Optimization of Alternating Current Imposing Period on Crystal Alignment of Sn–Pb Alloy Subjected to a Static Magnetic Field during Solidification

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For optimization of alternating current imposing period in the crystal alignment process in which both a static magnetic field and an alternating current are imposed on an alloy during solidification, experimental work has been done using a Sn–10mass%Pb alloy. Index of crystal alignment decreases with increase in the imposing period of the alternating current because the electromagnetic torque induced in the sample disturbs the alignment of solid particles suspended in the liquid phase. Electromagnetic torque is dominant as resistance of the crystal rotation to magnetically preferred direction when the size of the crystal is 21 μm or larger. For the optimization of the crystal alignment in this process, imposing period of the electric current and the magnetic field should be controlled as follows. The electric current and the magnetic field are simultaneously imposed on the alloy only in the initial stage of the solidification. Then the electric current is turned off and only the magnetic field is imposed during the solidification.

KEY WORDS: solidification; crystal alignment; electromagnetic processing of materials; magnetic field; electric current.

1. Introduction

A material with an anisotropic crystallographic structure shows anisotropy in the physical properties such as electric conductivity, magnetic susceptibility, thermal conductivity and so on. That is, alignment of the crystals in a material enhances the anisotropy of these physical properties. Therefore, the crystal alignment is an attractive method for fabricating functional materials. For example, grain-aligned magnetic steel with $\{110\}\langle 001 \rangle$ crystallographic structure has been developed, in which magnetically easy axis, $\langle 001 \rangle$ is aligned to the rolling direction, because this structure drastically reduces core loss comparing with the traditional one.^{1,2)} Crystal alignment is also useful tool to improve a magnetic property of a ferrite magnet because its residual magnetic flux density is in proportion to degree of the crystal alignment.³⁾ Graphite is suitable for a heat extraction material since its averaged thermal conductivity is about three or four times larger than that of aluminum and copper.⁴⁾ Because its thermal conductivity in a,b -axis is about two hundreds times larger than that in c -direction caused by hexagonal crystallographic structure,⁴⁾ its crystal alignment strongly intensify the ability as the heat extraction material.

However, conventional methods for the crystal alignment have some problems. A mechanical method for the alignment of crystals such as a rolling has been industrially

used,⁵⁾ though the crystal alignment in this process is limited to a specific direction. Slip casting in a magnetic field⁶⁾ can be applied for the crystal alignment of ceramics. But this process is not suitable for large mass production because of low productivity. Epitaxial growth⁷⁾ is used as a thin film production process though formation of a bulk material is difficult.

On the other hand, a static magnetic field has become a powerful tool for the crystal alignment under the development of the superconducting technology.^{8,9)} For the crystal alignment using a static magnetic field, environment in which crystals can easily rotate to magnetically stable direction is required. However, it is difficult to introduce this condition during solidification because a columnar solid usually forms from liquid phase.¹⁰⁾ To achieve the crystal alignment during solidification, we have proposed a new method in which both a static magnetic field and an alternating current are imposed on an alloy during solidification, and a primary phase of a Sn–10mass%Pb alloy are aligned to magnetically preferred direction by this process.¹¹⁾ In this study, optimization of alternating current imposing period in this process has been investigated using the Sn–10mass%Pb alloy.

2. Experimental Setup

Experimental apparatus is shown in **Fig. 1**. An Sn–

Fig. 1. Schematic view of experimental apparatus.

Table 1. Experimental condition.

Sample No.	Magnetic field	Current, I	Turning off time after recalescence, t_{off}
I	0T	80A(2kHz)	after solidification
II	7.5T	80A(2kHz)	10s
III	7.5T	80A(2kHz)	40s
IV	7.5T	80A(2kHz)	after solidification

10mass%Pb alloy of 0.3 kg poured into a glass rectangular vessel of 40 mm length and 25 mm width was set in the bore of a super-conducting magnet. To control temperature distribution in the sample, one of short walls of the vessel was heated while the other one was cooled. Furthermore, upper part of the copper electrodes was heated to reduce temperature disturbance caused by heat extraction through the electrodes from the sample. A vertical static magnetic field of 7.5 T was imposed during solidification of the sample. A couple of copper electrodes were inserted in the vicinity of the short wall of the vessel to supply an alternating current to the sample. Amplitude and frequency of the alternating current were 80 A and 2 kHz, respectively. The electrodes were covered by an electrical insulator except 5 mm · 5 mm square tip as shown in Fig. 1.

