GROWTH OF SINGLE CRYSTALS OF IRON
BY THE STRAIN-ANNEAL METHOD

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by

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Abstract

Large single crystals of iron were grown by the strain-anneal method. Two types of material were used, A.I.S.I. C1018 steel and Ferrovac E iron. The largest and most homogeneous crystals were grown by decarburizing and strain-annealing the C1018 steel. The crystals obtained by strain-annealing the Ferrovac E iron were not as large as those grown with C1018 steel.

The conditions determined for optimum grain growth in C1018 steel are an initial grain size of 180 grains per sq mm, obtained by decarburizing for 48 hours in wet hydrogen at 925°C, straining 3.0 percent, and annealing for 48 hours at 870°C.

Orientations of the largest crystals in the longitudinal plane of the bar were determined by x-ray analysis. A highly preferred orientation was noted with [100] developed along the direction of straining and [010] perpendicular to the plane in which the bar had been formerly rolled.

Contents

I. INTRODUCTION

The growing of large single crystals of iron by the strain-anneal method is a process of controlled recrystallization. Recrystallization is caused by introducing imperfection in the specimen and then raising the temperature high enough to form growth nuclei. If only one growth nucleus were formed, the specimen would be transformed to a single crystal.

The number of growth nuclei that appear in a deformed metal upon annealing depends upon the amount of strain, temperature, and initial grain size. The higher the temperature and greater the amount of strain, the greater the number of growth nuclei formed. Edwards and Pfeil (1)* found that the initial grain size also affects nucleation. In general, there will be a unique grain size for a given strain that will produce a minimum number of nuclei. If

*The figures appearing in parenthesis pertain to references appended to this paper.

the initial grain size is either larger or smaller than this critical size, the nucleation will be less favorable for the growth of large crystals. It is thus apparent that to grow a large single crystal of iron the optimum amount of strain, initial grain size, and critical temperature must be determined.

Two stages may be considered for the grain-growth process which occurs during recrystallization. During the first stage, more perfect crystalline material replaces the strained portion. When this process is complete, further growth must stem from differences in the surface free-energy of the grains. Orientational features also appear to effect the mobility of grain boundaries.

X-ray studies were made on crystals grown to determine if there were a preferred orientation after growth and if there were any affect from primary orientation due to the initial cold rolling.

This study was undertaken in an attempt to isolate the variables affecting single crystal growth in iron and to grow large crystals for use in further experiments for the determination of elastic constants of body-centered cubic materials. The study is developed under the following areas of investigation:

11. EXPERIMENTAL PROCEDURE

Two different materials were selected for this experiment in an attempt to isolate the variables affecting large crystal growth. These materials were C1018 steel and Ferrovac E iron.

The C1018 steel was selected because it contained a small amount of carbon and other impurities. The Ferrovac E iron was selected as representative of relatively high-purity iron.

The discussion of the two materials will be treated separately because the experimental procedure varied as a result of chemical composition.

A.I.S.I. C1018 Steel

The C1018 steel came in cold-rolled bars 1/8 x 1/2 in. x 20 ft. The nominal composition of this material in weight percent was as follows:

C	Mn	P (Max)	S (Max)
0.15-0.20	0.60-0.90	0.040	0.050

In order to eliminate cementite in iron-carbon alloys at the temperature of recrystallization, there must be less than 0.005 weight percent carbon present. It would be expected that such cementite would interfere with the growth process and, accordingly, small quantities of carbon should play an important role in retarding grain growth. Such effects have been noted by Chappell(2).

Decarburization was carried out by heating in an atmosphere of wet hydrogen. The furnace used was an electrically heated horizontal-tube furnace, with a ten-inch heated zone. Through the center of the furnace was a 1 1/2-in. diam, 30-in.-long mullite tube, the ends of which were closed by means of rubber bungs. Glass tubing, 5-in. long, extending through the bungs, allowed hydrogen to pass through the furnace. One bung, adapted with another opening, contained a thermocouple which was inserted into the center of the furnace.

Decarburization generally required about 72 hours but varied according to rate of hydrogen flow and temperature. Decarburization was considered complete when a polished and etched section appeared free from pearlite or cementite.

The specimens were strained on a 10,000-pound Dillon universal testing machine. A gauge length of 10 cm was used, and elongations were measured by means of a divider and steel rule.

