Substructures in Single Crystal Foils of Tin Grown from the Melt

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For one reason of the investigation of a relationship between the thickness and the formation of striated subboundaries in the crystals grown from the melt, and for the other reason of the direct observation of dislocations in thin crystals by Lang's X-ray diffraction method, a technique for producing single crystal foils of tin from the melt was devised[1]. The single crystal foils of tin grown by this technique, however, were pitted with small pox and had several cavities on their surfaces, because molten tin was sucked up into a special type of glass mould in air, the inner wall of which was previously coated with silicon oil. In addition, it was impossible by this method to produce the single crystal foils less than 100μ in thickness.

To improve the disadvantages mentioned above, after vacuum melting, molten tin was pushed into a special type of glass mould with a pressure of N_2 gas as shown in Fig. 1. Then this mould was sealed in an evacuated glass tube and was lowered in a vertical furnace of 25°C in temperature gradient. In this case, the seed crystal used was produced by a technique devised by one of the authors, in which the pre-existing striated subboundaries were eliminated by the glass tube tip inserted in the outer glass tube[2]. The glass tubes, T_1, T_2, were dissolved in HF solution after growing as shown in Fig. 2 and the glass plate, P, was also dissolved after cutting off both the ends of foil part with an acid saw. The foils thus prepared, were examined both by Berg-Barrett method and by a transmission type-bent-quartz monochromator**. By this technique, the single crystal foils of tin of 80μ in thickness will be obtained.

The foils examined in this study were 99.8 and 99.999% in purity, 100~200μ in thickness, and were grown in (001)-[110] orientation at the rate of 2~0.07 mm/min.

The Berg-Barrett photograph given in Fig. 3 shows an example of an incubation period prior to generation of striated subboundaries (in this specimen, growth rate, thickness, and purity are 2 mm/min, 100μ and 99.999% respectively).

Fig. 4 shows two sets of Berg-Barrett patterns which were reflected from (112) plane in the crystal grown at the rate of 0.31 mm/min (99.999% in purity). In this figure, two pieces of Berg-Barrett photographs taken from the different

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** CuKα was used.

parts on the surface of the specimen are patched together. The left photograph, (a), is that from the seed crystal part of 4 mm in diameter, and the right one, (b), is that from the single crystal foil of 200μ in thickness (the formation of pitted surface is attributed to silicon oil coated on the glass plate of mould). The wavy lines running obliquely which are indicated with a pair of arrows are the joints mentioned above. In (b), the striated subboundaries manifested by bright striate regions or by dark ones, are observed in parallel with each other after an incubation period—these subboundaries have penetrated the specimen to the reverse side—, whereas in (a) the irregular subboundaries are observed throughout the section, presumably for the migration of subboundaries after solidification***. In the case of low purity, however, the regular subboundaries are formed in parallel with the impurity subboundaries (corrugations) as shown in Fig. 5[3]. This figure shows a Berg-Barrett photograph taken from the sectional surface of the seed part in the specimen, in which the purity is 99.8% and the rate of growth is 0.1 mm/min.

The following results were also obtained in this study : the incubation period was longer, the misorientation of subboundaries increased less steeply, and the spacing between the subboundaries was wider respectively in the specimens of 200μ in thickness than in the specimens of 100μ.

If it is supposed that the supersaturated vacancies are diffused out from the flat surfaces of crystal foils, at the rate of 0.31 mm/min no dislocation should be expected in the crystals less than 1.4 mm in thickness under the assumption of Frank's theory[4]. In the specimen of 100μ in thickness, however, the striated subboundaries of 0.07 cm in spacing were observed. Furthermore, the dislocation density in the specimen of 100μ in thickness was larger than that in the specimen of 200μ.

An effect of stress induced by an interaction between the specimen surface and the inner wall of glass mould, may be an important clue to explain these experimental results.

The effect of an interaction between the specimen surface and the glass tube upon perfection of crystals, has been examined in bulk tin crystals by one of the authors[5]. He has suppressed the above mentioned effect upon perfection of crystals by coating the inner wall of glass with carbon films. With respect to the rod-like zinc crystals grown by the same method as in the case of tin, a similar avail has also been observed[6]. From these experimental illustrations, it seems that the efficiency of carbon coationg is remarkable.

