Grain Growth of Aluminium Sheets during Strain-Annealing Process in Temperature-Gradient Furnace

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Kohsuke TAGASHIRA, Masachika MASUDA* and kazuyuki KIKUCHI

Abstract

Grain growth behaviours in strain-annealing for commercial aluminium have been investigated in relation to the process parameters such as pre-annealing temperature (380－600℃), strain (0 － 7 %) and inserting velocity into the temperature-gradient furnace (1.88－40㎜/h). The results are as follows: (1) The growth modes have been determined on the strain-inserting velocity diagrams. According to these diagrams, the upper critical velocity for single-crystal growth mode has increased with strain. Bi-crystal growth mode has been occurred under the conditions of high strain and low inserting velocity. Whereas many fine matrix grains were remained within growing single crystals as island grains under both low strain and high velocity conditions.

(2) It has been made clear that grain boudary energy should play an important role for driving force for bondary migration as much as strain energy. (3) Orientations of growing grains have been always near (100) [011], being unaffected by strain and inserting velocity. In the temperature-constant furnace, hewever, these have been much dispersed from (100) [011] except unstrained specimens. (4) Both the temperature at the growing front of single cyrstal and the number of island grains have decreased with an increase in strain.

1. Introduction

Strain-annealing process in metallic materials is a series of treatment that stable fine matrix polycrystalline are strained by less than 10% and then annealed at reasonable temperature. This process is quite different from the ordinal annealing process for the heavily cold-rolled metal sheets, because during annealing stage the appearences of nuclei at grain boundaries, within grains or near surface layers are not usually occurred but do only the normal growth, or the abnormal one at times, of matrix grains by strain-induced grain boundary migration. So this is sometimes adopted to obtain large single crystals of metals or alloys[1),2)]. On such an occasion, the methods that specimens deformed in a few per cent are inserted into temperature-gradient furnace at very low uniform velocity is adopted in spite of being kept inside the box-annealing furnace.

The technique in order to obtain single crystals by this process seems to be easy at first sight, but the optimum conditions or the behaviour of single crystal growth have not been necessarily discussed clearly and systematically. For example, it is well known that (110) [001] oriented grains grow abnormally at the sacrefice of other oriented grains when commercial purity Fe-3.25%Si

* Graduate student, Present adress; Musashi Works of Hitachi Ltd, Tokyo.

alloy sheets are strained to a few per cent and then annealed. For the answer to the question why only the selective oriented grains grow on this process, the interpretation by Dunn and Nonken[3] has been regarded as reasonable that grain boundaries can usually move by means of strain energy difference between two adjacent grains (strain-induced grain boundary migration) and that some of the matrix grains having preferred orientation may happen to be low residual strain state and grow by absorbing adjacent fine matrix grains which are higher strain state. Against these explanation, one of the authors[4),5)] has suggested about strain-annealing in Fe–Si alloys having several kinds of impurity levels that the driving energy for grain growth might not always depend upon the difference of residual strain energy. They also suggested that the contribution of grain boundary energy should not be neglected and that the texture formation during strain annealing may be caused by the behaviour of dissolution and diffusion of impurities in large angle grain boundaries.

In this paper, the behaviours of grain growth in strain-annealing have been studied in relation to process parameters for commercial purity aluminium.

2. Experimental Procedures

Materials used here are commercial-purity aluminium sheets of 1.2mm thickness which have been cold rolled by 70% in thickness. The chemical compositions are shown in Table 1.

Al	Cu	Si	Fe	Mn	Mg	Zn	Cr	Ti
Bal.	0.15	0.10	0.53	0.01	tr.	0.01	tr.	0.02

Table 1 Chemical compositions of specimen (wt%)

These were cut to the rectangle sheets as the size of 15mm width, 125mm length and 1.2mm thickness. Longitudinal direction of these specimens were to be as same as cold rolling direction. These were annealed in vaccuum (2.7×10^{-3} Pa) for 2h at the temperature from 380 to 600℃. This heat treatment will be called as "pre-annealing" from now on. Then they were deformed in strain range from 0 to 7 % by Instron type tensile machine at the speed of 0.33mm/s and annealed again (secondary heat treatment) in either the constant-temperature furnace (for 2h at 630℃) or the temperature-gradient furnace (maximum temperature: 630℃). These secondary heat treatments were also carried out in vaccuum (2.7×10^{-3} Pa).