The furnace used for annealing was an 18-in. long electrically heated vertical-tube furnace. Through the furnace was a 1-in. diam, 30-in. long Vycor tube which was sealed by means of rubber bungs. The bung arrangement for the passage of hydrogen through the tube was the same as that for the decarburizing furnace. A thermocouple, used to maintain a constant temperature check, came in from the top of the furnace. The hydrogen gas used in the annealing process was dried by means of passing hydrogen through a drying chain consisting of several sections, the first of which was a 30-in. long pyrex tube packed with a commercial material called "Drierite." From this tube the hydrogen then went through a heated Vycor tube containing a nickel boat filled with titanium chips.

In the early experimental studies, tensile-test pieces were cut from the C1018 bars and machined to 6-in. lengths, the reduced section being 1/8 x 1/4 x 2 1/2 in. In later experiments the test pieces were not machined to shape, and the length was varied. The results obtained were equally as good as those obtained from the shaped specimens.

The first experiment was made with six test pieces elongated in 1-percent intervals, the first specimen being elongated 1-percent. These specimens were annealed at 870°C for 48 hours and allowed to cool in the furnace. The specimens were etched in a 10-percent nitric acid solution to determine whether macroscopic grain growth had occurred. With preliminary etching, it appeared as if no appreciable grain growth had occurred in any specimen. Upon further etching, however, a coarsely crystalline structure appeared on the surface of the specimens that were elongated 2, 3, and 4 percent. It is thus apparent that a thin film of small crystals was present over the entire specimen, masking the interior structure.

In the next experiment, four specimens received elongations of 2.5, 3.0, 3.25 and 3.5 percent respectively and were annealed at 870°C for 72 hours. When the specimens were etched, an outer layer of fine crystals again appeared. With prolonged etching the large internal crystals were revealed. The largest crystals obtained were found in the specimen that had been elongated 3.0 percent. In all of the large crystals grown, the width was equal to the maximum width of the test specimen. More experiments were carried out to see whether the critical strain could be determined more accurately. The results of these experiments were rather erratic; that is, the experimental results could not be duplicated consistently.

The only difference that could be determined from the various specimens was the time and temperature of decarburization. A microscopic examination of samples decarburized at different temperatures and longer times showed that there were marked differences in initial grain size. This difference in initial grain size will be discussed in detail under the section "Factors Affecting Crystal Growth."

As mentioned previously, there was a thin film of small crystals that masked the internal structure of the annealed specimens. Experiments were carried out to see whether this film could be consumed by the large crystals during prolonged annealing. The annealing time was extended from two to five days, but no appreciable change could be noticed. Specimens were then strained by rolling, and upon etching, a large crystalline structure was immediately produced. These results compare favorably with those of Edwards and Pfeil (1).

Ferrovac E Iron

Ferrovac E iron was selected for this experiment as representative of relatively high-purity iron. The nominal chemical composition in weight percent is as follows:

C	Mn	P	S	Si	Ni	Cr
0.007	0.001	0.002	0.006	0.009	0.05	0.009
V	N	O	Sn	Al	Co	Cu
0.004	0.0027	0.023	0.005	0.003	0.005	0.01

The material was received as cold-rolled bars, 7/8 in. in diameter and 20 ft long.

A decarburization process was not required for the Ferrovac E iron because of its low carbon content.

The specimens used in this part of the experiment were cut from bars and machined to a size of 1/4 x 1/2 x 6 in. After the specimens were machined, they were placed in a furnace and annealed at 500°C for 18 hours to give them a full anneal before straining.

Because an approximate critical strain for iron had been determined in earlier experiments with C1018, the problem of determining this strain was greatly reduced. Three test pieces were strained to 2.75, 3.0, and 3.25 percent elongation and annealed at 870°C for 72 hours.

Etching of the specimens produced a thin film of crystals on the specimen surface, but this film was very irregular and not as thick as that found on the C1018 test pieces. In some cases the layer of crystals was not present over the entire surface of the specimen, and large crystals appeared immediately upon etching. The largest crystal formed during this experiment, 1/2 x 1/4 x 1 in., was in the specimen that was elongated 3 percent. In further experiments the results were approximately the same. The longest crystals obtained were approximately 1 in. long and 3/8 in. in width, but the thickness was never uniform. The boundaries of these large crystals were not sharply defined, and many small crystals were imbedded in every face (Fig. 1). When these crystals were cross-sectioned and examined, "pockets" of small crystals were discovered in the center of the large crystal (Fig. 2).

