X-Ray Studies on the Cast Structure of High Purity Aluminium in the Light of Anisotropy of the Rate of Crystal Growth. (I)

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It has been well known that the columnar crystals in cast metals have strongly preferred orientations. An X-ray examination was performed, in the light of anisotropy of the rate of crystal growth, concerning the high purity aluminium ingot (99.993%) which was melted and cast in vacuum.

In the central portion of the ingot, those favorably oriented crystals which are near the [001] direction (direction of easy growth) survived by crowding out the unfavorably oriented crystals. In the outer portion of the ingot, however, even the unfavorably oriented crystals unexpectedly developed, probably owing to the more rapid cooling of the outer portion than the center.

I. INTRODUCTION

When the molten metal or alloy solidifies, starting at the cold walls of the container, a thin layer consisting of small equiaxed crystals of random orientation (chill crystals) is formed first. Adjacent to this skin a zone of large columnar crystals of strongly preferred orientation develops which either fills the rest of the casting completely, or is followed by another layer of small equiaxed crystals, again of random orientation. The preferred orientation of columnar crystals is related to its crystal structure. Schmid[1] has studied the relation between the preferred orientation and the crystal structure in various metals : e. g. in cubic metals the preferred orientation is parallel to the [001] direction, in tetragonal metals to the [110] direction.

Tammann[2] has studied in aluminium and nickel ingots the crystallographic direction perpendicular to the mould surface of crystal grains existing in each layer parallel to the mould surface. From his study it was made clear that the directions perpendicular to the ingot surface of the crystal grains existing in the columnar csystal zone were mostly parallel to the [001] direction.

One of the authors[3] has succeeded in preparing single crystals of nickel and silicon steels (3.2 and 4 % silicon) oriented in any crystallographic direction, by the Bridgman

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method, using the vacuum Tammann furnace. In this case, a result was obtained that the [001] direction was the direction of far easier growth than other direction.

When molten metals are poured into a cold mould, then the extraction of heat will take place through the mould walls. The layer of the melt close to the wall will solidify first and a large number of grains which nucleate at random along the walls will start to grow into the melt.

On account of the anisotropy of the rate of crystal growth, grains in which the direction of [001] is parallel to the direction of heat flow will grow faster and hence farther (e.g. in cubic metals). In so doing, they will crowd out the grains of different orientations (assumed to grow slower) and form a zone of ingot having a strongly preferred orientation.

In the light of this anisotropy of the rate of crystal growth, an X-ray examination[4] was previously performed with respect to the specimens cut off from a part of the ingot of four percent silicon steel (930 kg in weight) produced by the Kawasaki Iron Mfg. Co. However, the analysis was difficult as the used specimen contained various impurities and gases and its cast structure was also very complex. The high purity aluminium was therefore melted and cast in vacuum, and a method was devised in which the bottom of mould was cooled with water so that the columnar crystals grow perpendicularly to the bottom of ingot. The cast structure in the ingot produced by the method mentioned above, was examined in the same light as before and some of the results were already reported.[5] Further study is carried out here.

II. EXPERIMENTAL PROCEDURE

1. Preparation of Specimens

The purity of aluminium was 99.993 percent (silicon, 0.003 ; iron, 0.002 ; copper, 0.002 percent). The apparatus of melting and casting in vacuum is shown in Fig. 1 in which the inner walls of both the crucible and the path through the connecting portion between the melting part and the casting one, were preheated at 1000°C, after being covered with alumina powders mixed with water.

When the melt reached the desired temperature, the apparatus was so held up that the upper surface of the melt became parallel to bottom of mould, after lifting up the apparatus from the electric furnace and pouring the melt into the mould.

In order that columnar crystals grow perpendicularly to the bottom of mould, the outer surroundings of the mould were preheated with two gas burners before casting, and the bottom of mould was cooled with water. Therefore, the cast structure in ingot is assumed to be related with the temperature of the melt, the preheating temperature of mould and the cooling method with water. In this case, the melt was always cast at 800°C and the mould was simultaneously cooled after casting.