The apparatus including the temperature-gradient furnace is the similar one which was designed in order to make Fe–Si single crystals by one of the authors[6),7)]. The different point for both

apparatus is that the present one is able to be varied temperature-gradient by sliding the water cooling jacket in the vertical combustion tube. At the present experience, the temperature gradient has been settled as 150℃/cm at the point of 550℃ in the furnace without any specimens (maximum temperature: 630℃). Inserting velocity of the specimens into the furnace were selected from 1.88 to 40mm/h.

Textures after pre-annealing were determined by the X-ray diffraction method (Co-K_α, 30kV, 10mA). The optical gonio-microscopy were used in order to measure orientations of grown grains after secondary heat treatment.

3. Experimental results and discussions

3.1 Pre-annealing structure

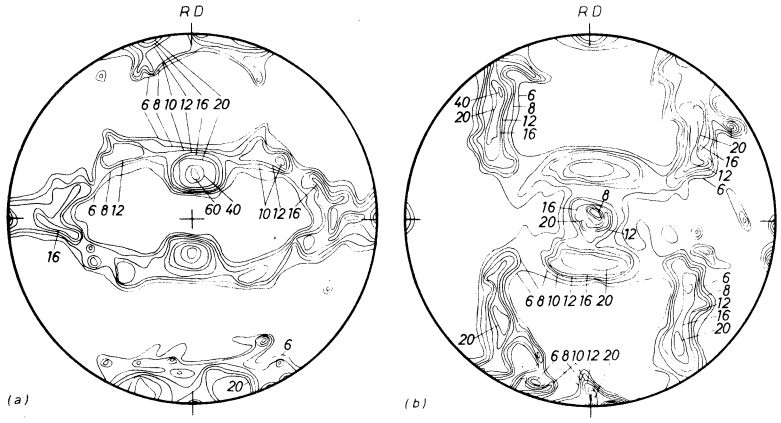

Fig. 1 Pole figures of matrix structure showing contours of equal pole density. Annealed for 2h at 400℃. (a) (100) pole figure, (b) (111) pole figure. RD: rolling and tensile direction.

Mean grain sizes in matrix structures after pre-annealed for 2h at 380 to 570℃ were almost same of 45 to 50 μm. For higher temperature than 590℃, however, abnormally grown grains were observed in matrix, hence mean grain sizes increased abruptly. Matrix texture formed by pre-annealing at 400℃ is shown in Fig. 1. It is really same as primary recrystallization texture of heavily cold rolled aluminium sheets. Some of the preferred orientations of matrix structure are (110) ⟨111⟩, (110) ⟨112⟩, (112) ⟨111⟩ and (456) ⟨121⟩. Although recrystallization textures of the specimens pre-annealed at 500℃ and 550℃ are not shown here, they are similar to one annealed at 400℃. (100) [011] orientation and its near ones were not so strongly detected by the X-ray diffraction method, but these existences were often certificated by the optical microscopic observation as shown in Fig. 2.

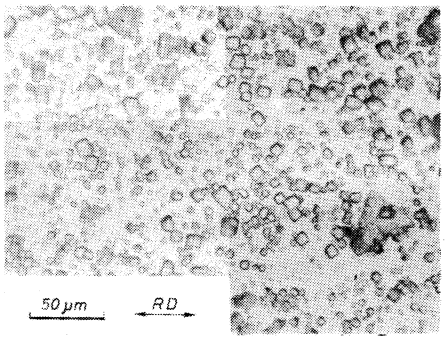

Fig. 2 Matrix structure after pre-annealing for 2h at 550℃. Coarse (100) [011] grain can be distinguished by orientation etch-pits pattern.