We solidified four samples under the different experimental conditions. The sample I was solidified without the static magnetic field and with the alternating current. For the samples II, III and IV, both the alternating current and the static magnetic field were imposed on them during a

Fig. 2. Magnetically preferred direction of tin crystal.

specific period of their solidification. Difference in these samples was the imposing duration of the alternating current. The alternating current was turned on for the every sample when the temperature was 250°C, which is 31°C higher than the liquidus temperature of the sample. After the certain time, t_{off} from the maximum undercooling temperature, the alternating current was turned off. It was 10 s for the sample II, 40 s for the sample III and after the end of the solidification for the samples I and IV, respectively. On the other hand, the magnetic field was imposed during the solidification for the samples II, III and IV. The experimental conditions are summarized in Table 1.

Since the composition of the primary phase in this experiment is very close to pure tin, we expect that physical properties of the primary phase is quite similar to that of pure tin. Because tin has a tetragonal crystal structure and magnetic susceptibility in a -axis is $2.476 \times 10^{-6} (-)$ and that in c -axis is $2.212 \times 10^{-6} (-)$, the c -axis is perpendicular to the direction of a magnetic field as shown in Fig. 2 if the magnetically anisotropic energy is larger than thermal fluctuation, $k_B T$ (k_B : Boltzmann constant, T : absolute temperature). The magnetically anisotropic energy is equal to the thermal fluctuation when the solid particle size is $7 \cdot 10^{-8}$ m in this research.

3. Experimental Results

3.1. Macrostructures

After the solidification, the sample was cut just below the electrodes tip as shown in Fig. 3 and the horizontal cut surface was polished by an emery paper of #2000 and polished by a buffing compound with 0.05 μm alumina particles. Then, it was chemically etched by a mixed liquid of distilled water, hydrochloric acid and iron chloride(III) for the observation of macrostructure and X-ray diffraction analysis.

Macrostructure solidified under the different experimental conditions is shown in Fig. 4. The macrostructure of the sample I is not uniform. Grains are refined around the current passing region while they are coarse in the other region. The macrostructures of the samples II, III and IV are essentially refined. The grain size of the sample II and III are slightly larger than that of the sample IV because the

Fig. 3. Observed surface of macrostructure and XRD analysis.

Fig. 4. Macrostructure solidified under different electromagnetic imposing condition.

imposing period of the electromagnetic vibration in these samples is shorter compared with the sample IV. That is, short imposing period of the electromagnetic vibration is not suitable for the structure refinement.

3.2. Temperature History

The temperature histories of the samples measured 20 mm above from the bottom at the center of the long wall are shown in Fig. 5. Undercooling of the sample I is larger than that of the other samples. We might say that the flow induced by the electromagnetic vibration carries solid particles to the neighborhood of the thermocouple since the measured undercooling depends on the electromagnetic vibration. On the other hand, imposition of the electromagnetic vibration induces nucleation because the solidified structure is refined. Therefore, small solid particles are introduced in the sample in the initial stage of the solidification in this process.

Fig. 5. Temperature curves.

Fig. 6. X-ray diffraction pattern.

3.3. Evaluation of Crystal Alignment

The X-ray diffraction analysis was used for evaluation of the crystal alignment, and the result is shown in Fig. 6. The X-ray irradiation area is about 10mm squares. In the case of the sample I (only the alternating current was imposed during the solidification), the first and second main peaks are (101) and (211) planes though the magnetically preferred peaks are planes parallel to the *c*-axis such as (200), (220) planes. That is, crystal alignment is not observed in this sample. In the case of the samples II, III and IV, the magnetically preferred peak of (200) plane is intensified and other peaks such as (101), (211) and (112) planes are suppressed. Furthermore, the (220) peak is also intensified in the case of the sample IV. Therefore, the crystals in these samples are aligned to the magnetically preferred direction.

For the quantitative evaluation of crystal alignment, ratio of the (200) plane peak intensity to the (101) plane peak intensity is calculated and the result is shown in Fig. 7. The calculated ratio for the sample I is smaller than the values for the samples II, III and IV in which the crystal alignment is observed. For the samples II, III and IV, the intensity

Fig. 7. Intensity ratio.

Fig. 8. Ratio of viscous effect to electromagnetic effect.

ratio decreases with increase in the imposing period of the alternating current. This means that the electromagnetic force induced in the sample disturbs the alignment of solid particles suspended in the liquid phase.