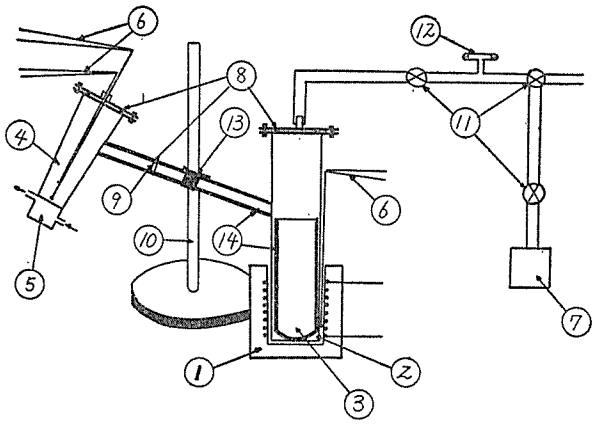

Fig. 1. Apparatus for melting and casting.

(1) Electric furnace, (2) Melting part (stainless steel), (3) Crucible, (4) Mould (stainless steel), (5) Cooler, (6) Thermocouple, (7) Vacuum pump, (8) Rubber packing, (9) Connecting part (stainless steel), (10) Stand, (11) Cock, (12) Geisler tube, (13) Handle, (14) Alumina lining.

In order to know the approximate rate of crystal growth in ingot, two sets of thermocouple held in the porcelain tubes of the outer diameter of 3 mm, were situated at the points P and Q in Fig. 2. Then the cooling curves were obtained,

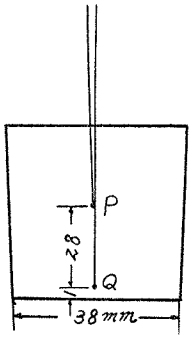

Fig. 2. Model figure of the longitudinal section of ingot at its center.

by measuring the change of temperature at the interval of five seconds.

The vacuum obtained by the rotary pump alone was about $10^{-1}\sim10^{-2}$ mm of Hg in the molten state.

The preparing conditions of the ingot A used for the X-ray analysis are as follows: The preheating temperature of the mould was 750°C and 730°C at the points P and Q in Fig. 2 respectively. Further, the mould was immediately cooled after casting.

The cooling curves obtained are given in Fig. 3. Considering the temperature

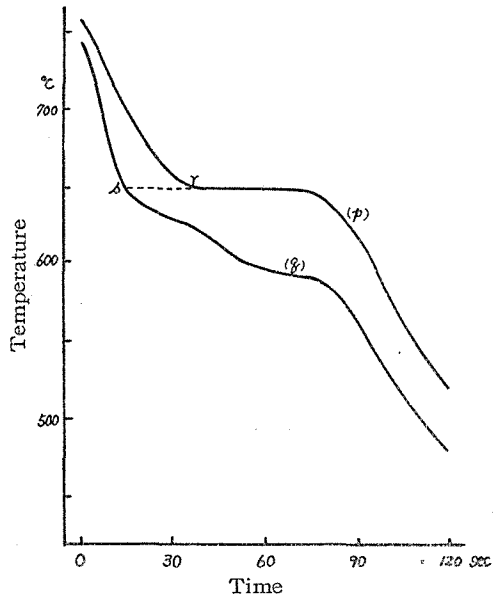

Fig. 3. Cooling curves at the points P and Q in Fig. 2.

of the horizontal part in the cooling curve p at the point P as the solidification temperature, the time difference (r-s) between the time r in which the curve p started to become horizontal and the time s in which the cooling curve q at the point Q passed through the solidification temperature mentioned above, was 25 seconds. The distance between the points P and Q was 28 mm. The approximate rate of crystal growth in the ingot A becomes, therefore, about 67 mm per minute from the above values.

2. Experimental Method

As shown in Fig. 4, the plate-form specimen C was cut off perpendicularly to

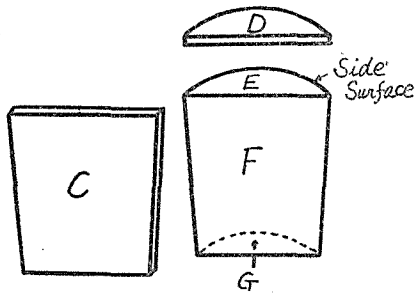

Fig. 4. Model figure.

the bottom of ingot from the half of the ingot cut perpendicularly to the bottom of ingot at its center. The plate-form specimen D was also cut off to be parallel to the bottom, from the upper part of the residual half of the ingot, after cutting off the specimen C. Then the specimens C and D were polished with emery

papers and etched in cupric chloride solution.

