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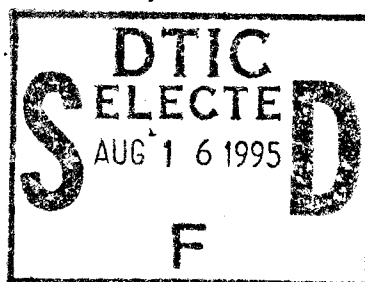
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UNITED STATES ATOMIC ENERGY COMMISSION

PREPARATION OF ALPHA URANIUM SINGLE  
CRYSTALS. PART I. PHASE TRANSFORMATION  
METHOD

Final Report—Metallurgy Program 2.1.4

By  
E. S. Fisher



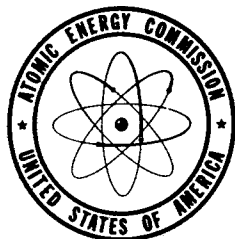
November 18, 1953

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ARGONNE NATIONAL LABORATORY  
P. O. Box 299  
Lemont, Illinois

PREPARATION OF ALPHA URANIUM SINGLE CRYSTALS  
Part I  
PHASE TRANSFORMATION METHOD

by

E. S. Fisher

Final Report - Metallurgy Program 2.1.4

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## PREFACE

The following report is Part I of a series of 3 reports covering Metallurgy Program 2.1.4, Uranium Single Crystal Investigations. The object of the program was to develop a method for preparing single crystals of alpha uranium. Three methods were investigated, the results of which appear in separate reports as follows:

Part I - Phase Transformation Method, ANL-5159

Part II - Strain-Anneal Method, ANL-5160

Part III - Grain Coarsening Method, ANL-5021

Some highly imperfect single crystals were prepared by the phase transformation method. A large number of good quality single crystals have been prepared by the grain coarsening method, which has proved to be reliable although not fully developed. The availability of high purity uranium is responsible for the success of this method. The strain-anneal study consisted of a large number of experiments with specimens of differing purity. No single crystals were produced by strain-anneal techniques; however, the data show the large influence of impurities on grain growth and recrystallization in alpha uranium. Because the data offer a background for explaining the grain coarsening method, the strain-anneal report appears as Part II.

# PREPARATION OF ALPHA URANIUM SINGLE CRYSTALS

## PART I PHASE TRANSFORMATION METHOD

by

E. S. Fisher

### I. INTRODUCTION

The solid state phase transformations which occur when cooling uranium from the melting point to room temperature precludes the utilization of the various methods for preparing single crystals by controlled cooling from the liquid state. Any single crystal of gamma phase uranium which could be formed would be susceptible to recrystallization in cooling through the transformation temperatures of gamma to beta and beta to alpha at approximately 770°C and 660°C, respectively. An attempt has been made to utilize a modification of the controlled cooling methods in which solid specimens were heated to and slowly cooled from the beta phase. The method investigated was a modification of the Bridgman<sup>(2)</sup> technique for growing single crystals from a melt. Specimens which were pointed at one end were gradually moved through a gradient of decreasing temperature from temperatures at which the beta phase is stable in order to encourage a single nucleus of alpha phase to form at the extreme point and serve as a seed for growth of a single alpha crystal from the transforming aggregate of beta grains.

The object of this investigation was to determine the feasibility of preparing uranium single crystals similar in structural perfection to those prepared by the grain coarsening procedure (see Part III, ANL-5021). In this respect, the investigation has not been completed. Although it was found feasible to prepare single crystals of relatively large dimensions, the structural perfection of the single crystals was far from desirable for most purposes.

Besides the conditions which are inherited in adopting the Bridgman method, such as material composition, rate of specimen travel, and degree of temperature gradient, the size, shape, and penultimate structure of the specimens were also varied. With the apparatus used, the degree of structural perfection could not be significantly changed. The bulk of this investigation was concerned with determining the conditions under which the imperfect single crystals could be made so large as to fill the cross section of the rod.

Among the 18 experimental specimens, only one specimen of high purity material was included. The results with this specimen indicated that material composition is a large factor. As the effects of most of the other

variables were investigated using lower purity material, the conclusions indicated by the results may not be completely valid when considering high purity material.

In the following report a general description of the structures produced by this treatment is presented. This is followed by a summary of the indicated effects of the variables on the structures. A small amount of information obtained by X-ray diffraction is presented.

## II. APPARATUS AND EXPERIMENTAL PROCEDURE

Most of the experiments were carried out in liquid NaK in a furnace which was constructed primarily for this investigation but which was also used in the investigation of the strain-anneal method. Three other types of furnaces were used for isolated experiments to investigate the effects of varying the temperature gradient.

### IIa: Liquid NaK Furnace

This furnace was designed so that uranium specimens could be lowered at various speeds through a column of NaK in which a gradient of temperature existed along its length. There are several advantages to the use of NaK alloy<sup>(3)</sup> as a liquid metal medium for heat treating uranium. Firstly, the melting point of NaK alloys over a wide range of compositions remains below room temperature, and the vapor pressures of the constituents are relatively low at temperatures up to 750°C. Secondly, uranium is apparently not attacked by NaK during heating in this temperature range. Thirdly, the similarity of the thermal conductivity coefficients of NaK and uranium provide a means of indirectly measuring the temperature of the specimen as it passes through a temperature gradient, since it can be assumed that the temperature of the uranium at any point is similar to that in the surrounding NaK. Since NaK is not corrosive to stainless steel<sup>(4)</sup> there is no container problem.

The design of the NaK furnace is shown in Figure 1. The furnace consists of a vertical cylindrical tank, 10 in. in diameter x 13 in. in length, through the center of which passes a 1 in. diameter specimen transfer tube approximately 48 in. long. The tank and center tube are both partially filled with NaK alloy of eutectic composition (23 w/o Na), which is liquid between 12.5°C and 760°C at one atmosphere. The NaK in the tank is in direct contact with the outside of the center tube. A "Calrod" resistance heater is immersed in the tank of NaK and wound about the center tube at the inside base of the tank. A second "Calrod" heater is wound about the outside of the tank.

The NaK in the center tube is heated by conduction from the tank through the tube wall. A coil of 3/8 in. diameter copper tubing is wound about and soldered to the center tube as it protrudes from the base of the tank. This coil is in direct contact with the base of the tank so that during operation the NaK just below the heated length is subjected to vigorous cooling.

The NaK level in the tank is within 3 in. of the top and is covered with an argon atmosphere. To prevent convection currents within the transfer tube, the NaK volume in the transfer tube at room temperature was selected so that the top level of the NaK was always below that in the tank.

The space above the NaK in the tube is filled with argon to prevent boiling of the NaK and to protect against oxygen entrance through probable small system leaks. A gate valve is used to close off the tube system when inserting or extracting specimens. Cooling coils are used to prevent temperature increases at the valves and gages.

The tank temperature and, consequently, the maximum temperature of the NaK in the tube, was controlled from a thermocouple in a well between the tube and tank walls. Because of the large heat losses to the cooling water, a high current-low current control, rather than on-off type, was used to prevent gross temperature fluctuations. A thin-walled tube passing through the NaK column of the transfer tube and out through the tube walls was used for passing thermocouples to record the temperature distribution in the NaK column.

In the phase transformation experiments the pointed specimens were placed in tantalum supports (Figure 2) so that the tapered end rested in a similarly tapered hole in the lower tantalum disc. The specimen was held vertical by inserting the top of the specimen through a hole of slightly larger diameter in the upper tantalum disc. The tantalum discs were held horizontally parallel by wire supports which did not touch the specimens. The pointed ends of the specimens generally protruded at least 1/8 in. below the lower tantalum disc.

The power to the Calrod heaters in the NaK furnace was controlled so that the maximum temperature of the NaK bath in the transfer tube was regulated at approximately 750°C. As the top level of the NaK in the transfer tube was at approximately 30 in. below the furnace top and was in the maximum temperature zone, a temperature gradient in the transfer tube was obtained in the gas above the maximum temperature zone. The temperature distribution along the transfer tube is shown in Figure 3. The maximum temperature was maintained at 750°C for all experiments. Under this condition the temperature gradient between 670°C and 650°C was approximately 244°C per inch in the region below the maximum temperature zone. It should be noted that the temperature gradient could have been varied by varying the maximum temperature.



Before heating the furnace, the specimen in the tantalum holder was inserted into the transfer tube so that the point of the specimen would be at the desired starting temperature when the maximum NaK temperature was reached. The tantalum holder was supported by a nichrome wire which extended out of the system through a lubricated gum rubber vacuum seal to a metal spool, about which the top end of the wire was wound. After the NaK was heated to temperature, the spool was rotated by a Telechron clock motor so that the specimen descended through the hot NaK to the cold zone at a desired constant rate.

To remove the specimens from the transfer tube the furnace was first cooled to room temperature, so that the specimen could be lifted back through the NaK column to the chamber above the valve, the valve was closed, and the chamber removed. The adhering NaK was removed by bathing the specimens in butyl alcohol or water.

#### Temperatures of specimens prior to lowering through NaK furnace

In the 16 experiments carried out in the NaK furnace, the specimens were generally positioned so that the points were at 575°C to 600°C prior to the start of travel. Consequently, the specimens traveled into the beta phase through a temperature gradient. One specimen was placed in the NaK so that the full length was heated to 750°C simultaneously prior to the start of travel.

#### Various speeds of lowering in furnace

By varying the Telechron motor speed or by varying the diameter of the attached spools, any desired rate of specimen lowering could be attained. The travel rates used in the different experiments varied from 0.1 in./hr to 10 in./hr. For purposes of possible comparison with other systems, the range of cooling rates can be expressed as 24° per hr to 2440° per hr respectively on the basis of a 244°C per in. temperature gradient between 670°C and 650°C (the temperature of transformation).

#### IIb. Nichrome Wound Tube Furnace

A schematic sketch of this simple furnace and accompanying apparatus is shown in Figure 4. By passing a constant current through a coil of nichrome wire wound about a 12 in. long x 1 in. diameter ceramic tube, a natural temperature gradient of approximately 50°C per in. was obtained when the maximum temperature was maintained at 710°C. Using this furnace and the above conditions, a specimen was sealed in an evacuated Vycor capsule, and the capsule was gradually lowered through the full length of the furnace at 0.6 in./hr (30°C per hr) by a Telechron clock system as described above.

### IIc. Uranium Filament Furnace

A third type of apparatus was partially developed in an effort to obtain sharper temperature gradients than was feasible in the NaK furnace. In this apparatus the heat was produced by passing an electric current through the uranium specimen in an assembly similar to the "Vacuum Filament Crystal Device" designed at KAPL<sup>(5)</sup>. Some small changes of the KAPL design were introduced; the apparatus was constructed and assembled, and some experiments were made to investigate the operating procedure. Although it was found feasible to produce sharp temperature gradients, there were many factors that required investigation before adequate control of the procedure could be developed. It was difficult to justify the time necessary to make these investigations in light of more promising approaches to the general problem; consequently, work on this apparatus was halted. As the feasibility of achieving the purpose of the apparatus has been somewhat established, a description of the apparatus and the state of development is reported here.

