an equilibrium or near equilibrium condition, Figure 2d. Therefore, the time required for complete homogenization can be taken at the point in the anneal where resolution of K_{α_1}, K_{α_2} doublet first becomes apparent, It must be emphasized that at this point the X-ray line broadening technique may become limiting in this application. That is, although the data indicate complete homogenization, it may be somewhat less than

complete. Despite this possible limitation, the line broadening technique does yield a reproducible end point for study of the process of homogenization and was therefore of great value in this work. An important supplement to the line broadening data was the microstructural evidence of state of homogenization. Note the coring in the as-cast condition, Figure 2a; and absence of coring in the homogenized microstructure, Figure 2b.

Effect of Anneal Temperature - Since rate of homogenization will be dependent on anneal temperature it was decided to determine the optimum anneal temperature for use in subsequent work. Portions of Heat 23-1-7 were annealed for 1, 2, and 24 hr intervals at temperatures between 400 and 610 C. X-ray line broadening and density increase after a 150,000 psi compression were used to evaluate the degree of homogenization achieved during the anneals. The data obtained are illustrated in Figure 5 and Table III.

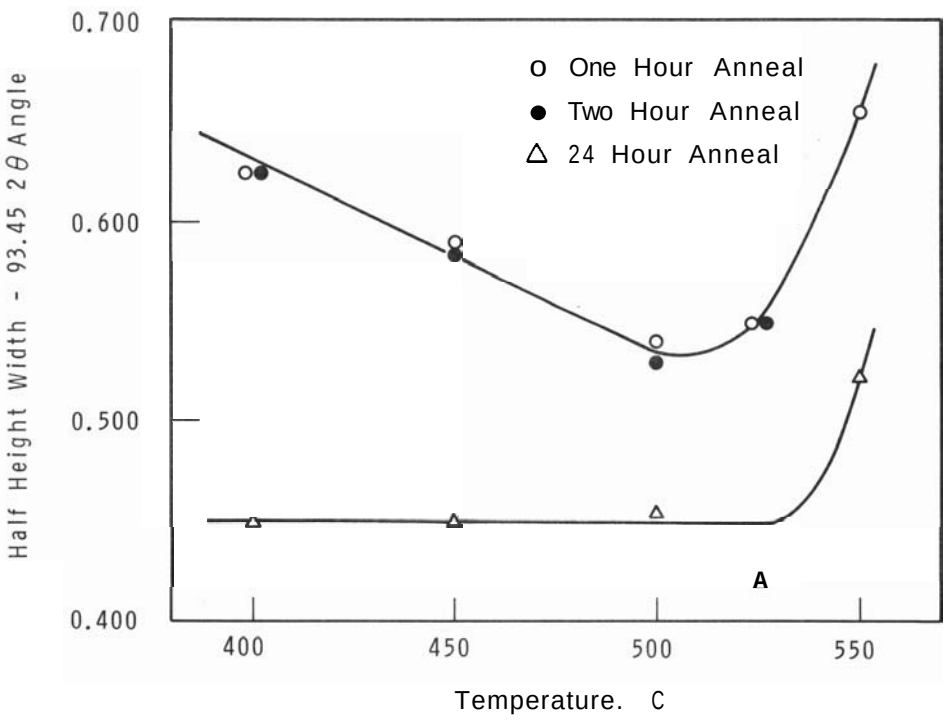


FIGURE 5

Effect of Anneal Time and Temperature on X-Ray Line Broadening in an As-Cast, Nonhomogenized Pu-1.0 wt% Ga Delta Stabilized Alloy, Heat 23-1-7 As-Cast Half-Height Width: 0.610

TABLE III

EFFECT OF TEMPERATURE FOR A 24 HOUR ANNEAL ON BOTH DENSITY INCREASE AFTER A 150,000 psi COMPRESSION AND X-RAY LINE BROADENING, HEAT 23-1-7

Sample Number	Anneal Temperature, C	After Anneal Density, g/cc	Density After 150,000 psi, g/cc	Thickness Reduction During Compression, %	Half-Height Width (331) Peak, °2θ
As-Cast	---	15.74	16.10	52.1	0.610
2411	400	15.74	15.85	52.0	0.440
2412	450	15.74	15.77	51.0	0.450
2418	500	15.74	15.87	51.0	0.455
2410	525	15.74	15.93	53.7	0.420
2419	550	15.75	16.01	48.0	0.525
3180	570	-----	-----	----	0.805
3159	610	-----	-----	----	0.790

As temperature is increased for the 1 and 2 hr anneals, the decrease in line broadening indicates that the rate of homogenization is increased, as it should, considering its diffusion dependent nature. After a 2 hr anneal at both 500 C and 525 C, homogenization is 53% complete compared to the 24 hr anneal data and an as-cast half-height width of 0.610.

For the 400 to 550 C, 24 hr anneals, the density data after the 150,000 psi compression indicate that a 450 C anneal for 24 hr provides the best homogenization. Microstructures after the 400 and 450 C anneals are identical to that for the 425 C anneal illustrated in Figure 2b. The X-ray data do not indicate any appreciable difference in degree of homogenization between the anneals at 525 C and lower temperatures. The 550 C anneal temperature is in the epsilon plus delta phase region where coring occurs during cooling and no homogenization should be expected. Density and X-ray data bear this out and a cored microstructure was observed, Figure 6.

During metallographic examination of the 500 and 525 C annealed specimens, a two-phase structure was evident, Figure 7. It is suggested that this indicates the existence of a delta plus delta-prime phase region in the equilibrium diagram as illustrated schematically in Figure 8. Coring should occur in this phase region and the density data after compression confirm this observation. The coring, however, is apparently not of sufficient magnitude to be detected by X-ray line broadening techniques.

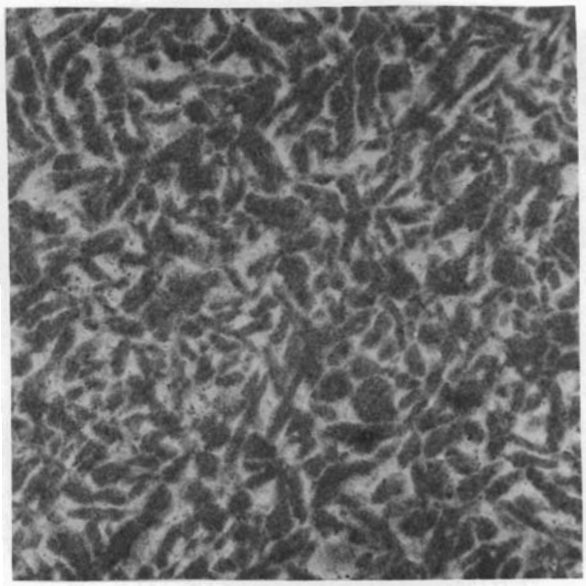


FIGURE 6

Heat 23-1-7. Annealed for 24 hr at 550 C and Furnace Cooled As-Polished, 150 X

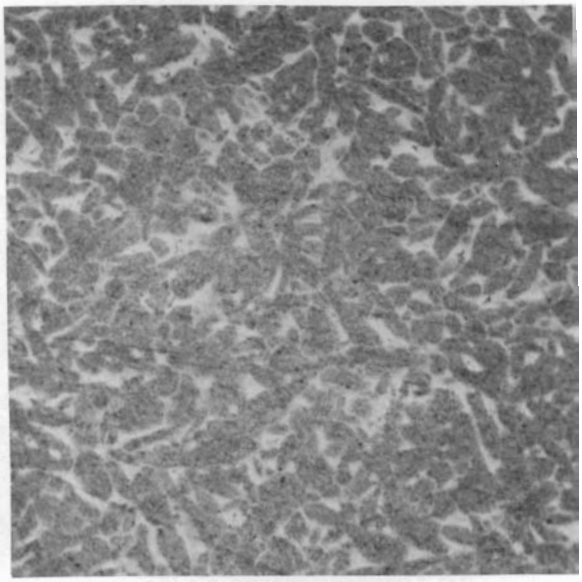


FIGURE 7

Heat 23-1-7. Typical Structure After 500 or 525 C Anneal for 24hr and Furnace Cooled. As-Polished, 150 X

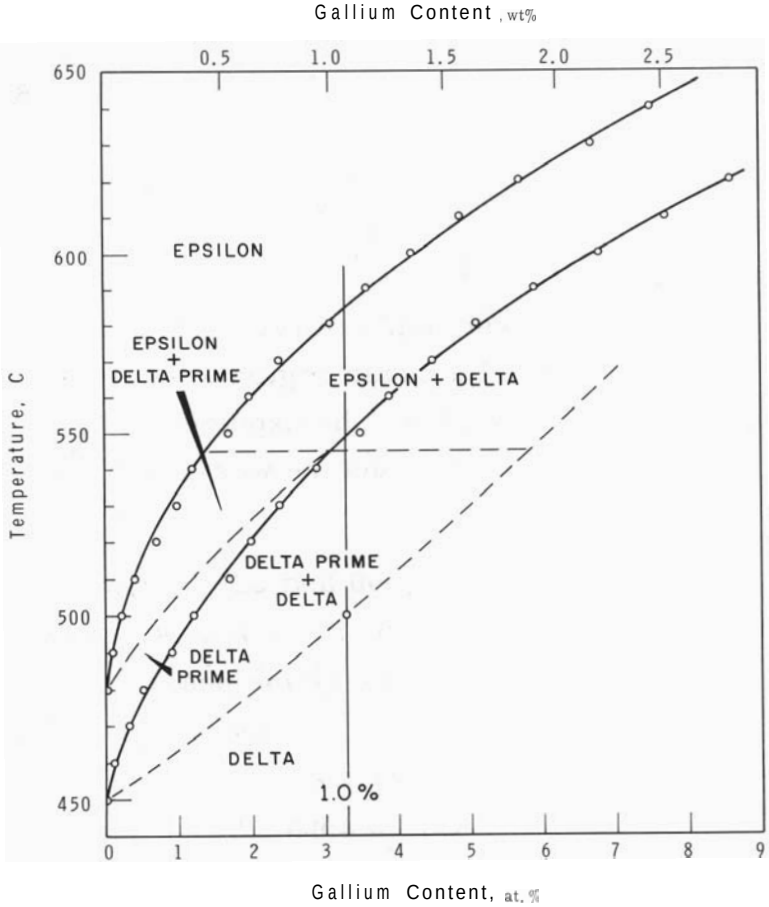


FIGURE 8

Possible Modification to Portion of Pu-Ga Equilibrium Diagram

At the 570 C anneal temperature, cored grains of two general sizes were observed, Figure 9. This is considered to be typical of an anneal in the two-phase epsilon plus delta region followed by furnace cooling. During an anneal at 610 C and furnace cool, the structure illustrated in Figure 10 was obtained. It is considered to be typical of an anneal in the epsilon phase.

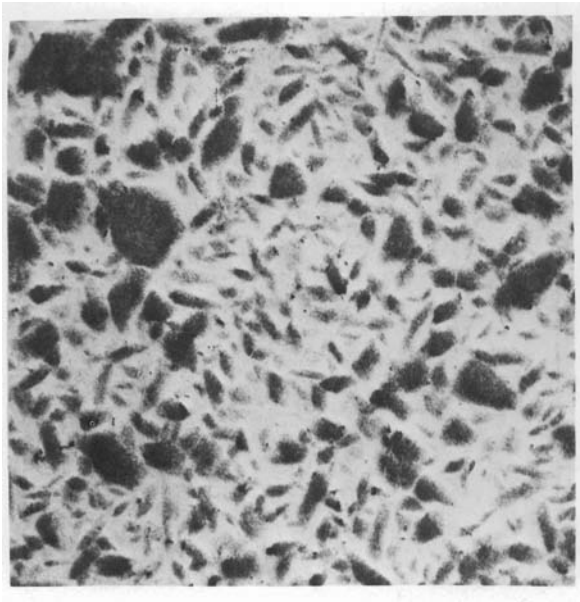


FIGURE 9

Heat 23-10-11. Annealed for 20 hr at 570 C and Furnace Cooled As-Polished, 150 X

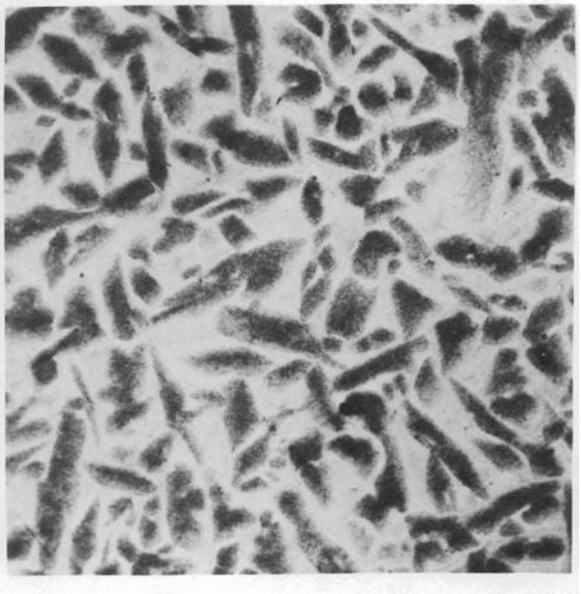


FIGURE 10

Heat 23-10-11. Annealed for 20 hr at 610 C and Furnace Cooled As-Polished, 150 X

Liquid Plus Epsilon and Epsilon Plus Delta Coring

To isolate the effect of coring in each of the two-phase regions, alloy was melted in MgO crucibles to form discs 1 1/2 in. in diameter by 1. 4 in. thick which were then subjected to a variety of cooling sequences. Cooling rate in both two-phase regions was varied by in vacuo oil quenching, furnace cooling, program cooling, or a combination of these. Rate of homogenization determinations, using X-ray line broadening and metallographic techniques, were made on portions of the cooled discs which had been annealed at 450 C for various times. Typical broadened X-ray diffraction patterns for the range of experimental conditions produced in

this study are illustrated in Figures 11a and llb. A typical resolved doublet pattern indicating near or complete homogenization was illustrated earlier in Figure 2d.

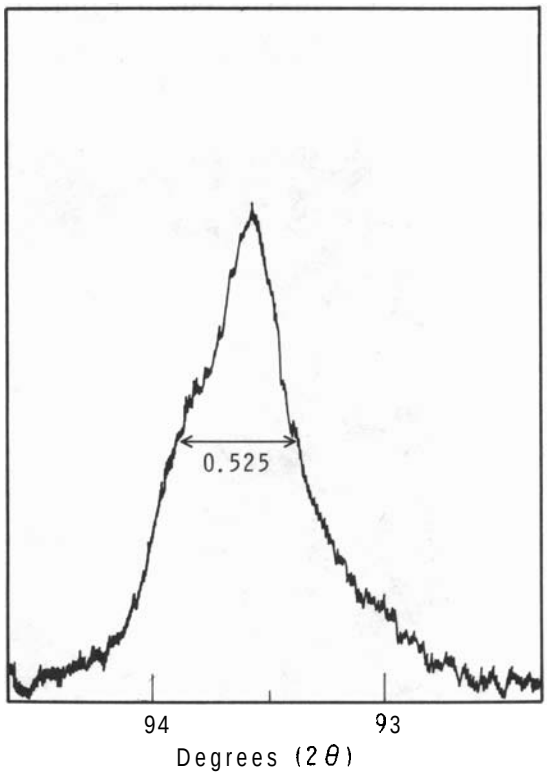


FIGURE 11a

Typical Broadened (331) Peaks for As-Cooled Conditions Illustrated in Figures 12, 15, 16a, 16b, and 17

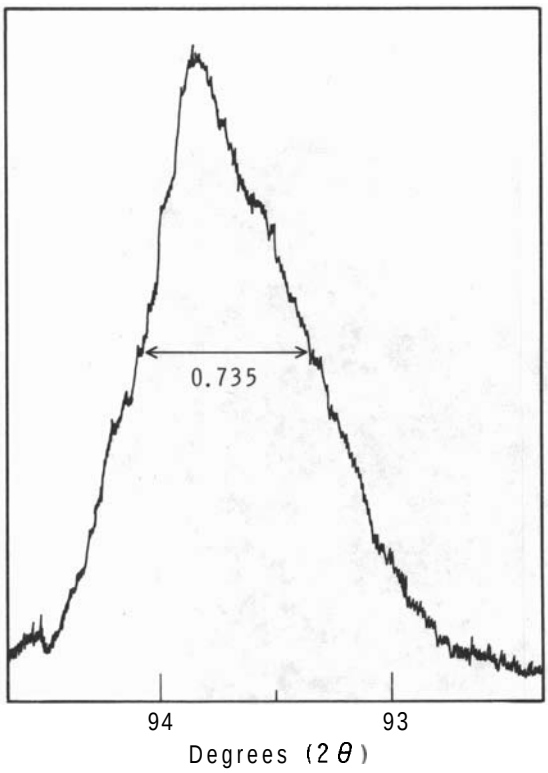


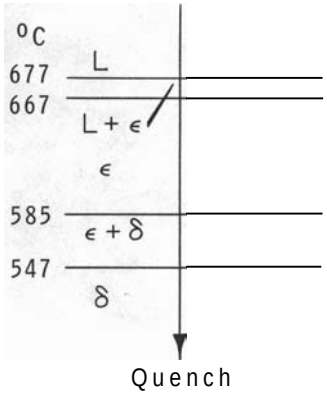
FIGURE 11b

Typical Broadened (331) Peaks for As-Cooled Conditions Illustrated in Figures 13, 14, 18, 19, 20, 21, and 25

To isolate the effect of coring in the liquid plus epsilon region, a specimen was quenched from the melt into oil, Figure 12. Homogenization was very rapid after this treatment, occurring in approximately 1 hr during a 450 C anneal. The quenched specimen was then heated to 640 C and cooled at 1 C/min through the epsilon plus delta region to 515 C followed by furnace cooling, Figure 13. The 0. 10 mm diameter-cored grains obtained in this cooling sequence require an anneal of 215 hr at 450 C for complete homogenization. It is interesting to note that the same result was obtained by a 1 C/min cool through both liquid plus epsilon plus delta regions,

Figure 14. This indicates that the coring occurring in the liquid plus epsilon region probably homogenizes rapidly and does not have a compounding effect on the epsilon plus delta coring.

