Investigation of the Melt Flow on Solidified Structure by a Levitation Technique Using Alternative and Static Magnetic Fields

Hideyuki YASUDA, Itsuo OHNAKA¹, Rintaro ISHII², Satoru FUJITA² and Yuki TAMURA²

Department of Adaptive Machine Systems, Osaka University, Osaka 565-0871 Japan. E-mail: yasuda@ams.eng.osaka-u.ac.jp
1) Formerly at Department of Adaptive Machine Systems, Osaka University, now at Department of Entrepreneur Engineering, Osaka Sangyo University, Osaka 574-8530 Japan. 2) Graduate student, Department of Adaptive Machine Systems, Graduate school of Engineering, Osaka University, Osaka 565-0871 Japan.

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A levitation method using the simultaneous imposition of an alternating field and a static magnetic field was applied to study effect of the melt flow on the microstructure of Cu, Cu–1at%Ag alloys, a middle carbon steel alloys and Fe–10at%Ni alloys solidified from the undercooled melt. Convection in the levitated melts was remarkably reduced when the imposed static magnetic field exceeded 1 T. Reduction of the melt flow did not affect the nucleation temperature, but caused the morphological transition from the equiaxed grains to the columnar grains for the Cu–Ag, the steel and the Fe–Ni alloys. The experimental results indicated that the melt flow in the mushy region caused the dendrite fragmentation and consequently resulted in formation of the equiaxed grains at the low undercooling region.

KEY WORDS: containerless process; nucleation; columnar structure; equiaxed structure.

1. Introduction

It has been well known that containerless processes were useful to investigate solidification from the undercooled melts¹ and to measure thermophysical properties.² Levitation methods have been developed by using the electromagnetic force due to the alternating magnetic field,²–⁴ by using the aero-acoustic waves,⁵ and by using the electrostatic force.⁶ Recently, the magnetization force that originates in interaction between the magnetization and the external magnetic field was also used to levitate diamagnetic materials stably.⁷–⁹ Since the magnetization is a body force, the pseudo-microgravity condition can be achieved and the diamagnetic materials are passively levitated.

The electromagnetic force due to the alternating magnetic field is generally expressed by the two terms. One is a non-rotational term, which causes a lift force against the force of gravity. The other is a rotational term, which results in the electromagnetic stirring. Even when the first term becomes dominant at high frequencies, the second term still exists. Thus, the levitated melt usually oscillates, and rather strong convection occurs in the melt. In contrast, the levitation method using the static magnetic field can avoid the violent vibration and the strong convection, because the magnetization force is a body force. However, the stable levitation can be achieved only for diamagnetic materials.⁷–⁹ From a technical aspect, it is required to develop a new levitation technique in which the various melts are levitated without convection.

In general, the melt flow may affect the nucleation. Furthermore, the solidified structure can be significantly affected by the melt flow. If one can control intensity of the oscillation and the convection in the levitated melts, the experimental technique is a powerful tool to understand how the melt convection affects the nucleation and the solidified structures. Thus, from a fundamental aspect, it is of interest to develop an electromagnetic levitation method that enables to levitate the melt without convection.

It has been reported that titanium was statically melted in the cold crucible when a high static magnetic field was imposed.¹⁰,¹¹ We have developed a levitation technique using the alternating and the static magnetic fields.¹²–¹⁴ In this technique, intensity of the melt flow is reduced by the static magnetic field. This paper reports effect of the melt flow on the nucleation and the solidified structure of pure copper, Cu–Ag dilute alloy, Fe–Ni alloy and a conventional middle carbon steel alloy by using the levitation method using the alternating and the static magnetic fields.

2. Experimental Procedures

Figure 1(a) shows a schematic illustration of the experimental setup to levitate a melt by the simultaneous imposition of the alternating and the static magnetic fields. An RF generator with a frequency of 200 kHz and a power of 20 kW was connected to the levitation coil, which causes a lift force. Detail of the setup was mentioned in ref.¹⁴ Figure 1(b) shows configuration of the levitated sample, the RF coil and the bore of the superconducting magnet. A copper shield was inserted into the bore to shield the superconducting coil from the alternating magnetic field. A static magnetic field was imposed by a cryogen-free superconducting
magnet (Maximum field: 10 T, bore size: 100 mm). Melts were levitated in Ar atmosphere at the maximum position of the static magnetic field. After keeping the melt for 30–60 s, the specimens were cooled by He or He–5%H₂ gas flow.

Temperature of the levitated melt was measured by a two-color pyrometer located above the melt as shown in Fig. 1(a). A CCD camera (250 frames/s) was also placed above the levitated melt to record motion of the levitated melt and inclusions.

The samples used in the present study were pure Cu (99.999%), Cu–1at%Ag, Fe–10at%Ni and a conventional middle carbon steel (S45C). Typical weight of the sample was 1 g and diameters of the levitated melt were approximately 6 mm. Inclusions on the levitated melt were traced to examine the melt flow. The nucleation temperature as a function of the static magnetic was measured for the pure copper and the Cu–Ag alloy, since the He–H₂ gas sufficiently avoided formation of the oxide film that may act as a nucleation site. Nucleation temperatures were measured by using the specimen in which the inclusions were not observed during the levitation experiment. Solidified structures were observed with an optical microscope and a conventional SEM. Grain structure was observed by electron backscatter diffraction (EBSD).