Figure 5 is a schematic drawing of the assembly at the final stage of development. The specimen was supported vertically by clamping the upper end to a copper electrode. The mercury pool acted as the lower electrode when in contact with the specimen. Three tungsten electrodes which were sealed through the Pyrex carried the current between the mercury in the system and an outer mercury pool which was contained in a copper cup. The copper cup was connected to the upper copper electrode through cables which formed the secondary windings of a transformer. A high capacity rheostat was used to vary the primary input from a constant voltage transformer which eliminated line voltage fluctuations.

As the copper electrode was cooled by water flowing through a hollow center well and the mercury was in contact with the water-cooled Pyrex walls, only that part of the specimen between the copper electrode and the mercury pool increased in temperature significantly during passage of the current. The height of the mercury column was regulated by varying the pressure head in the mercury reservoir through the valve orifices. The space above the mercury pool was continuously evacuated to maintain a constant pressure and to prevent oxidation of the specimen surface during heating. The temperature distribution was recorded by 6 chromel-alumel thermocouples spot welded to the specimen at 1/4 in. intervals along the length.\* The thermocouple leads were extended out of the system through Stupakoff insulated tubes which were soldered to the flange at the top of the copper electrode. Apiezon wax was used to insulate the bare lead wires from the metal tubes and to prevent the leakage of air into the system.

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\*The very short welding time was insufficient to cause any changes in the microstructure beneath the surface of the specimen. A brittle phase was observed at the very point of thermocouple-uranium contact.

It was planned to use the apparatus for both the transformation method and for the strain-anneal method. In the preliminary tests for the transformation method, the height of the mercury pool was first adjusted so that only the lower  $1/4$  in. length of the specimen was immersed (the specimens were approximately 3 in. in length). The specimens were then heated by passing sufficient current so that the maximum temperature of the specimen (at a section midway between the mercury and the copper rise) was  $700^{\circ}\text{C}$  to  $725^{\circ}\text{C}$ . For specimens of 0.150 in. diameter, approximately 175 amp were necessary. Under these conditions, approximately  $1-1/2$  in. of the specimen were in the beta phase, and the temperature gradient between  $700^{\circ}\text{C}$  and  $600^{\circ}\text{C}$  was approximately  $350^{\circ}\text{C}$  per in. The mercury level was then forced upwards in steps of  $1/2$  in., and thermocouple readings recorded at each  $1/2$  in. step. Also at each step, the current was increased to maintain a  $700^{\circ}\text{C}$  maximum temperature; this increased current was necessary to compensate for decrease in electrical resistance and for increased cooling effects. It was found that the length of specimen between  $700^{\circ}\text{C}$  and  $600^{\circ}\text{C}$  decreased as the mercury level rose, so that a  $700^{\circ}\text{C}$  per in. temperature gradient existed when  $1-1/2$  in. of the specimen length were above the mercury pool.

This stepwise operation was, of course, not suitable for growing single crystals, since the full length of the specimen was cooled almost simultaneously into the alpha phase each time the mercury level was raised and before the current was increased. Satisfactory operation would require a gradual movement of the section of the specimen undergoing transformation. To attain these conditions, a system was planned whereby the current would be gradually increased simultaneously with gradual elevation of the mercury level. This system was to consist of a controlled slow leak of gas into the space above the mercury reservoir, for gradual elevation of the mercury pool, and a program operated variable transformer for increasing the current as a function of time. The current-time program was to be first determined for a specific rate of mercury elevation and for specific specimen dimensions. For reasons noted above, the plans were not carried out.

#### IId. Cadmium Bath Unit

To obtain a smaller temperature gradient than was possible with the NaK furnace and to incorporate the desirable features of heating in contact with a liquid metal, without the inconveniences of a NaK system, a tube was constructed by which the pointed specimens were to be drawn out of a bath of liquid cadmium which was uniformly heated to approximately  $675^{\circ}\text{C}$ . The cadmium was heated by inserting the tube vertically into a lead bath in a pot furnace. The space above the cadmium was evacuated and filled with argon gas prior to heating. The system was sealed by a neoprene gasket between a top screw cap and a flange on the tube. Although the neoprene gasket was approximately 15 in. above the hot lead bath, it was necessary to cool the top of the tube by passing water through 3 turns of copper coil wound above the top of the tube.

The specimens were transported by a system similar to that used in the NaK furnace. The nichrome wire supporting the specimen holder extended out of the tube through a gum rubber seal at the top end of the tube and was wound about a spool of a clock motor. The specimens could be pulled out of the cadmium bath, point first, at any desired rate.

Although the system did not provide for the transformation to take place within the liquid metal medium, as in the NaK furnace, the temperature distribution was such that the lower end of a specimen, which was over 1-1/2 in. in length, would be in the liquid metal when the point started to transform. Under these conditions a temperature gradient of approximately 10°C per in. would exist along the specimen length. Because of the large heat capacity of the system, including the lead bath, the temperature fluctuations in the cadmium bath were approximately 2°C.

Before deciding on the use of cadmium a number of other low melting metals were considered from the point of view of corrosion of the uranium specimens. Cadmium showed the least rate of attack after 66 hr at 720°C. A very slight attack, which was considered insufficient to preclude the use of cadmium as a heating medium, was observed.

The first and only experiment with this system failed because of the condensation of the cadmium vapors at the top of the tube and about the nichrome supporting wire. During the test the condensed metal prevented passage of the wire through the gum rubber seal and the wire fractured.

No further experiments have been made with this system. Some means of preventing the condensation of cadmium on the supporting wire is necessary for successful operation.

### III. MATERIAL

The uranium specimens used in this study were machined from rods of 4 different materials.

Specimens Lots A-1 and B were relatively impure, containing high carbon concentrations and appreciable quantities of other constituents. Lot J was relatively pure (compared to Lots A-1 and B) with respect to constituents other than silicon. Lot G-1 was relatively pure with respect to all impurity constituents.

The rods from which Lot A-1 was obtained were prepared from a piece of uranium cut from a large biscuit. The round piece was heavily reduced in cross-section (87%) by rolling at 300°C. Lot B was obtained from a rod which was prepared by rolling of a Hanford slug at 300°C.

The specimens of Lot J were machined directly from a small ingot (approximately 200 g) made by liquation melting of solid pieces of biscuit metal.

Lot G-1 consists of only one specimen. This specimen was machined from a rod which was produced by rolling a small ingot of high purity uranium, prepared by vacuum melting of electrolytically prepared uranium crystals.<sup>(6)</sup>

#### IV. SPECIMEN SIZES AND SHAPES

Three types of specimen shapes were used in this investigation. Most of the specimens were pencil-shaped rods. Five of the specimens were machined to form constricted areas above a larger diameter lower end, for reasons which are discussed below. One specimen was ground to form a rectangular cross-section above a taper of circular section.

The degree of taper to form the points remained the same regardless of specimen shape or diameter. The tapers were machined so that the point formed a 30° included angle.

The diameters of the pencil-shaped specimens varied. The 2 specimens of Lot B were of 0.375 in. diameter. The specimens of Lot A-1 were 1/4 in. and 1/8 in. in diameters. The specimens of Lot J were of 0.185 in. diameter. The high purity specimen was of 0.210 in. diameter. With the exception of the high purity specimen, which was 1-1/2 in. long, the specimens machined from rolled rods were 2 in. lengths. The specimens machined from the ingot for Lot J were 1 in. in length.

The contoured specimens were machined from pencil-shaped specimens of Lot A-1 of 0.200 in. diameter. These specimens were shaped so that a constriction existed above the tapered end. One set of specimens consisted of 3 tapered sections as shown in Figure 7a, b, and c, whereas, the other set consisted of 2 tapers (Figure 7e). In both sets the constricted lengths were 1/16 in. in diameter.

The specimen of rectangular cross-section was approximately 1/8 in. x 3/32 in. x 2-1/2 in. in dimensions (Figure 11).

#### V. EXAMINATION PROCEDURE

##### Va. Visual Examination

To observe the surface grain size resulting from experiment, the specimens were macro-etched electrolytically using an aqueous electrolyte consisting of trichloroacetic acid (500 g/l) and hydrochloric acid (20 cc/l).

Although this type of etching procedure revealed large differences in orientations, the degree of sub-structure was not readily revealed. Some specimens were sectioned and metallographically polished for micro-examination of the sub-structure using polarized light.

To reveal the sub-structure for macroscopic examination of polished surfaces, some specimens were anodically etched in an aqueous citric-nitric acid electrolyte.<sup>(7)</sup> As shown in Figure 8d this type of etch reveals small differences in orientation. A slight oxide surface film after this type of etching further emphasizes small orientation differences.

#### Vb. X-ray Technique

A number of back reflection Laue photograms were taken from various grains and at various points within the crystals. The unfiltered radiation from a copper target tube, operating at 45 kv, was collimated through a 0.6 mm pinhole. The specimen to film distance and exposure times were 3 cm and 2 hr, respectively. Under these conditions the Laue spots were of low intensity and in some cases were not sufficient in number or clarity for orientation determinations. The orientations which were determined indicated that the citric-nitric acid etchant revealed orientation differences in the order of  $2^\circ$ . The orientations of the grains and appearance of the Laue spots are discussed below.

### VI. RESULTS

#### Vla. General Description of Results

Although the number of grains and the degree of sub-structure varied as a result of experiments under different conditions, the grain size distributions within the specimens, the shapes of the grains, and the nature of the sub-structure were generally similar. Variations from the general pattern and the observed effects of varying conditions are described in later sections.

**General Appearance of Grains:** The grains in the lower tapered sections of the specimens, i.e., that part of the specimen which was first to pass through the transformation, were of small size and appeared to be of widely varying orientations (Figure 6b). The number of grains generally decreased with distance from the point. The grains which grew out of the tapered section were of elongated shape (Figure 6a). Micro-examination of longitudinal sections revealed that the grain boundaries were generally parallel and were aligned in directions from  $10^\circ$  to  $30^\circ$  from the rod axis. In some specimens, grains which did not originate in the tapered sections were observed. These grains appeared to have nucleated at the specimen surfaces and to have grown in directions parallel to the long axes of grains originating in the tapered sections.

