

THE GROWTH OF LARGE SINGLE CRYSTALS OF IRON

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Due to the increasing importance and demand as compared to the availability and cost of the large single crystals of metals and their oxides for the studies of metallurgy, corrosion, electronics etc. and their application in various new devices, the techniques of the large single crystal production have become of significant commercial importance. In this work the technique for the growth of large single crystals of 'Ferrovac E' and carburized zone-refined iron is described. The method was not successful in growing single crystals from zone-refined iron or nitrided zone-refined iron. Attempts to grow single crystals from 'Ferrovac E' were 90% successful.

Methods of growing single crystals of high purity iron have been reported in the literature.¹⁻⁴ However, each of these methods are not completely reliable when used on materials having slightly different chemical composition. Except for the method of Dunn⁴ it has not been possible to re-orient single crystals of iron to give any desired orientation. The method to be described has made it possible to grow single crystals from 'Ferrovac E' and to orient the crystals with respect to growth direction and surface. Dunns method of re-orientation was not successful when applied for zone-refined iron.

Starting Materials

The materials used during this investigation were (i) a vacuum melted high purity iron marketed by the Vacuum Metals Corporation designed 'Ferrovac E' and (ii) a zone-refined pure iron supplied by the Battelle Memorial Institute. The original composition of the two materials in (ppm) determined by mass spectrometry is shown in Table 1.

Procedure

The starting materials were cold rolled to a thickness of approximately 0.5 mm. The thin sheets were cut into strips of suitable dimensions. The specimens were then degreased and electropolished in 95% acetic acid/5% perchloric acid at 17°C. The zone-refined iron specimens were either carburized or nitrided in a quartz tube at

600°C for 30 min. Carburization was done in the presence of acetylene at an initial pressure of 2 mm; whereas nitriding was carried out in a continuous flow of dry nitrogen (at a rate of approximately 10 cm³/min). Specimens were then annealed at 700°C for 10 hr in a continuous flow of dry hydrogen. The annealed specimens were then strained by tensile elongation at a rate of 0.0002 in/min. Single crystals were grown successfully up to a straining speed of 0.003 in/min without any inhomogenous deformation. Total strains normally applied were 2.5 to 4.5%. When large strains were used it became difficult to restrict the nucleation of new grains in the matrix, which competed with the seed crystal for the growth of a single crystal. With strains less than 2% continuous growth of a single crystal in the whole specimen became difficult. After straining the portions of the specimen that were in the grips of the tensile machine were etched off in 50% HNO₃+10% HF+40% H₂O by covering the rest of the specimen with a thin layer of a lacquer. After etching the specimen was cleaned with acetone.

The temperature-gradient furnace employed for growing single crystals is shown in Fig. 1. Tap water was circulated through the copper jacket to maintain the temperature-gradient. The temperature-gradient in the furnace after setting at 875°C is shown in Fig. 2.

The strained specimen was then placed in the quartz tube as shown in Fig. 1. and the quartz

TABLE E. 1.

	Mg	Mn	Si	Cr	Ni	Al	Cu	Co	Sb	C	Sn
Ferrovac 'E'	1	3	50	30	20	10	100	20	5	70	3
Zone-refined	1	0.3	5	30	10	2	10	5	3	3	1

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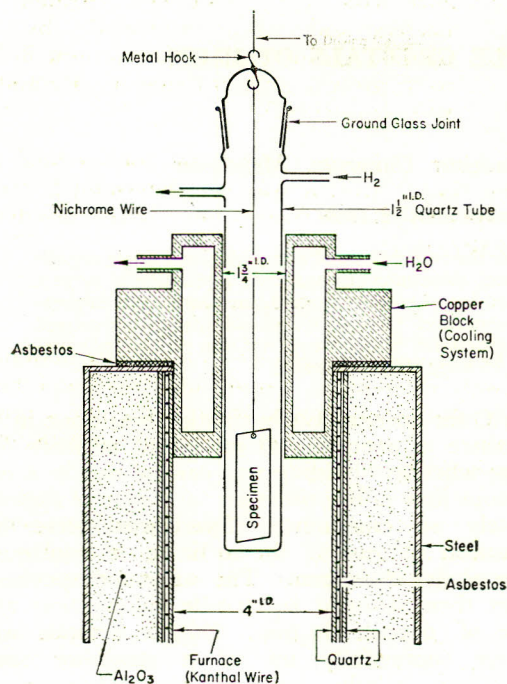


Fig. 1.—Cross-section of the temperature-gradient furnace 19 in high.

