

## 2. X-Ray Studies on Cast Structure of 4% Si-Steel in the Light of Anisotropy of the Rate of Crystal Growth

Hideo TAKAKI, Masashige KOYAMA and Hidekiyo FUJIHIRA\*

(H. Takaki Laboratory)

*Received January 24, 1958*

It was assumed by one of the authors that the growth rate in the  $[001]$  direction would be far larger than that of others on the preparation of the single crystals of Si-steel. On the other hand, it is well known that the direction of growth of columnar crystals in cast metals which form the cubic lattice is always parallel to the  $[001]$  direction preferentially. Therefore, it seems very interesting to study the relationship between the above two phenomena.

An X-ray examination was performed by both the Laue and the rotation methods from the view-point above mentioned. From this X-ray examination on the cast structure of 4% Si-steel, the following results were obtained :

(1) The practical columnar structure of this Si-steel was complex, as the condition was not so simple as in the case of the preparation of single crystals.

(2) The direction of growth of columnar crystals was parallel to the  $[001]$  direction.

(3) At the ingot surface, very small crystals grown by severe supercooling were arranged at random. But it seemed that among these, the crystals whose  $[001]$  directions were not parallel to that of thermal gradient, were gradually superseded by the ones, the  $[001]$  directions of which were parallel, during growth and the so-called columnar structure of  $[001]$  finally resulted in at the inner part of ingot.

In addition to the X-ray examination above mentioned, the most part of crystals contained in the specimen was separated each other, by electrolyzing the specimen in the potassium dichromate solution. The length of the longest one among these crystals thus separated was 25 mm.

The following results were also obtained :

(1) Although the microscopic structure should show only the ferrite at the equilibrium state, the result obtained showed unexpectedly the "Widmanstätten Structure" of pearlite, probably owing to the partial super-saturation of carbon.

(2) Fractures formed by cutting the ingot with acetylene gas were not intergranular but crystalline and it was found by X-ray analysis that the fracture planes

---

\* 高木秀夫・小山昌重・藤平秀清

were parallel to the plane (100) or (112).

### Introduction

It is well known that the preferential growing direction of cast structure of metals and alloys is particular about their crystal structures, but the reason with respect to this fact has not yet been accounted for. It has been reported by E.Schmid *et al.*<sup>1)</sup> that the preferential growing direction of columnar crystals in the cast silicon-steel forming the bodycentred cubic lattice, is parallel to the [001] direction. On the other hand, from the consequence of the investigation on the preparation of single crystals of the silicon-steel which was previously performed by one of the authors<sup>2)</sup>, it seems that the growth rate of the [001] direction is maximum, namely the preferential growing direction of cast structure in the former investigation<sup>1)</sup> may be relative to the result in the latter one<sup>2)</sup>. Accordingly, from both the X-ray method and the observation on the numerical change of the crystal grains contained in the various parts of specimen, an investigation on the preference of the growing direction in cast silicon-steel was carried out. In this X-ray examination, the heterogeneous X-rays emitted from Fe anticathode was utilized. The results thus obtained will be briefly described below.

### Specimens

As for specimens, one part of the ingot of 4% silicon-steel (930 Kg. in weight) produced at the Kawasaki Iron Mfg. Co. (bottom poured) was used and the size of it is shown in Fig.1. Two smaller blocks were cut off by acetylene gas from the position A and B shown in Fig.1, towards the direction perpendicular to the ingot surface and that of the ingot centre. The parts effected by acetylene gas were then sharpened away with the lathe.

### Experimental Results and Discussions

#### (a) Macro-Structure

The side section surface of the position A in Fig.1 was polished and etched with the 50% aqua regia. The macro-structure thus obtained is shown in Fig.2. Furthermore, the same surface of the specimen was etched by the three etching solutions: 50% hydrochloric acid, 3% alcohol solution of picric acid and 10% nitric acid. Nevertheless the so-called dendritic structure was not observed. While, on the specimen in Fig.2, the numbers of crystals contained in the lines parallel to the ingot surface were counted by eye piece, in which case each line was drawn to be parallel to the ingot surface towards the inner part of ingot at the distance of 1 mm. As the result, it seems that the number of grains firstly decreases little by little but hardly does at the part of columnar zone far from the ingot surface.

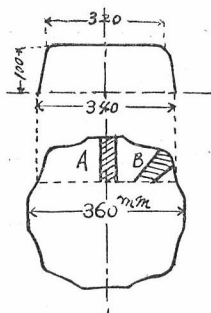


Fig. 1.