**General Appearance of Sub-structure:** With the exception of some grains in one specimen, all of the grains microscopically observed contained sub-structure. No attempt has been made to critically study the sub-structures, but there appeared to be 2 general types of sub-grains within the more definite grain boundaries. The predominant type of sub-grain was columnar in shape with boundaries aligned in the general directions of the rod axis (Figure 6c). In some grains, the sub-boundaries were quite definite in at least one position of the microscope stage, whereas, the sub-structure in other grains was revealed only by careful observation while rotating the microscope stage. As most grains contained twin bands, the sub-structure was also revealed by abrupt changes in direction of the twins at the sub-boundaries. This phenomenon is shown in Figure 6g. Some of the sub-boundaries extended the length of the grain boundary, but, in most cases, the sub-grains were of relatively short length.

A sub-structure consisting of equi-axed small blocks was observed less often (Figure 6h). The experiments which produced this type of structure were somewhat abnormal with respect to the bulk of the experiments and are described below.

**Specimen Bending:** The specimens of diameters below 0.150 in. were very obviously macroscopically bent as a result of traveling through the temperature gradient from the beta phase. In the pencil-shaped specimens, the bend was found at different points along the lengths of different specimens. In the specimens which contained constrictions, the bending was found to have occurred at the smallest diameter. Figures 7a, b, and c show the nature and degree of bending in the constricted specimen. All 3 photos are of the same specimen. For Figures 7a and c, the specimen was positioned so that the bend was not apparent, whereas, for Figure 7b, the profile formed by the bend is apparent.

The cause of the bending is apparently not connected with a physical restraint by the specimen holder, as the specimens which were heated as a filament in the apparatus shown in Figure 5 also showed pronounced curvature as a result of heating a portion of the specimen to temperatures above the transformation. In this apparatus, the specimens were chucked in a vise grip only at one end. The other end of the rod was immersed in a cold mercury bath so that no restraint to expansion was possible. Furthermore, those specimens which were heated to temperatures in the alpha phase by this method showed no bending.

#### VIIb. Effects of Various Factors

In discussing the number of grains which occupied a specimen it is necessary to state a basis for differentiating between sub-grains and grains. In the following discussion the number of grains refers to the number of orientations which varied sufficiently to cause a sharp boundary to exist as

a result of macro-etching in the trichloroacetic electrolyte. Based on correlation of etched appearances and orientation studies by X-ray diffraction (see below, VIc), this basis allows for a spread in orientation within a grain of approximately  $10^\circ$

The conditions which were varied in carrying out the 18 experiments were as follows:

1. Purity of Metal
2. Grain size prior to experiment
3. Temperature of point prior to start of travel
4. Specimen shape and dimensions
5. Rate of specimen travel
6. Temperature gradient
7. Maximum temperature of specimen.

In all of the experiments which were carried out in the NaK furnace the temperature gradient and the maximum temperature were held constant (conditions 6 and 7 above). As only 2 experiments were carried out to completion in other apparatus, in which these latter conditions varied, there is not sufficient data to warrant discussion of these factors.

The observed effects of changing the other variables are summarized below.

Effects of Material Composition: Of the conditions which were varied, the metal purity appears to be of greatest importance; this conclusion is based on the results obtained with one specimen of high purity material. The differences among the lower purity lots were not marked. Only 2 specimens of the low purity metal contained single crystals, filling the whole cross-section, and these crystals were of relatively small dimensions. In contrast, the one specimen of high purity consisted of what is considered a single crystal of relatively large dimensions.

Figure 8a shows the macro-etched surface of the pencil-shaped specimen of high purity material (Lot G-1) after the first passage through the NaK furnace transfer tube at 0.3 in. per hr. The point of the specimen was at  $600^\circ\text{C}$  prior to start of specimen travel. The specimen consisted of approximately one orientation with 2 attached grains of relatively small size. One of the attached grains can be seen in Figure 8a, and the other grain existed at the flat end of the specimen as shown in Figure 8b. This



specimen was again passed through the NaK furnace tube under conditions similar to the first experiment. After the second passage, the specimen consisted of one grain with regions which differed in orientation so that the surface showed patches of dark areas in a light colored matrix (Figure 8c). The dark patches appear to have a sharp outline in the photograph, but this condition was not readily apparent on visual examination. The orientation of this crystal and the appearance of the Laue spots are discussed below in VI.

This specimen was 0.210 in. in diameter and approximately 1-1/2 in. in length. Specimens of similar dimensions made from lower purity lots were definitely polycrystalline after similar treatments.

Effect of Specimen History: The specimens used in this investigation were of the following 4 different types prior to experiment:

1. As rolled
2. As recrystallized below 600°C
3. As solidified from melt (Lot J)
4. Large columnar grains produced by prior experiment.

In addition to the above initial structures, the history prior to passing through the beta to alpha transformation was varied by starting the specimen travel from different levels of the NaK furnace transfer tube.

The initial micro-structure did not appear to affect the number of grains produced by experiment. However, repeated experiments with the same specimen appeared to decrease the number of grains in the tapered ends of the specimens and, to some small extent, to decrease the number of columnar grains in the body of the specimens. This effect has been described above in connection with the high purity specimen (Figures 8a and c) and was also found in repeated experiments with lower purity specimens.

The temperature of the specimen point prior to start of travel appeared to have some effect on the number and shape of the grains produced by this type of treatment. Two specimens of the same history were similarly treated, with the exception of the positions in the transfer tube prior to initiation of lowering. The specimen which was gradually lowered into and out of the beta phase consisted of columnar shaped grains (Figure 9a), whereas the specimen which was heated uniformly to 750°C prior to the start of travel to the alpha phase consisted of irregular shaped grains of smaller sizes (Figure 9b).

Effect of Varying Rate of Specimen Travel: The rate of specimen travel through the NaK furnace transfer tube appeared to be of relatively small importance with respect to the number of grains formed in the upper three-fourths of the impure specimens. There appeared to be some effect of the rate of lowering on the number of grains formed in the pointed ends.

On the basis of 4 experiments it appears that a lowering rate of 0.1 in./hr was too slow for preparing large grains. Speeds of 0.3 in./hr to 10 in./hr yielded large grains, with little significant differences in this range. Figures 10a and b show the similarities between the results using 0.6 in./hr and 2.5 in./hr, respectively. Although fewer grains exist in the pointed section of the slower specimen, the number of grains in the body of the specimens is approximately the same.

Effect of Specimen Size: Whereas a single crystal was obtained from a 0.125 in. diameter specimen of the low purity metal, larger diameter specimens of the same material were less successful. Although a number of grains formed in the pointed ends of all the impure specimens, the number of grains which grew along the specimen length increased with specimen diameter. (Compare Figure 6e with Figure 6f.)

Effect of Specimen Shape - Constrictions: In order to further encourage the growth of a single grain to a cylindrical shape 2 types of constricted specimens were used as shown in Figures 7a and c. The intended function of the constriction was to reduce the number of grains which grew out of the point ends of the specimen. If the specimen design was successful in permitting only one grain to grow through the constriction, it was presumed that this grain could continue to grow as a single crystal in the upper end of the specimen. Consequently, the first type specimens were designed so that the specimen diameters increased gradually with distance above the constriction.

Of the 3 experiments carried out with this type specimen, only one showed a single crystal in the constricted section. One specimen contained 2 grains and a third specimen contained 3 grains in the constriction. The specimen which consisted of a single crystal at the constriction contained 3 grains in the section above the constriction, as shown in Figures 7a, b, and c. Two "new" grains apparently nucleated at the surface and grew alongside the original crystal. Figure 7d shows the grain boundary between the crystal which grew from the constriction (light color) and the "new" grain.

A fourth specimen was machined so that the upper section of the specimen was of the same diameter as the constriction. In this specimen 2 grains grew into the constricted diameter and only these grains appeared at the top end of the specimen as shown in Figure 7e.

Effect of Specimen Shape - Rectangular Section: The sizes and the shapes of the grains which were formed in the one experiment with a pencil type specimen of rectangular cross-section were markedly abnormal. Although the grains in the tapered end, which was of circular cross-section were elongated in the rod axis direction, the grains in the lower end of the shaft of the specimen appeared to have grown in directions almost perpendicular to the axis as shown in Figure 11b. The structure appeared to have been formed by grains growing from the edges. The upper length of the shaft consisted of a number of large and small grains. The large grains were somewhat elongated in shape, but the small grains were of regular shape as shown in Figures 11b, c, e, d, and f.

The peculiarity of this structure was emphasized by the existence of various degrees of sub-structure within the grains of the specimen as noted in the photographs. Some of the small grains showed a lack of sub-structure under microscopic examination, whereas the other grains showed a large degree of the cell type sub-structure (Figure 11e).

It was noted that this specimen was markedly bent, as shown in Figure 11a.

#### VIc. Study of Sub-Structure in Single Crystal Prepared from High Purity Metal

The single crystal which was prepared by passing a pencil type specimen of high purity material through the NaK furnace is shown as macro-etched in Figure 8c. This specimen was mounted in a cold setting plastic mount for grinding and polishing of a longitudinal surface for microscopic and X-ray examination.

Figure 8d shows the appearance of the section after etching in citric-nitric aqueous electrolyte. The structure appeared to consist of a number of bands of differing orientations. At this section, the region close to the pointed end appeared to be very close to a single orientation. The structure above this area appeared to be predominantly of a different orientation (dark appearing) with small areas of the light color orientation exposed. The area of dark etching orientation crossed the specimen in a direction tilted from the rod axis.

Upon examination in polarized light the differences in orientations corresponding to the dark and light etching regions, discussed above, were quite pronounced. Figure 8e shows the micro-appearance at the boundary between the dark structure and adjacent light regions at the left boundary of the specimen. The stage was rotated so that the maximum contrast was obtained for photographing. The twin bands extended across the boundary with a slight change in direction. Some elongated sub-grains could be seen within the larger structure.

Figure 8f shows the micro structure at the end of the specimen on a face perpendicular to the longitudinal section described above. At this section the sub-structure consisted of thin but wide areas of differing orientations. The boundaries between the differing orientations tended toward straightness and appeared somewhat parallel. The direction of the twin bands, which were continuous across orientation boundaries, indicated that the dark and light regions corresponded to nearly similar orientations.

Back reflection Laue patterns taken from approximately 25 points in the upper 1 in. length of the specimen showed a spread in orientation of approximately  $10^\circ$  throughout this structure. The X-ray beam was focussed on points corresponding to the different shades revealed by etching the longitudinal section (Figure 8d). The X-ray beam diverged from the 0.6 mm pinhole to cover an area of approximately 1 mm in diameter. Approximately 20 of the patterns were sufficiently clear for an orientation determination. The polished longitudinal surface by chance was within  $2^\circ$  to  $6^\circ$  of the (302) plane. Although most of the patterns consisted of diffuse spots, the  $\{111\}$ ,  $\{110\}$ ,  $\{310\}$ , and  $\{313\}$  reflections and the hyperbolas formed by the crystallographic zones containing the  $\{001\}$  reflections were generally discernible. The orientation spread and the mean orientation of the crystal are shown in stereographic projection in Figure 12. Seven orientations including the extremes are noted.