The variables affecting crystal growth, namely, annealing temperature, amount of strain, initial grain size, and time of anneal were all varied in different combinations; but the results were never comparable to those of the C1018 steel.

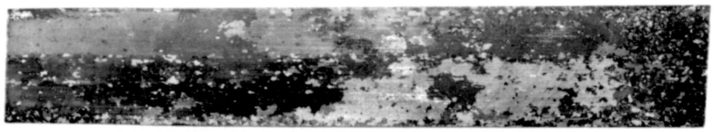

Figure 1. Ferrovac E iron strained 3.0 percent and annealed at 870°C for 48 hours. Actual size.

Figure 2. Cross section of bar in Figure 1. Magnification 4X.

III. FACTORS AFFECTING CRYSTAL GROWTH

The influence of annealing temperature, amount of strain, initial grain size, time of anneal, and presence of foreign elements on crystal growth were evaluated and are treated separately in the following section.

Annealing Temperature

The temperature of recrystallization is directly dependent upon the amount of strain present in the metal. The more strain, the lower the temperature of recrystallization, and the more growth nuclei formed. The growth of large single crystals of iron necessitates finding the critical strain that will cause recrystallization at the maximum temperature below the A3 transformation. The A3 transformation in pure iron occurs at 910°C. If impurities such as carbon and manganese are present, the transformation point is lowered.

The transformation temperature can be established on a dilatometer. The expansion of a heated specimen can be compared to that of a standard bar. This expansion is essentially linear up to the transformation temperature; as the crystal lattice changes from body-centered cubic to face-centered cubic, there is a sudden contraction compared to that of the standard bar, and the temperature of this discontinuity is measured on a precision potentiometer. The transformation point for the decarburized C1018 steel was determined to be 776°C. The primary element that caused this lowering of the transformation point was manganese.

Numerous attempts were made to grow crystals by passing the specimen through a temperature gradient. Because this procedure was not successful, one may assume that the right temperature gradient was never attained, for theoretically this is the ideal situation to form only one nucleation center.

Amount of Strain

The amount of strain necessary to cause grain growth is dependent on temperature and initial grain size. If the temperature is below 400°C, no amount of strain will cause significant grain growth.

The dependency of initial grain size on critical strain was determined by varying the initial grain size and straining followed by annealing. From this procedure it was determined that for a test bar with an initial grain size of approximately 200 grains per sq mm the minimum elongation for abnormal growth was 2.75 percent. The minimum elongation for maximum grain growth in a specimen having an initial grain size of 60 grains per sq mm was 3.75 percent. The largest crystals were obtained from the specimen that had an initial grain size of approximately 180 grains per sq mm and that was strained 3.0 percent.

From these experiments it is apparent that the critical strain for maximum grain growth is increased as the initial grain size is increased. Also, the higher the annealing temperature, the less strain required.

The relationship of strain to grain growth for a specific grain size is shown in Fig. 3, 4, 5, 6, 7, 8. All specimens had an initial grain size of approximately 180 grains per sq mm and were elongated 2.25, 2.50, 2.75, 3.0, 3.25, and 3.5 percent, respectively.

Initial Grain Size

The initial grain size in C1018 steel was controlled by the time and temperature of decarburization; whereas in

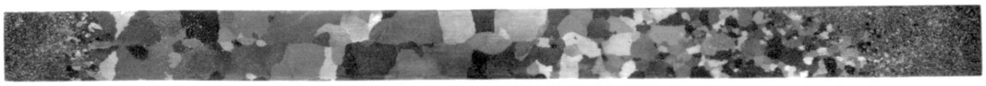

Figure 3. C1018 bar strained 2.25 percent and annealed at 870°C for 48 hours. Reduced to 0.81 of actual size.

Figure 4. C1018 bar strained 2.5 percent and annealed at 870°C for 48 hours. Reduced to 0.81 of actual size.

Figure 5. C1018 bar strained 2.75 percent and annealed at 870°C for 48 hours. Reduced to 0.81 of actual size.

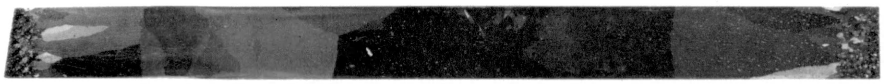

Figure 6. C1018 bar strained 3.0 percent and annealed at 870°C for 48 hours. Reduced to 0.81 of actual size.