Fig. 2. Macro-Structure  $\times 3/4$

#### (b) X-Ray Examinations

It is generally known that the so-called chill crystals formed by the severe supercooling are arranged at random at the ingot surface and then the so-called columnar crystals, the  $[001]$  directions of which are parallel to that of thermal gradient, grow towards the direction perpendicular to the ingot surface but the growth of these columnar crystals is suddenly stopped and the large equiaxed crystals are again formed at the far inner part of ingot. Now, X-ray examinations were carried out by means of the following four methods, under the consideration that among chill crystals, those, the  $[001]$  directions of which are not parallel to that of thermal gradient, will be superseded gradually by the ones whose  $[001]$  directions are parallel, during growth and that only the columnar crystals of the  $[001]$  directions will remain finally.

(1) X-rays were transmitted perpendicularly to the two thin plates which were respectively cut off perpendicularly to the direction of thermal gradient at the sections 1 and 2 shown in Fig. 2.

The results of this X-ray analysis by the Laue method are shown in Fig. 3. It may be seen from the figures that on the specimen of section 2, the stereographic plots are assembled near to the  $[001]$  direction in the stereographic triangle diagram, but on the other specimen the plots are at random.

(2) Rotation photographs were taken with the plate specimen of 1 mm. in thickness which was cut off from the position 3 in Fig. 2, perpendicularly to the plane of photograph and towards the direction of thermal gradient (here, the rotation axis was

preferred to be parallel to the direction of thermal gradient).

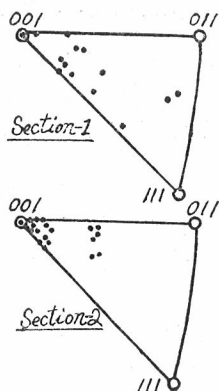


Fig. 3.  
●, the stereographic plot of the crystal direction parallel to the direction of thermal gradient.

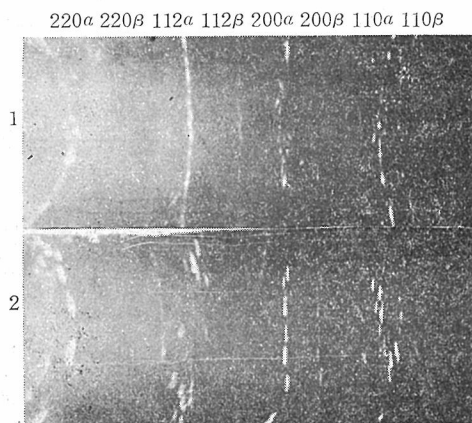


Fig. 4. 1 - Ingot surface of Part 3.  
2 - Inner part of Part 3.

The diffraction patterns thus photographed from the various parts on the specimen, are shown in Fig.4. It may be seen from the photographs that the diffracted points from the planes (110), (200), (112) and (220) are all arranged uniformly on the photograph obtained from the part of ingot surface but those from the planes (110), (200) and (220) are passably assembled on the equatorial line on that obtained from the inner part of specimen.

(3) Rotation photographs with the rod specimen of 1.5 mm. in diameter which was cut off from the position 4 in Fig.2 towards the direction of thermal gradient, are shown in Fig.5 (rotation axis was preferred to be parallel to the direction of thermal gradient).

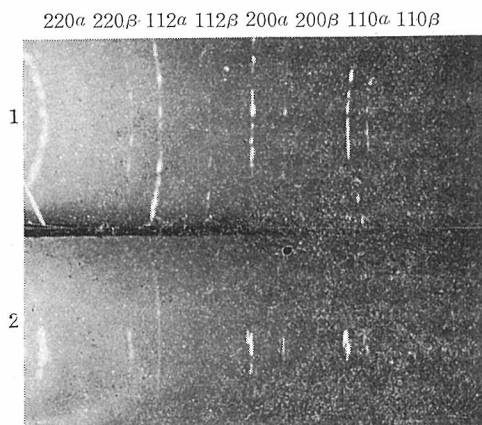


Fig. 5. 1 - Ingot surface of Part 4.  
2 - Inner Part of Part 4.