The positions upon which the X-ray was focussed for the individual patterns were selected by macro-examination of the etched longitudinal section. Figures 13a, b, c and d are examples of the patterns obtained. Only one pattern contained reflections from a nearly single orientation. The individual spots in this pattern were split but relatively sharp. The other patterns (those which are shown in Figure 13) generally contained reflections from 2 distinctly different orientations and individual spots of various shapes from intermediate orientations. The spots appeared to be divided so that the more intense spots from equivalent planes were connected by smaller spots of lower intensities. There did not appear to be any consistent direction of spread in orientation.

Based on metallographic examination and the X-ray patterns, it appears that the structure of this crystal consisted of thin plates which varied in orientation so that a general spread of  $10^\circ$  existed within the crystal. The plates were, in turn, sub-divided into smaller size sub-structure.

#### VId. Orientations of Columnar Grains

A number of back reflection Laue photograms were obtained from large grains in the less pure specimens. Most of the large grains examined were those which existed in the constricted sections of the shaped specimens shown in Figure 7. The purpose of this investigation was to determine whether the crystallographic directions parallel to the rod axis were similar, i.e., to determine whether a texture existed in the polycrystalline specimens.

Under the conditions of irradiation used (see VIb) only 4 patterns were obtained from which orientations could be determined. In all the patterns the spots were very diffuse and a large spread in orientation was indicated. The 4 patterns which were sufficiently clear contained Laue spots similar in diffuseness to those shown in Figure 13d.

The 2 grains which occupied the constriction shown in Figure 10b were of widely different orientations. The  $[100]$  direction of one grain was approximately  $10^\circ$  from the rod axis. In the other grain the  $[010]$  direction was approximately  $20^\circ$  from the rod axis.

The long dark grain in the small diameter portion of the specimen of Figure 7e also was oriented so that the  $[010]$  direction was approximately  $15^\circ$  from the rod axis. In the long dark grain shown in Figure 6f (right hand end) the  $[010]$  direction was approximately  $20^\circ$  from the rod axis.

In addition to the above data, the mean orientation of the single crystal in the high purity specimen is shown in Figure 12. In this crystal the  $[010]$  directions were approximately  $25^\circ$  from the rod axis.

Unfortunately, no effort was made to determine the orientations with respect to the long directions of the grains so that no definite conclusions can be made with regard to a possible preferred growth direction. However, since the grain boundaries were observed to be somewhat parallel to each other, although tilted from parallelism with the rod axis, it is probable that the  $[010]$  direction does correspond to a preferred growth direction. As only 5 orientations are included in this data it cannot be stated that other textures are improbable. As stated, above, one large grain showed the  $[100]$  direction close to the rod axis.

#### VIe. Summary of Results

Eighteen experiments were carried out in attempts to grow single crystals of alpha uranium by the transformation method. Sixteen of these were made in the NaK furnace. Pointed specimens of various diameters and shapes were lowered at various rates through a tube filled with liquid NaK. The specimens traveled point first from  $750^\circ\text{C}$  to  $20^\circ\text{C}$  through a temperature gradient which was approximately  $240^\circ\text{C}$  per in. between  $670^\circ\text{C}$  and  $650^\circ\text{C}$ . The results are summarized below.

a. The pointed ends of the low purity specimens contained a number of small grains. Grains elongated in the general direction of the rod axis were generally found above the tapered ends. With the exception of some grains in one specimen of rectangular cross-section, all the grains contained sub-grains which were generally elongated in the direction of specimen travel.

b. One cylindrical single crystal of 0.210 in. diameter was prepared in a specimen of high purity material. This was the only high purity specimen used in this investigation. The structure of this crystal consisted of a number of plates of slightly differing orientations. The spread in orientation was approximately  $10^\circ$ .

c. Although 2 single crystals were obtained from the lower purity material, the crystals were of small size.

d. The rate of travel of the specimen through the temperature gradient did not appear to be critical with regard to the number of large elongated grains formed in low purity specimens.

e. Fewer grains were formed when the specimen was lowered through a temperature gradient into the beta phase prior to further lowering into the alpha phase. Specimens which were subjected to repeated experiments showed a decrease in the number of grains as a result of repetition.

f. The one specimen of rectangular cross-section showed a markedly different grain distribution and sub-structure appearance than the cylindrical specimens.

g. A bend in the specimens as a result of experiment was noticeable in specimens of small diameters.

h. The orientations as determined by X-ray diffraction from 5 elongated grains indicates that the [010] direction corresponds to a preferred direction of alpha grain growth under the experimental conditions.

## VII. DISCUSSION OF RESULTS

### VIIa. Interpretation of Results

The increase in grain size with distance from the point of the impure specimens can be interpreted as indicating a strong influence of differences in rates of grain growth in different crystallographic directions. Because of the impurities in a specimen or because of other adverse experimental conditions a number of alpha phase nuclei formed as the point of the specimen reached a sufficiently low temperature. These nuclei were of different orientations. As the specimen was gradually lowered the nuclei tended to grow preferentially in certain crystallographic directions. As only a small number of nuclei were oriented so that a preferred growth direction was at a small angle to the rod axis, most of the nuclei grew rapidly to the surface of the tapered section and at a lesser rate in the long direction of the specimen.

Because of the relatively slow growth rates in the less preferred directions in the initial grains, the nuclei formed in the subsequently transformed material were permitted to increase in size and resist being consumed by these earlier formed alpha grains. As the volume of metal transforming per unit time increased, from the point of the specimen, as the specimen was lowered, more nuclei of favorable orientations were formed. These nuclei grew in the general direction of the thermal gradient and were eventually in sufficient number to prevent more nuclei from forming or from growing to unconsumable sizes. These grains continued to grow and they co-existed all along the rest of the specimen.

The number of favorably oriented nuclei decreased with specimen diameter, so that, with decreased competition, the probability of a single crystal existing in the upper part was increased in specimens of small diameter. In larger diameter specimens the probability of the formation of a single crystal was decreased by the larger number of favorably oriented nuclei and because of the growth of nuclei which apparently formed at the upper surfaces.

The successful experiment with the high purity specimen can be explained by assuming a decrease in the number of nuclei which were formed in the point and a faster rate of growth in directions other than a preferred growth direction. Dilatometric studies<sup>(8)</sup> have shown that impurities generally tend to decrease the temperature of the transformation on cooling from the beta phase. The higher temperature of the transformation in the high purity specimen suggests a lower rate of formation of alpha nuclei. The higher temperature and fewer grain growth inhibiting particles would permit a more rapid rate of grain growth in all directions. Consequently a nucleus which was not of the most favorable orientation could still grow to form a single crystal in the upper part of the specimen. Although this particular specimen was of relatively large diameter no grains were found to have nucleated at the surface of the upper part, as was observed in the large diameter specimens of low purity.

Although the rate of specimen travel did not appear to be an important factor in the results with impure metal, because of the copious nucleation caused by a lower transformation temperature and the slow growth rates, this variable may be of importance in experiments with high purity materials.

The observed benefit of subjecting the high purity specimen to a second passage through the transfer tube indicates a possible effect of the beta phase grain size, assuming that the beta grain size is in turn dependent upon the prior alpha grain size.

## VIIb. Feasibility of Preparing More Perfect Crystals

Considering the results with the high purity specimen, it appears to be feasible to grow large single crystals by this method, but the high degree of sub-structure precludes the use of these crystals for many purposes. Because of the complex nature of the grain coarsening procedure which has been developed for growing more perfect single crystals, it would be of great advantage if single crystals of similar perfection could be prepared by the phase transformation method.

The existence of noticeable bends in most of the specimens, and in all the specimens to at least a small degree, indicates that appreciable stresses are produced when a temperature gradient exists in specimens undergoing transformation. It is conceivable that such stresses are caused by the co-existence of the low density beta phase and high density alpha phase<sup>(9)</sup> and/or by differences in the directions of thermal contractions of the columnar alpha grains during cooling in the alpha phase. Based on the fact that the high purity specimen which was essentially a single crystal was slightly bent (Figure 8) it would appear that the latter factor is of small importance, since it should not lead to bending in a single crystal. Assuming the transformation stresses to be predominating, the stresses would then be present as soon as some alpha grains have formed. At these high temperatures alpha grains deform rather easily by slip and kinking to produce cells of differing orientations.<sup>(10)</sup> The columnar nature of the cells found in the elongated grains would indicate that the sub-structure did form during the early stages of alpha grain growth and tended to grow in parallel directions, the direction of most rapid growth depending on the orientation.

Whether conditions can be found by which single crystals lacking sub-structure can be prepared by this method is still to be determined. If the size of cells formed by high temperature deformation is a function of the strain rate and deformation temperature, as proposed by Wood and Rachinger,<sup>(11)</sup> it may be possible to decrease the rate of specimen travel so that the sizes of cells formed would approach the grain size. This possibility would be enhanced if the specimens are of high purity material since the temperature at which the initial stresses occur would be higher, i.e., the transformation would occur at higher temperatures.