Average Width of Acicular Cored Grains, 0.002 mm. As Polished, 150X



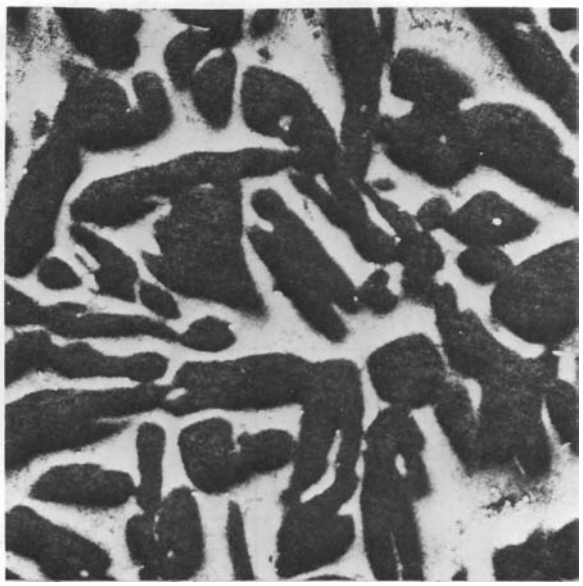
Cooling Schedule

Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

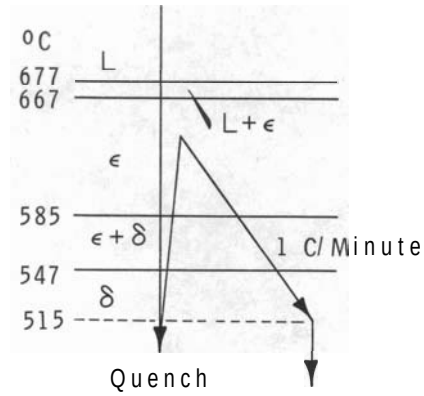
Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of Ka_1, Ka_2 Doublet
0	0.490	No
1	0.485	Yes
3	0.480	Yes

FIGURE 12

Pu–1.0% Ga. Alloy Oil-Quenched from Melt at 750 C to Ambient



Average Width of Acicular
Cored Grains, 0.100 mm
As Polished, 150X



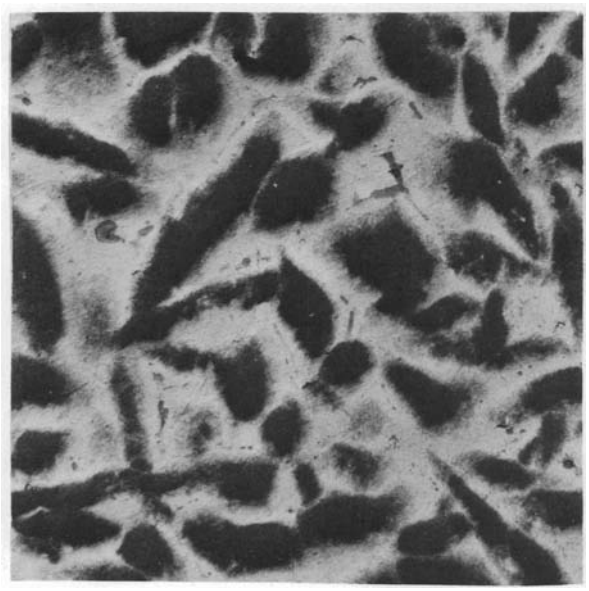
Cooling Schedule

Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

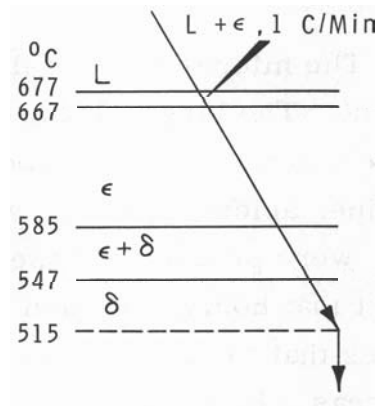
Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha_2$ Doublet
0	0.735	No
1	0.740	No
3	0.720	No
7	0.655	No
24	0.585	No
44	0.550	No
56	0.545	No
62	0.555	No
72	0.570	No
82	0.530	No
150	0.495	Yes
215	0.465	Yes, Better Than 150 Hrs.

FIGURE 13

Pu–1.0 wt% Ga Alloy, Oil Quenched from Melt at 750 C to Ambient. Reheated to 640 C, Cooled at 1 C/min to 515 C and then Furnace Cooled to Ambient



Average Width of Acicular
Cored Grains, 0.100 mm.
As Polished, 150X



Furnace Cool
To Room Temp.

Cooling Schedule

Effect of Anneal Time at 450 C on X-Ray
Line Broadening of (331) Peak

Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha_2$ Doublet
0	0.790	No
1	0.805	No
3	0.810	No
7	0.755	No
24	0.620	No
44	0.545	No
56	0.545	No
62	0.555	No
72	0.575	No
82	0.555	No
150	0.500	Trace
215	0.465	Yes

FIGURE 14

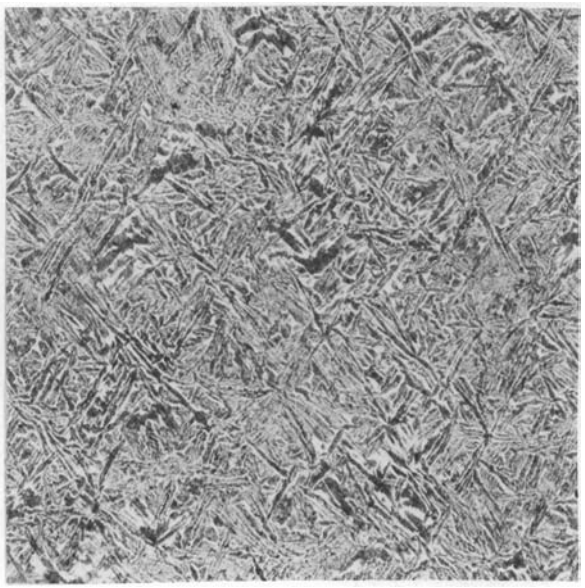
Pu–1.0 wt% Ga Alloy, Cooled at 1 C/min from Melt at 750 C to 515 C and Then Furnace Cooled to Ambient

To isolate the effect of coring in the epsilon plus delta region, specimens which had been cooled at rates of 1 C/min and 15 C/min through the liquid plus epsilon region to 640 C were either oil quenched immediately or held at 640 C for periods of 15 min and 4 hr before oil quenching, Figure 15, 16a, 16b, and 17. During a 450 C anneal, it was determined that all specimens were completely homogenized in 1 hr, indicating a very rapid rate of homogenization.

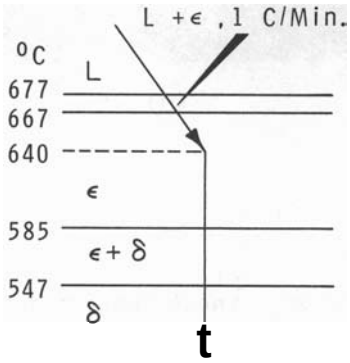
The microstructure illustrated in Figure 16 is worthy of additional comment. The large, dark, dendritic-appearing grains in Figure 16a are the result of the 15 C/min cool through the liquid plus epsilon region. The much finer acicular grains, which also appear within the liquid plus epsilon grains, were produced by the quench through the epsilon plus delta region. The fact that homogenization of this duplex structure occurs rapidly at 450 C indicates that the gallium concentration gradient is minimal within the large, dark areas. Furthermore, as illustrated in Figure 16b, the large, dark dendritic areas dissappear during a 15 min anneal at 640 C. These data support the original contention that the liquid plus epsilon coring homogenizes rapidly during cooling through the liquid plus epsilon and epsilon regions.

Further evidence for a rapid rate of homogenization in the epsilon phase is provided by the treatments illustrated in Figures 18, 19, and 20. No increase in rate of homogenization is gained by holding for 1.5 and 4 hr at 640 C, Figures 18 and 19, compared to an uninterrupted furnace cool from the melt to room temperature, Figure 20.

It can be concluded that epsilon plus delta coring produces the major effect on gallium concentration gradient. Furthermore, cooling rate within this region while it may not affect the overall gallium concentration gradient certainly does affect grain size and hence rate of homogenization. The slower the cooling rate, the larger the grain size and the lower the rate of homogenization. Using the structures in Figures 12, 13, 14, 18, 19, and 21, a relationship was established between grain diameter or grain width, for acicular grains, and the reciprocal of the square root of the cooling rate, Figure 22. Cooling rate was determined for the 540 to 590 C temperature range in the epsilon plus delta region.



Average Width of Acicular
Cored Grains, 0.002 mm.
As Polished, 150X



Quench

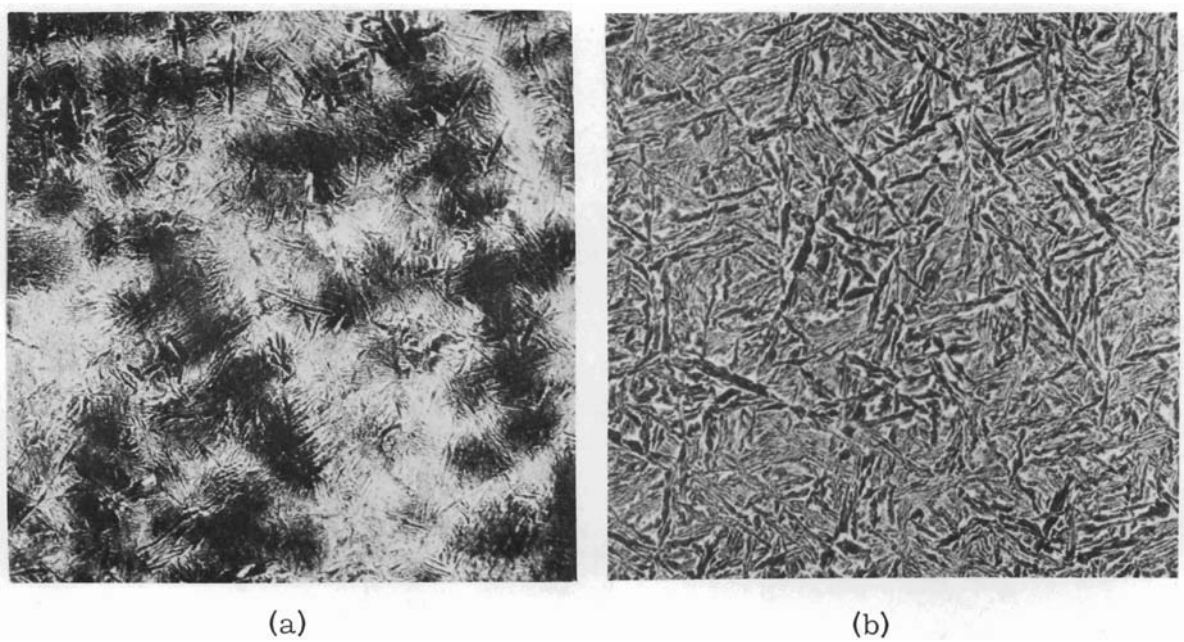
Cooling Schedule

Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha_2$ Doublet
0	0.520	Trace
1	0.460	Yes

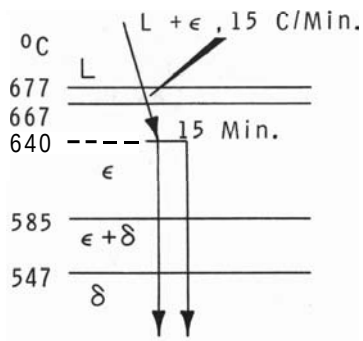
FIGURE 15

Pu-1.0 wt% Ga Alloy Cooled at 1 C/min from Melt at 784 C to 642 C and Oil Quenched to Ambient



Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha$ Doublet
0	0.525	Trace
1	0.505	Yes
3	0.455	Yes
24	0.460	Yes



Cooling Schedule

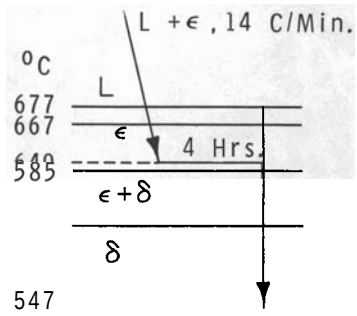
FIGURE 16

Pu–1.0 wt% Ga Alloy, Furnace Cooled from Melt at 800 C to 640 C

(a) Oil Quench from 640 C to Room Temperature. As-Polished, 150 X

(b) Hold at 640 C for 15 min and Oil Quench to Room Temperature, As-Polished, 150 X

Average Width of Acicular
Cored Grains, 0.002 mm.
As Polished. 150X



Quench
Cooling Schedule

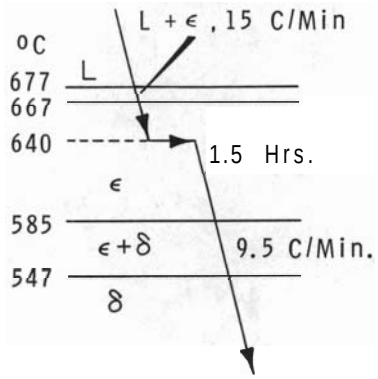
Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha_2$ Doublet
0	0.515	No
1	0.465	Yes
3	0.465	Yes
7	0.450	Yes

FIGURE 17

Pu–1.0 wt% Ga Alloy, Furnace Cooled from Melt at 785 C to 640 C and Hold 4 hr Before Oil Quenching to Ambient

Average Cored Grain
Diameter, 0.034 mm.
As Polished, 150X



Furnace Cool
To Room Temp.

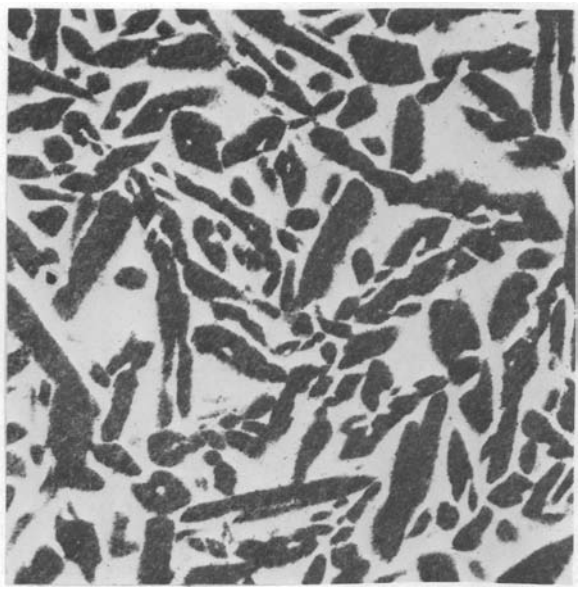
Cooling Schedule

Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

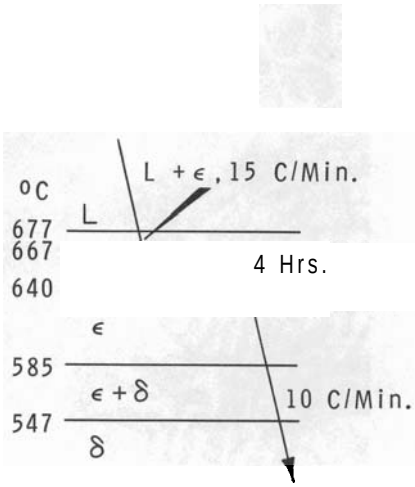
Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha_2$ Doublet
0	0.875	No
1	0.730	No
3	0.700	No
7	0.605	No
24	0.515	Trace
31	0.500	Trace
36	0.460	Yes

FIGURE 18

Pu–1.0 wt% Ga Alloy, Furnace Cooled from Melt at 765 C to 640 C and Held 1.5 hr Followed by Furnace Cooling to Ambient



Average Width of Acicular
Core Grains, 0.038 mm
As Polished, 150X



Furnace Cool
To Room Temp.

Cooling Schedule

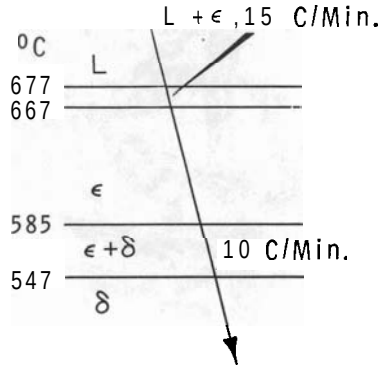
Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha_2$ Doublet
0	0.790	No
1	0.770	--
3	0.705	No
7	0.635	No
24	0.520	Trace
36	0.455	Yes

FIGURE 19

Pu-1.0 wt% Ga Alloy, Furnace Cooled from Melt at 760 C to 635 C and Held 4 hr Followed by Furnace Cooling to Ambient

Average Width of Acicular
Cored Grains, 0.034 mm
As Polished. 150X



Furnace Cool
To Room Temp.

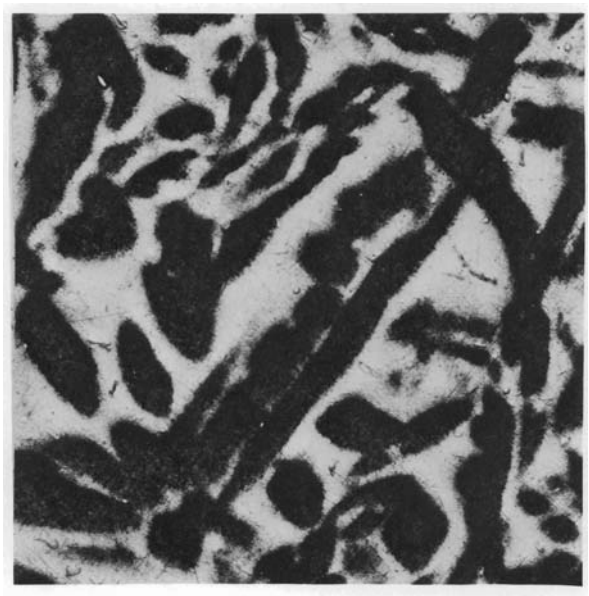
Cooling Schedule

Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

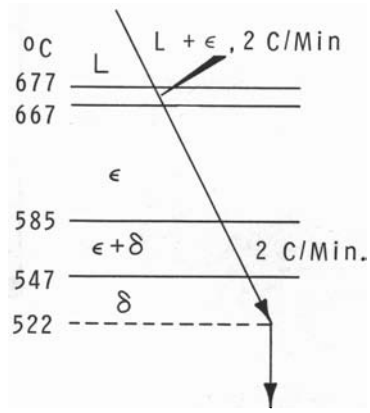
Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha_2$ Doublet
0	0.770	No
1	0.685	No
3	0.630	No
7	0.590	No
28	0.500	Trace
32	0.485	Almost
36	0.460	Yes

FIGURE 20

Pu-1.0 wt% Ga Alloy, Furnace Cooled from Melt at 827 C to Ambient



Average Width of Acicular
Cored Grains, 0.074 mm
As Polished, 150X



Furnace Cool
To Room Temp.

Cooling Schedule

Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

Anneal Time (Hours)	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1$, $K\alpha_2$ Doublet
0	0.715	No
1	0.690	No
3	0.720	No
7	0.680	No
24	0.525	Trace
40	0.480	Trace
44	0.515	Trace
48	0.510	Trace
56	0.505	Trace
66	0.500	Trace
76	0.500	Trace
108	0.495	Trace

FIGURE 21

Pu–1.0 wt% Ga Alloy, Cooled at 2 C/min from Melt at 810 C to 522 C Followed by Furnace Cooling to Ambient

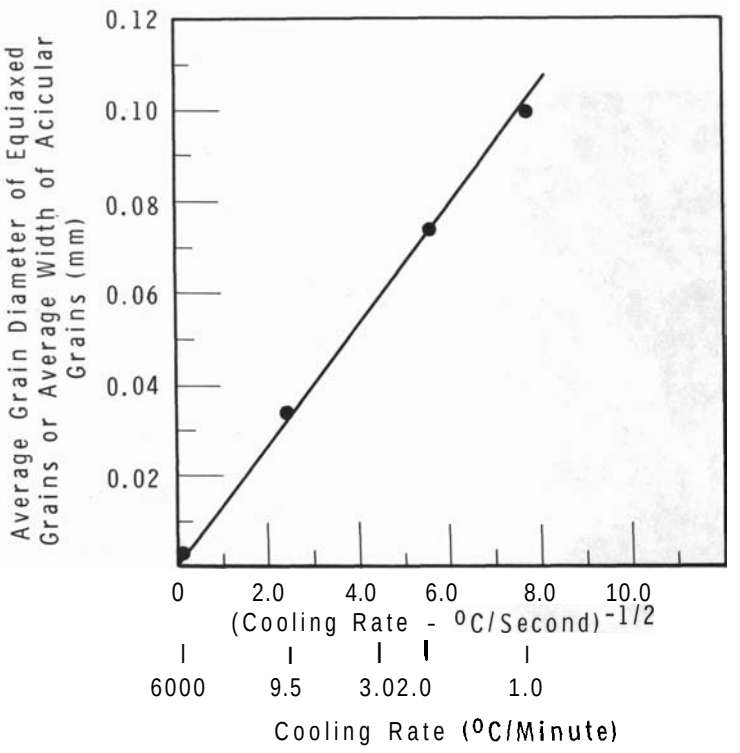
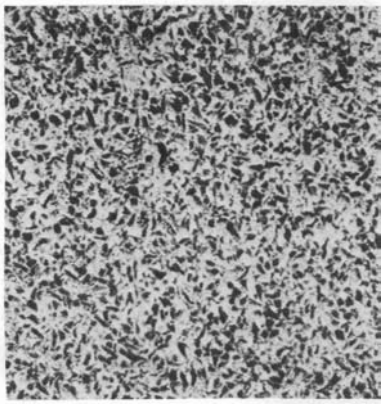


FIGURE 22

Relation Between Cooling Rate for 540 to 590 C and Grain Size in Cored Pu–1.0 wt% Ga Delta Stabilized Alloy

The data in Figures 12, 13, 14, 18, 19, 22, 23, and 24 were used to establish a relationship between grain size and time required for 25, 50, 75, and 100% homogenization, Figure 26. The time for complete homogenization ranges from 1 hr for a grain width of 0.002 mm to 215 hr for a grain width of 0.10 mm. A comparison of the data in Figures 18 and 20 indicates that the configuration of the cored grains, i.e., equiaxed or acicular, does not have any measurable influence on rate of homogenization.

