The Effect of Jetting Temperature on the Fabrication of Rapidly Solidified Fe-Si-B Systems Using Single-Roller Melt Spinning*1

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Fe-Si-B systems amorphous alloys were rapidly solidified from the melt by the single roller method. In this process the alloys were melted in a crucible and the molten alloy is jetted through a small orifice by gas pressure or gravity. The effects of the jetting temperature of the melting alloy on the ribbon thickness of rapidly solidified Fe79Si10B15 and Fe79.5Si8.5B12 alloys were examined. The rapidly solidified Fe79Si10B15 and Fe79.5Si8.5B12 alloy ribbons’ mean thickness decreased continuously with the increased jetting temperature. However, the temperature dependence of the ribbon thickness in the Fe79Si10B15 alloy was greater than that of the Fe79.5Si8.5B12 alloy.

The results demonstrate that the viscosity of liquid Fe-Si-B systems alloys influences the ribbon thickness. Moreover, when the contact conditions between the rotating roll comes and puddle are good, the ribbon thickness decreases. [doi:10.2320/matertrans.M2010321]

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1. Introduction

Rapidly solidified Fe-Si-B ribbons have outstanding soft magnetic properties, such as high saturation flux density, which have promoted their practical application as iron cores for transformers today. Many studies aimed at improving their thermal stability and magnetic properties have been reported.1–4) Further improvement in performance of rapidly solidified Fe-Si-B ribbons is expected in the future to meet increased demand for transformers and application to magnetic core materials for various motors for large transport machinery such as automobiles, as a countermeasure against global warming.5)

The planar flow casting (PFC) method,6) among single-roll melt spinning methods, is commonly used for fabrication of uniform and wide rapidly solidified ribbons. The PFC method is the most effective procedure for industrial production of rapidly solidified Fe-Si-B ribbons because the method’s use of large nozzles can accommodate a wide variety of ribbon widths. Fabrication of ribbons with uniform shape using the PFC method requires control of the gap distance between a nozzle orifice and a roll (nozzle gap), the roll speed, molten metal temperature, and other parameters. Control of ribbon thickness is particularly important for fabrication of a precise ribbon because the ribbon thickness strongly affects the ribbon cooling rate. Hillman and Hilzenger7) studied the preparation of rapidly solidified ribbon using the single-roll process with a single-hole nozzle. They reported increased ribbon thickness accompanied with a molten metal puddle extended along the roll rotation direction as a factor affecting the ribbon thickness. Sato et al.8) reported that the molten metal puddle length along the roll rotation direction must be uniform over the ribbon width to obtain a wide ribbon of good quality and uniform thickness. The amount and shape of molten metal supplied from a nozzle orifice, and the interaction between molten metal and a rotating cooling roll at contact, are considered to affect ribbon thickness remarkably in any case. However, sufficiently detailed studies have yet to be conducted; particularly, the effect on ribbon thickness of jetting temperature, which is significantly related to molten metal and its undercooled state, has seldom been reported.9)

This study analyzed the processing conditions of rapidly solidified Fe-Si-B ribbons by single-roll melt spinning using a melt-quenching apparatus: NAV-A3. The ribbon thickness at each jetting temperature was investigated carefully, with roll speeds mainly of 26 m/s, 31 m/s, and 36 m/s; the jetting temperature of molten metal varied within 1573–1773 K. Moreover, micro-Vickers hardness measurements and transmission electron microscope (TEM) observations were conducted of the rapidly solidified ribbons for mechanical property analyses and microstructural observation.

2. Sample and Experimental Procedure

2.1 Preparation of mother alloys

High-purity Fe79Si10B15 (hypereutectic alloy) and Fe79.5Si8.5B12 (eutectic alloy) were used for this study. A micro-vacuum arc melter (NEV-AD03; Nissin-Giken Corp.) was used for mother alloy preparation. High-purity raw materials (99.99% Fe, high purity ferroboron, and 99.999% Si) were fused using the arc melting method in Ar atmosphere. Raw materials were melted completely and repeatedly several times in a water-cooled hearth. Then the Fe79Si10B15 and Fe79.5Si8.5B12 mother alloys were prepared into button-shaped pellets (about 20 mm diameter, about 10 g).

2.2 Fabrication of rapidly solidified Fe-Si-B ribbon

Rapidly solidified Fe-Si-B alloy ribbons were produced using planar flow casting (PFC) as described above using a melt quenching apparatus (NAV-A3; Nissin-Giken Corp.) A mother alloy button of about 20 mm diameter was cut into pieces and put into a rectangular quartz nozzle of about

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10 mm diameter (hereinafter designated as a nozzle). The inside of the melting chamber of the melt quenching apparatus was evacuated to a high vacuum condition of \(2 \times 10^{-3}\) Pa, and converted to Ar atmosphere. Then the mother alloy in the nozzle was melted with RF melting. Once a predetermined temperature was reached, molten metal was jetted from the nozzle orifice promptly onto the copper roll surface rotating at high speed. Then the rapidly solidified ribbon was fabricated continuously. Spacing between the nozzle orifice and the roll surface (nozzle gap) was set to 0.3 mm in this experiment. The copper roll was rotated at a speed of 23–39 m/s; the mother alloys were molten in the nozzle with RF melting at a temperature of 1473–1773 K. The surface temperature of molten metals on the roll was measured using a radiation thermometer (IR-CAQ; Chino Corp.) installed at the upper part of the apparatus. The surface temperature of molten metals on the roll was measured as around 285 K (room temperature: about 13°C) using a non-contact laser thermometer ( emissivity: about 0.95; Raytek Corp.).