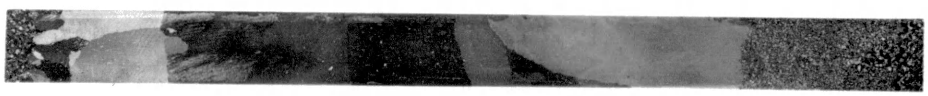

Figure 7. C1018 bar strained 3.25 percent and annealed at 870°C for 48 hours. Reduced to 0.81 of actual size.

Figure 8. C1018 bar strained 3.5 percent and annealed at 870°C for 48 hours. Reduced to 0.81 of actual size.

Ferrovac E iron this variable was controlled by the same method but in an anneal prior to straining and annealing.

In the experiments performed, the specimens were heated to temperatures between 730°C and 1000°C. The higher the temperature, the greater the grain size.

Further differences in grain size were obtained by varying the time from 24 hours to five days. The longer the time, the greater the grain size.

The heating and cooling rates also were very important in duplicating results. Because both types of material used contained an unknown amount of cold work, the temperature had to be raised slowly to prevent growth and to ensure that the end product would contain equi-axed grains. Also, when the decarburization temperature went above the A3 transformation point, the specimen had to be cooled slowly back through this point, or abnormal grain growth would occur.

The method used to obtain the desired initial grain size of approximately 180 grains per sq mm was to heat the specimen to 925°C for 48 hours and cool to room temperature in 12 hours.

Time of Anneal

According to Cottrell (3), a plot of grain size versus time at constant temperature would be as shown in Fig. 9. It will be noted that the grains do not begin to grow at once but wait for a short interval of time. The time at which the grains begin to grow is known as the time of nucleation. From zero time to the time of nucleation is the incubation period for nucleation.

After nucleation has begun, the rate of grain growth is very rapid, and the grain size increases linearly with time. When several large grains come in contact with each other the rate of growth is slowed considerably as is shown by the flattening of the curve in Fig. 9, and, when equilibrium is reached, the grains cease to grow.

In the specimens used for this experiment, the time for maximum growth was found to be approximately 48 hours.

Presence of Foreign Elements

The presence of an appreciable amount of carbon in steel restricts the grain size by forming pearlite which retards the mobility of the boundary.

Because the largest crystals were grown in the impure C1018 decarburized steel and not in the Ferrovac E iron, impurity atoms, present in some quantity in the steel and not in the iron, possibly aided grain growth by retarding the number of nuclei formed.