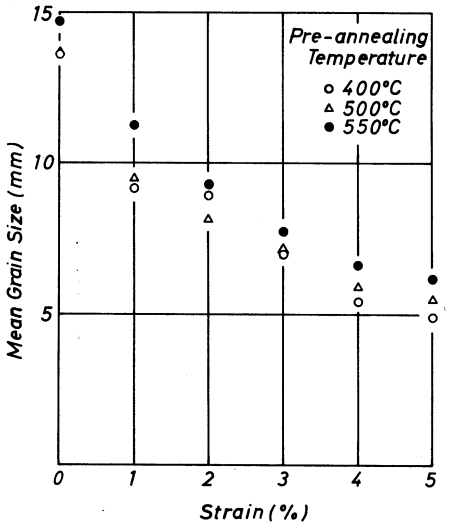

Fig. 3 Relationship between strain and grain size. Secondarily annealed for 2h at 630℃ in the temperature-constant furnace after pre-annealed at various temperatures and strained.

3.2 Grain Growth in Constant-temperature Furnace

Mean grain sizes of the specimens which were pre-annealed for 2h at 400, 500 and 550℃, strained to 0 to 5 % in tension and then carried out secondary heat treatment in the constant temperature furnace are presented in Fig. 3 . According to this figures, grain sizes are always largest on the specimens which were not strained after pre-annealing and these decreased with an increase in strain. It may be thought that the generation frequency of coarsening grains (seed grains) in matrix structure increased with strain. This phenomenon is completely different from grain growth in Fe–3.25%Si sheets[8]. In the case of Fe–Si alloys which were molten in vaccuum, the grains after strain-annealing process in constant-temperature furnace were generally coarsened within the strain range of 0 to 4 %[1]. It can be applied therefore to guess the optimum condition to obtain large single crystals in the temperature-gradient furnace. Similar tendency has been acknowledged on the commercial purity Fe–Si sheets[9] . But Matsumura and Kamada[10] reported that seed grains were unable to grow on strain-annealing process in such a low strain level as a few percent when starting from Fe–3.25%Si decarburized in atmospheric air.

(100) pole figures for specimens pre-annealed at 550℃, strained to 0 , 3 and 5 % in tension and then annealed again at 630℃ were indicated in Figures 4 (a), (b) and (c) respectively. For unstrained specimens the orientations of growing grains are almost near (100) [011], but it become apparently dispersed from (100) [011] by strain ratio. This tendency were basically similar to other pre-annealing temperatures (400 and 500℃). The dispersion from (100) [011] orientation for unstrained specimens has turned narrower with an increase in pre-annealing temperature.

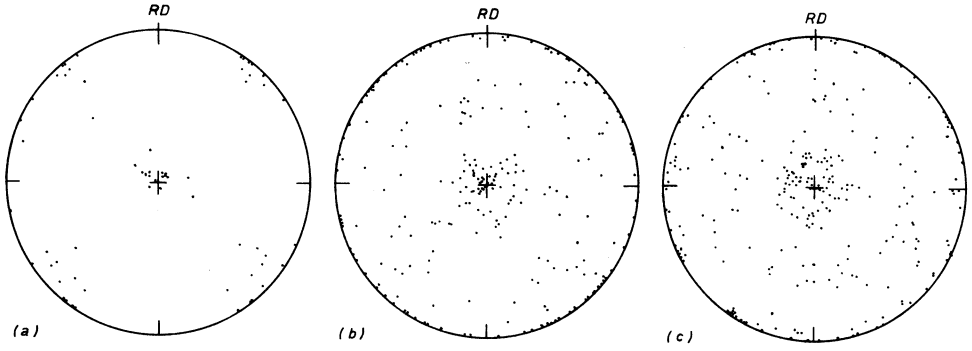

Fig. 4 (100) pole figures. Pre-annealed for 2h at 500℃, strained to (a) 0%, (b) 3% and (c) 5% and then annealed for 2h at 630℃ in the temperature-constant furnace.