Of interest is a comparison of rate of homogenization data dbtained by X-ray line broadening and by density increase after a 150,000 psi compression

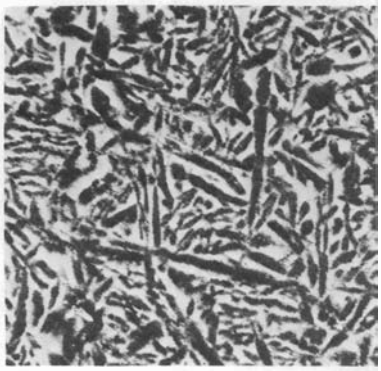


Effect of Anneal Time at 450 C on X-Ray Line Broadening of (331) Peak

Anneal Time Hrs.	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1, K\alpha_2$ Doublet
0	0.665	No
2	0.525	Trace
3	0.505	Trace
4	0.455	Yes

FIGURE 23

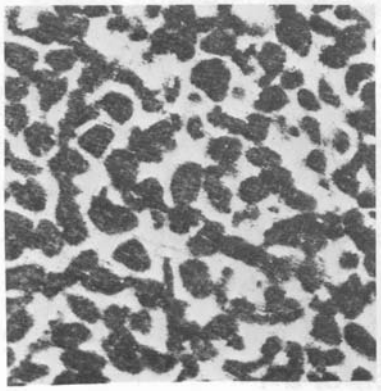
Heat 23-10-11. Melt Pouring Temperature 885 C. Graphite Mold Temperature 390 C, 0. 134 in. Thick Sheet
Average Grain Width 0.005 mm. As-Polished. 150 X



Anneal Time Hrs.	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1, K\alpha_2$ Doublet
0	0.630	No
4	0.505	Trace
6	0.460	Yes

FIGURE 24

Heat 5-16. Melt Pouring Temperature 800 C. Graphite Mold Temperature 495 C, 5/16 in. Diameter Rods
Average Acicular Grain Width 0. 008 mm. As-Polished 150 X



Anneal Time Hrs.	Half-Height Line Breadth (2θ)	Resolution of $K\alpha_1, K\alpha_2$ Doublet
1	0.810	No
2	0.785	No
4	0.695	No
8	0.595	No
18	0.575	No
24	0.535	Trace
40	0.470	Yes

FIGURE 25

Heat G. Melt Pouring Temperature 890 C, Graphite Mold Temperature 625 to 650 C.
Average Grain Diameter 0.037 mm. As-Polished. 150 X

Figure 27. The degree of correspondence between the two curves is excellent and instills further confidence in the use of the line broadening technique for rate of homogenization determinations. The cored microstructure for the material used is illustrated in Figure 25.

METASTABILITY IN ALPHA-DELTA PHASE MIXTURES

Ad indicated earlier, in cored Pu-Ga alloys, pressure will cause the delta phase containing less than 1. 2 wt% gallium to transform to alpha phase. Of interest were, (a) the effect of anneal time and temperature on the transformation of the metastable alpha phase thus produced, and (b) the stability of the resultant product during storage at room temperature.

Cored Alloy

Cored Pu–0. 94 wt% Ga alloy of 15. 75 g/cc density was cold-rolled to 94% reduction forming approximately 10% alpha phase and then annealed for times up to 24 hr at temperatures up to 300 C, Table IV. The data indicate

TABLE IV

EFFECT OF ANNEAL, TIME AND TEMPERATURE ON DENSITY*'

IN 94% COLD-ROLLED Pu–0. 94 wt% Ga ALLOY

Anneal Time, hr	As-Rolled Density, g/cc	Density after 150 C, g/cc	Density after 200 C, g/cc	Density after 250 C, g/cc	Density after 300 C, g/cc
0. 167	16.08	16.05	15. 92	15. 86	15. 75
6. 0	16. 08	16. 05	15. 91	15. 86	15. 75
24	16. 19	16. 12	16.02	15. 84	15. 75

*Determined at room temperature, after anneal.

that a 10 min anneal is sufficient to produce the full magnitude of density change, i. e., phase transformation that can occur at any given temperature. In addition, densities after the 150, 200, and 250 C anneals indicate that either beta and gamma phases are being retained to room temperature or the amount of transformation of alpha phase to delta phase is proportional to anneal temperature. To test the retention of beta and gamma idea, a 0. 37 wt% Ga cored alloy was prepared by dilution of a Pu–11. 1 wt% Ga master alloy. A 150, 000 psi compression at room temperature produced a density of 18. 20 g/cc indicating that approximately 60% alpha phase was present.

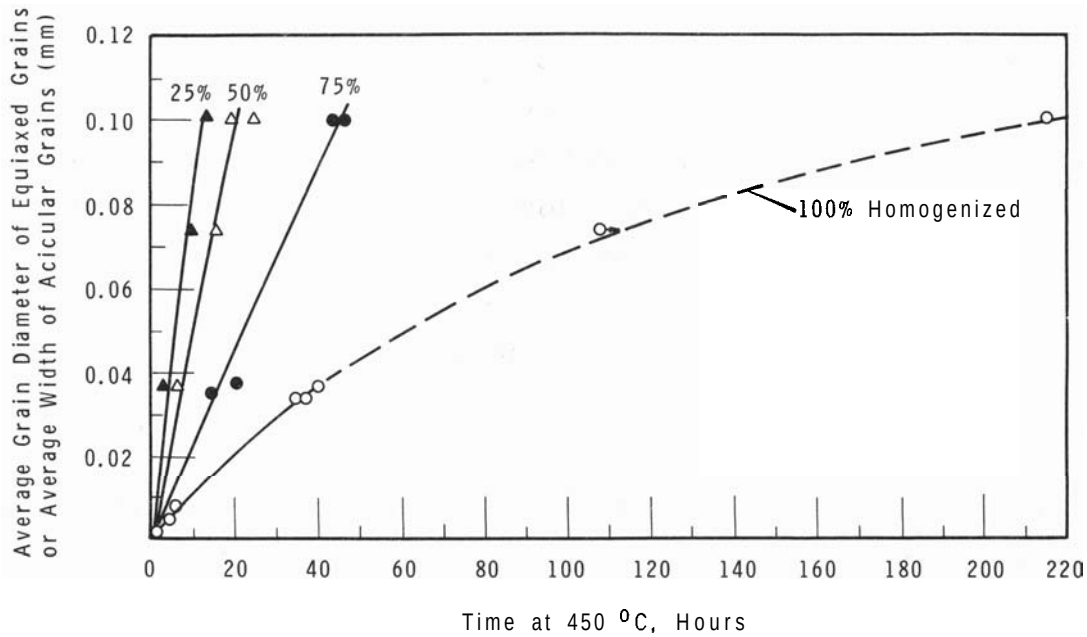


FIGURE 26

Relation Between Grain Size and Anneal Time Required for Homogenization in a Cored Pu–1.0 wt% Ga Delta Stabilized Alloy

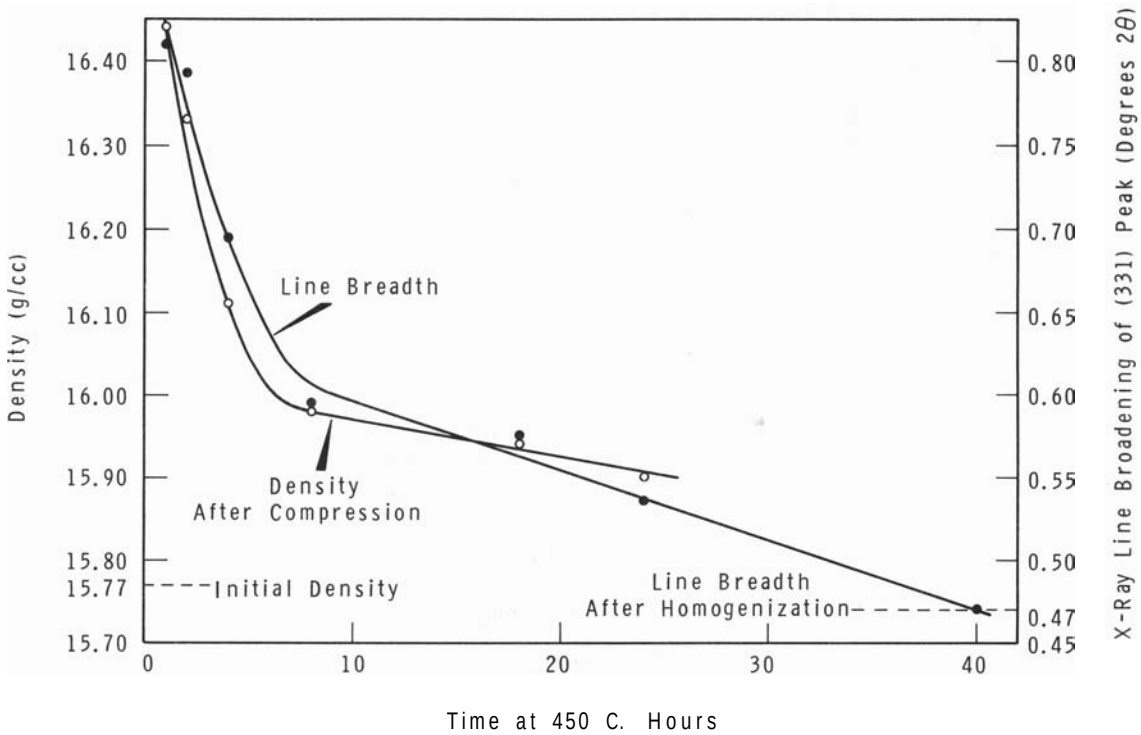


FIGURE 27

The Effect of Anneal Time at 450 C on Rate of Homogenization as Indicated by X-Ray Line Broadening and Density After a 150,000 psi Compression in Pu–1.0 wt% Ga Alloy

During a 10 min anneal at 200 C the density decreased to 16. 27 g/cc which corresponds to the presence of 24% beta phase. During phase identification studies using X-ray diffraction, no beta phase could be detected. A similar study after a 250 C anneal to form gamma phase indicated that gamma phase was not being retained to room temperature. Since beta and gamma phases are not retained to room temperature after appropriate anneals, it was decided to perform additional annealing experiments, on alpha-delta phase mixtures, designed to provide data leading to an explanation of the phenomena.

Portions of the cored Pu–0. 94 wt% Ga, 94% cold-rolled alloy used earlier in this study were given 10 min anneals at temperatures from 200 to 300 C. The density data obtained at room temperature after the anneal decrease with increasing anneal temperature up to approximately 280 C where the normal delta phase density is achieved, Figure 28. The following

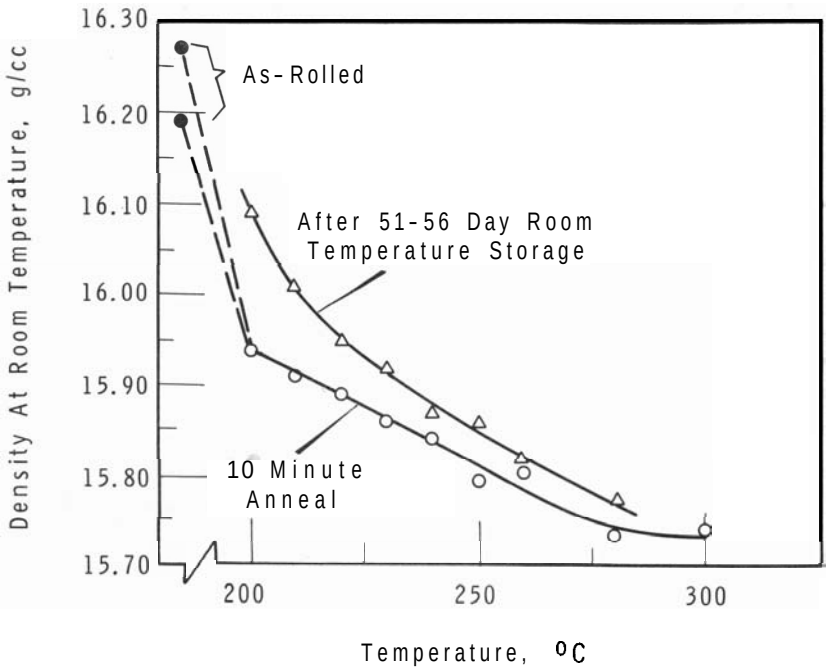


FIGURE 28

Effect of Anneal Temperature and Storage Time on Density of Pu–1. 0 wt% Ga Cold-Rolled, Cored Alloy

mechanism is proposed to account for the observed relationship. At temperatures up to 280 C, the alpha phase originally present transforms to beta or gamma phase plus delta phase. The amount of delta phase formed in this way is apparently related to temperature. As temperature increases the proportion of beta or gamma transforming to delta phase increases until at approximately 280 C the transformation is complete. During cooling to room temperature from temperatures below 280 C the remaining beta or gamma phases transform back to alpha phase while the delta phase which formed from beta or gamma remains delta phase, at least during the interval of density determination. The immediate net result is a decrease in density of the alpha plus delta phase mixture, the higher the anneal temperature the greater the decrease in density. A question of interest, as indicated earlier, is the degree of stability of the annealed and cooled alloy.

Accordingly, the above annealed specimens were stored at room temperature and in the case of the 200 and 250 *C* anneals, subjected to up to three cycles of cold treatment from room temperature to -54 C, Figure 29. The initial density decrease, from 16.30 - 0.31 g/cc to

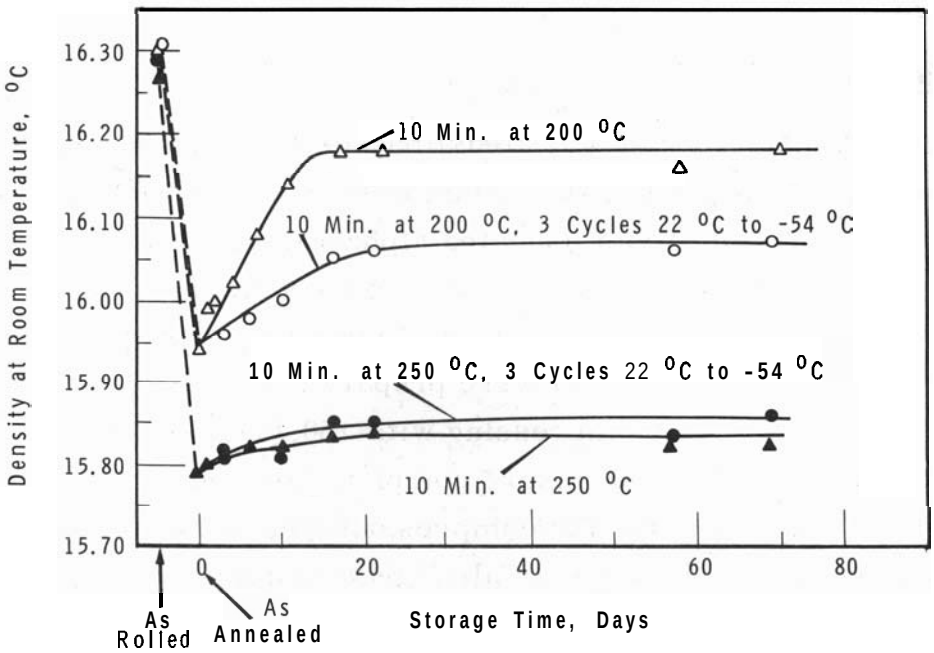


FIGURE 29

Effect of Anneal Temperature, Cold Treatment and Storage Time on Delta to Alpha Transformation

15.94 - 0.95 g/cc, indicates that approximately 5% alpha phase was present at room temperature after the 200 C anneal. After the 250 C anneal, however, less than 1% alpha phase was present. The rate of density change was much higher after the 200 C anneal than after the 250 C anneal, indicating that a larger amount of the delta phase formed from the beta phase at 200 C is exhibiting metastable behavior. To speculate briefly, the lower gallium areas in the cored alloy most probably have the greatest tendency to exhibit metastable delta to alpha transformation. As annealing temperature is increased from 200 to 250 C; sufficient diffusion apparently occurs to decrease the amount of delta phase capable of exhibiting metastable behavior during room temperature storage. The three cold treatment cycles improved stability after the 200 C anneal, but did not measurably affect stability after the 250 C anneal.

Looking again at Figure 28, the densities obtained after 51 to 56 days of room temperature storage are expressed as a function of anneal temperature. The decrease in density after storage with increasing anneal temperature up to 280 C is further evidence for the diffusion consideration discussedabove. .

Homogenized Alloy

Since the instability of annealed alpha-delta phase mixtures is believed to be related to the amount of low gallium areas in the cored grains, it was decided to study the stability of annealed alpha-delta mixtures in homogenized systems. To produce large and varying amounts of alpha phase in homogenized alloy during the application of pressure, a series of Pu-Ga alloys ranging from 0,37 to 1.26 wt% Ga were prepared by dilution of a Pu–11.1 wt% Ga master alloy. Prior to compressing with 150,000 psi, the alloys were annealed for 150 hr at 450 C to insure complete homogenization. The density data after the anneal and after the compression are presented in Table V with the approximate percentages of alph phase produced. For compositions greater than 0,99 wt% Ga the amount of alpha phase produced was 1% or less.

The effect of anneal temperature on percent alpha phase retained to room temperature in homogenized systems is illustrated in Figure 30. Again

TABLE V

ALPHA PHASE FORMATION DURING COMPRESSION OF HOMOGENIZED Pu-Ga ALLOYS

Analyzed Composition, wt%	Density after Anneal, * g/cc	Density after Anneal;;: at 150,000 psi Compression, g/cc	Percentage Alpha Phase after Compression, %	Minimum Anneal Temperature Required for Completion of Reaction, °C
0. 37	15.83	18. 11	62. 0	250
0. 62	15.81	17.08	35. 0	260
0. 78	15.78	16. 40	17. 0	230
0. 99	15.76	15. 92	4. 5	240
1. 12	---	15. 75	---	---
1. 26	15. 73	15.78	1. 0	---

* 150 hr at 450 C.

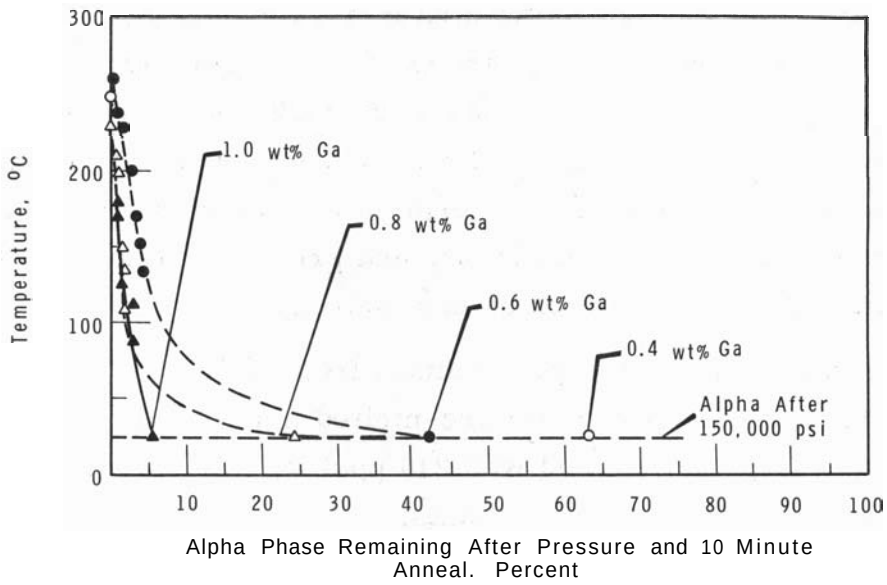


FIGURE 30

Effect of 150,000 psi Pressure on Alpha Phase Formation and Effect of Anneal Temperature on the Alpha–Delta Transformation Initial Condition - Cast and Annealed for 150 hr at 450 C Before Compression

the proportion of delta phase forming from alpha, beta, and gamma phases is proportional to anneal temperature. Complete transformation to delta phase occurs only after heating to approximately 260 C. In contrast to the cored alloy, it was found that the delta phase thus formed in

homogenized systems containing as little as 0.37 wt% Ga is stable, i.e., no measurable density change, with respect to storage at room temperature for times up to 3 months.