It may be seen from the photograph taken from the part far from the ingot surface, that the diffraction points from the planes (110), (200) and (220) are all perfectly assembled on the equatorial line: *i.e.* the growing direction of columnar crystals contained in the inner part of ingot, is parallel to the [001] direction. In comparison with the experimental result examined by E. Schmid *et al.*,<sup>1)</sup> the growing direction of our columnar crystals seems to be more exactly parallel to the [001] direction.

(4) Laue photographs were taken from many crystals contained in the plate specimen which was cut off from the position B in Fig.1 perpendicularly to the ingot

surface and to be parallel to the direction of thermal gradient towards the inner part of ingot.

In this case, X-rays were transmitted perpendicularly to the plate specimen. The arrangement figures of crystals are shown in Fig.6 and the stereographic plots of crystal directions parallel to the direction of thermal gradient, in Fig.7. Three specimens E, F and G in Fig.6 were originally one specimen but it was divided into

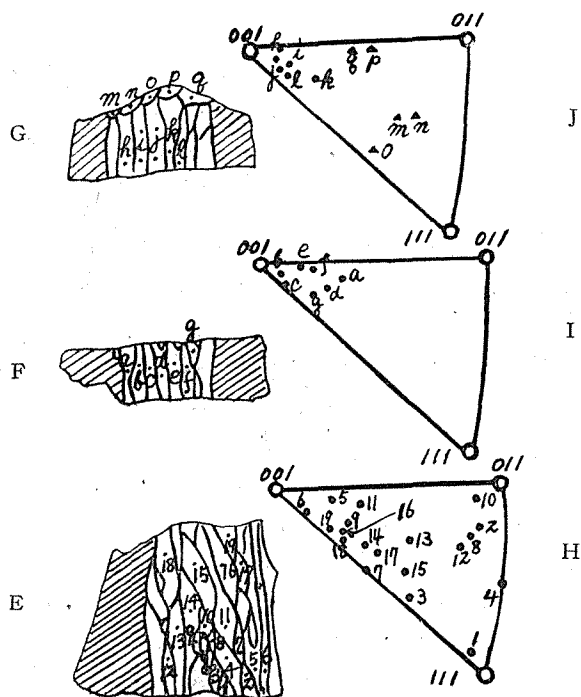


Fig.6.

Fig.7.

- ①. the same plot as in Fig.3.
- ②. the stereographic plot of the larger equiaxed crystal formed at the for inner part of columnar zone, the direction of which is parallel to the direction of thermal gradient in the columnar zone.

three sheets, and as each sheet was etched by aqua regia of 50% to make these sheets suitable for X-ray exposure, the relationship among these sheets became indistinct. In Figs.6 and 7, E, F and G are correspondent to H, I and J respectively, E having been taken from the nearest part to the ingot surface among these sheets. It may be seen from I and J in Fig.7 that the crystal directions parallel to the direction of thermal gradient are near to the  $[001]$  direction on the whole, concerning the crystals contained in the inner part of columnar zone. However, the stereographic plots of the crystals contained in the specimen are arranged at random (H in

Fig.7).

Under the minute inspection of the result with respect to the specimen E, the following fact is observed: Longer crystals (*e.g.* crystals 5,6) are near to the  $[001]$  direction but shorter crystals (*e.g.* crystals 1, 2, 3, 4, 8, 10, 17, *etc.*) are very far from the  $[001]$  direction. On the other hand, the crystals 13, 15, are far from the  $[001]$  direction despite of their longer length. However, if their stereographic plots are transformed to the projection parallel to the direction of crystal growth, these plots turn to be comparatively near to the  $[001]$  direction. Still more, it seems that the larger equiaxed crystals formed at the far inner part of columnar zone are arranged at random.

### (C) Separation of Each Crystal

In order to observe the more minute appearance of crystal growth, the same electrolysis examination as that of W.E. Ruder<sup>2)</sup> was performed with the plate specimen of 2 mm. in thickness, in which case this specimen had been cut off from the lower part of specimen E at the position B in Fig.2. After annealing the above specimen at 800°C for 17 hrs. in air and leaving the oxides formed on the surface of specimen, most part of crystals contained in the specimen was separated each other by electrolysis this annealed specimen in the 1 *N* potassium dichromate solution with current density of 0.4 A/cm.<sup>2</sup> for 5 hrs. Among the crystals thus separated, the length of the longest one was 25 mm. and the mean length of the small crystals separated from the part near to the ingot surface of specimen was 2.6 mm.