3.3 Grain Growth in the Temperature-Gradient Furnace

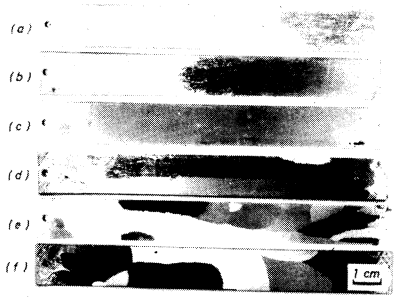

Fig. 5 Growing grains by strain-annealing process in the temperature-gradient furnace at the inserting velocity of 10 mm/h. Pre-annealed for 2h at 400℃ and then strained to (a) 0%, (b) 1%, (c) 2.5%, (d) 3.5%, (e) 4% and (f) 5%. Left-hand of specimens is low temperature side.

Macroscopic structures of specimens annealed in the temperature-gradient furnace are shown in Fig. 5 . These specimens were inserted into the furnace at 10mm/h after being pre-annealed at 400℃ and then deformed to several kinds of strain ratio. It is observed on the specimens strained to 0 , 1 and 2.5% that only one grain, so called, single crystal is growing from the tip of specimen (higher temperature side) to matrix (lower temperature side). On undeformed specimens, however, a lot of fine matrix grains (seen as bright spots in Fig. 5 (a)) are remained as island grains within single crystal (dark area). At the strain ratio of 3.5 and 4 %, a few seed grains are simultaneously growing parallel to the longitudinal direction of specimens (bi-crystals). At the strain of 5 %, progress of seed grains to the longitudinal direction seems to be restrained, therefore the growth mode became in polycrystalline. It may be also pointed out from these photographs that the growing front of single crystal, which is just the boundary between single crystal and its adjacent matrix grains, has changed its position from higher temperature side to lower one with an increase in strain of less than 2.5%. It will be referred at next paragraph.

As mentioned above, lots of matrix grains have been unabsorbed and remained as island grains within growing single crystals in the case of extra-low strain ratio. Its macroscopic structure is shown in Fig. 6 . The proportion of single-crystallized grain growth S (total length of single-

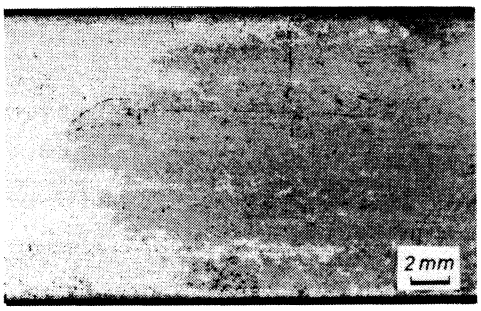

Fig. 6 Fine island grains (light) remained in single crystal (dark). Pre-annealed at 400℃, strained to 1% and then inserted into the temperture-gradient furnace at the velocity of 2.5mm/h.

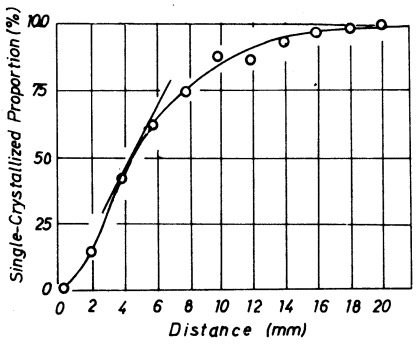

Fig. 7 Relationship between distance (x) and Single-crystallized proportion (S) related to Fig.6.

crystallized part per unit width in transverse direction of specimen) is also shown as function of distance x along longitudinal direction in Fig. 7. Incidentally the relationship between strain and gradient $G = dS/dx$ at the 50% single-crystallized location are shown in Fig. 8 for specimens pre-annealed at 400℃ and inserted into the furnace at 5 mm/h.