X-rays were transmitted perpendicularly to the specimens C and D, and the direction of each crystal normal to the bottom of ingot, was stereo projected.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

1. Macrostructure

The macrostructures of the specimens C and D etched in cupric chloride solution, are shown in Figs. 5 and 6. In Fig. 5 some crystals disappear soon after germinating

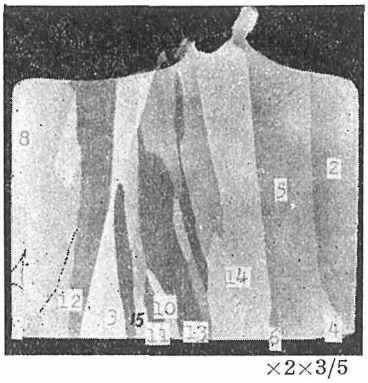

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Fig. 5. Macrostructnre of the specimen C.

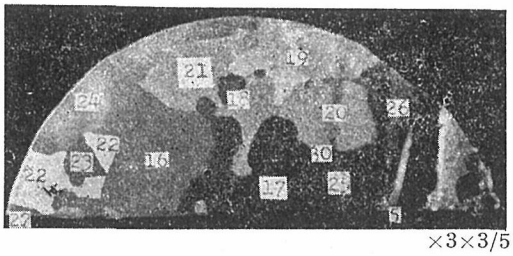

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Fig. 6. Macrostructure of the specimen D.

but others develop strongly.

The macrostructures of the upper portion E, the sectional surface F, the bottom surface G and the side surface in Fig. 4 (each was etched in dilute hydrochloric acid for a long time), are shown in Figs. 7~10 in which crystals are denoted with numbers.* Some crystals not denoted with number, were omitted from the X-ray transmission Laue analysis, owing to their superposition with another crystals.

The specimen shown in Fig. 9 (corresponding to the primary period of the crystal growth) consists of much more crystals than those in the specimen shown in Fig. 6 (corresponding to the last period of the crystal growth). Therefore, the crystals in the former specimen are smaller than those of the latter. This may be attributable to the preferential growth. The dendrite structure seen in the outer round of the specimen in Fig. 7 and also in the upper parts in Fig. 10, will be discussed in the next report.

2. X-Ray Analysis

In Figs. 11 and 12 are given the results of X-ray analysis of the specimens C and D shown in Figs. 5 and 6. The crystals 3,5 and 14 which are near the direction

* Some crystals in the specimen shown in Fig. 10 have the same numbers, because of the fact that three photographs were taken at the side surface, owing to its round shape.

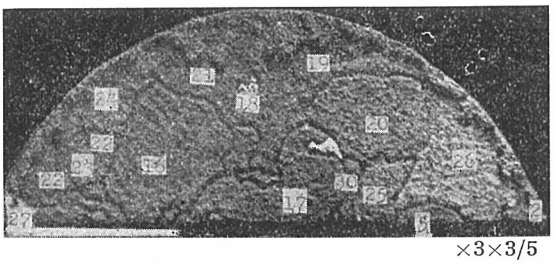

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Fig. 7. Macrostructure of the surface E in Fig. 4.

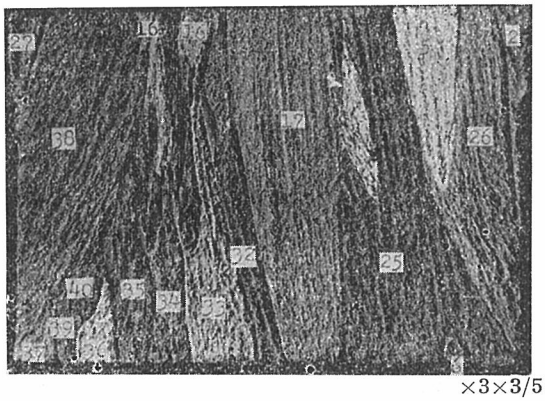

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Fig. 8. Macrostructure of the surface F in Fig. 4.

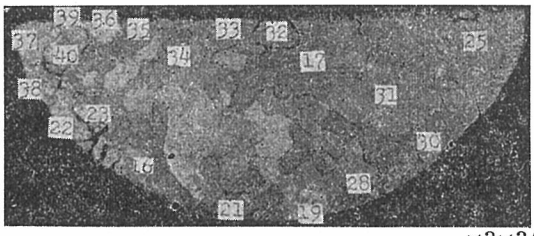

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Fig. 9. Macrostructure of the bottom surface G in Fig. 4.