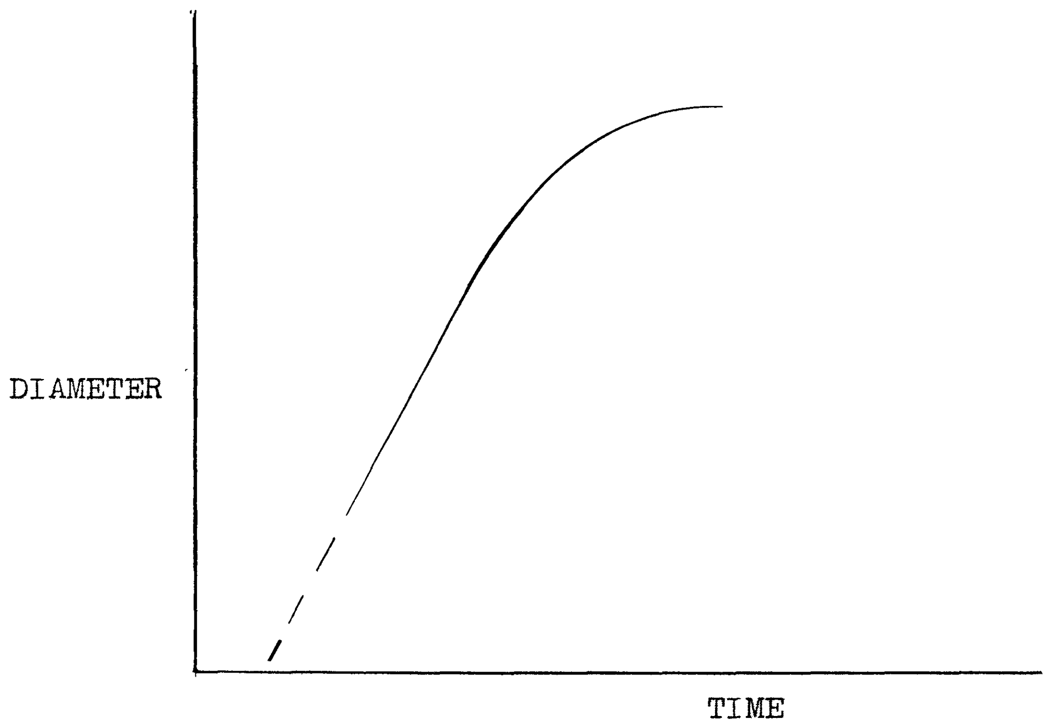

Fig. 9. Increase in the size of a growing crystal during recrystallization.

IV. ANALYSIS OF SINGLE-CRYSTAL ORIENTATION

The nine largest crystals grown were used to determine if there were a preferred orientation of crystal growth. The orientation of the faces of the crystals in the plane of the bar was established by the Laue back-reflection method, discussed in detail in Appendix I.

Once the orientations of the faces of the crystals are established in a reference system, the poles of the {100} planes of the crystal are plotted graphically by means of a stereographic net to obtain a pole figure (Fig. 10). This plot shows a preferred orientation of the (100) poles in the center, and the (010) poles clustered around the tension and primary rolling direction. Barrett (4) describes this orientation as the "cube texture," (100) [001] which has been produced only in face-centered cubic materials. He also states that the cube texture is often, though not always, associated with a tendency for the recrystallized grains to coalesce into extremely large

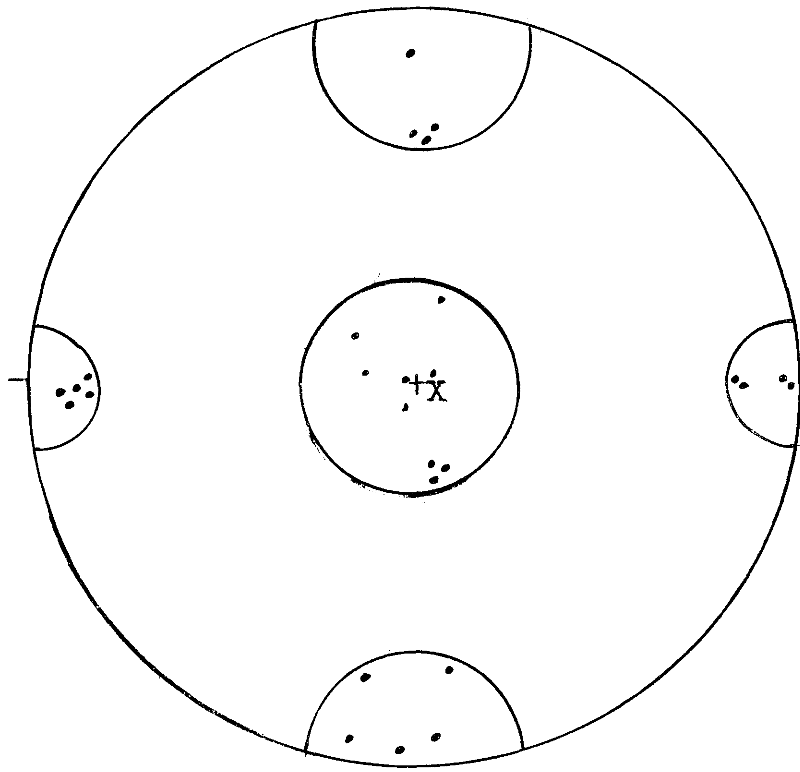

Fig. 10. Orientations of (100) poles of nine largest crystals grown. The plane of rolling is located at point X.

grains.

The initial rolling texture of cold-rolled steel is chiefly one in which [110] directions of the grains lie along the direction of rolling and {001} planes lie in the plane of the rolled sheet or bar (5).

Kurdjumow and Sachs (6) states that the primary rolling recrystallization texture of steel is (100) in the rolling plane and 15° from [110] in the rolling direction.

The recrystallization textures reported by Kurdjumow and Sachs differ from those found by the author.

V. CONCLUSIONS

Large single crystals of iron can be grown from decarburized A.I.S.I. C1018 steel by the strain-anneal method. Crystals grown from Ferrovac E iron by comparable techniques are neither as large nor free of minute sub-crystals.