EFFECT OF COMPOSITION ON LATTICE PARAMETER, DENSITY, HARDNESS, AND PRESSURE METASTABILITY

As indicated in the Experimental Procedure Section, p. 7, care must be exercised during the preparation of Pu-Ga alloys to prevent the formation of Pu_xGa_Y compounds which reduces the gallium available for solid solution stabilization of the delta phase. Inspection of the phase diagram, Figure 1, indicates that to successfully prepare the alloy using induction or resistance heating facilities, melt temperatures in the range of 1100 to 1300 C must be achieved. Because of the inherently high metal temperatures achieved during arc melting, this method of alloy preparation also prevents the formation of Pu_xGa_Y compounds. In general, in compositional studies performed by other investigators, neither of the indicated methods were used to prepare the alloy. It was therefore decided to determine the compositional dependence of lattice parameter, density, hardness, and pressure metastability in the Pu-Ga system with alloy prepared by arc melting.

Pu-Ga alloys ranging in composition from 0.37 to 1.26 wt% Ga were prepared by dilution of 11.1 wt% Ga arc-melted master alloy. For the alloys, the average total impurity content was 216 ppm with 50 ppm aluminum and 90 ppm carbon as the major contaminants.

Lattice Parameter

To provide homogenized alloy for lattice parameter determinations, the specimens were annealed for 150 hr at 450 C. Since the cored grain size was of the order of 0.005 to 0.007 mm, the anneal was more than sufficient to achieve complete homogenization.

Two specimen preparation methods were used to obtain lattice parameter versus composition data on a Norelco X-Ray Diffractometer with CuK_α radiation. In the first method, the specimens were prepared as indicated in the Line Broadening Section, p. 8. When the minimum line width for the

(331) peak was approached, scans of the (331), (420), (422), (511/333), (440), and (531) peaks were made at 0.25°/min. Lattice parameters were calculated and graphed against the Nelson-Riley function. It was found that the best estimates of a_o could be obtained either from the Nelson-Riley extrapolation or from the lattice parameter of the (531) plane. In the second method, developed by Hays,[6] powder was filed from the specimens in an nitrogen atmosphere glove box and stress relieved in vacuo at 300 C for 3 hr. After mounting the powder in a recessed lucite holder covered with cellophane tape, a diffractometer scan of the (511/333), (440), and (531) peaks was made using the fixed count method. The time necessary to register 25,600 counts on a diffractometer scaler was recorded at 0.05° 2θ increments for each diffraction peak. Using the method of Koisteinen and Marburger,[7] a parabola was fitted to each of these diffraction peaks. The vertex of the parabola being taken as the diffraction angle. The data points used in fitting the parabola were at least 90% of the maximum peak intensity. The intensity readings were initially corrected for Lorentz-polarization and background, however this caused little or no change in the final value of lattice parameter and was discontinued.

The powder data by Hays are in good agreement with Ellinger,[1] Figure 31. The data of Gardner are displaced by roughly a constant amount from Hay's and Ellinger's. The presence of a nonrepresentative specimen surface in the solid specimens used by Gardner was felt to be the main source of error. Other than a possible difference in impurity content, no explanation can be given for the large difference between the data of Hays and Ellinger and that of Lee.[8]

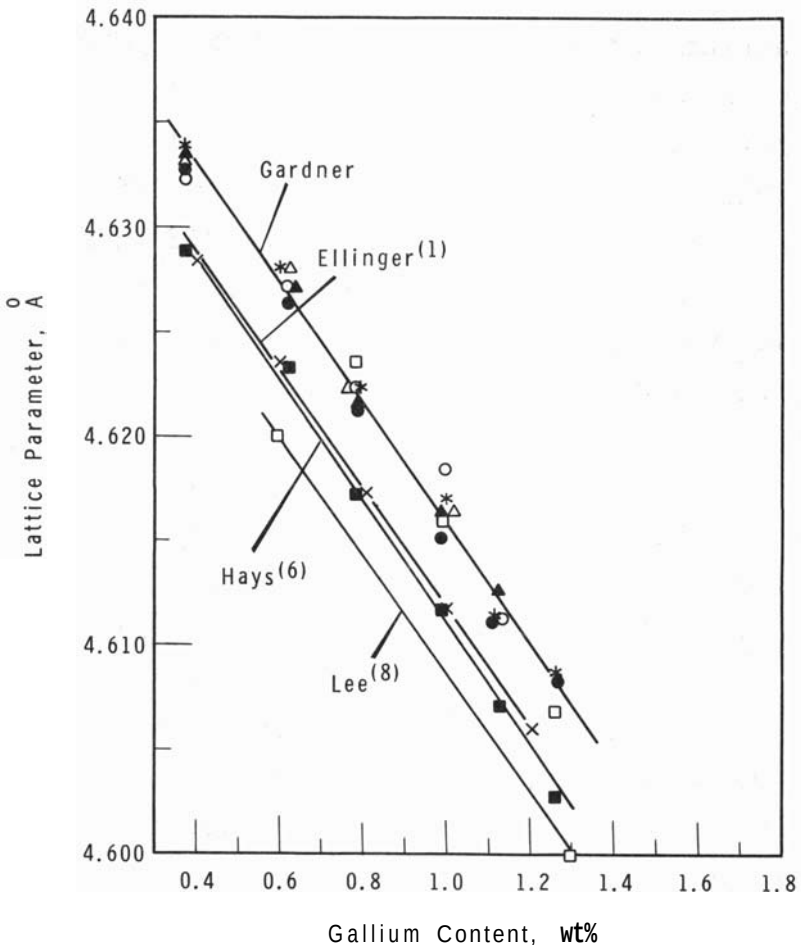


FIGURE 31

Effect of Gallium Content on Lattice Parameter

Density

Density determinations were made at room temperature using a fluid displacement technique(9) with FC-75* as the displacement medium. Comparison of density data before and after a 150 hr, 450 C anneal, indicate that alpha phase was present at room temperature in the cored as-cast alloy when compositions were 0.99 wt% Ga or less, Table VI. The density versus composition data of three investigators presented in Figure 32 are in reasonably good agreement. Density decreasing linearly with increasing gallium content.

*FC-75 is an inert fluorochemical consisting principally of isomers of perfluoro cyclic ether: $C_8F_{16}O$.

TABLE VI

EFFECT OF GALLIUM CONTENT ON DENSITY, HARDNESS, AND PRESSURE METASTABILITY

Analyzed Composition, wt% Ga	As-Cast Density, g/cc	Density after Anneal,* g/cc	As-Cast Hardness (DPH- 2 kg Load)	Hardness after Anneal* (DPH- 2kg Load)	Density after Anneal* and 150,000 psi Compression, g/cc	Percentage Alpha Phase after Compression, %
0.37	16.12	15.83	53	32	18.11	62.0
0.62	15.95	15.81	42	36	17.08	35.0
0.78	15.83	15.78	48	38	16.40	17.0
0.99	15.79	15.76	46	40	15.92	4.5
1.12	----	----	49	44	15.75	---
1.26	15.71	15.73	49	46	15.78	1.0

* 150 hr at 450 C.

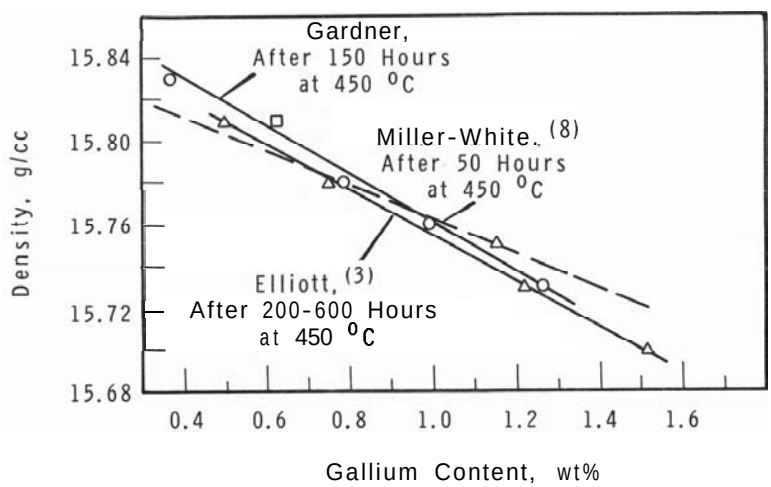


FIGURE 32

Density Versus Composition

Effect of Composition and Pressure On Density - To achieve a near isostatic pressure, specimens approximately 1/8 in. thick with an initial area of 1 in.^{2} were used. The effect of pressure on the density-composition relationship is illustrated in Figure 33. As gallium content increases, the effect of pressure on density diminishes at a rapid rate. Similar data obtained earlier by Elliott and Gschneidner[3] generally show the same result except for a substantial vertical displacement. Since the pressures used by both investigating teams were nearly the same the difference between the two curves may be the result of a lower effective gallium

content in the Elliott-Gschneidner specimens since they were not prepared by either arc melting or high melt temperatures.

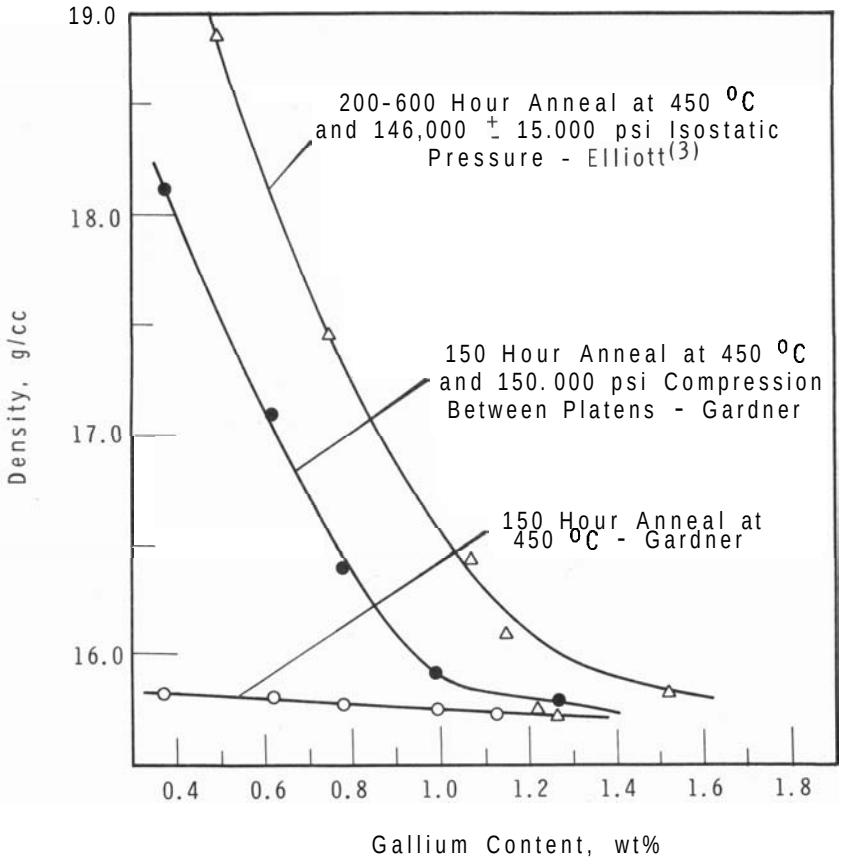


FIGURE 33

Effect of Composition and 150,000 psi Pressure on Density in Pu-Ga Alloys

Hardness

Hardness data were obtained on a Wilson "Tukon" Microhardness Tester using a 2 kg load. Data for both the as-cast and homogenized conditions are presented in Table VI. In the homogenized alloys, the linear increase in hardness with gallium content caused by solid solution strengthening, indicates that no alpha phase is present and that no alpha phase forms during the test. In the as-cast, cored condition, however, the lower gallium content alloys contain alpha phase which results in higher hardnesses. In addition, during hardness testing, with its inherent compressive loading, alpha formation occurs further increasing the apparent

hardness over the homogenized value. As gallium content increases in the cored alloys the amount of alpha phase present and the alpha forming tendency both decrease resulting in a lower rate of increase of hardness with composition than in the homogenized alloys. It is expected that the as-cast cored, and homogenized hardness curves will eventually coincide at the point where alpha formation no longer occurs in the cored area of minimum gallium content.

A comparison of hardness data obtained by three investigators is illustrated in Figure 34. The Miller-White[8] and Gardner data compare very well, however, the Elliott-Gschneidner[3] data are in substantial disagreement. The formation of alpha phase during the hardness test could account for the discrepancy if an excessive load were used. The fact that the Elliott-Gschneidner hardness versus composition curve is less steep than the others seems to substantiate the alpha phase formation consideration in light of the discussion in the preceeding paragraph.

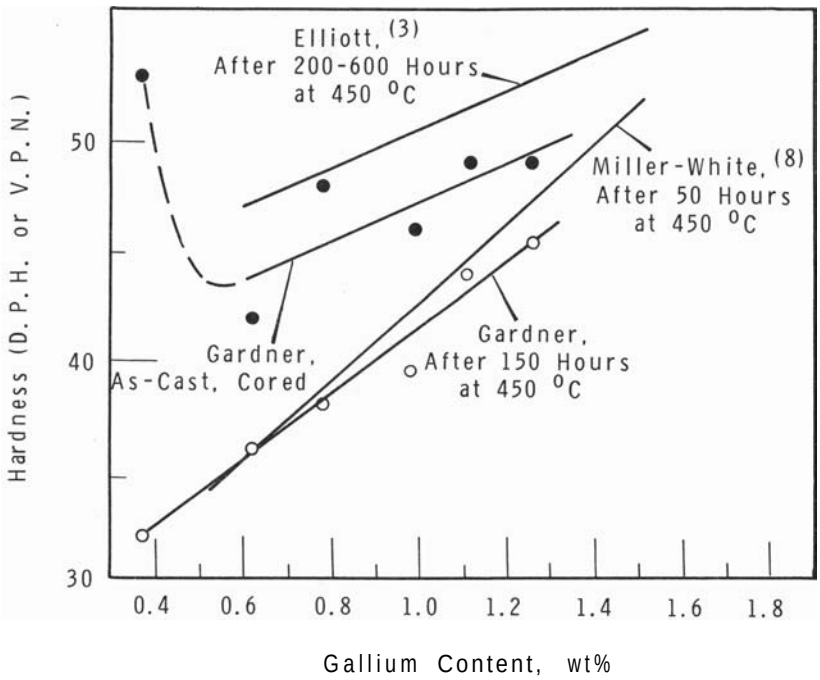


FIGURE 34
Hardness Versus Composition

RECRYSTALLIZATION

Recrystallization in a Pu-0.94 wt% Ga alloy was studied on both a homogenized, wrought condition and a nonhomogenized, wrought condition. To obtain the homogenized, wrought condition, alloy was cast into a mold at 450 C, furnace cooled to room temperature, annealed for 192 hr at 425 C and cold-rolled to 94% reduction. The resultant hardness was 56 DPH and with a density of 15.75 g/cc indicating that very little, if any, alpha phase was present. Since the as-homogenized hardness was 41 DPH, the 56 DPH after rolling with no increase in density indicated that appreciable strain 'hardening had taken place. The nonhomogenized, wrought condition was achieved simply by rolling the as-cast alloy to 94% reduction. Here the hardness increased to 80 DPH and the density to 16.08 g/cc. The density increase indicated that approximately 10% alpha phase had formed. The large hardness increase indicated that alpha formation causes strengthening over and above that obtained from strain hardening alone.

To prepare material for the recrystallization studies anneals were conducted at 100, 150, 200, 250, 300, and 425 C for various periods of time on both conditions of the alloy.

In the homogenized, wrought condition, no change in hardness occurred during the 100 C anneal while the 150 C anneal caused a slight increase in hardness, possibly the result of age hardening, Figure 35. At 200 C, the age hardening affect appears to have been accelerated, however, at the longer anneal times, the hardness is approximately the same as that obtained during the 150 C anneal. Recrystallization occurs during the 250 C anneal and and hardness gradually decreases to the dead soft value of 41 DPH. At 300 C, recrystallization occurs rapidly, the dead soft hardness was obtained after a 10 min anneal.

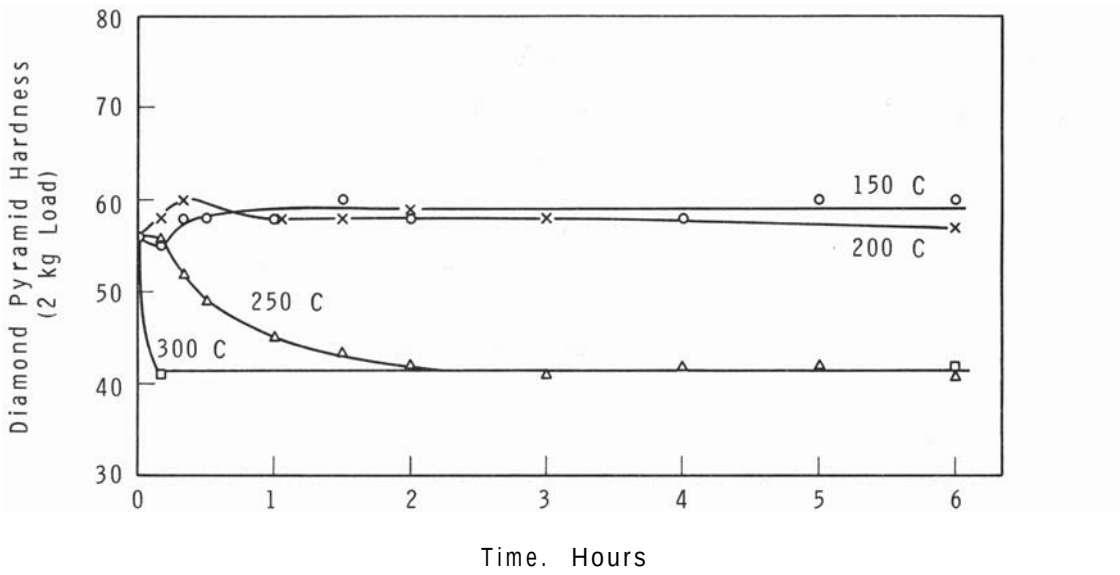


FIGURE 35

Effect of Annealing Time and Temperature on Hardness of a Pu-1 wt% Ga Delta Stabilized Alloy (Cast, homogenized for 192 hr and rolled 94% reduction in thickness before annealing.)

In the nonhomogenized, wrought condition, again no change in hardness occurred during the 100 C anneal. At 150 C, the hardness decreased initially followed by an increase back to the as-wrought hardness level, Figure 36. Stress relief followed by age hardening is a suggested explanation. At 200 C, the stress relief-age hardening effect seems to have accelerated. In addition, a substantial decrease in level of hardness occurred. Since the 200 C anneal did not have much effect on hardness in the homogenized wrought condition, the decrease in hardness after this anneal in the nonhomogenized, wrought condition was attributed to the transformation of alpha phase to delta phase. This phenomena has been discussed in detail in the section on Metastability in Alpha-Delta Phase Mixtures (p. 31). Recrystallization occurs during the 250 C anneal, however, hardness does not decrease to the dead soft level as in the homogenized, wrought condition since the gamma phase in the gamma plus delta phase mixture at 250 C transforms back to alpha phase during cooling. The resulting alpha plus delta phase mixture has a higher hardness, of course, than the delta

phase alone. At 300 C, recrystallization occurs rapidly and a dead soft hardness is obtained during a 10 min anneal. All alpha phase originally present has transformed to delta phase and does not reform immediately on cooling to room temperature.