### VIII. ACKNOWLEDGEMENTS

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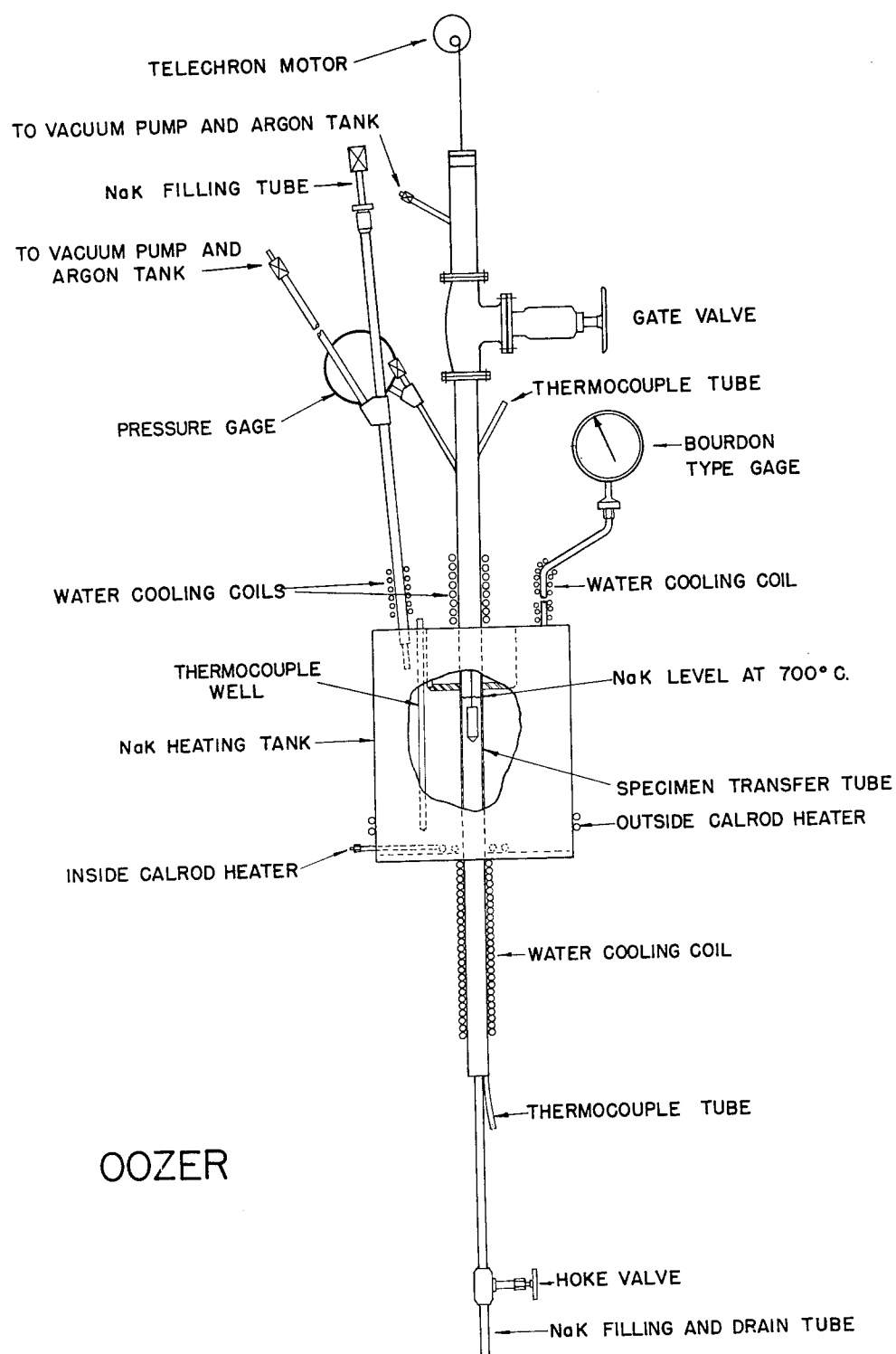
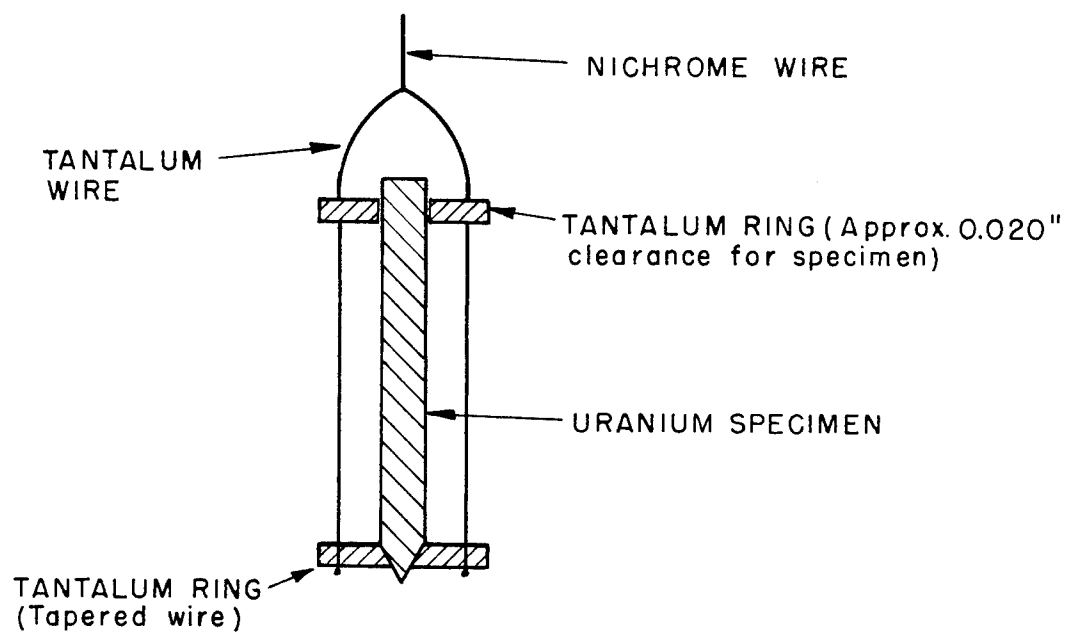


Fig.1 Schematic Drawing of NaK Furnace

FIGURE 2  
SPECIMEN SUPPORT

PHASE TRANSFORMATION SPECIMEN IN ASSEMBLY  
FOR TRANSPORTING THROUGH NaK FURNACE TRANSFER TUBE



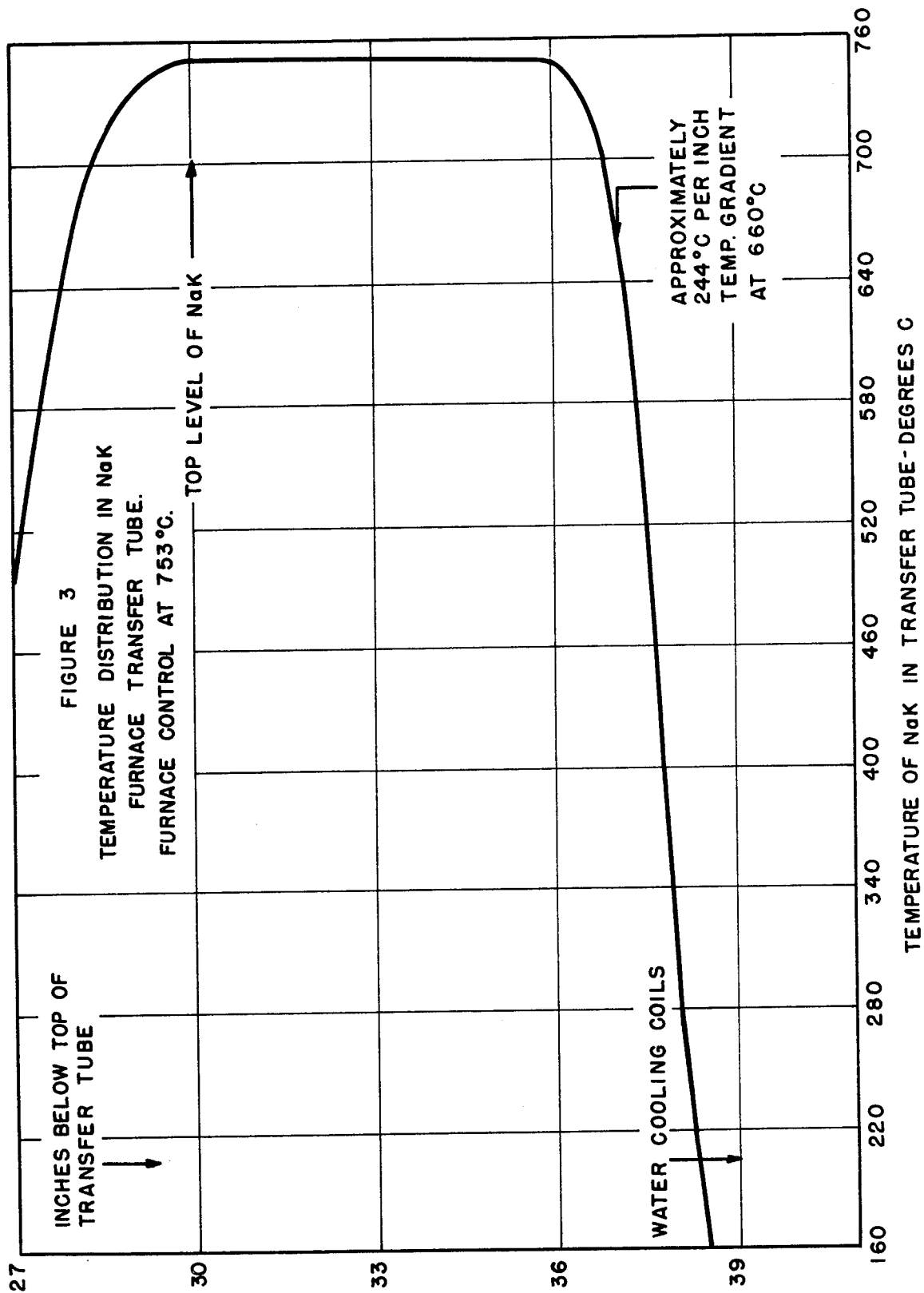


FIGURE 4  
NICHROME WOUND TUBE FURNACE  
SCHEMATIC DRAWING OF ASSEMBLY  
FOR 50°C PER INCH TEMPERATURE GRADIENT  
HEAT TREATING

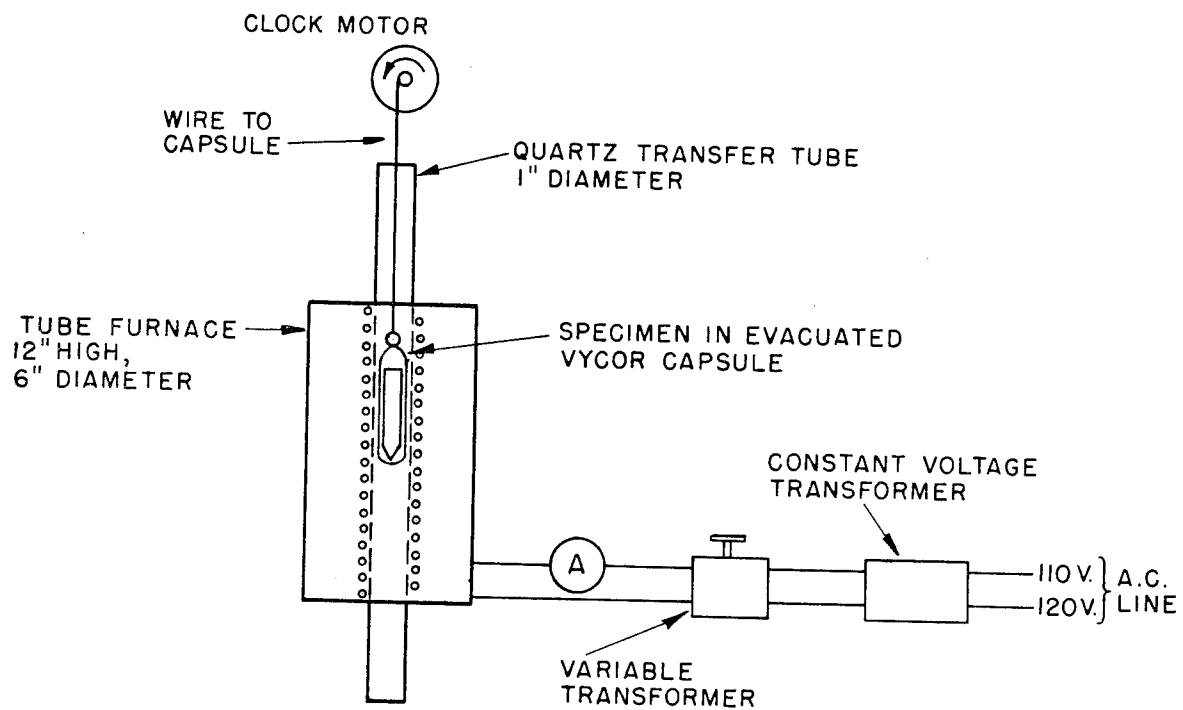


FIGURE 5  
 URANIUM FILAMENT FURNACE APPARATUS FOR ANNEALING  
 SPECIMEN AS FILAMENT  
 (Similar to Vacuum Filament Crystal Device-KAPL)