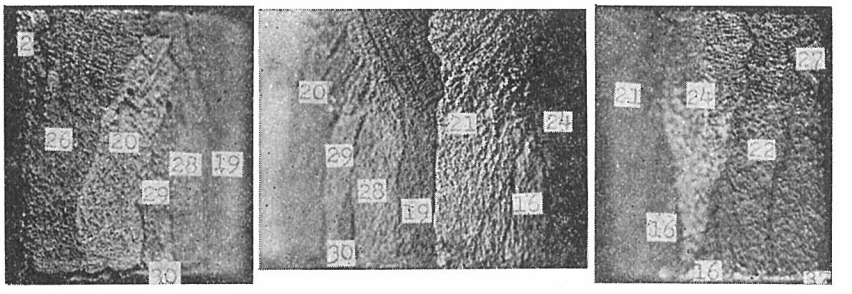

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Fig. 10. Macrostructures of the side surface in Fig. 4.

of [001] develop strongly, whereas the crystals 4, 6, 7, 10, 11 and 13 which are

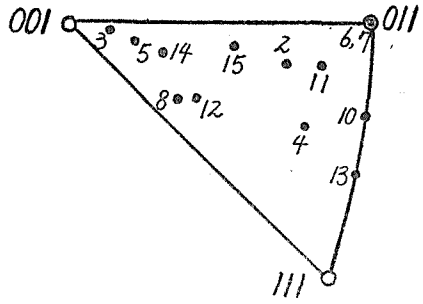

Fig. 11. Stereographic projection points of the columnar crystals in Fig. 5 whose directions are perpendicular to the bottom of ingot.

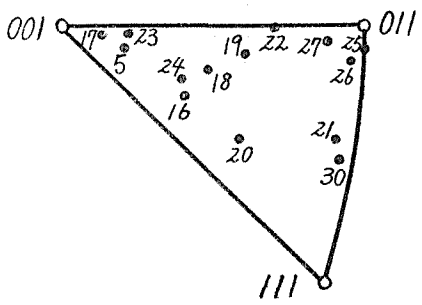

Fig. 12. Stereographic projection points of the columnar crystals in Fig. 6 whose directions are perpendicular to the bottom of ingot.

far from the [001] direction disappear soon after the growth. The crystals 12 and 15 disappear at the intermediate period (the crystal 15 is a little longer behind the surface than on the figure). The crystals 15, 11, 10 and 13 which become far from the direction of [001] in this order, crowd out each other in the inverse order. The crystal 2 develops strongly despite its being very far from the direction of [001] , and this abnormal phenomenon will be discussed afterwards.

In Fig. 12, the large crystals 5 and 17 are oriented very nearly in the direction of [001] . The crystal 23 is small, despite its being oriented very nearly in the direction of [001] . But as seen in Fig. 9, this crystal is very small at its start and is a little larger than its size at the primary period. The crystals 25 and 27, far from the direction of [001] , are small, whereas the crystals 21, 22 and 26 very far from that of [001] , are very large. This abnormal phenomenon may contradict the consideration of the authors. However, all of these abnormal crystals germinated at the outer round of the ingot and it seems that under the present condition the outer round of the ingot was more rapidly cooled than its center. If so, the interface between the solid phase and the liquid one should be concave to the direction of growth. Therefore, following the idea proposed by Chalmers,[6] the crystals which germinated at the outer round of the ingot should develop towards the center of the ingot, even if these directions are very far from the [001] direction (e.g. the crystal 2).

In order to ascertain the above mentioned phenomenon, the residual block after the specimens·C and D were cut off from the ingot shown in Fig. 4, was sectioned to be parallel to the bottom of ingot at the positions 4, 8, 15 and 22 mm from the bottom respectively. Figs. 13, 14, 15 and 16 show the macrostructures of these upper sectional surfaces and the changes of both the size and the shape of each crystal can be clearly observed from these figures. The crystals denoted by the same numbers as in Figs. 6, 7 and 9 are the same ones. Fig. 17 shows the