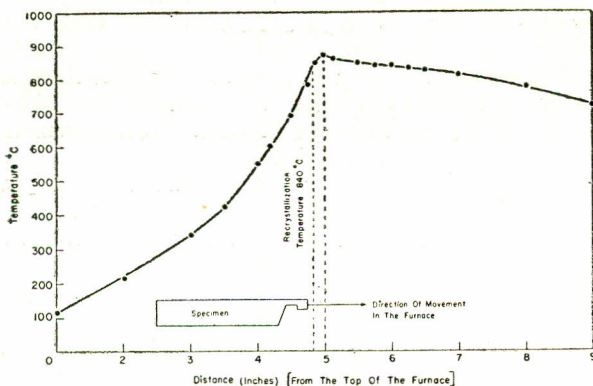


Fig. 2.—Temperature gradient in the furnace set 875°C.

tube was itself connected to a driving mechanism. The atmosphere inside the tube was flowing dry hydrogen. The tube was moved in the furnace through the maximum temperature region at a rate of 1 cm/4 hr. After about 2 cm of the specimen had passed the maximum temperature region, the specimen along with the tube was taken out. Usually three or four grains started at the leading end of the specimen. The orientation of each grain was determined by using the back-reflection Laue method. Only one grain was selected for re-orientation. The technique of Dunn⁴ was used to

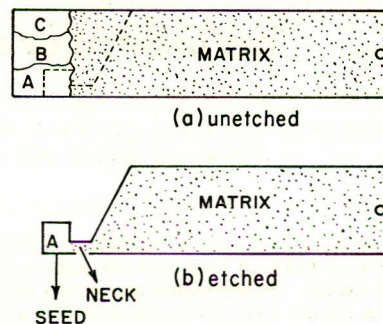


Fig. 3.—Shape of the specimen, (a) before and (b) after isolated seed crystal.

orient the crystal in both direction and the plane of growth. The selected grain and the rest of the specimen was covered with a thin layer of a lacquer and the extra grains etched out, as shown in Fig. 3. The specimen was cleaned with acetone. By use of a stereographic projection and a Wulff net the desired plane and direction of growth were located and the necessary rotations to bring the seed crystal to the desired orientation were determined. To facilitate bending the neck joining seed crystal and the matrix was maintained at about 700°C, by heating with a torch. After applying the required rotation, the specimen was passed through the temperature gradient furnace set at 875°C, at a speed of 1 cm/4 hr.

Results

Single crystals of Ferrovac 'E' having the plane (100) (110) (112) (111) (123) and several others, and the directions of [110] [001] [111] and a few others, were grown successfully. The single crystals of zone-refined carburized iron grown were of random orientations and it was not possible to re-orient them by the Dunn's technique. Because of the texture in the annealed metal certain orientations grow more readily than others and it was observed that certain crystals were more imperfect in some orientations. The suggestion that a minimum amount of carbon is necessary for the single crystal growth of iron⁵ was found to be true in zone-refined pure iron.

Figure 4 shows a few of the specimens after various treatments.

- (a) Ferrovac 'E', 3.5% strained. (110) plane [001] direction. Small end portion is polycrystalline, this portion was not passed through the highest temperature region.

- (b) Ferrovac 'E', 6% strained. After applying such a high strain it was not possible to control the nucleation of another crystal.
- (c) Ferrovac 'E' 1.8% strained. A polycrystalline portion is left; probably the applied strain was not enough for continuous growth.
- (d) Ferrovac 'E', 3.2% strained. This specimen was electropolished after the seed crystal growth. Polycrystalline portion and non-uniform growth probably suggests that some of the necessary strain was removed during electropolishing.
- (e) Zone-refined iron, 3.5% strained. Carburized, annealed and strain-annealed (without growing seed crystal).
- (f) Pure iron, 3.5% strained. Nitrided, annealed and strain annealed. By this method grains only of a few mm in dia were grown, and nitrogen as a impurity was not successful.

Carbon concentration in the zone-refined iron after crystal growth was not determined, but it is expected that it would have been considerably reduced.¹

Summary

The critical requirements for the growth of single crystals of iron are: annealing treatment below the growth temperature before straining, a critical strain of 2.5–4.5% and a growth temperature in between 825–890°C.

For a continuous growth of a crystal in a specified plane and direction the initial growth of a seed crystal and its re-orientation is necessary.

The presence of carbon in a critical amount as impurity is necessary.

Use of nitrogen as impurity for single crystal growth is not successful.

References

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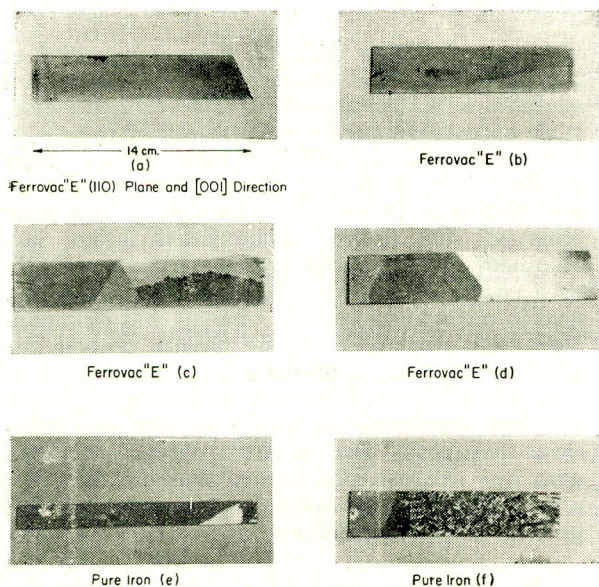


Fig. 4.—Crystal growth in Ferrovac E and pure iron.