4. Discussion

The rotation time for a single crystal under a static magnetic field is calculated using the following equation.¹²⁾

$$\tan \theta = \tan \theta_0 \exp\left(-\frac{t}{\tau}\right) \quad (1)$$

$$\tau = \frac{30\eta + r^2\sigma B^2}{5\Delta\chi B^2} \quad (2)$$

where θ_0 is an initial angle between the magnetically easy axis of the crystal and the magnetic field direction, $\Delta\chi$ is the magnetic susceptibility difference, η is viscosity, r is the radius of the single crystal, σ is electrical conductivity and B is the intensity of the magnetic flux density, τ is alignment time, respectively.

The second term in the numerator of the right hand side of the Eq. (2) is caused by the electromagnetic effect while the first term is due to the viscous effect. We introduce the ratio of the viscous effect to the electromagnetic effect, R as following.

$$R = \frac{30\eta}{r^2\sigma B^2}, \quad \frac{1}{R} = \frac{r^2\sigma B^2}{30\eta} = \frac{1}{30} B^2 r^2 \frac{\sigma}{\eta} \quad (3)$$

This ratio is calculated under the condition of the 7.5 T magnetic field and the result is shown in Fig. 8. The radius is 21 μm when the ratio is unity. Therefore, the viscous effect is dominant as resistance of the rotation motion for a

Fig. 9. Rotating angle of a crystal with different radius.

crystal whose radius is less than 21 μm while the electromagnetic effect is dominant for a crystal whose radius is larger than 21 μm . On the other hand, the Hartmann number defined as the Eq. (4) indicates the square root of the ratio of the Lorentz force to the viscous force.

$$Ha = BL\sqrt{\frac{\sigma}{\eta}}, \quad Ha^2 = B^2 L^2 \frac{\sigma}{\eta} \quad (4)$$

where L is the characteristic length.

From the similarity between the Eqs. (3) and (4), the characteristic length in the Hartmann number is related with the radius as follows.

$$L = \frac{r}{\sqrt{30}} \approx \frac{r}{5.5} \quad (5)$$

Therefore, the characteristic length in the Hartmann number and that in the ratio, R is the same order though the former is about 5.5 times smaller than the latter.

The rotation time for a tin crystal under a 7.5 T static magnetic field is calculated using the Eq. (2) and the result is shown in Fig. 9. The initial angle in this calculation is set to 89 degrees. As the crystal radius increases, the rotation time increases. Crystal rotation almost finishes within 0.01 s when the crystal radius is a few μm . This means that the solid particles align to the magnetically preferred direction in the initial stage of the solidification. However, the alignment formed in the initial stage might be disturbed by the electromagnetic vibration. Recovery time of the alignment disturbed by the electromagnetic vibration increases with increase in the solid particle size. Therefore, degree of the crystal alignment decreases with increase of the imposition time of the electromagnetic vibration.

5. Conclusion

For optimization of alternating current imposing period in the crystal alignment process in which both a static magnetic field and an alternating current are imposed on an alloy during solidification, experimental work has been done using the Sn–10mass%Pb alloy.

The ratio of the (200) plane peak intensity to the (101) plane peak intensity in XRD analysis which is index of crystal alignment to magnetically preferred direction decreases with increase in the imposing period of the alternating current because the electromagnetic force induced in the sample might disturb the alignment of solid particles. The electromagnetic torque is dominant as resistance of the rotation when the size of the crystal is 21 μm or larger in this experiment. For crystal alignment in this process, im-

posing period of the electric current and the magnetic field should be controlled as follows. The electric current and the magnetic field are simultaneously imposed on the alloy only in the initial stage of the solidification. Then the electric current is turned off and only the magnetic field is imposed during the solidification.

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Nomenclature

- B : Magnetic flux density (T)
 L : Characteristic length (m)
 R : Ratio of the viscous effect to the electromagnetic effect
 r : Radius (m)
 t : Time (s)
 t_{off} : Turning off time of current after the maximum cooling temperature (s)
 $\Delta\chi$: Difference of magnetic susceptibility in each crystal axis (—)
 μ_0 : Magnetic permeability in vacuum (H/m)
 χ : Magnetic susceptibility parallel to the imposed mag-

netic field (—)

- θ : Angle between magnetically easy axis and magnetic field direction (rad)
 θ_0 : Initial angle between magnetically easy axis and magnetic field direction (rad)
 η : Viscosity (Pa · s)
 σ : Electrical conductivity (S/m)
 τ : Alignment time defined in Eq. (2) (s)

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