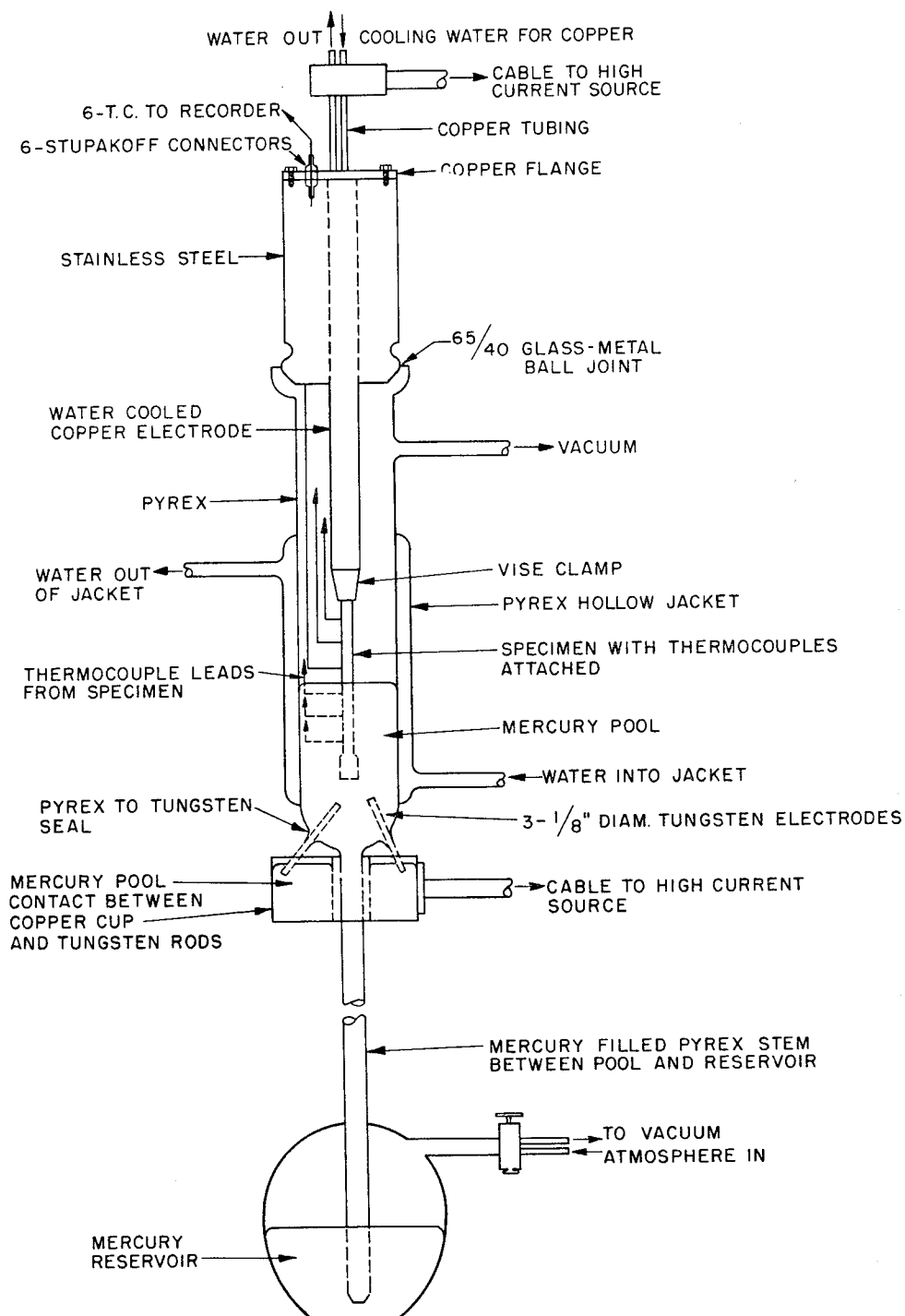
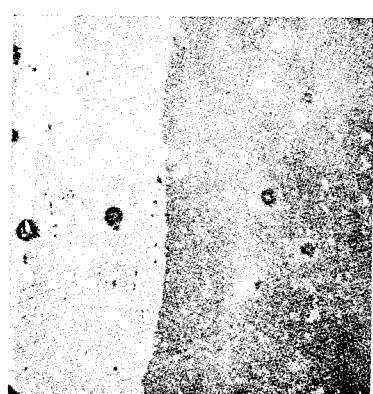
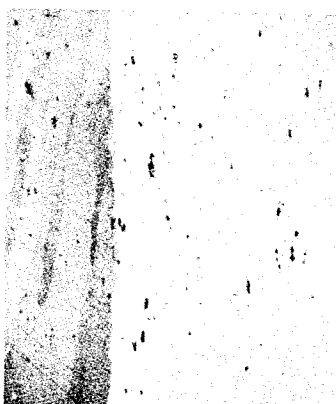


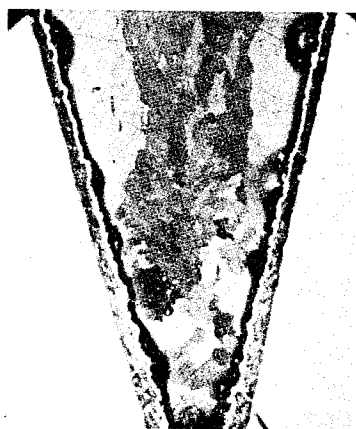
Figure 6. Typical Grain Size Distributions Resulting From Phase Transformation.



7663 a. 25x



7658 c. 25x



7664 b. 25x



7657 d. 25x



4644 e. 2x

Figure 6 - a, b, c, and d. Longitudinal microstructures at upper sections (a & c) and tapered ends (b & d) of two 1/8" diameter specimens of Lot A-1, both lowered through NaK furnace at 0.3" per hour from 600°C to 750°C to 20°C.

Figure 6 - e. Surface of a 1/8" diameter specimen as macroetched after above treatment. Light colored section at top was a single crystal with large scatter in orientation.

Figure 6 (continued)



4620

f.

2x

Figure 6-f. Surface of 1/4" diameter specimen as macroetched after treatment similar to that described for above figures.



10,001

g.

50x



7564

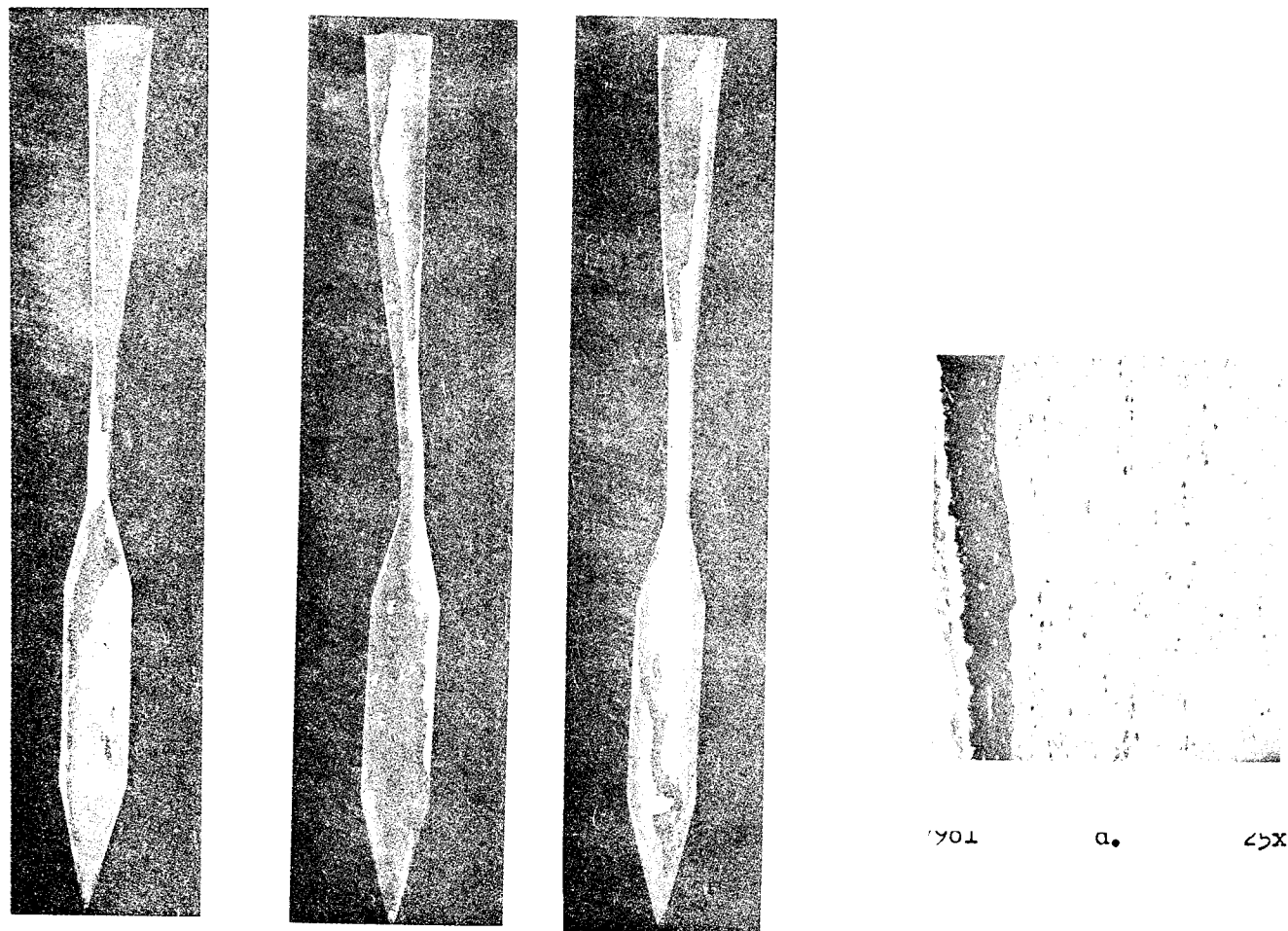
h.