The carbon content of 1/8 in.-thick A.I.S.I. C1018 steel may be reduced to less than 0.005 weight percent by a 48 hour treatment at 925°C in an atmosphere of wet hydrogen.

The critical amount of strain for maximum grain growth in a specimen that has a grain size of 180 grains per sq mm is 3-percent elongation by tension.

The best annealing temperature for C1018 steel is just below the A3 transformation temperature of 876°C. The time for maximum crystal growth at this temperature is 48 hours.

When the optimum conditions for grain growth are employed, a thin film of small surface crystals surrounds the large interior crystal. It is possible to avoid these surface crystals by inducing the strain for recrystallization by rolling. However, the recrystallization caused by rolling does not lead to grains as large as developed by the former method.

In the largest crystals grown, a highly preferred orientation was noted. This orientation has a [100] developed along the direction of straining and [010] perpendicular to the plane in which the bar had been previously rolled.

Appendix I

Determination of Crystal Orientation by X-Ray Analysis

The purpose of this appendix is to describe in detail the method used to determine crystal orientations.

A Greninger net (Fig. 11) was used to read angular relations of the back-reflection patterns. This net is reproduced in the proper size for a three-cm separation of specimen and film. Any row of spots on the film (Fig. 12) can be made to coincide with a hyperbola extending across the chart horizontally by turning the film about its center. The angle of inclination of the zone axis of the crystal to that of the pattern axis is given directly, and the degrees of rotation of the film that are required to get a row to coincide with a horizontal hyperbola are measured with respect to a vertical reference line through the center of the film.

The pattern is usually not very transparent and a copy of it can be made on tracing paper. When making this tracing one must be sure that it is copied from the side

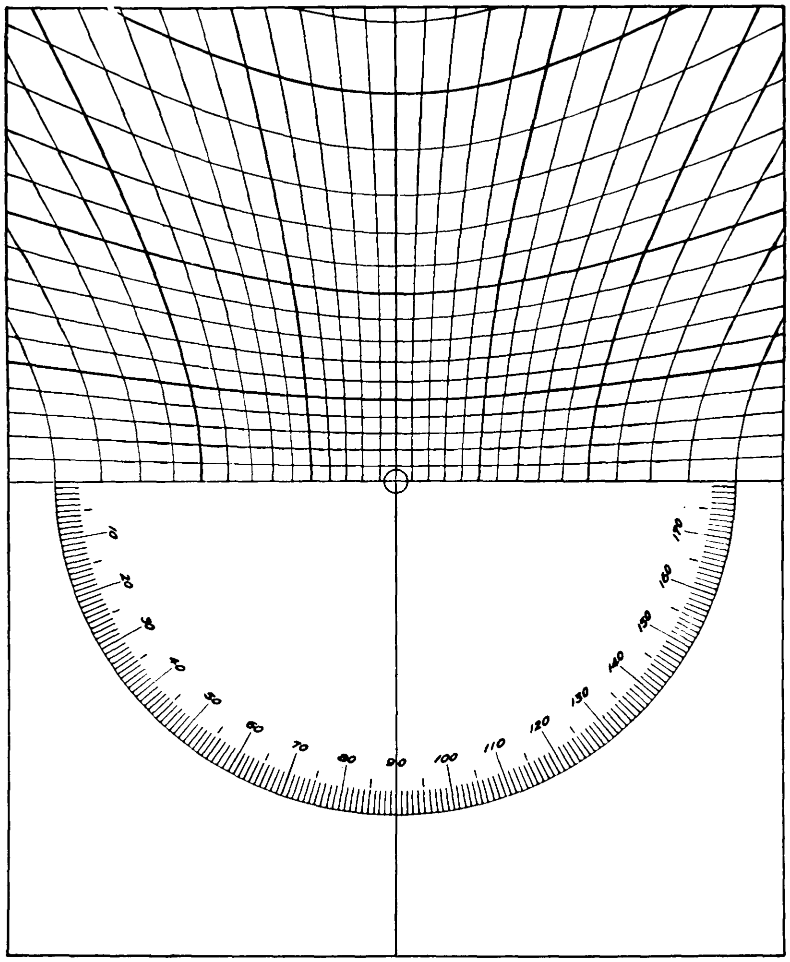

Figure 11. Greninger net. Printed for 3-cm. distance from specimen to film; graduated in 2° intervals.