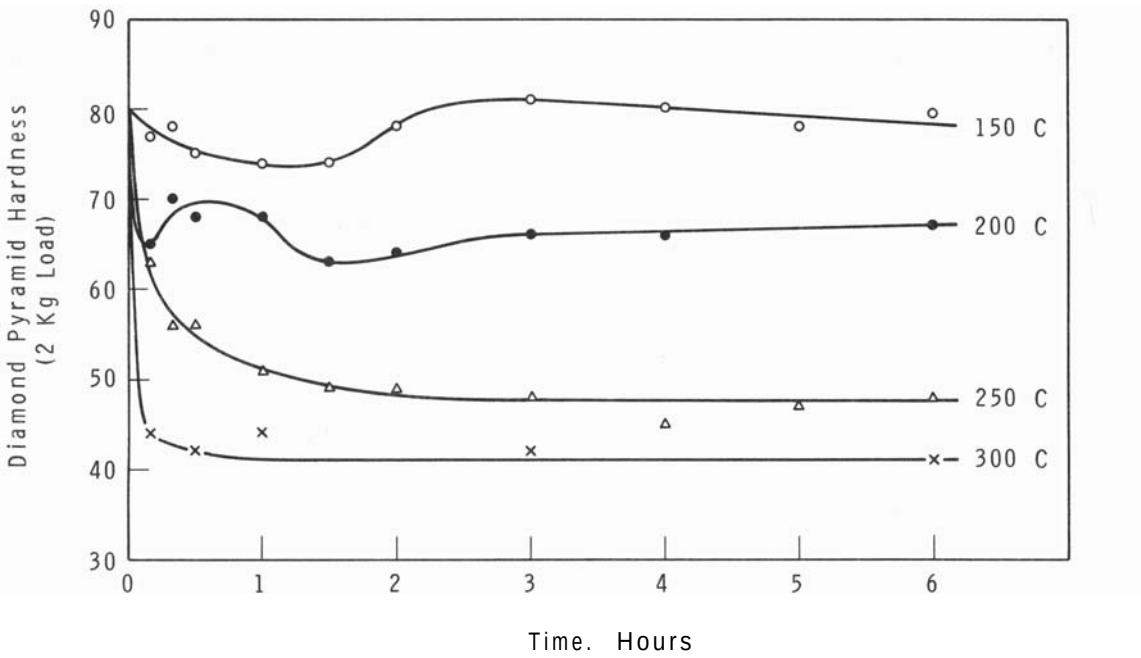


FIGURE 36

Effect of Annealing Time and Temperature on Hardness of a Pu–1.0 wt% Ga Delta Stabilized Alloy
(Cast with no homogenization and rolled 94% reduction in thickness before annealing.)

No microstructural evidence for recrystallization could be observed after the 100, 150, and 200 C anneals in either material condition. At 250 C and higher temperatures, recrystallized structures were observed, Figures 37 and 38. Note that in the nonhomogenized, wrought condition the 150 and 200 C anneals produced what appear to be extremely fine equiaxed grains along the elongated rolled grains. Since these fine equiaxed grains are absent after corresponding anneals of the homogenized, wrought material, it is suggested that they are a product of either the alpha→delta or alpha ⇄ beta transformation.

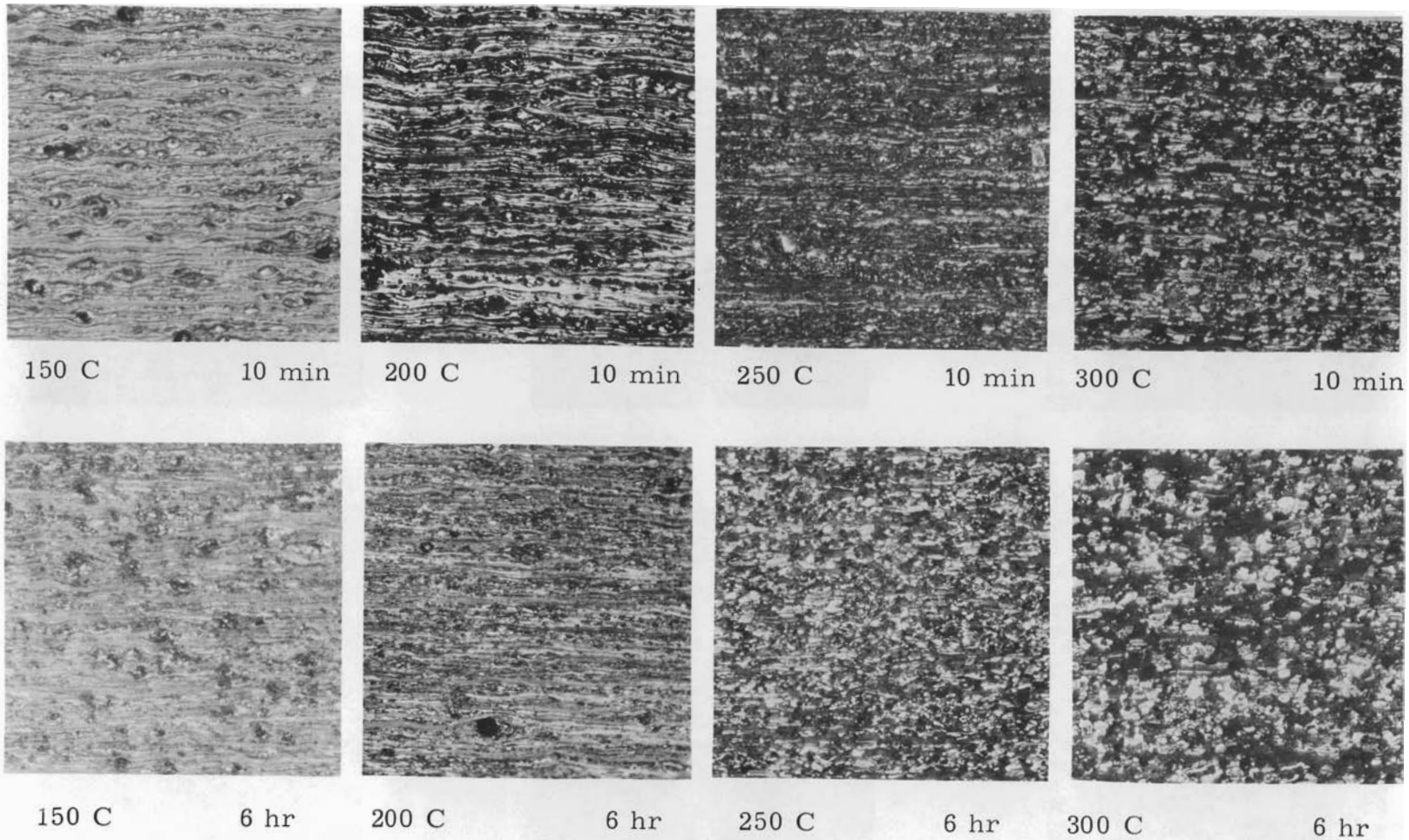


FIGURE 37

Effect of Annealing Time and Temperature on Recrystallization in the Nonhomogenized and 94% Rolled Condition. Electropolished, Polarized Light 150 X

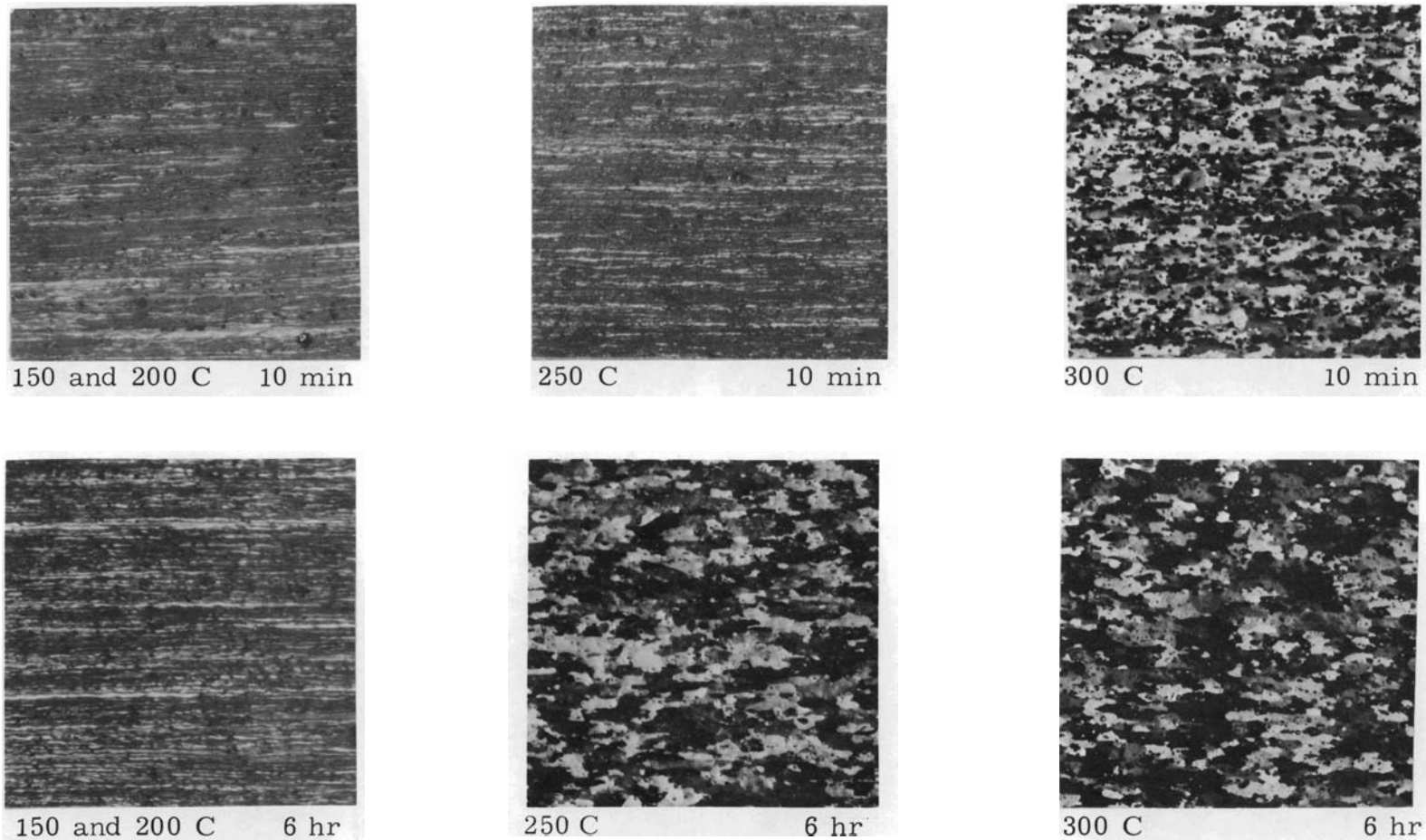


FIGURE 38

Effect of Annealing Time and Temperature on Recrystallization in the Homogenized and 94% Rolled Condition. Electropolished, Polarized Light. 150 X

Recrystallized grain sizes are coarser in the homogenized, wrought condition than in the nonhomogenized, wrought condition, Figures 37 and 38. Transformation of alpha to gamma and delta phases before recrystallization starts in the nonhomogenized wrought condition may furnish a larger number of nuclei resulting in a finer grain size. In addition, the coring present could act as a barrier to grain growth resulting in a finer grain size than in the homogenized, wrought sheet.

The nonhomogenized, wrought material has been annealed at 425 C for times up to 975 hr. Grain size increases generally with annealing time from 0.020 mm for 10 min at 425 C to 0.080 mm at 200 hr, Table VII. No reason can be given for the finer grain size after the 975 hr anneal.

TABLE VII

THE EFFECT OF MATERIAL CONDITION AND ANNEALING TEMPERATURE AND TIME ON GRAIN SIZE* OF Pu-0.94 wt% Ga DELTAL STABILIZED PLUTONIUM

Time, hr	Nonhomogenized, Wrought Temperature, C			Nonhomogenized, Wrought Temperature, C		Nonhomogenized, As-Cast Temperature, C
	250	300	425	250	300	425
As-Cast	-----	-----	-----	-----	-----	0.030
0.17	-----	0.007	0.020	-----	0.020	-----
0.33	-----	-----	0.020	-----	-----	-----
1.5	-----	-----	0.020	-----	-----	-----
2	0.007	-----	-----	-----	-----	-----
3	-----	-----	0.020	-----	-----	-----
4	-----	-----	-----	-----	-----	0.030
6	0.007	0.013	-----	0.023	0.027	0.030
24	-----	-----	0.033	-----	-----	0.030
48	-----	-----	0.080	-----	-----	0.067
96	-----	-----	0.053	-----	-----	0.040
200	-----	-----	0.080	-----	-----	0.080 - 0.570
975	-----	-----	0.040	-----	-----	3.0 - 5.0

*Average grain diameter in mm.

Grain Coarsening

As indicated in the Recrystallization Section, p. 43, grain coarsening occurs in the wrought Pu-0.94 wt% Ga alloy. This effect has been observed on a much greater scale in as-cast material. Starting from a grain size of 0.030 mm in the as-cast, nonhomogenized condition, Figure 39, grains

3 to 5 mm in diameter have been produced during an anneal at 425 C for 975 hr, Figure 40, Table VII. Grain coarsening is evident after a 24 hr anneal, Figure 41, while a 200 hr anneal produces grains up to 0.57 mm in diameter, Figure 42. The grain coarsening in the 24, 200 and 975 hr anneal specimens is felt to be the result of a critical strain near the sheared edge of the specimen. Note the fine grains at the sheared edge in Figure 42, then coarse columnar grains growing into a fine-grained area. It is suggested that very large grain sizes, perhaps even single crystals, could be obtained by the application of the strain-anneal technique to this alloy.

In strain-free specimens it was determined that anneal temperature between 400 and 525 C had no apparent effect on grain size during a 24 hr anneal.

Void Formation

Grain boundary void formation has been observed after 24 hr and longer anneals of the Pu-0.94 wt% Ga alloy. At 425 C, a 24 hr anneal appears to be sufficient to produce grain boundary voids, Figure 43; while after 975 hr at 425 C, extensive grain boundary voids are observed, Figures 40 and 44.

In a 94% wrought alloy, no voids have been observed after annealing at 425 C for times up to 975 hr. In addition to studying the mechanism of void formation, it would be pertinent to determine the level of reduction required to inhibit void formation during annealing.

MECHANICAL PROPERTIES

The tensile and compressive properties of delta stabilized alloys ranging from 0.9 to 1.3 wt% Ga have been determined as a function of temperature and testing speed. Except for the early work on the Pu-0.9 wt% Ga alloy, material preparation consisted of dilution of arc melt Pu-11.1 wt% Ga master alloy. The Pu-0.9 wt% Ga alloy used early in the study was prepared by the direct addition of gallium during casting in a resistance furnace at a melt temperature of 900 C. It has been determined that this method of preparation promotes the formation of Pu_xGa_Y compounds, therefore reducing the amount of gallium available for stabilization. Some of the inclusions appearing in Figure 45 are believed to be Pu_xGa_Y compounds.

150X FIGURE 39

Heat 23-1-7. As-Cast, No Homogenization. Average Grain Diameter, 0.030 mm. As-Polished, Bright Field.

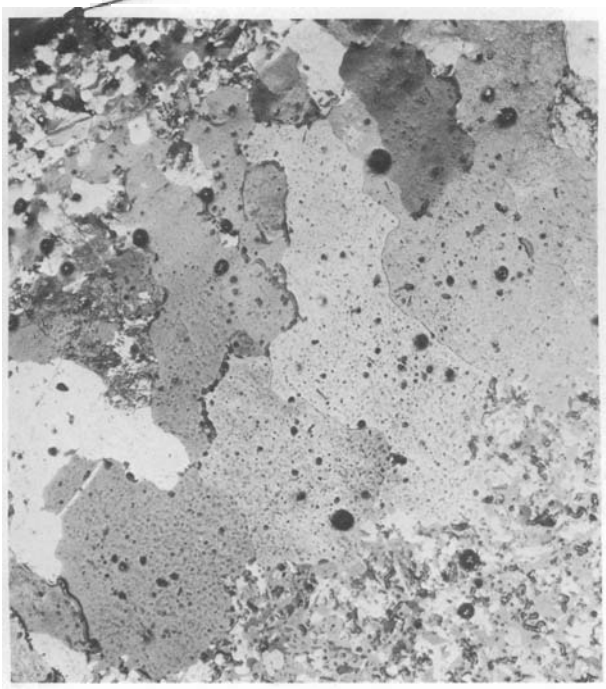
FIGURE 40 6X

Heat 23-1-7. Annealed for 975 hr at 425 C and Furnace Cooled. As-Polished, Bright Field. 6X

150X FIGURE 41

Heat 23-1-7 Annealed for 24 hr at 425 C and Furnace Cooled As-Polished, Bright Field

Sheared Edge



50 X FIGURE 42

Heat 23-1-7. Annealed for 200 hr at 425 C and Furnace Cooled As-Polished, Bright Field

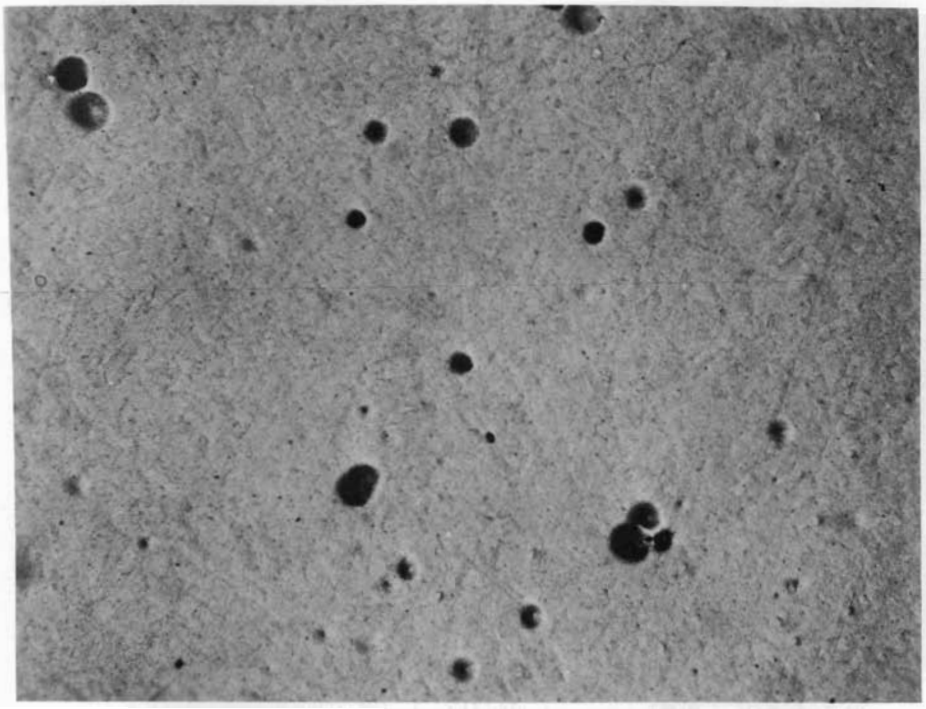


FIGURE 43

Voids Present After 24 hr Anneal at 425 C
As-Polished 75X

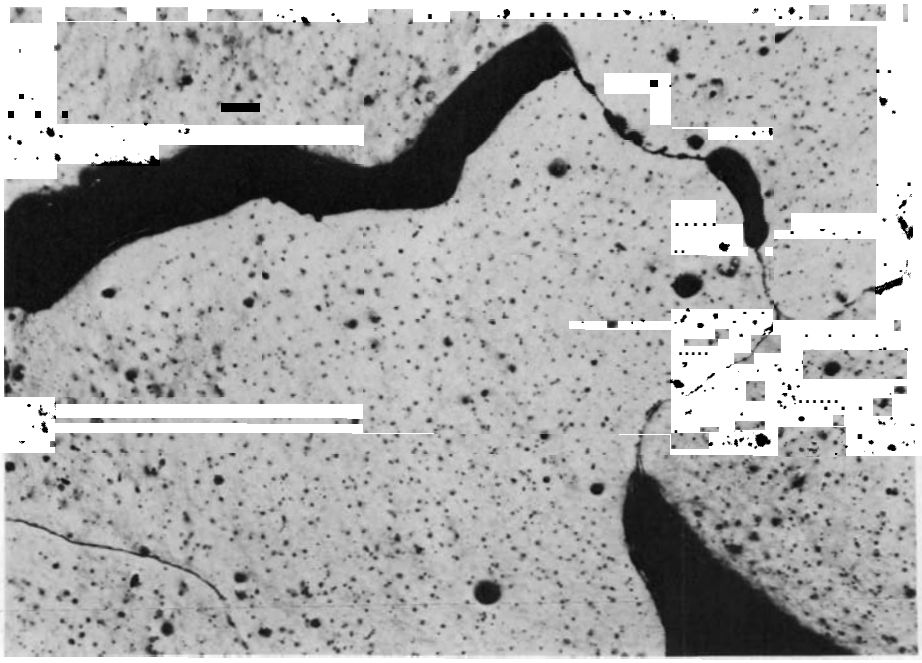


FIGURE 44

Grain Boundary Voids Present After 975 hr Anneal at 425 C
As-Polished 75X

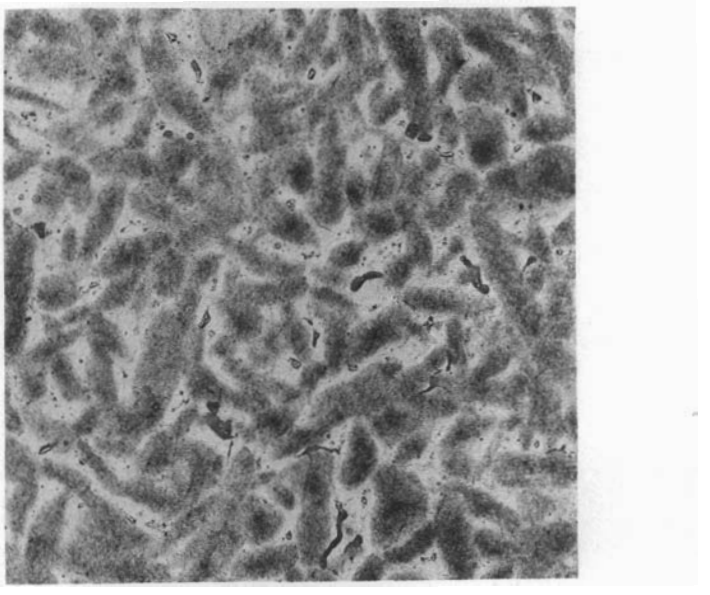


FIGURE 45

Pu-0.9 wt% Ga Alloy Prepared by Resistance Melting Plutonium and Gallium
(Note inclusions believed to be Pu_x-Ga_y compounds.)
As-Polished 150 X

Tensile

Contoured castings for tensile specimen fabrication were obtained by pouring in vacuo from 900 C into a MgO coated graphite mold which was heated to 475 - 500 C. To reduce the potential for alpha phase formation, the cast specimens were held in the mold for 1 to 2 hr at 425 - 475 C before cooling to room temperature. Average impurity content and density data are presented in Figure 46, along with a typical microstructure. The 2 hr anneal at 425 - 475 C after casting was sufficient to achieve possibly 15% homogenization.