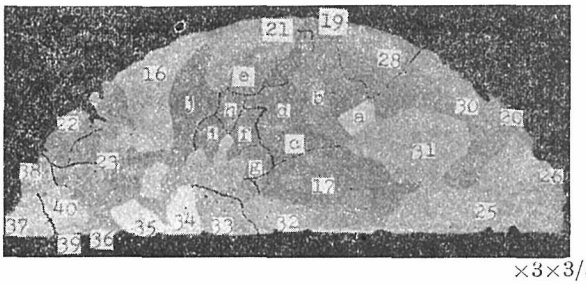

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Fig. 13

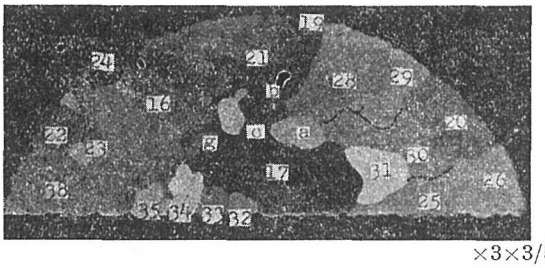

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Fig. 14

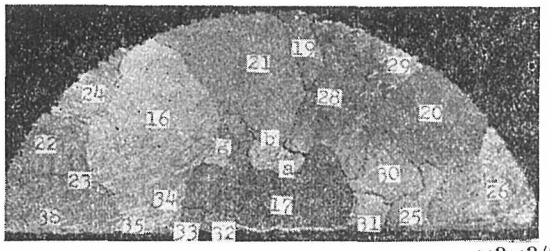

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Fig. 15

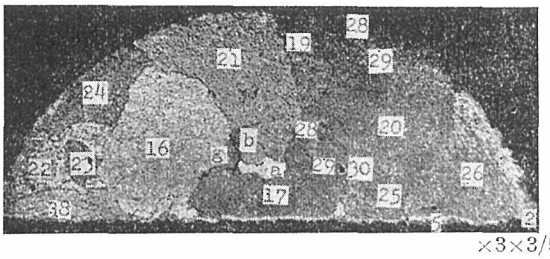

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Fig. 16

stereographic projection points of these crystals perpendicular to the bottom of ingot. The crystals denoted by the underlined numbers are those which have already germinated at the bottom of ingot and survived up to the upper surface. At the start, the crystals 17, 25, 30 and 31 are large and the crystals 16, 21, 28 and 33 are fairly large, whereas the crystals 19, 22, 32, 34, 35, 36, 37, 38, 39 and 40 are small. The crystals 20, 24, 26, 27 and 29 which germinated midway, not at the start,

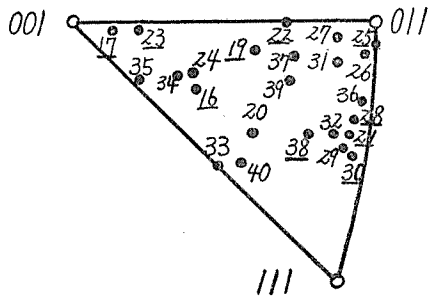

Fig. 17. Stereographic projection points of the columnar crystals in Figs. 13~16 whose directions are perpendicular to the bottom of ingot.

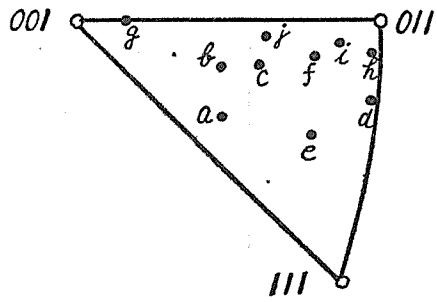

Fig. 18. Stereographic projection points of the crystals a~j in Fig. 13 whose directions are perpendicular to the bottom of ingot.

gradually developed inward. However, these crystals except the crystal 24 are all very far from the [001] direction. The crystals 25, 30 and 31, far from the [001] direction, are suppressed by the crystals 20 and 26 germinated at the outside, while the crystal 29 is suppressed by the crystals 19 and 20 nearer to the [001] direction than the former, in spite of its germination at the outer round of ingot.*

The crystal 23 is small in spite of its situation near the direction of [001] but it was far more smaller at the start and it would have been suppressed by the crystal 22 germinated at its outer position if it was far from the [001] direction. It developed after a fashion, owing to its situation near the direction of [001] .

Fig. 18 shows the stereographic projection points of the small crystals a~j in Fig. 13 perpendicular to the bottom of ingot. It can be considered from this figure that most of the crystals soon disappeared after the germination.

Further studies concerning these problems will be treated in the next report.

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* The crystals 28 and 29 are divided by the crystal 20 on the way.