So, a further examination has now been carried out by coating the inner walls of glass, T_1, P, in Fig. 1 with carbon. For example, Fig. 6 shows two pieces of Berg-Barrett photographs which were reflected from the etched surface of the specimen grown by using a carbon-coated glass mould. In this case, the growth condition was the same as in the specimen shown in Fig. 4 (b). The generation of striated subboundaries is very slighter and the incubation period is very longer (about 55 min) than in Fig. 4 (b). In this carbon coating techni-

*** This longitudinal section was obtained by an acid polishing machine.

que, the thickness of carbon film seems to be an important factor.

Furthermore, the spacing between the striated subboundaries in the specimens of 99.8% in purity seems to be wider than in those of 99.999% as in the case of impurity subboundaries. The detailed results will be published in future.

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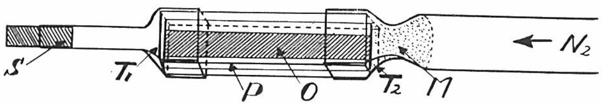


Fig. 1. A special type of glass mould ; S : seed crystal, T_1, T_2 : glass tubes, P : glass plate, M : molten tin, O : opening for producing foil.

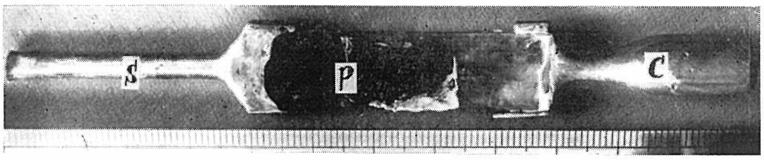


Fig. 2. As the glass tubes, T_1, T_2, were dissolved in HF solution ; S : seed crystal, P : glass plate, C : tin crystal.

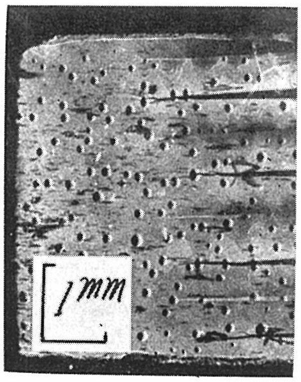
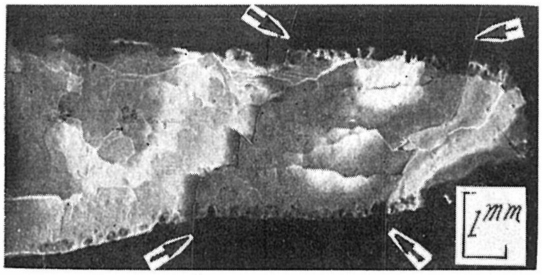
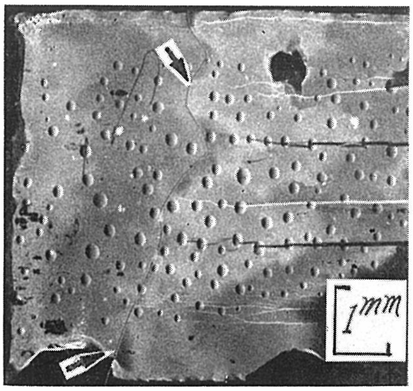


Fig. 3. A Berg-Barrett photograph taken from the surface of foil grown at the rate of 2 mm/min (100μ in thickness, 99.999% in purity).



↑←
7.5 mm from the left end of foil

(a)



(b)

Fig. 4. Two sets of Berg-Barrett photographs taken from the sectional surface of the seed part (in (a)) and from the foil part (in (b)) in the specimen grown at the rate of 0.31 mm/min (200μ in thickness, 99.999% in purity).

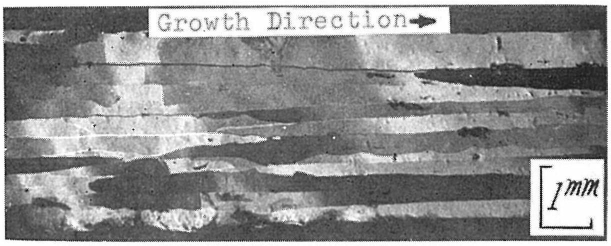


Fig. 5. A Berg-Barrett photograph taken from the sectional surface of the seed part in the specimen grown at the rate of 0.1 mm/min (99.8% in purity).

→↑
17.2 mm from the left end of foil

Fig. 6. Two pieces of Berg-Barrett photographs from the specimen grown by using a carbon-coated mould under the same condition as in Fig. 4 (b).