A portion of a stress-strain curve for the Pu-0.9 wt% Ga alloy is illustrated in Figure 47. Stress is proportional to strain up to 4500 psi and there is no well-defined yield point. The curve is quite similar in appearance to that obtained on aluminum.

AVERAGE IMPURITY CONTENT AND DENSITY
OF Pu-0.9 wt% Ga AND Pu-1.3 wt% Ga ALLOYS

Element	Analysis (ppm)	Density (g/cc)
Al	22	----
Ca	14	----
Cr	44	----
Cu	7	----
Fe	202	----
Mg	49	----
Mn	37	----
Ni	72	----
Si	6	----
C	237	----
Total	690	----
Ga	0.92 wlo	15.76
Ga	1.24 wlo	15.73

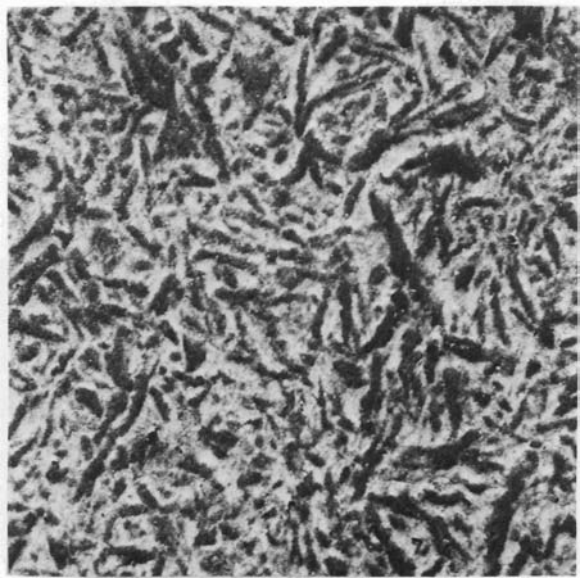


FIGURE 46

Typical Microstructure for Pu-1.0 to 1.3 wt% Ga Tensile Specimens after 1 to 2 hr at 425 to 475 C
(Average width of acicular cored grains, 0.017 mm.)
As-Polished 150 X

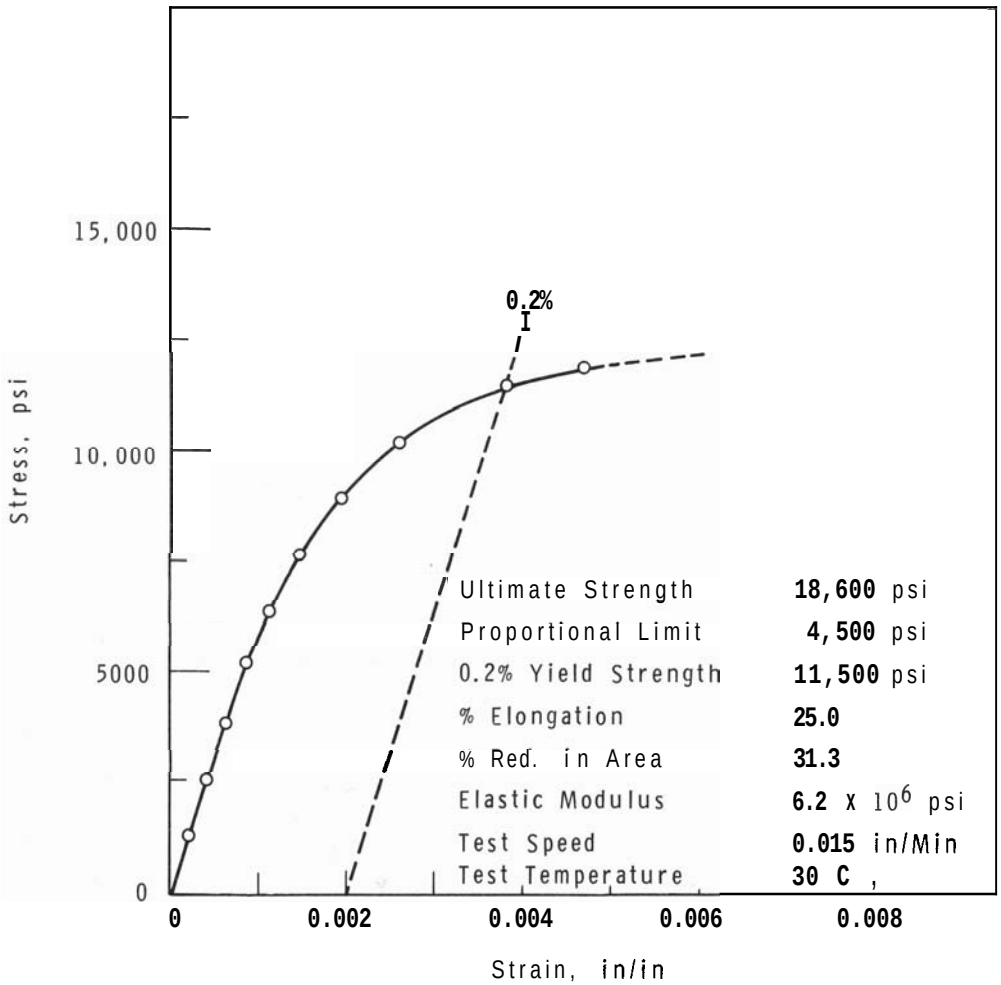


FIGURE 47

Stress-Strain Curve for Pu-0.9 wt% Ga Delta Stabilized As-Cast Plutonium, 0.250 in. Diameter Specimen

Effect of Temperature and Iron Content - The effect of temperature and iron content on the tensile properties of the Pu-0.9 and Pu-1.3 wt% Ga alloys are illustrated in Figure 48. The 1.3 wt% alloy is stronger than the 0.9 wt% alloy at all test temperatures because of the solid solution strengthening effect of the additional gallium. Both alloys decrease in strength as temperature increases, while ductility increases with temperature up to 300 C. Between 380 and 450 C, large variations in ductility are observed. Specimens containing 930 ppm iron exhibit no ductility and a decrease in strength, i.e., a typically hot short behavior. As iron content decreases, ductility in general

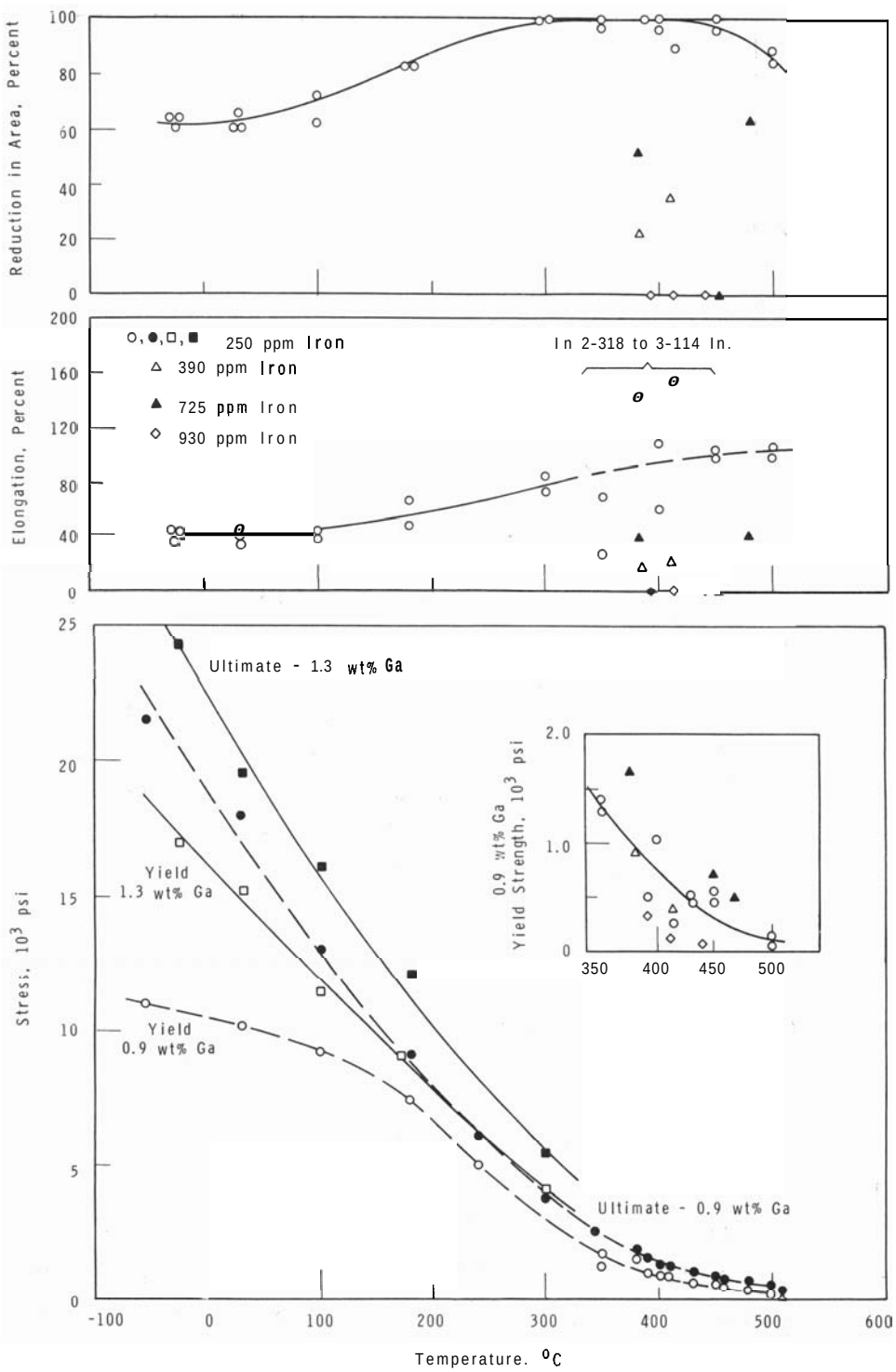
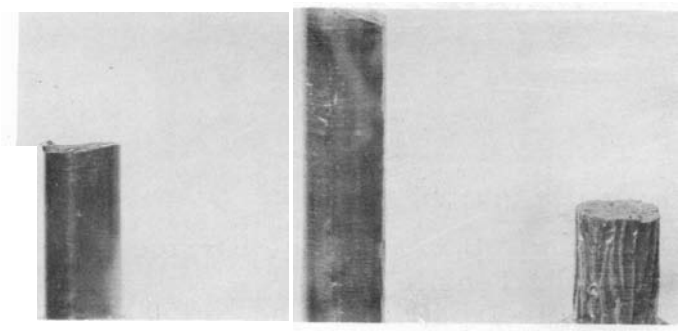


FIGURE 48

Effect of Temperature on Tensile Properties of Pu-0.9 and 1.3 Ga Delta-Stabilized Plutonium Alloys
Testing Speed, 0.015 in./min;

improves, however, even when iron content is less than 250 ppm, large variations in elongation occur which can probably be attributed to iron.

Specimens containing 930 ppm iron tested at 390, 410, and 440 C are illustrated in Figure 49. Note the vertical ridges on the specimen tested at 440 C. These ridges are formed by an accumulation of Pu-Pu_6-Fe eutectic as illustrated in Figure 50. The migration of Pu-Pu_6-Fe eutectic to exterior surfaces has been observed in unalloyed plutonium by Gardner[10] and is attributed to the existence of a delta plus liquid Pu-Pu_6Fe region between 413 and 425 C at the plutonium-rich side of the plutonium-iron equilibrium diagram. During the heating of either unalloyed or alloyed plutonium containing, for example, 1000 ppm iron, the Pu-Pu_6Fe eutectic expands when it melts. In a heated section, molten eutectic appears at the surface initially since it reaches the melting temperature first. As heating continues, the eutectic, expanding as it melts, is forced to exterior surfaces. It is expected that this transfer would be promoted by slow heating rates and minimized by fast heating rates. Microstructural examination of the specimen containing ridges indicate that only the first 0.004 in. of surface was involved in the eutectic transfer. The mechanism for the accumulation of eutectic into ridges aligned in the direction of testing is unknown. Since the loss of ductility in the high iron Pu–1.0 wt% Ga alloy occurs from 380 to 450 C, it is suggested that the presence of gallium may result in enlarging the temperature range of the delta plus liquid region.



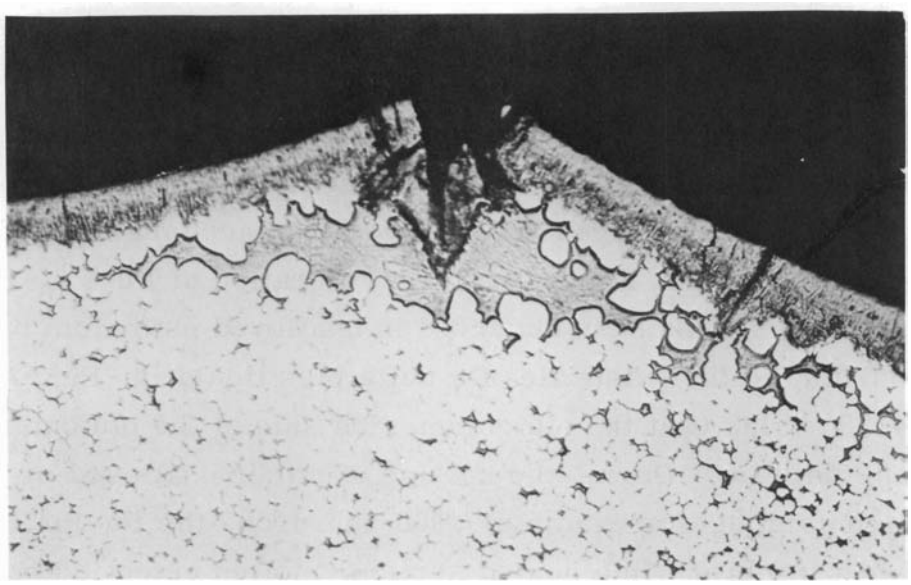
#19 390 C | #20 410 C | #17 440 C

High Iron

FIGURE 49

Pu–1.0 wt% Ga, 0.505 in. Diameter, Tensile Specimens Tested at Indicated Temperatures (Note vertical ridges formed by migration of Pu-Pu_6Fe eutectic, aligned with tensile axis on specimen tested at 440 C.)

A



B

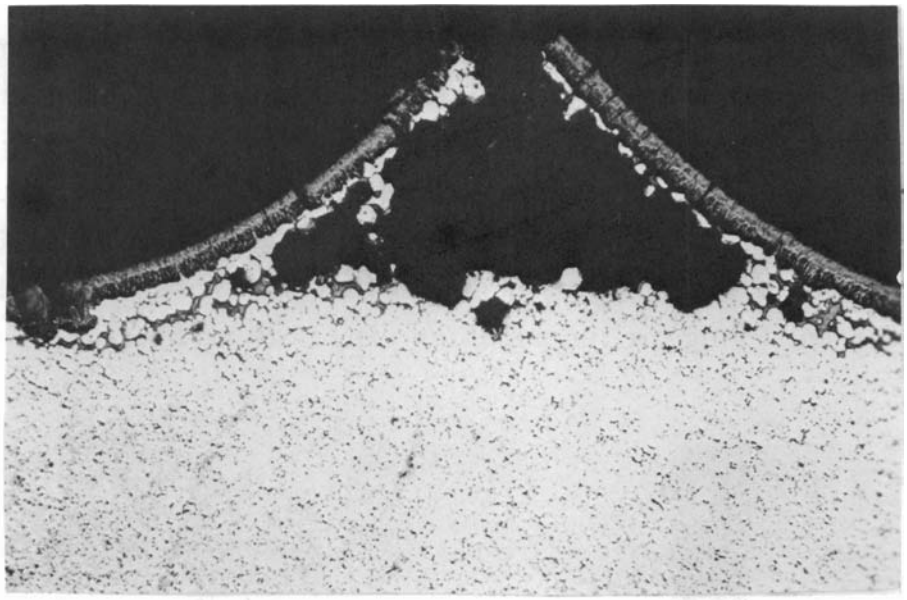


FIGURE 50

A. Accumulation of Pu-Pu_6Fe Eutectic Under Nitride Surface Layer in Ridge Aligned Along Tensile Axis
Specimen Tested at 440 C, Electroetched 5 sec, 150X

B. Ridge After Escape of Pu-Pu_6Fe Eutectic. Electroetched 5 sec, 75X

Effect of Testing Speed - The effect of testing speed on the tensile properties of the 1.3 wt% Ga alloy is slight in the -32 to 300 C temperature range investigated, Figures 51 and 52.