Besides, another specimen of 8 mm. in thickness was electrolysed under the same condition as before. As a result, it was found that the length of the crystals 8, 9, 17, *etc.* shown in E of Fig.6 did not denote the transversal planes formed by cutting the long crystals obliquely, but their real lengths on the whole : *i.e.* small crystals are formed even at the part of columnar zone.

The width of single crystals thus separated varies according to its parts and its boundary is slightly wavy. Still more, wherever crystals may exist in the columnar zone, the common fact between the two crystals grown in parallel each other is that the crystal arranged at the more inner part of columnar zone gradually increases its width but the width of that at the more outer part decreases gradually.

Our aforesaid consideration on the cast structure is inferred to be reasonable on the whole according to the results of (a), (b) and (c) above mentioned. However, the crystals, the  $[001]$  directions of which are parallel to that of thermal gradient or near, do not laterally develop so much and this result seems to be due to the fact that the crystal growth is governed by the thermal gradient very much; while it may be also inferred that the impurities (*e.g.* carbon) contained in the specimen interrupt the crystal growth.

**(d) Abnormal Structure**

According to the microscopic examination of the specimen etched by 10% alcohol solution of nitric acid, the pearlite structure was unexpectedly observed along the grain boundaries or sometimes the so-called "Widmanstätten Structure" of pearlite was observed in grains as shown in Fig.8. After examining minutely, this pearlite structure was hardly observed at the part near to the ingot surface but gradually formed along the grain boundaries and then on the columnar zone it was visible even at the inner parts of grains; while, on the equiaxed crystals this structure was also found at the both portions above stated. However, after annealing this specimen at 850°C (a temperature above its eutectoid one) for 16 hrs. in air and cooling slowly in the furnace, the pearlite structure was not observed.

Originally at the perfect equilibrium state, the microscopic structure of this specimen should show only the ferrite; nevertheless, the fact that the pearlite structure is observed is very incomprehensible. As the part effected by cutting with acetylene gas was sharpened, it may not be inferred that this is the cause of this structure. Accordingly, this abnormal structure probably seems to be due to the partial super-saturation of carbon by rapid cooling, and this irregular distribution of carbon may be inferred to be one of the cause of the tendency that the single crystals do not develop towards their transversal directions.

**(e) Fracture Planes**

Lastly, when the ingot was cut by acetylene gas to take out specimens, many cracks were induced in specimens by thermal stress. One of these specimens was then divided by hammering along these cracks and its macro-structure is shown in Fig.9. The fractures thus formed were not intergranular but crystalline, and it was

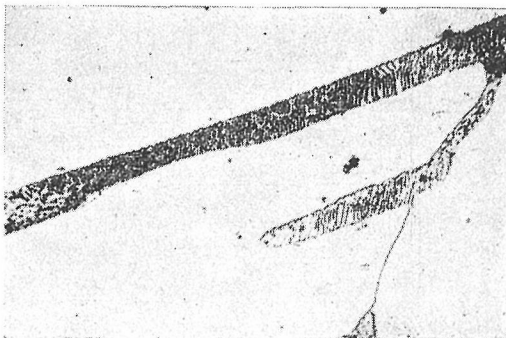


Fig. 8. Pearlite Structure  $\times 350$



Fig. 9. Fracture Plane (Natural Size).

found by X-ray analysis utilizing the back reflexion method that the fracture planes were parallel to the plane (112) or (001). This result is coincided with that obtained by C.F.Tipper<sup>4)</sup>.

### Conclusion

In this investigation, no minute examination was carried out with respect to the equiaxed crystals which were suddenly formed at the far inner part of columnar zone.

Our consideration on the columnar structure seems to be applicable to the specimen used in this examination on the whole. However, as this specimen contains many impurities a more minute examination will be next carried out with the super pure metal which can be easily melted and cast in our laboratory, under the simple condition.

In conclusion, the authors wish to express their best thanks to Dr.M.Imai of the Kawasaki Iron Mfg. Co., who supplied them with the blocks of Silicon-steel used in this experiment.

### References

- (1) F.C.Nix and E.Schmid: *Z. Metallkunde*, **21**, 286 (1929).
- (2) H. Takaki and Y. Nakamura: Unpublished.
- (3) W. E. Ruder: *J. Ind. Eng. Chem.*, **5**, 452 (1913).
- (4) C. F. Tipper and A. M. Sullivan: *Trans. Amer. Soc. Metals*, **43**, 906 (1951).