Judging from the macroscopic observations, G−values have been always more than 9.0 for single crystals excluding island grains, while the G−values have been less than 6.0 for imperfect single crystals containing much island grains. In the case of G−values between 6.0 and 9.0, appearences of single crystal growth are not always corresponding to the G−value. G−values tend to rise in proportion to strain. It may be supported by the fact that driving force for migration of single crystal front should be increased with strain ratio. Though the inserting velocities into the furnace are not particularly entered in Fig. 8, the higher the inserting velocity the less the G−value for the same strain ratio.

The growth modes after secondary heat treatment are arranged by relating to strain ratio and inserting velocity as shown in Figures 9 (a), (b) and (c) for three kinds of pre-annealing temperature. In these figures, single crystal mode (marked by •) is corresponding to G−value being more than 9.0, while inperfect single crystal mode containing lots of fine island grains (marked by ∘) are corresponding to G−values of 3.5 to 6.0. The area surrounded by two dashed lines and the coordinate axis are conformed with the condition to obtain single crystals without any island grains. Upper critical inserting velocity for the growth mode of complete single crystals have become increased with an increase in strain ratio having no connection with pre-annealing temperature. It should be supposed that driving force for boundary migration in unstrained specimens may be chiefly dependent on only grain boundary energy. On the other hand, as increasing strain ratio the effect of strain energy due to dislocdtion introduced within fine matrix grains by deformation must

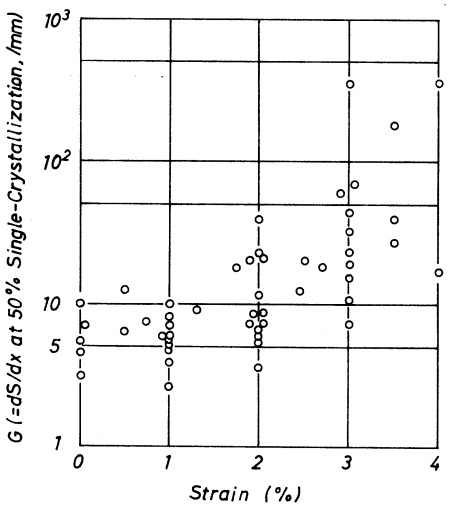

Fig. 8 Relationship between log G ($=dS/dx$ at 50% single-crystallization) and strain.
S:total length of single-crystallized part per unit width in transverse direction of specimen, x:distance.

be added to the effect of grain boundary energy. Under the strain being more than 4.5%, growth mode has changed to polycrystalline mode (marked by □) except in specimens pre-annealed at 550℃ and then inserted at extra-low inserting velocity. It might be caused by an increase in the generation frequency of seed grains in matrix structure.

Grain boundary migration may be generally expressed by the rate equation,

$$V = M \times P, \quad \cdots\cdots(1)$$

where M is the boundary mobility and P is the driving force.

M is expected in some way to depend on the structure of grain boundary, temperature and impurity level but not on the driving force. If the matrix grain structure for strain annealing process is quite stable, M may be constant for a given temperature. If so, V must depend on P for high purity specimens. Disregarding the effects of impurities, M and P are given by the equations,

$$M = A \exp(-Q(\theta)/kT) \quad \cdots\cdots(2)$$

$$P = (\gamma_b/R) + \mu b^2 \Delta N_d + (\Delta\gamma_S/t) \quad \cdots\cdots(3)$$

Here T is the absolute temperature, $Q(\theta)$ is the activation energy for grain boundary migration depending on the misfit angle θ (orientation difference) of the moving boundary, and A is contant being independent of temperature, γ_b is the grain boundary energy, μ is sheat modulus, b is Burgers' vector of dislocation. ΔN_d is dislocation density difference between growing single crystal and each matrix grains and also γs is surface energy difference between them. Third term of right hand side in Eq. (3) may be negligible when specimen thickness t is remarkably large as compared with average matrix grain size R. Orientation differences between single crystal and matrix grain are considered to be not so varied with each matrix grain, hence V may be decided by P rather than M. For the present specimens, R, μ, b and t are about 5×10^{-5} m, 2.5×10^{10} N/m^2, the order of 3×10^{-10} m and 1.2×10^{-3} m, respectively. Average value of grain boundary energy should be approximately 2.5×10^{-1} mJ/m^2.[11]. If dislocation density difference could be estimated as 10^5 and 2×10^6 /mm^2 coresponding to strain ratio of 0 and 5 %[11], then values of V should be $5.2 \times 10^2 \times$ M and $9.5 \times 10^2 \times$ M (m/s), respectively. Comparing these values with the upper