3. Results and Discussion

3.1. Melt Flow in the Levitated Melts

Figure 2 shows traces of the inclusions floating on the levitated copper melts. The inclusions randomly moved in the levitated melts under a static magnetic field of 0 T. The random motion indicated that the melt was stirred by the electromagnetic force. Magnitude of the velocity fluctuation of the inclusions was approximately of the order of $10^{-2}$–$10^{-1}$ m/s at a magnetic field of 0 T. At a static magnetic field up to 0.1 T, effect of the static magnetic filed was not detected in the inclusion motion. At a static magnetic field of 0.5 T, rotation of the melt was clearly recognized from the traces, although the random movement of the inclusions was still observed. At a static magnetic field exceeding 1 T, the traces indicated the inclusions exhibited the circular motion, since the inclusions on the droplet have the same angular velocity each other.

Motion of the levitated melts is classified into the several categories such as oscillation, convection, rotation and movement of the center of the gravity. The oscillation means the surface vibration due to the surface tension of the melt, while the convection means the circulating melt flow in the levitated melts. There are two modes in the rotation. One is the rotation of which axis is perpendicular to the static magnetic field, and the other is the rotation of which axis is parallel to the static magnetic field. Although each
the oscillation and the convection originate in nature of the fluids, the rotations and the movement are not associated with the melt flow. In the rotations and the movement, relative position in the levitated melt is the same. Thus, effect of the rotations and the movement on the solidification differs from that of the melt flow. The rotation of the melt that was observed at magnetic fields exceeding 1 T was the same as the rotation of the hard sphere, and the melt flow in the melt was not detected by the present observation.

The flow velocity of the melt was not estimated from the movement of the inclusions, since the inclusions often slipped on the melt surface due to the centrifugal force and the gravity force. In addition, movement of the center of the gravity of the levitated melt caused a serious error in the estimation of the velocity. Thus, degree of effect of the static magnetic field on the convection velocity was qualitatively explained in this paper.

3.2. Nucleation of Cu and Cu–1at%Ag Alloys

Figure 3(a) shows undercooling to nucleate in the levitated melt of the pure copper. Cooling rates in the present study ranged from 10 to 100 K/s. Maximum undercooling was approximately 250 K. The maximum undercooling did not clearly depend on intensity of the static magnetic field. As shown in Fig. 3(b), addition of Ag into Cu reduced the nucleation undercooling and the maximum undercooling was 100 K at most. Effect of the static magnetic field on the nucleation undercooling was not clearly observed for the Cu–1at%Ag alloys.

The magnetic energy was negligible for pure copper and the Cu–Ag alloy, since the magnetic susceptibilities were extremely small. The present results indicate that the oscillation and the convection do not affect the nucleation in the electromagnetically levitated melt. Pressure change due to the convection and the oscillation would be sufficiently small, comparing to the activation energy for the nucleation. Therefore, effect of the static magnetic field on the solidified structure is attributed to effect of the melt flow during the solidification.

3.3. Solidified Structure of Cu and Cu–1at%Ag Alloys

Figure 4 shows the levitated Cu melts observed by the CCD camera. The sequence started immediately after the recalescence. Growth of the dendrite arms was observed due to difference of the emissivity between the liquid and the solid phases. In the case of the higher undercooling, i.e. 70 K (Fig. 4(a)), the dendrites rapidly grew over the specimen. The alignment of the dendrite arms suggested that the observed dendrites had the same crystallographic orientation and formed into a grain after the solidification. In the case of the lower undercooling, i.e. 0 K (Fig.4(b)), the dendrite arms were clearly observed. A single dendrite grew and covered whole of the specimen. The dendrite evolution did not depend on intensity of the static magnetic field.
Solidified structures of the pure copper are shown in Fig. 5. Only a few grains were observed for each specimen, and the static magnetic field did not affect the solidified structure. The solidified structure was consistent with the observation of the dendrite evolution. For the pure copper, the melt flow was not essential factor to evolve the solidified structure.

Figure 6 shows the solidified structures of the Cu–Ag alloys solidified under magnetic fields of 0 T and 10 T. In the specimen solidified under a static magnetic field of 0 T, fine equiaxed grains are observed in whole of the sample. In contrast, dendrite arms were clearly observed and much porosity was formed at the center of the solidified samples solidified under a static magnetic field of 10 T. The melt flow in the mushy region, which is induced by the solidification shrinkage, was suppressed by the static magnetic field and consequently porosities are formed in the center region.

Figure 7 shows the grain structures of the Cu–1at%Ag alloys. The fine equiaxed grains were clearly observed in the Cu–1at%Ag alloy solidified under a magnetic field of 0 T. On the other hand, coarse columnar grains were observed in the Cu–1at%Ag alloy solidified under a magnetic field of 10 T. Imposition of the static magnetic field mainly contributed to reduction of the melt flow, but it did not change the cooling rate. Thus, the experimental result implied that the reduction of the convection in the levitated melt contributed to formation of the columnar grains.