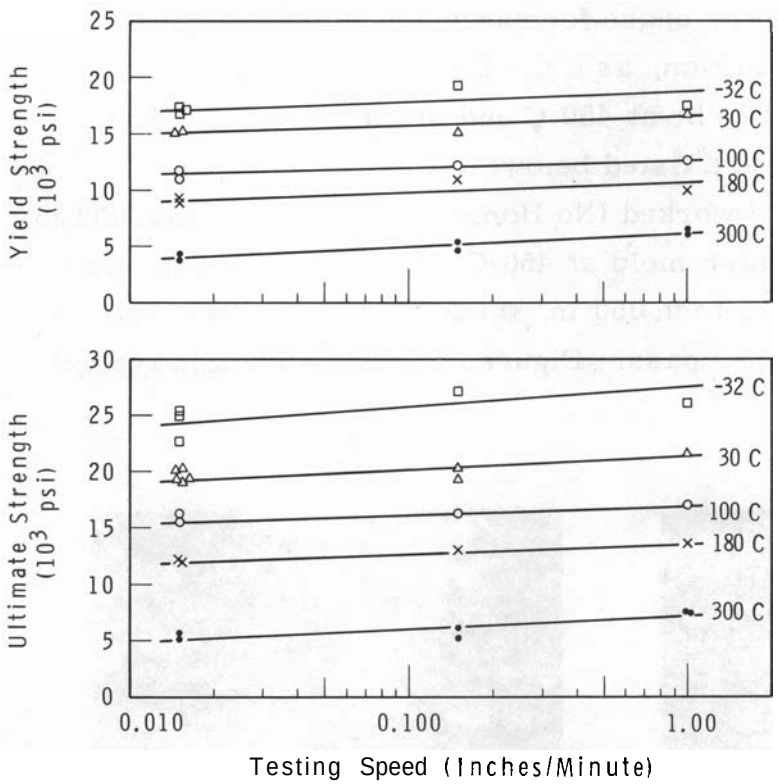


FIGURE 51

Effect of Testing Speed and Temperature on Tensile Strength in a Pu-1.3 wt% Ga Delta Stabilized Alloy

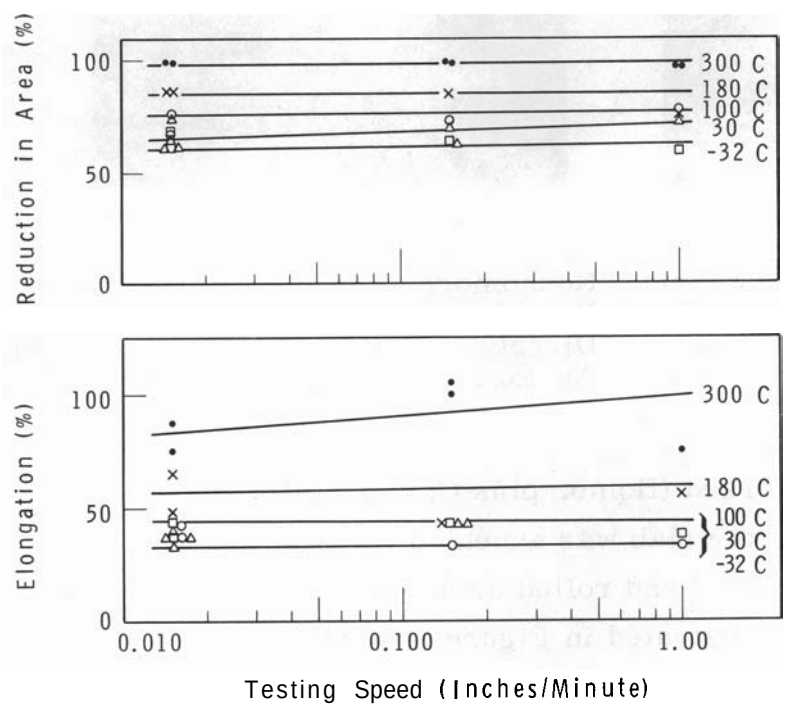


FIGURE 52

Effect of Testing Speed and Temperature on Ductility in a Pu-1.3 wt% Ga Delta Stabilized Alloy

Effect of Cold Work and Alpha Phase Formation on Tensile Properties

Rolled to 94% Reduction - The two conditions of the Pu-0.94 wt% Ga alloy used in the Recrystallization study (p. 43) were also used to determine the effect of cold work and alpha phase formation on tensile properties, density, and hardness. In addition, as a third condition, both initial conditions were annealed for 24 hr at 450 C and furnace cooled. For clarity, the three conditions are listed below:

1 Nonhomogenized, cold-worked (No Homo. plus C. W.). A 0.833 in. thick slab was cast into a mold at 450 C, furnace-cooled to room temperature and rolled to 0.050 in. thick sheet for a 94% total reduction, 0.030 in. per pass. Figures 53 and 54 contain typical microstructures.

FIGURE 53

Nonhomogenized, 94% Rolled Sheet View Perpendicular to Rolling Direction Across Width of Sheet. As-Polished, Bright Field, 150 X

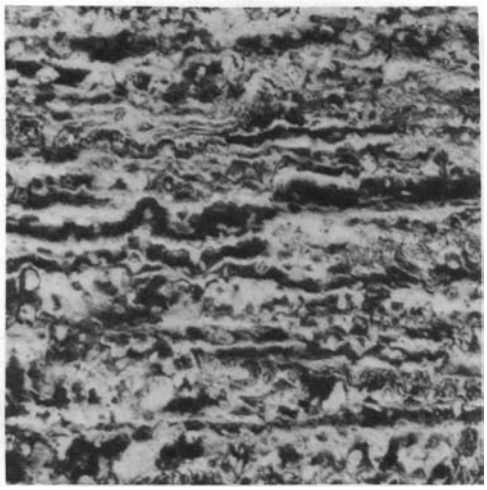


FIGURE 54

Nonhomogenized, 94% Rolled Sheet View Perpendicular to Rolling Direction Across Thickness of Sheet. As-Polished, Bright Field, 150 X

2. Homogenized, cold-worked (Homo. plus C. W.). After casting as in the first condition, the slab was annealed for 200 hr at 425 C, Figure 55, furnace-cooled and rolled as above. Representative microstructures are illustrated in Figures 56 and 57.

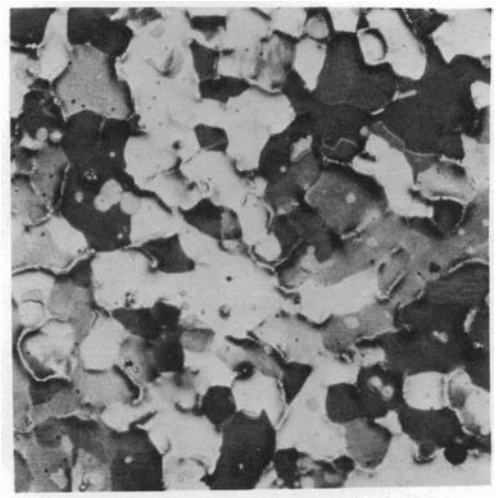


FIGURE 55

As-Cast, Homogenized for 200 hr at 425 C,
Furnace Cooled. Average Grain Diameter, 0.050 mm
Electropolished, Bright Field, 150 X

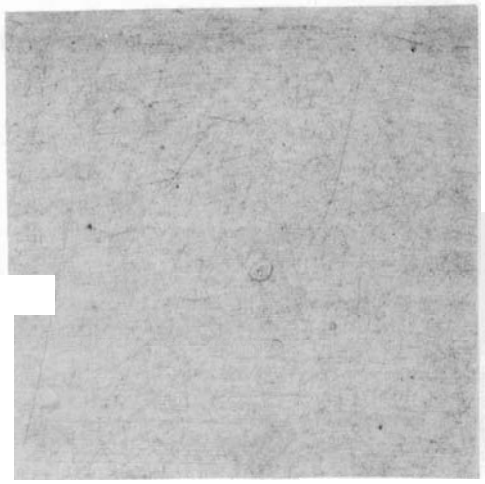


FIGURE 56

Homogenized and 94% Rolled Sheet.
As-Polished, Bright Field, 150 X

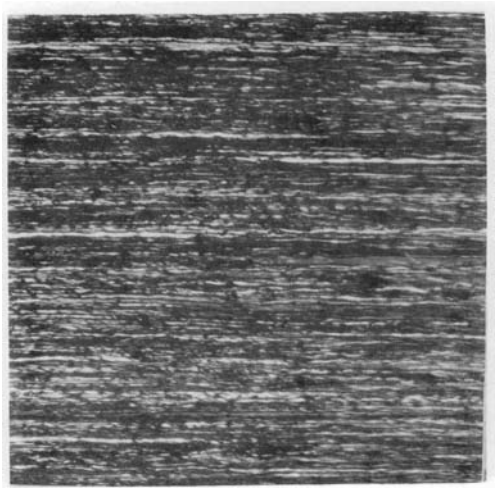


FIGURE 57

Homogenized and 94% Rolled Sheet.
Electropolished, Polarized Light,
150 X

3. Annealed (Ann.). Rolled sheet of both above conditions were annealed for 24 hr at 450 C and furnace-cooled. Typical micro structures are illustrated in Figures 58 and 59.

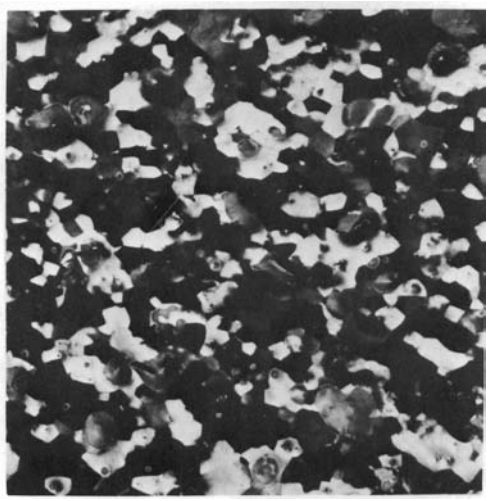


FIGURE 58

Nonhomogenized, 94% Rolled and Annealed for 24 hr at 450 C. Average Grain Diameter, 0.027 mm. Electropolished, Bright Field, 150 X

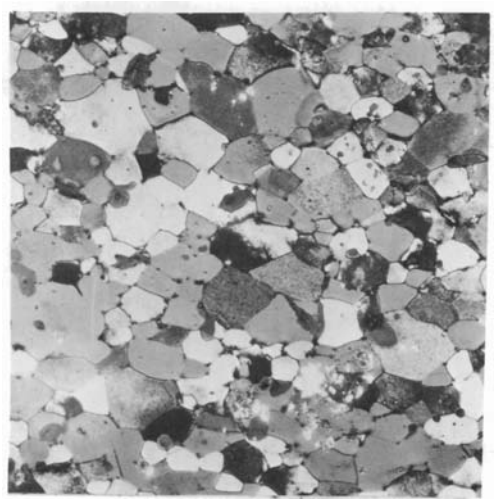


FIGURE 59

Homogenized, 94% Rolled and Annealed for 24 hr at 450 C. Average Grain Diameter, 0.040 mm. Electropolished, Bright Field, 150 X

Pertinent mechanical and physical properties of these different metallurgical conditions are illustrated in Figure 60 and Table VIII. Because of both strain hardening and alpha phase formation, the non-homogenized, cold-worked condition had the greatest strength, hardness, and density. Since no alpha phase formed during rolling the homogenized, cold-worked condition, the strengthening exhibited was produced by strain hardening alone. Both cold-worked conditions were dead soft after the anneal, yield strength decreased by a factor of three and ductility increased by a factor of 3 to 4. The elastic modulus was lowest in the annealed condition. The modulus in the nonhomogenized, cold-worked condition is higher than in the homogenized, cold-worked condition because of alpha phase formation.

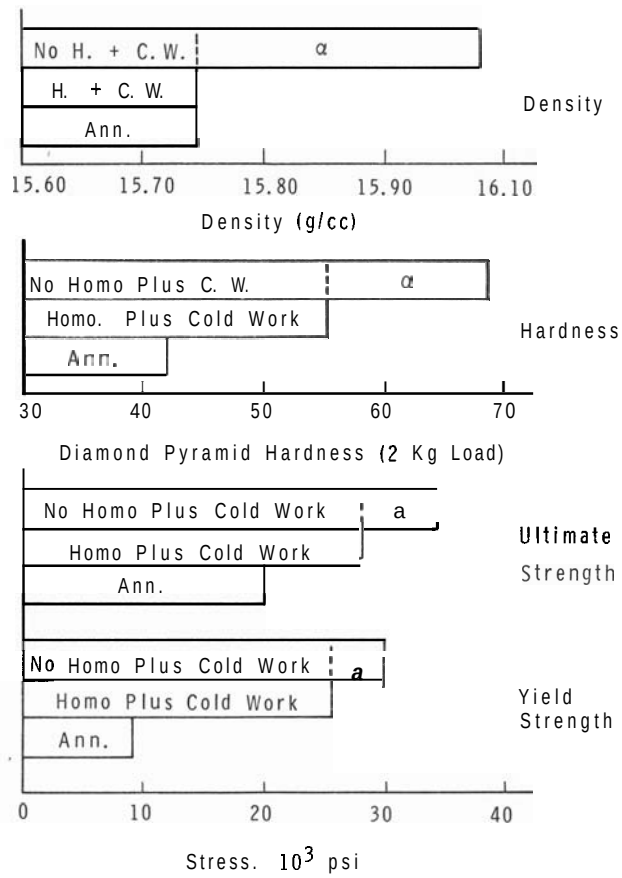


FIGURE 60

Effect of Cold Work and Alpha Formation on Properties of Pu-0.9 wt% Ga Alloys Rolled to 94% Reduction in Thickness

TABLE VIII

EFFECT OF COLD WORK, ALPHA PHASE FORMATION AND ANNEALING ON PROPERTIES OF Pu-0.94 wt% Ga ALLOY[a] ROLLED TO 94% REDUCTION[b]

Property[c]	Condition A, cored nonhomogenized, rolled to 94% reduction	Condition B, homogenized[d] rolled to 94% reduction	Increase in properties because of alpha phase formation	Conditions A and B after a 24 hr anneal at 450 C
Ultimate strength, psi[e]	35, 500	30, 300	5, 200	16, 200
0.2% yield strength, psi[e]	32, 200	28, 200	4,000	10, 900
Elongation in 1 in., %[e]	9.4	7.6	---	32.4
Elastic modulus, 10^6 psi[e]	8.1	6.8	1.3	5.5
Hardness, DPH - 2 kg load	69	55	14	42
Density, g/cm^3	16.08	15.74	0.34	15.74

(a) Total impurity content 676 pprn with 180 pprn carbon, 150 pprn chromium, 150 pprn nickel, 850 pprn iron and 50 pprn aluminum as major contaminants. As-cast density 15.74 g/cc.

(b) Final sheet thickness 0.050 in.

(c) Determined at 23 C.

(d) Annealed 200 hr at 425 C and furnace cooled.

(e) Testing speed: 0.015 in./min.

Rolled to 30% Reduction - Pu–0.89 wt% Ga sheet cast to 0.077 in. thickness was rolled to 30% reduction in both the as-cast and the homogenized, i.e., 24 hr at 450 C conditions. Properties determined are presented in Table IX. Rolling the as-cast sheet produced slightly more

TABLE IX

EFFECT OF A 30% REDUCTION ON THE PROPERTIES OF Pu–0.89 wt% Ga ALLOY[a, b]

Property	Cored, nonhomogenized rolled to 30% reduction	Homogenized,[d] rolled to 30% reduction
Ultimate strength, psi[e]	21,800	20,900
0.2% yield strength, psi[e]	20,500	19,700
Elongation in 1 in., %[e]	14.0	12.5
Elastic modulus, 10^6 psi[e]	6.7	6.4
Hardness, DPH - 2 kg load	53	51
Density, g/cm^3	15.84	15.80

(a) Total impurity content –700 ppm. As-cast density 15.77 g/cm^3

(b) Final sheet thickness: 0.055 in.

(c) Determined at 23 C.

(d) Annealed for 24 hr at 450 C and furnace cooled.

(e) Testing speed: 0.015 in./min.

strengthening than rolling the annealed sheet. This can be attributed to the formation of a greater amount of alpha phase in the cast and rolled sheet. It is interesting to note, however, that alpha formation occurred in both conditions during rolling. It is apparent that roll pressures exceeded the value critical for alpha formation in even the annealed and rolled sheet. A comparison of the properties of the 30 and 94% rolled alloy is illustrated in Figure 61. For the homogenized conditions, a 30% reduction produces a 93% increase in yield strength while a 94% reduction produces an increase in yield strength of 164%.

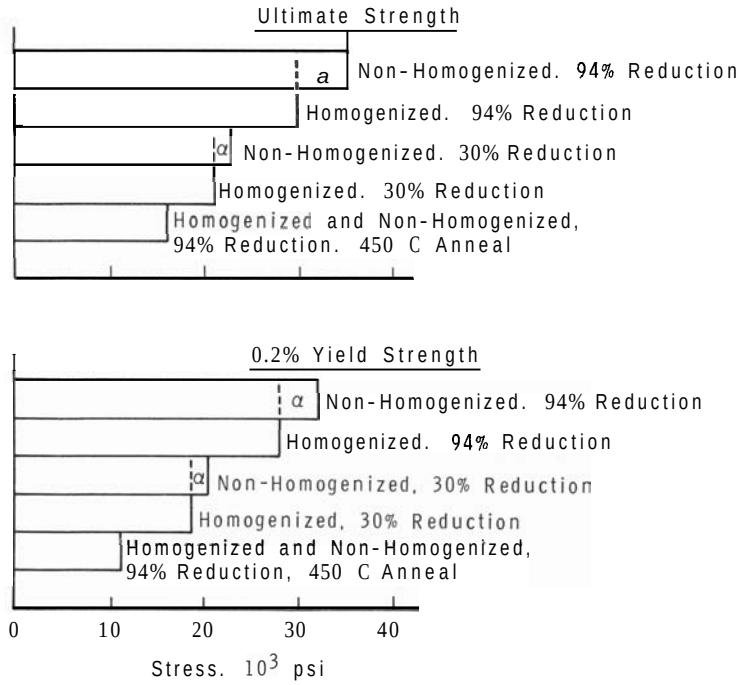


FIGURE 61

Effect of Cold Work and Alpha Formation on the Tensile Properties of Rolled Pu-0.9 wt% Ga Alloys

Compressive

Effect of Temperature and Testing Speed - The compression properties of 0.9 and 1.3 wt% Ga alloys prepared by dilution of master alloy have been determined up to 300 C, Figure 62. Again strength decreases and ductility increases as temperature increases. Testing speed in compression, except for the -32 C test temperature, does not affect yield strengths appreciably in the two alloys, Figure 63.