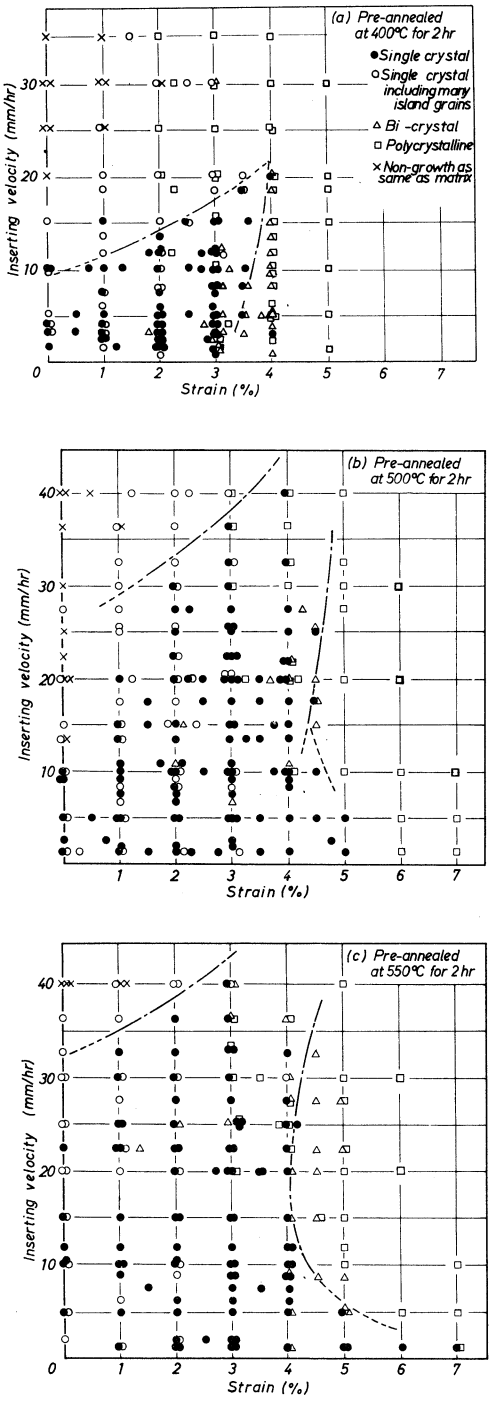

Fig. 9 Growth modes related to strain and inserting-velocity into the temperature-gradient furnace.

dashed line in Fig. 9, M should be calculated as 1.4×10^{-9} m^4/J·s.

Dunn and Walter[12] calculated M values for high-purity aluminium on account of the experimental results about normal grain growth by Beck and Sperry[13]. According to their caliculation, M was 2×10^{-10} m^4/J·s for the strong matrix texture case, while M was 2×10^{-9} m^4/J·s for the weak matrix texture case. These values are similar to the above our values. Though there are still remained some questions against the estimation for dislocation density difference and other, grain boundary energy should play an important role for the driving force of boundary migration as much as strain energy in the present specimens.

The average matrix grain size was almost same about 50 μm which is independent of pre-annealing temperature as mentioned in 3.1. It is curious that the range of complete single crystal mode have become wider as shown in Fig. 9 with pre-annealing temperature in spite of similar matrix grain size. But size of (100) grains which have been rarely observed in pre-annealed structures are at least twice the mean diameter of matrix grains as described in Fig. 2. Orientations of these coarse matrix grain were ascertained to be almost near (100) [011] as a result of accurate measurement of micro-orientation etch-pits. If the dislocation density in fine mat-

rix grains should be supposed to be higher than in coarse ones after low plastic strain ratio, it may be understood that only a few (100) coarse grain of low strain energy level will be able to be grown selectively to single crystal by absorbing other oriented fine matrix grains. Agreement on the relation of grain size to strain ratio between two different kinds of secondary heat treatment could not be found so much.