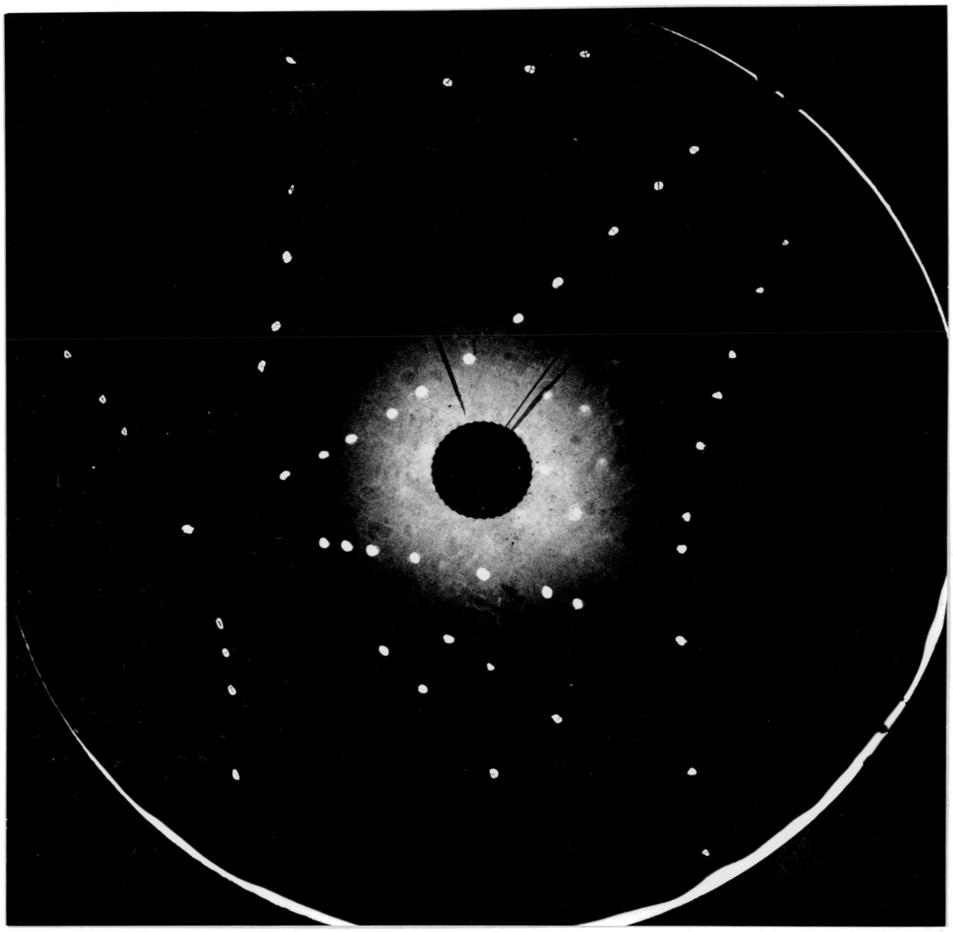

Fig. 12. Laue Back-reflection pattern of crystal No. 6.

opposite to that on which the reflected rays were incident, thus giving a projection of the crystal that corresponds to viewing it from the position of the x-ray tube.

The angles obtained for a zone axis from the Greninger chart are then used to plot the pole of the zone on tracing paper overlying a standard Wulff net. The procedure is to select a point of the pattern that appears to have symmetry corresponding to a (100), (110), or a (111) point and to plot on the Wulff net all the poles of the zone axis that pass through the point. The point itself, then plotted on the Wulff net, is rotated into the center; and all the poles of the zone axis rotate out to the circumference of the sphere. Then standard orientations such as (100), (110) and (111) are superimposed on the Wulff net to see which orientation will index all the poles of the zone axis that have been rotated to the edge of the net. Because this is a trial-and-error method, possibly none of the projections will index the points, thus indicating that the selected point is not indexed as one of the standards used. Other points are then tried until a fit is determined, and the point selected is indexed. Once one point is known, other points on the film can be indexed by measuring angles between the known and unknown points by checking these angles with a standard table. When several points on a film have been determined, it can be assumed that the orientation of the crystal is completed; only one point is not sufficient. With the orientation assured, a (100) point is rotated into the center of the Wulff net, and the crystal face is rotated out to appear on a standard (100) projection.

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