50x

Figures 6 - g and h. Microstructures showing change in direction of twin bands at sub-grain boundaries (g) and block type sub-grain (in grain left of center in h).



Figure 7. Bending and Grain Size in Tapered Specimens.



7962 a. 2x 7963 b. 2x 7964 c. 2x

Figures 7 - a, b, c, d. Specimen lowered through NaK furnace transfer tube at 0.9" per hour from 600°C to 750°C to 20°C. Photos taken at different profiles of specimen so that maximum bending shows only in Figure b. Single crystal at constriction is 3/16" long. Figure d shows growth of grain from surface above constricted section as observed macroscopically in other photos.



7947

e.

2x

Figure 7-c. Second type of constricted specimen, processed as above.

# Figure 8. Transformation Experiments with High Purity Uranium.

Appearance of macroetched outer surface and polished longitudinal section of single crystal in specimen of high purity material twice lowered through NaK furnace transfer tube from 600°C to 750°C to 20°C at 0.3" per hour.

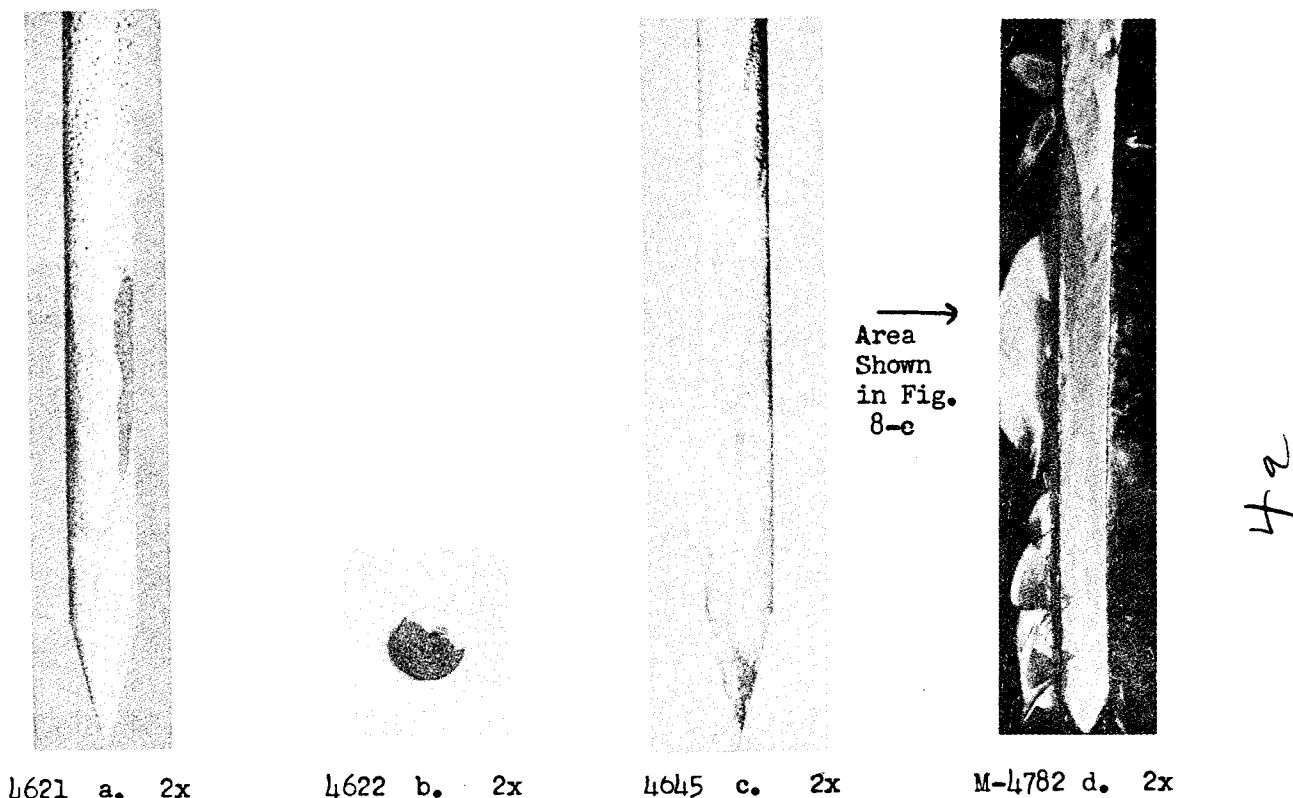
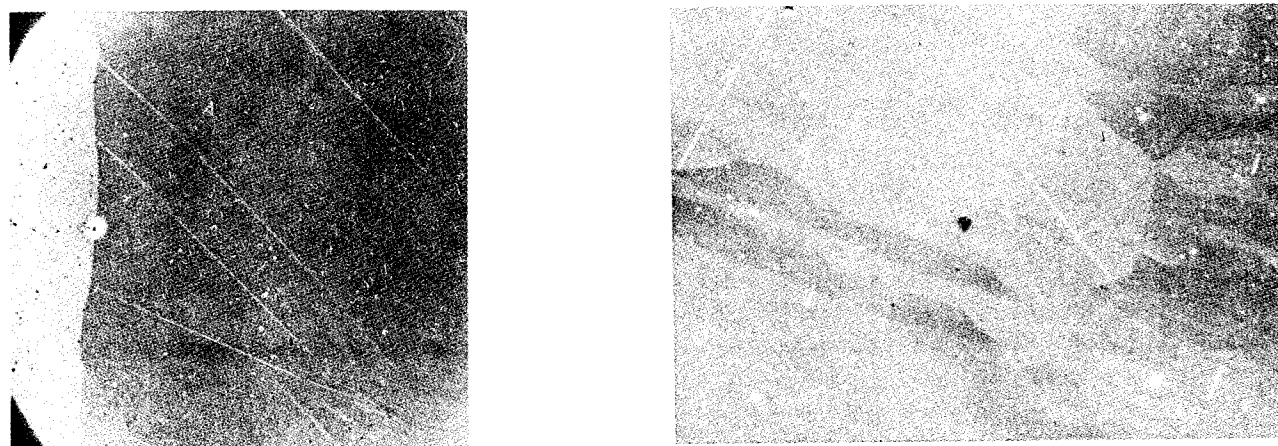


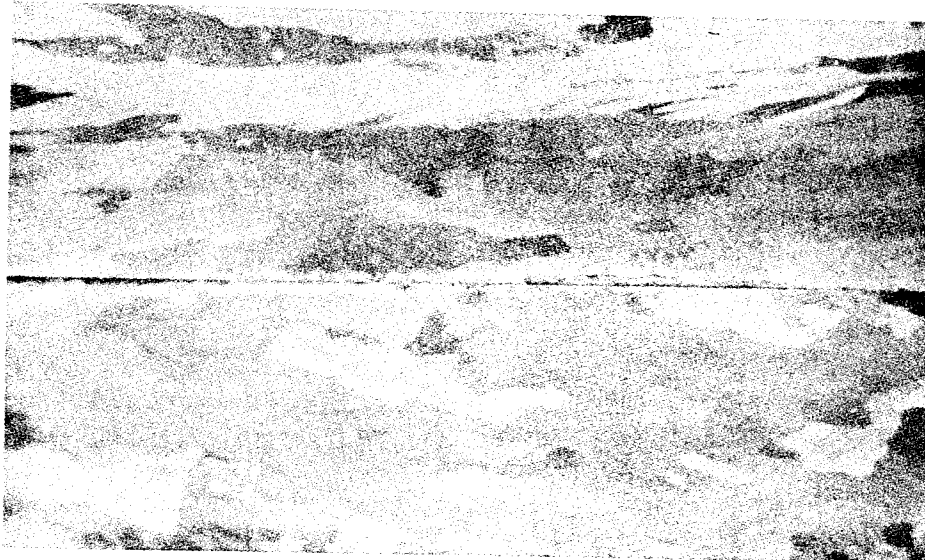
Figure 8-a. High purity specimen after first pass through  $\beta$  to  $\alpha$  transformation.  
 b. Grain boundary at rod end.  
 c. Same specimen after second pass from  $\beta$  to  $\alpha$ .  
 d. Polished and etched longitudinal section of specimen in c.



7726 e. 25x      7808 f. 50x

Figures 8-e and f. Sub-structure revealed by polarized light study of polished surfaces of section shown in d (e) and end of specimen perpendicular to axis (f).

Figure 9. Effect of Gradual Lowering Into  $\beta$  Phase During Specimen Travel.



M-4788

M-4787

10x

Figure 9-a and b. Specimen of Lot J, lowered from 600°C to 750°C to 20°C (a); other specimen (b) lowered from 750°C to 20°C. Both lowered at 0.9" per hour through NaK furnace.

Figure 10. Insignificance of Rate of Specimen Travel.