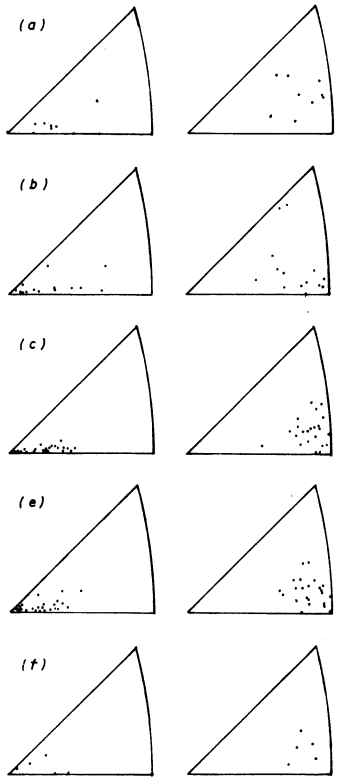

Fig. 10 Inverse pole figures of grown grains under the following pre-annealing temperatures and strains.
(a) 400℃, 2%, (b) 400℃, 3%, (d) 550℃, 3%, (e) 550℃, 5%,
Left; plane normals,
Right; tensile directions

In the case of pre-annealing temperature at 550℃, high quality single crystals have been grown even under the conditions of higher strain ratio of 5 to 7 % and extra-low inserting velocity (1.88mm/h). It should be assumed from these results that dislocation density in matrix grains which were just going to be absorbed by the growing grains in temperature-gradient furnace might become lower by no means of recrystallization but recovery in the consequence of extra-low inserting velocity, then coarse matrix grains which ought to be essentially activated to grow themselves to polycrystalline mode might have been grown to single crystal mode by each containment phenomenon.

Orientations of grown grains by strain-annealing are shown in Fig. 10 for several combinations of strain and inserting velocity. These are almost near (100) [011] having no connection with pre-annealing temperatures and strains. These (100) oriented grains must have been some of the matrix grains and grown preferrencially by absorbing the other oriented matrix fine grains. But it is unexplained yet whether this phenomenon may be similar to the case of (110) [001] grains grown during strain annealing stage of Fe–3.25%Si alloys or not. These are quite different from the results in the temperature-constant furnace that orientations have been much dispersed from (100) [011] with the increase of strain ratio as mentioned above paragraph. It is interesting that crystallographic indices of migrating directions are limited to near [011]. It is quite different from the case of solidification of aluminium in which growing directions parallel to columner structure are ⟨100⟩[14]. On the other hand, by studies on the directional solidification of aluminium alloys, such as Al–CuAl, Al–Al_3Ni etc., growing direction was generally [011][15),16)], and it is of the

same as the direction of preferred migration in the present strain-annealing experience. But whether exact growth mechanism of both cases are similar or not are still unknown.

3.4 Effects of Strain Ratios and Impurities on Temperature at Growing Front of Single Crystal

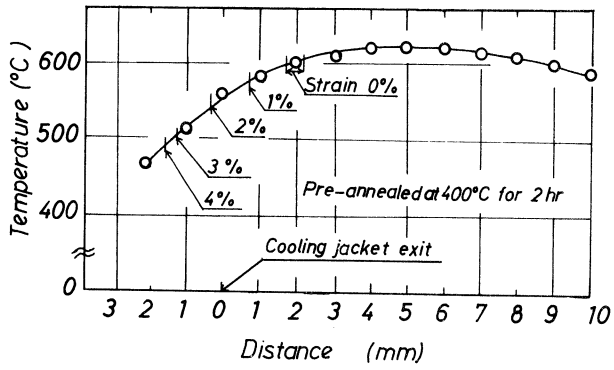

Fig. 11 Temperature distribution curve on the semi-infinite length specimen in the temperature-gradient furnace (in vaccuum). The growing front positions of single crystals are shown by arrows for each strain from 0 to 4%.