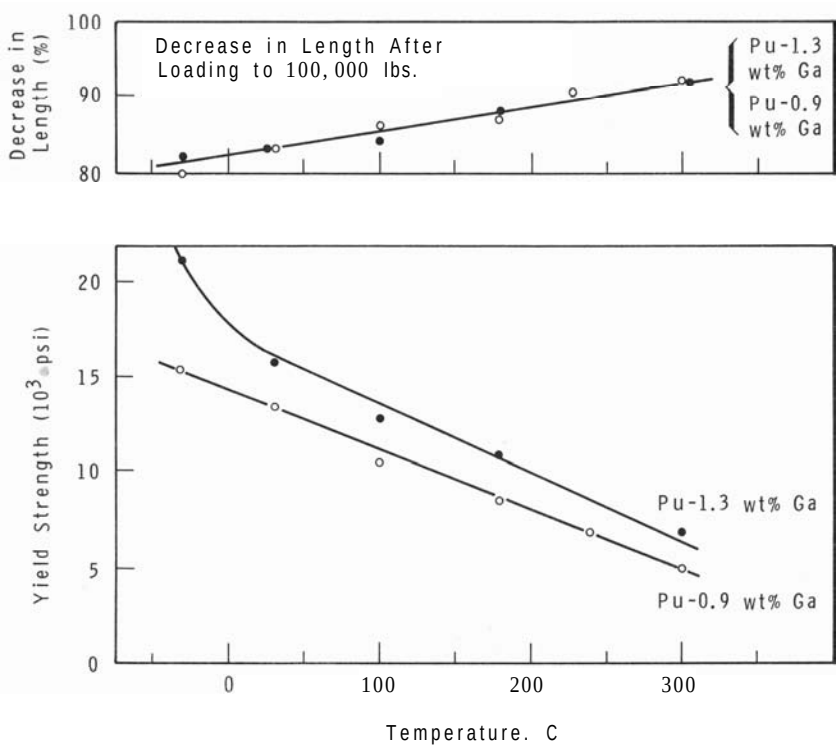


FIGURE 62

Effect of Temperature on Compression Properties of Alloys Testing Speed: 0.015 in./min

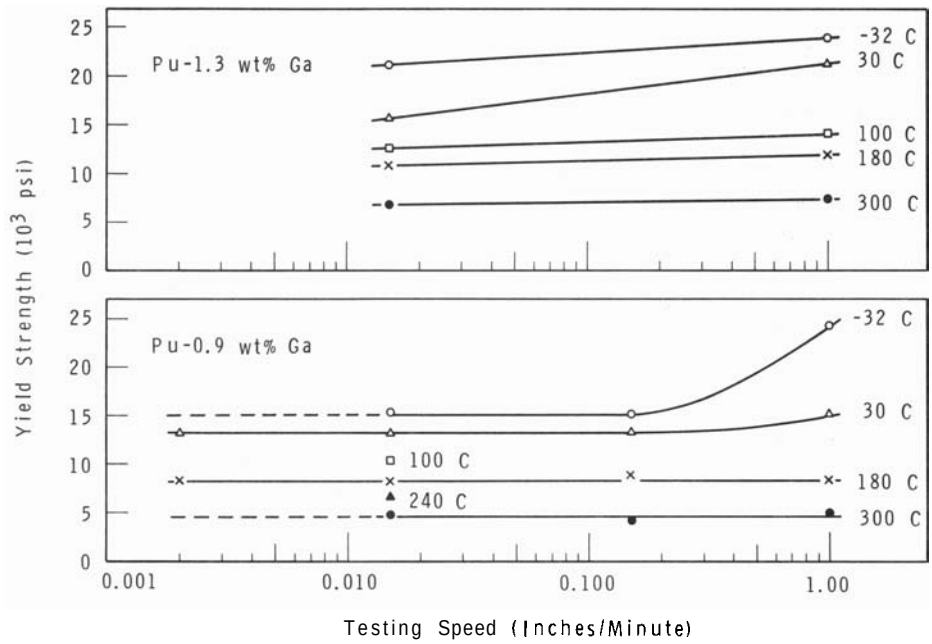


FIGURE 63

Effect of Testing Speed on Compressive Yield Strength of Alloys

Fatigue

Room temperature fatigue testing was done in the mean stress testing apparatus previously used for unalloyed plutonium.[11] Briefly, the fatigue specimen is vibrated in flexure by air pressure similar to a reed in the harmonica. The resultant stress cycle ranges from $\sigma_c + \sigma_m$ to $\sigma_c - \sigma_m$ where σ_c is the cyclic stress and σ_m is the mean stress. The cyclic stress was determined from the following relation:[12]

$$a_c = \frac{hE\,(\cos rl + \cosh rl)}{2(1-\nu^2)\,(\sin rl \sinh rl)}\; r^2 X,$$

where

h = thickness of fatigue specimen, 0.050 in.,

E = modulus of elasticity,

ν = Poisson's ratio,

rl = 1.875 radians,

l = free length of specimen, 1.57 in.,

X = one-half amplitude of vibration.

The mean stress was determined from the equation for a uniformly loaded cantilever beam:

$$\sigma_m = \frac{2hEX_o}{(1 - \nu^2)\, l^2},$$

where X is the constant deflection from horizontal of the free end of the speciment during vibration.

The specimen is nominally 0.050 in. thick, 0.625 in. wide, with a constant cross section, free length of 1.57 in. Unless specified otherwise, all specimens were taken with their longitudinal axis of symmetry parallel to the rolling direction.

Fatigue failure was taken at the point on the vibration frequency-time curve where the frequency had decreased 0.4%, i.e., one cycle/sec. Here, although failure had started, no visible crack could be detected when the specimens were forced to fracture by mechanical bending. Specimens allowed to vibrate until they stopped exhibited typical fatigue fractures. These originated in most cases at a corner near the start of the radius on the free length of the specimen where the maximum bending stress, $\sigma_c + \sigma_m$, was produced.

As will be seen in the discussionof the S-N curves for the different metallurgical conditions, no definite fatigue limit was observed. This is generally the case for those nonferrous materials which do not exhibit strain aging.[13] To provide a basis for comparison of the different metallurgical conditions studied, fatigue strength will be defined as the cyclic stress below which failure does not occur between 10^7 to 10^8 cycles.

To evaluate the effect of metallurgical conditions on the fatigue behavior of the Pu–1.0 wt% Ga alloy, the following condition; were fatigue tested:

- Cored, as-cast, milled to size, average grain size 0.005 mm,
- Cored, 86% cold-rolled,
  - (1) Milled specimens, contains 15% alpha phase,
  - (2) Punched specimens, contains 10% alpha phase.

- Cored, 86% cold-rolled, specimens punched, longitudinal axis of symmetry, transverse to rolling direction, contains 12% alpha phase.
- Cored, 86% cold-rolled, annealed 0.5 hr at 300 C, furnace cool, specimens punched, no alpha phase,
- Homogenized for 192 hr at 425 C, 94% cold-rolled, no alpha phase,
- Homogenized for 192 hr at 425 C, 94% cold-rolled, annealed for 0.5 hr at 300 C, no alpha phase.

The properties which may influence fatigue behavior for the above conditions are presented in Table X. Note that 0.1% yield strength ranges from 10,300 to 30,400 psi for metallurgical conditions ranging from as-cast to 94% cold-rolled. Modulii of elasticity above 7.3×10^6 psi indicate that alpha phase with its 14×10^6 psi modulus is present. The density data confirm this observation. Poisson's Ratio for the wrought alloy is roughly 25% lower than in the as-cast or annealed alloys, indicating a directionality effect. Total impurity contents and surface finish for the conditions tested are comparable.

The S-N curve for cored, as-cast, and milled alloy is illustrated in Figure 64. Despite the scatter existing, the data suggest a fatigue strength of 10,000 psi. Internal casting defects observed on radiographs of some of the specimens are believed to be at least part of the reason for the scatter in the data. The 15.81 g/cc density on the specimens instead of the expected 15.76 g/cc value indicates that alpha phase formation occurred on the surface during milling. The formation of alpha phase results in strengthening and a volume contraction. The surface therefore may be in a complicated state, i.e., residual compressive stresses from milling and/or residual tensile stresses plus strengthening from alpha formation. To further complicate the picture, the 5,840 psi average mean stress during testing is 53% of the 10,300 psi 0.1% yield strength which by itself would tend to lower the fatigue strength.[14] As will be shown later the 10,000 psi fatigue strength for homogenized, wrought, annealed and punched specimens indicate that the 10,000 psi fatigue strength for the as-cast, cored and milled alloy may be slightly high.

Two S-N curves for the cored, as-rolled alloy are presented in Figure 64. One curve is for milled specimens and the other for specimens punched

TABLE X

PROPERTIES OF METALLURGICAL CONDITIONS TESTED

Material Condition	Approx. 0.1% Yield Strength, psi	Approx. Ultimate Strength, psi	Cyclic Fatigue Strength, psi	Average Mean Stress, psi	Modulus of Elasticity,[a] 10^6 psi	Poisson's Ratio[b]	Density, g/cc	Diamond Pyramid Hardness, 2 kg Load	Gallium Content, wt%	Total Impurity Content, ppm	Surface Finish Range (RMS)	Average Vibration Frequency, cycles/sec
Cored, as-cast, milled to size, average grain size.	10, 300	16, 200	10,000	5, 840	6. 0	0. 245	15.81	46	0. 92	215	30-70	205
Cored, 86% cold-rolled												
Milled specimens	30, 400	35, 500	15,000	6, 740	8. 2	0. 160	16. 31	60	0. 88	355	20-25	218
Punched Specimens	30, 400	35, 500	14, 000	5, 400	7. 5	0. 166	16. 12	72	0.95	340	7-20	234
Cored, 86% cold rolled, specimens punched, longitudinal axis of symmetry, transverse to rollingdirection.	30, 400	35,500	13,000	-----	7. 4	0. 199	16. 20	72	0. 91	1364	8-18	232
Cored, 86% cold rolled, annealed 0. 5 hr at 300 C, furnace cool, specimens punched.	10, 300	16, 200	13, 000	6, 290	6. 1	0. 251	15. 77	43	1. 00	340	8-28	212
Homogenized 192 hr. at 425 C, 94% cold rolled	27, 400	30, 300	12,000	5, 670	7. 3	0. 183	15. 74	56	0. 92	352	10-25	228
Homogenized 192 hr at 425 C, 94% cold rolled, annealed 0. 5 at 300 C.	10, 300	16, 200	10, 000	6, 000	5. 5	0. 227	15. 74	43	0. 92	352	13-27	218

(a) Determined both with a linear differential transformer and 90° rosette strain gage.

(b) Determined with a 90° rosette strajn gage.

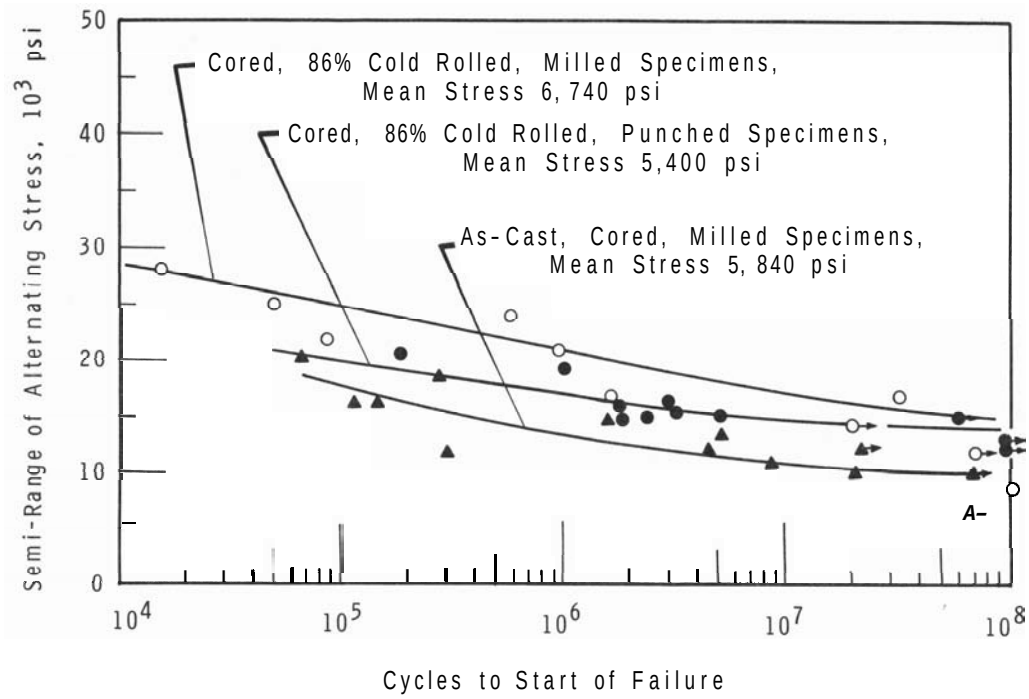


FIGURE 64

S-N Diagram for Pu–1.0 wt% Ga Alloy in As-Cast and Rolled Conditions

from as-rolled sheet which had been rolled to final thickness. The fatigue strengths for the milled and the punched specimens are approximately 15,000 and 14,000 psi, respectively. Since both fabrication methods produce comparable surface finishes, the higher fatigue strength for the milled specimens may be the result of their slightly larger amount of alpha phase, particularly on the milled surface. The mean stresses during testing were 6,740 and 5,400 psi, respectively, for the milled and the punched material. Since these are only approximately 20% of the 30,400 psi, 0.1% yield strength it is not expected that mean stress influenced the fatigue results.[14]

A comparison of S-N data from specimens punched both parallel and transverse to the rolling direction from the same sheet is illustrated in Figure 65. The fatigue strengths for both are essentially the same which is not unusual for a wrought cubic structure with a minimum of inclusion stringers,.

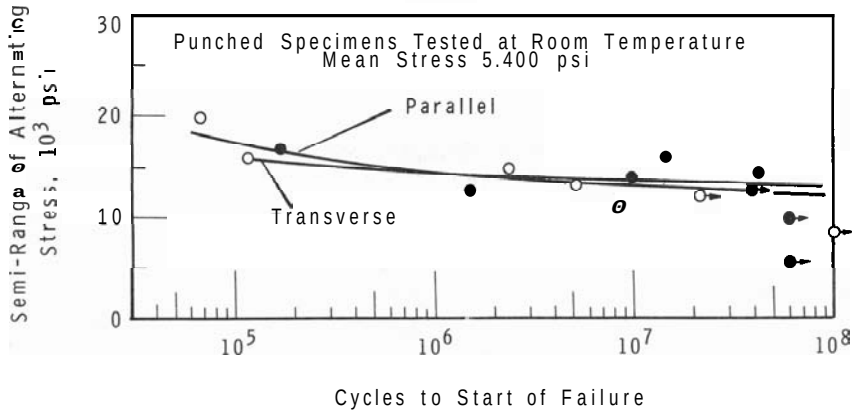


FIGURE 65

S-N Diagram for 86% Cold-Rolled, Cored Pu–1.0 wt% Ga Alloy Tested Parallel and Transverse to Rolling Direction

Annealing the cored, as-rolled alloy in vacuo for 30 min at 300 C produces a dead soft delta phase condition with no alpha phase. The 0.1% yield strength decreased from 30,400 to 10,300 psi and the hardness decreased from 72 to 43 DPH. The fatigue strength, however, decreased only slightly from 14,000 psi before the anneal to 13,000 psi after the anneal, Figure 66. This indicates that fatigue strength is relatively insensitive to magnitude of yield strength in the alloy. The 6,380 psi mean stress in the annealed condition is 62% of the 10,300 psi 0.1% yield stress and as in the as-cast alloy may have had an effect on the indicated fatigue strength.

The homogenized, i.e., 192 hr at 425 C, and 94% rolled alloy had a 12,000 psi fatigue strength which was only slightly higher than the 10,000 psi fatigue strength for the same material after the 300 C, 30 min anneal, Figure 66. Both fatigue strengths were somewhat lower than those obtained on similarly fabricated and annealed alloy in the cored condition. In the cored alloy, either the presence of alpha phase or its formation from the compressive stresses introduced during fatigue testing may have caused strengthening and increased fatigue resistance. In the homogenized and rolled condition the 5670 psi mean stress was only 20% of the 27,400 psi, 0.1% yield stress; therefore, having little, if any, effect on fatigue strength. In the annealed condition the mean stress was 54% of the 10,300 psi, 0.1% yield stress which could have an adverse effect on fatigue strength as suggested for previous dead soft conditions.

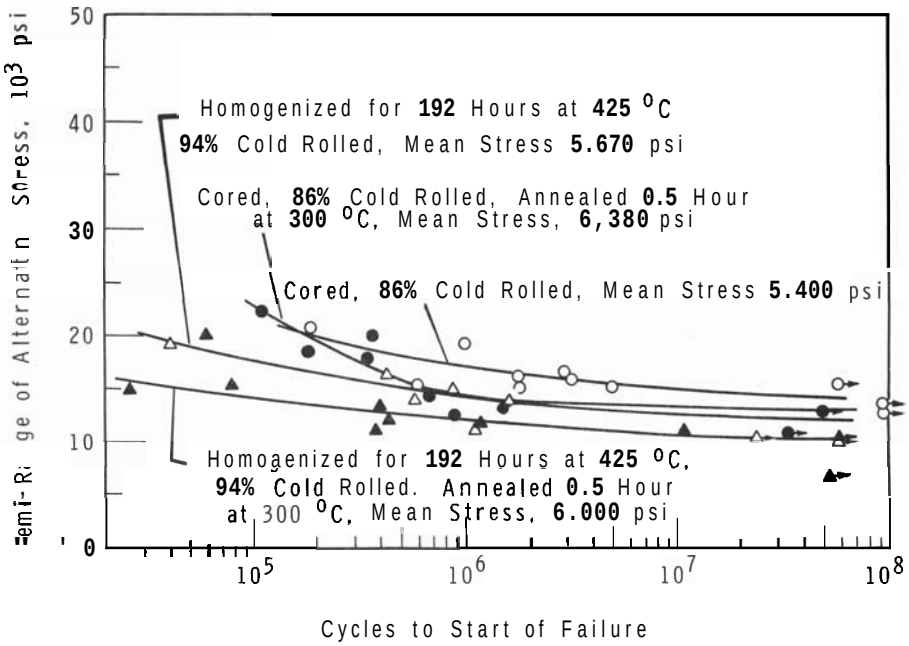


FIGURE 66

S-N Diagram for Rolled Pu–1.0 wt% Ga Alloy in Cored, Homogenized and Annealed Conditions, Punched Specimens Tested at Room Temperature

From a visual standpoint, oxidation of the specimens during testing was minimal. The normal oxide tarnish which forms soon after machining was not visibly affected.

It can be concluded that fatigue strength is relatively insensitive to yield strength, i.e., metallurgical condition. Over a 0.1% yield strength range of 10,300 to 30,400 psi, fatigue strength varied only from 10,000 to 15,000 psi. Fatigue strength is slightly higher in both conditions of the cored, wrought alloy. This is possibly attributable to either the presence of alpha phase or its formation by the cyclic compressive stresses during fatigue testing. In either case the alpha phase imparts additional strengthening.

ACKNOWLEDGMENT

My appreciation is extended to I. B. Mann for his outstanding support in all phases of this work. The rate of accomplishment was greatly increased by his efforts. Thanks are due W. C. Rossiter for his fine contribution in casting and testing the elevated temperature tensile specimens.

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2	NATIONAL LEAD COMPANY OF OHIO
1	NAVAL POSTGRADUATE SCHOOL
3	NAVALRESEARCH LABORATORY
1	NRA. INC.
1	NUCLEAR MATERIALS AND EQUIPMENT CORPORATION
1	NUCLEAR METALS, INC.
1	NUCLEAR UTILITY SERVICES, INC.
1	OFFICE OF ASSISTANT GENERAL COUNSEL FOR PATENTS (AEC)
2	OFFICE OF NAVAL RESEARCH
1	OFFICE OF NAVAL RESEARCH (CODE 422)
1	OHIO STATE UNIVERSITY
1	PETROLEUMCONSULTANTS
4	PHILLIPS PETROLEUM COMPANY (NRTS)
1	PHYSICS INTERNATIONAL, INC.
1	PICATINNY ARSENAL
1	POWER REACTOR DEVELOPMENTCOMPANY
3	PRATT AND WHITNEY AIRCRAFT DIVISION
1	PURDUE UNIVERSITY
1	RADIOPTICS, INC.
1	RAND CORPORATION

Ptd.	Standard Distribution
1	REACTIVE METALS. INC.
1	REACTIVE METALS. INC., ASHTABULA
1	RENSSELAER POLYTECHNIC INSTITUTE
1	SAN FRANCISCO OPERATIONS OFFICE
2	SANDIA CORPORATION. ALBUQUERQUE
1	SANDIA CORPORATION, LIVERMORE
1	SOUTHWEST RESEARCH INSTITUTE
1	STANFORD UNIVERSITY (SLAC)
1	SYLVANIA ELECTRIC PRODUCTS, INC.
1	TENNESSEE VALLEY AUTHORITY
1	TRWSPACE TECHNOLOGY LABORATORIES (NASA)
1	UNION CARBIDE CORPORATION. CLEVELAND
2	UNION CARBIDE CORPORATION (ORGDP)
5	UNION CARBIDE CORPORATION (ORNL)
	UNION CARBIDE CORPORATION (PADUCAH PLANT)
2	UNITED NUCLEAR CORPORATION (NDA)
1	U. S. GEOLOGICAL SURVEY. DENVER
1	U. S. GEOLOGICAL SURVEY, MENLO PARK
1	U. S. GEOLOGICAL SURVEY, WASHINGTON
1	U. S. PATENT OFFICE
2	UNIVERSITY OF CALIFORNIA, BERKELEY
2	UNIVERSITY OF CALIFORNIA, LIVERMORE
1	UNIVERSITY OF PUERTO RICO
1	WESTERN RESERVE UNIVERSITY (MAJOR)
4	WESTINGHOUSE BETTIS ATOMIC POWER LABORATORY
1	WESTINGHOUSE ELECTRIC CORPORATION
1	WESTINGHOUSE ELECTRIC CORPORATION (NASA)
325	DIVISION OF TECHNICAL INFORMATION EXTENSION

*New listina a change in old listina.