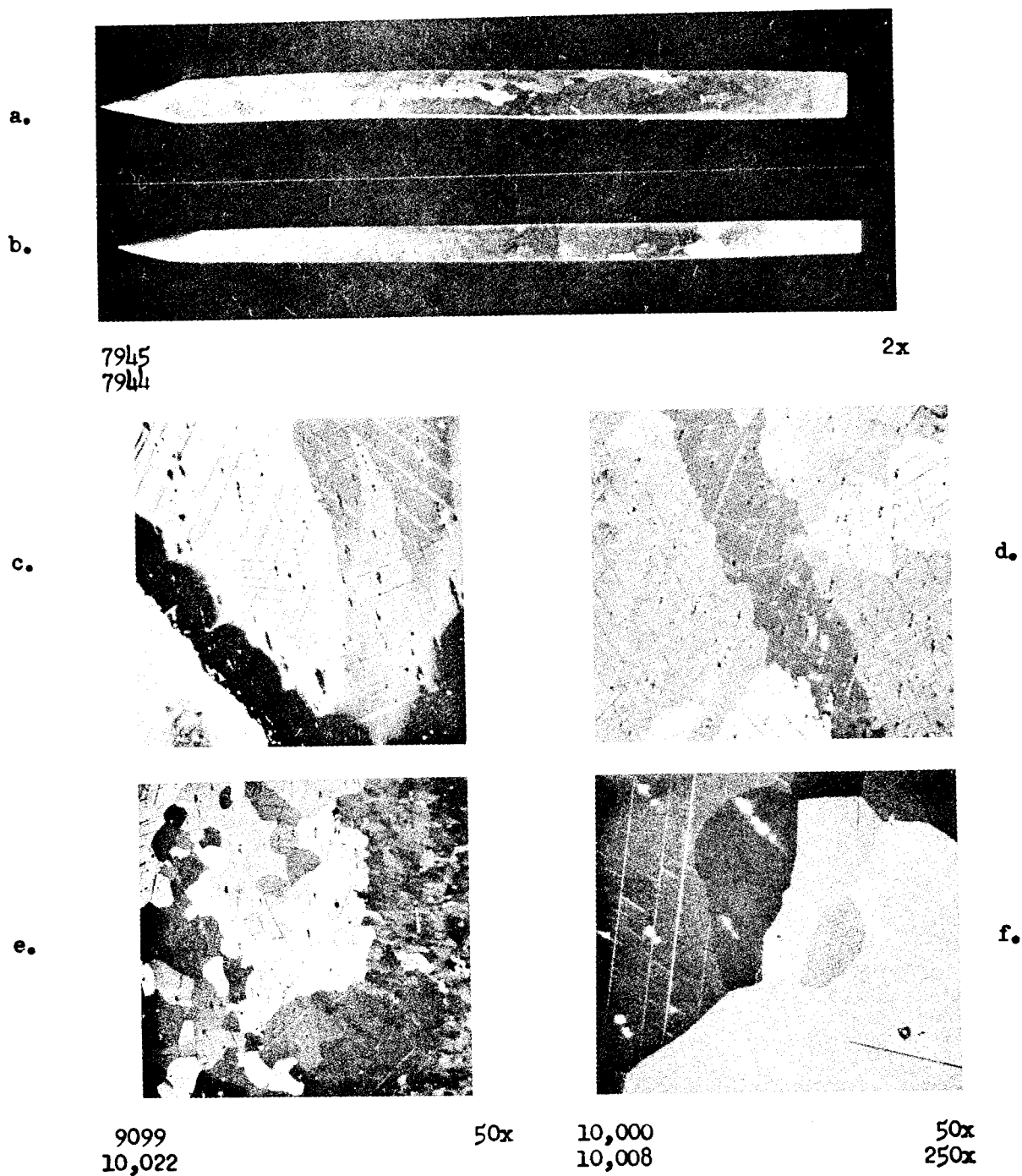
7906

7911

2x

Figure 10-a and b. Constricted specimens lowered from 600°C to 750°C to 20°C at 0.6" per hour (a) and 2.5" per hour (b).

Figure 11. Appearance of Grains in Specimen of Rectangular Section.



Figures 11-a and b. Appearance of grains in rectangular section specimen after passing through NaK furnace at 0.3" per hour from 600°C to 750°C to 20°C. c: Elongated grains in point. d: Microstructure at half length. e: Microstructure close to upper end. f: Appearance of small equi-axed grains in e at higher magnification.

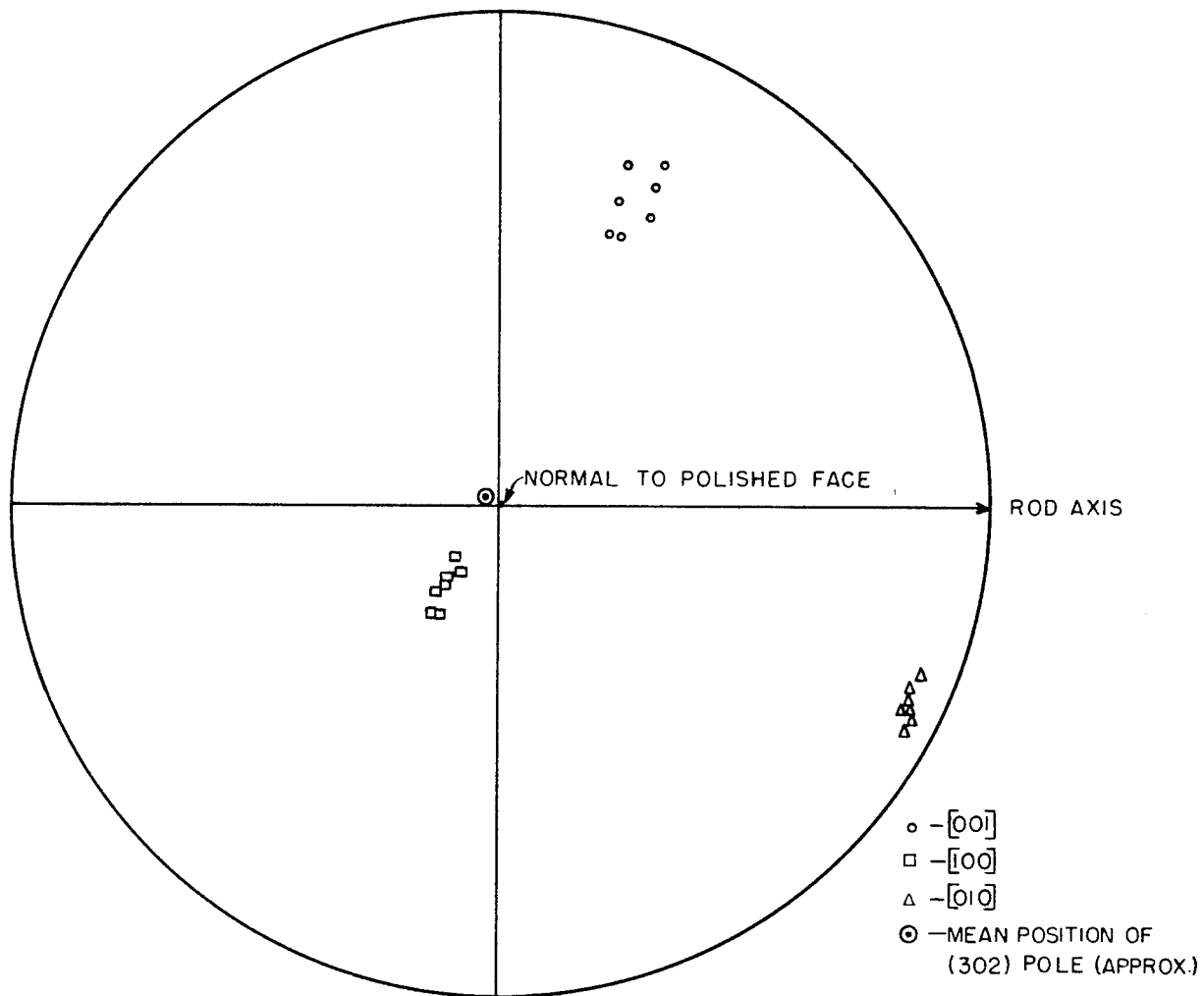


FIGURE 12  
 DIFFERENCES IN ORIENTATION BETWEEN SCATTERED  
 POINTS OF CRYSTAL FORMED IN HIGH PURITY SPECIMEN  
 BY PHASE TRANSFORMATION METHOD.

Figure 13. Laue Photograms of Structure in Single Crystal Formed in High Purity Specimen.

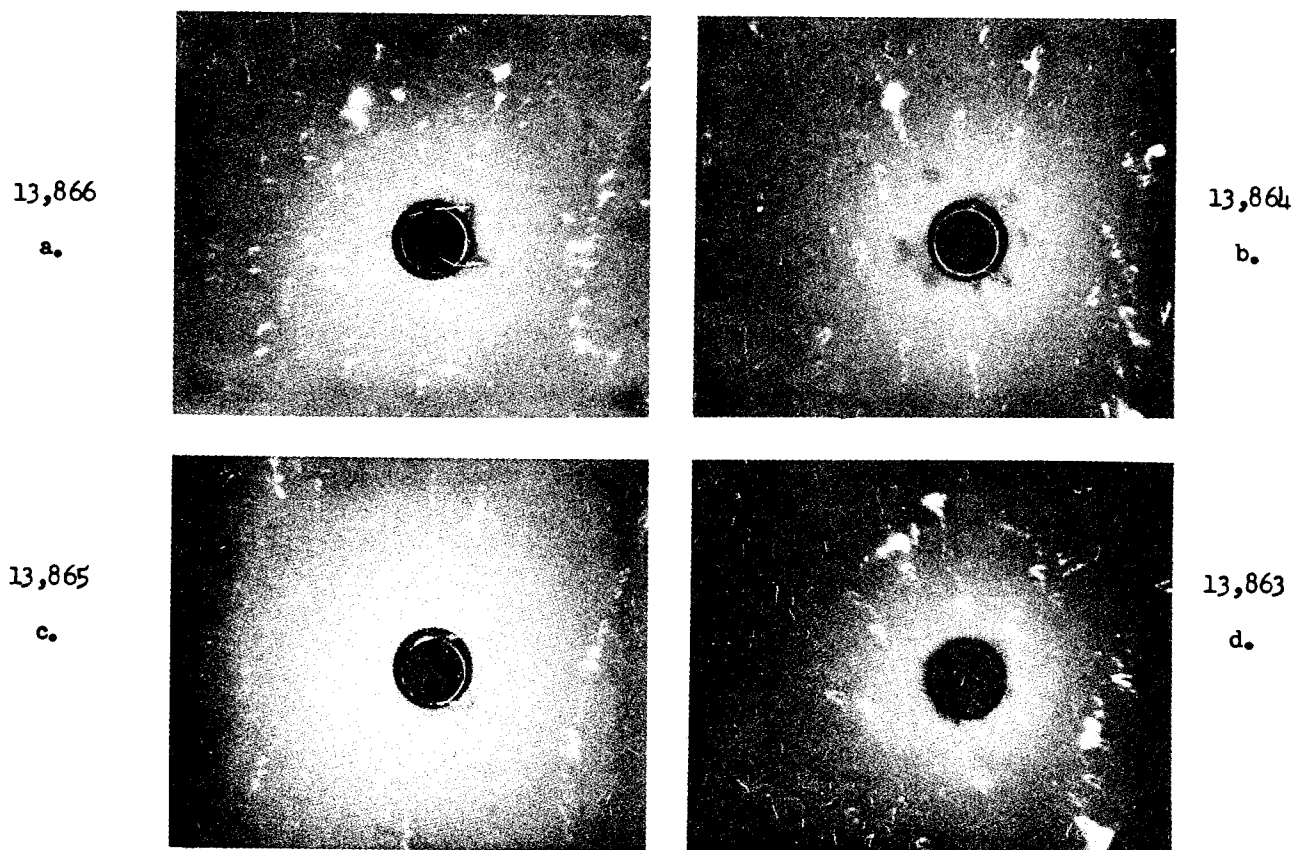


Figure 13. Photograms showing the variety of shapes of Laue spots and spread in orientations at points of approximately 1 mm diameter at longitudinal section shown in Figure 8-d. Cu target radiation, 2 hour exposure, 0.6 mm pinhole, 3 cm distance.