As mentioned in the above paragraph (3.3), the higher the strain ratio, the lower the temperature at the location of growing front of single crystal, that is, the boundary between growing single crystal and matrix grains. Besides the morphology of these growing front is not in straight but in zig-zag. Temperature distribution along the longitudinal direction of specimen being regarded as simi-infinite length is indicated in Fig. 11 for the present temperature-gradient furnace (in vaccuum). Judging from both the experimental results and this figure, actual temperature at growing front may be estimated to be 600, 585, 560, 515 and 490℃ for the specimens which were pre-annealed at 550℃ and strained in 0 , 1 , 2 , 3 and 4 % respectively and then inserted into the furnace at the velocity of 10㎜/h. In the specimens deformed in less than 1 % the location of growing front was defined as a 50% single-crystallized location because of being unable to measure accurate location due to the existence of many unabsorbed matrix grains.

The reason why the location of growing single crystal front in the temperature-gradient furnace is switched to lower temperature side by strain might be considered to be due to an increase in driving force by strain energy and interactions between impurity atoms and dislocation. It is well known that impurity atoms existing in compounds such as sulfides, nitrides, carbide, etc., are effective to disturb the recovery of both strain and the grain boundary migration. These impurities are usually effective to recovery and recrystallization only under a certain temperature as clarified by Tagashira et al.[5], that is the same as dissolving temperature of their compounds. In the present aluminium specimens, however, restraint temperature for grain growth decreased with strain.

So the role of impurity atoms does not play as compounds but probably as solid solutions. If lattice diffusion of impurity atoms in Al such as Cu, Si, may be aided by using the dislocations as

high diffusivity paths. These atoms may be able to diffuse easily from matrix grains (lower temperature side) to single crystal (higher temperature side). During this process, impurity atoms may stay temporalily along the boundary between single crystal and its adjacent matrix grains, but next instance they may be diffuse into single crystal as substitutional atoms by means of vacancies thermo-equilibriumly generated inside the single crystal. Thus boundary migration may be promoted by the release from impurity inhibitions. It should be more considered in details.

4. Conclusion

The effects of pre-annealing temperatures, strain ratios and specimen inserting velocities into temperature-gradient furnace on the strain-annealing behaviour has been studied in commercial purity aluminium. The following results have been obtained.

(1) growth modes have been determined on the diagram of strain and specimen inserting velocity. According to these diagrams, upper critical velocity for single crystal mode has increased with an increase in strain. It may be estimated that grain boundary energy should play an important role for the driving force of boundary migration as much as strain energy.

(2) Orientations of grown grains by strain-annealing have been always near (100) [011] being no affected by strain and inserting velocity. Whereas in the results of strain-annealing in the temperature-constant furnace these have been much dispersed from (100) [011] excepting undeformed specimens.

(3) As the pre-annealing temperature was raised, the condition for single crystal growth mode spreaded on the strain–inserting velocity diagram. After all, for pre-annealing at higher temperature single crystals excluding any island grains could be grown even at rapid inserting velocity in the region of higher strain and extra-low inserting velocity.

(4) Mode for poor quality single crystals containing lots of island grains was obtained under the conditions of lower strain and higher inserting velocity, while bi-crystal mode was generally done under higher strain and lower inserting velocity.

(5) Both the temperature at the front of growing single crystal and the mumber of remained fine island grains within single crystal were decreased by strain. It has not been observed in strain-annealing process on Fe– 3.25%Si alloy including certain kinds of impurities.

Acknowledgements

The authors gratefully acknowledge the useful advices of Professor H. Nakae of the Hokkaido University at the time and also Professor K. Ikawa of Tohoku University.

(Received 17th May, 1985)

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