3.4. Solidified Structure of the Middle Carbon Steel and the Fe–Ni Alloys

The solidified structure of the middle carbon steel was shown in Fig. 8. Maximum undercooling was 70 K in the present experimental condition. The undercooling did not depend on intensity of the static magnetic field. When the static magnetic field was not imposed, the equiaxed grains were obtained except the outer region of the solidified specimen. The dendrite grew into the center of the specimen when a static magnetic field of 10 T was imposed. The dendrite arm spacing increased from the outer side to the center, since the growth rate decreased as the solidification proceeded. However, the fragmentation did not occur. As a result, the coarse columnar grains were formed under a static magnetic field of 1 T. Reduction of the convection changed the morphology from the equiaxed grains to the columnar grains for the middle carbon steel as well as the Cu–Ag alloys.

Figure 9 shows the solidified structure of the Fe–10at%Ni alloys. In this alloys system, the maximum undercooling was approximately 250 K. At the lower undercooling region (<100 K), the equiaxed grains were observed in the specimen solidified under a static magnetic field of 0 T. The dendrite arms were observed in the specimen solidified under a static magnetic field of 1 T, indicating formation of the columnar grains. The same transition was observed at the higher undercooling (>100 K), although it was rather difficult to detect the dendrite arms due to the solute trapping during the rapid solidification from the highly undercooled melt.

3.5. Morphological Transition by the Static Magnetic Field

Table 1 summarized the morphological transition induced by the imposition of the static magnetic field. The results showed that the transition from the equiaxed grains to the columnar grains by reducing the melt flow is a universal phenomenon. There are different mechanisms to explain the grain refinement due to the equiaxed grain formation. For example, the copious nucleation in which the nucleation occurs ahead of the solidifying front and the fragmentation of the primary dendrite arms which supplies seeds of the equiaxed grains have been proposed. It was pointed out that a single nucleation event was sufficient to complete
the solidification for the metals and the metallic alloys that exhibited a high solidification rate. Thus, the possible mechanism is the fragmentation of the dendrite arms in the present case.

In the fragmentation, the primary dendrite arms are fragmented due to the instability of a cylindrical shape. The surface tension drives the shape change from a cylindrical...
shape to the spherical shape to minimize the interfacial energy in the system. In the model, the equiaxed grains are expected to be obtained when the dendrite break-up time is shorter than the solidification time. Since the shape change of the dendrite arms is controlled by solute diffusion around the arms, the solute transfer can significantly affect the fragmentation.

In the Ni–Cu system, a morphological transition from the equiaxed grains to the columnar grains occurred at a lower critical undercooling, and the other transition from the columnar grains to the equiaxed grains occurred at the higher critical undercooling.\(^{17,18}\) They evaluated the fragmentation of the dendrites during the period following the recalescence on the basis of the model.\(^{17,18}\) The estimated morphology was qualitatively agreed with the experimental results. However, they also pointed out that the lower critical undercooling was not observed in the present experimental conditions. Here, we discuss the transition of the solidified structure by the static magnetic field at the lower critical undercooling region.

The equiaxed grains was always obtained for the Cu–Ag alloys, the middle carbon steel alloys and the Fe–Ni alloys when the melt was stirred by the electromagnetic force without a static magnetic field (conventional levitation). The formation of the equiaxed grains at the low undercooling region is consistent with the previous result of the Ni–Cu alloys.\(^{11}\) In contrast, the columnar grains were obtained when the static magnetic field (1 T or 10 T) was imposed. This transition induced by the magnetic field indicated that the melt flow significantly promoted the solute transfer in the mushy region during the period after the recalescence and consequently caused the fragmentation of the dendrite arms. At the lower undercooling region, the melt flow has to be taken into account to understand the formation of the equiaxed grains.

The solidification in the lower undercooling region is not far from that in the conventional casting processes. The melt flow in the mushy region dominantly contributed to the fragmentation of the dendrite arms and is required to form the equiaxed grains at the lower undercooling region. Since the morphological transition at the higher undercooling region was observed only for the Fe–Ni alloys, further studies are required to understand the equiaxed grain formation at the higher undercooling region.

4. Conclusions

The new levitation method by using the simultaneous imposition of the alternating and the static magnetic fields was used to investigate effect of the melt flow on nucleation and solidified structure of the pure copper, Cu–1at%Ag alloys, the conventional middle carbon steel alloys, Fe–10at%Ni alloys.

The nucleation temperature did not depend on intensity of the static magnetic field for pure copper and the Cu–Ag alloys. The turbulent flow and the oscillation induced by the electromagnetic force did not affect the nucleation event. Imposition of the static magnetic field resulted in the morphological transition from the equiaxed grains to the columnar grains at the lower undercooling region (typically less than 100 K) except the pure copper. Since the melt flow was required to cause the fragmentation of the dendrite arms, the melt flow was a dominant factor to form the equiaxed grains at the lower undercooling region.

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