2.3 Thickness measurement of rapidly solidified Fe-Si-B alloy ribbons

Thickness of the rapidly solidified Fe-Si-B alloy ribbons was measured at about 100 points on a ribbon with about 10 m total length using a micrometer (Mitutoyo Corp.). Then the average thickness was computed.

2.4 Hardness measurement of rapidly solidified Fe-Si-B alloy ribbons

Hardness of the rapidly solidified Fe-Si-B alloy ribbons was measured at room temperature at a load of 1.96 N using a micro Vickers hardness tester (HM-200; Mitutoyo Corp.). Measurements were conducted at 15 points each. The maximum and minimum data were removed in each measurement. Then the average and standard deviation of the remaining 13 points were computed.

2.5 TEM observation inside rapidly solidified Fe-Si-B alloy ribbons

Microstructure observation inside rapidly solidified Fe-Si-B alloy ribbons was performed using a TEM (JEM-2000EX; JEOL) at an acceleration voltage of 200 kV. Ribbon samples for TEM observation were prepared as follows: small pieces were cut out from a rapidly solidified ribbon of about 10 m length; then samples were prepared by electropolishing (polishing conditions: polishing solution (450 mL of acetic acid (CH\(_3\)COOH) and 50 mL of perchloric acid (HClO\(_4\)), TenuPol 3; Struers A/S) at a polishing liquid temperature of 283 K.

3. Results and Discussion

3.1 Relation between roll speed and ribbon thickness in fabrication of rapidly solidified Fe\(_{75}\)Si\(_{10}\)B\(_{15}\) ribbons (hypereutectic alloy)

Figure 1 portrays the previously reported relation between roll speed and average thickness of rapidly solidified Fe\(_{75}\)Si\(_{10}\)B\(_{15}\) ribbons, with ejection temperatures of 1673 K, 1723 K, and 1773 K\(^{(10)}\) respectively. The ribbon thickness decreases gradually as the roll speed increases at every jetting temperature. The minimum ribbon thicknesses at respective ejection temperatures are 37 \(\mu\)m, 31 \(\mu\)m, and 23 \(\mu\)m at 1673 K, 1723 K, and 1773 K. Previously, the relation between roll speed and ribbon thickness was considered as follows for fabrication of rapidly solidified aluminum alloy ribbons: as the contact condition between a roll and molten metal improves, the heat transfer rate to the roll from molten metal or a ribbon also improves; also, the cooling rate rises and the ribbon thickness decreases as the roll speed increases.\(^{(11)}\) It is therefore inferred that the ribbon thickness decreases concomitantly with increasing roll speed in the alloy used in this study.

In addition, as the ejection temperature increases, the curve in Fig. 1 decreases: the ribbon thickness decreases in this alloy. Therefore, this point is investigated, and the result is discussed below.

3.2 Relationship between jetting temperature and ribbon thickness in fabrication of rapidly solidified Fe\(_{75}\)Si\(_{10}\)B\(_{15}\) alloy ribbon (hypereutectic alloy)

Figure 2 presents the obtained relation between jetting temperature and ribbon thickness of rapidly solidified Fe\(_{75}\)Si\(_{10}\)B\(_{15}\) ribbons (hypereutectic alloy) with roll speeds of 26 m/s, 31 m/s, and 36 m/s. The ribbon thickness is almost constant at jetting temperatures of 1623–1648 K for every roll speed. Then, with increased jetting temperature, the ribbon thickness decreases gradually. After the minimum, it increases again for every roll speed. The minimum thicknesses are, respectively, 33.8 \(\mu\)m, 30.8 \(\mu\)m, and 28.3 \(\mu\)m for roll speeds of 26 m/s, 31 m/s, and 36 m/s respectively. Consequently, in fabrication of rapidly solidified Fe\(_{75}\)Si\(_{10}\)B\(_{15}\) ribbons (hypereutectic alloy), with increased jetting temperature, the ribbon thickness decreases and the minimum is reached. Results also show that the minimal value falls as the roll speed increases. However, the minimum ribbon thickness that is given at different jetting temperatures for each roll speed differs. Therefore, jetting temperature dependence of the ribbon thickness is not evident.

Yamazaki \textit{et al.}\(^{(12)}\) studied the composition dependence of the viscosity of molten Fe-Si-B alloys. They reported that the...
viscosity and its temperature dependence of each alloy composition vary considerably with slight changes of the alloy composition for melting temperatures of 1500–1700 K. They particularly examined the viscosity of hypereutectic and eutectic compositions and reported that the viscosity of Fe$_75$Si$_{10}$B$_{15}$ alloy of hypereutectic composition is higher than that of any other alloy composition, and that the magnitude of the rate of viscosity decrease caused by the increase in melting temperature is also the greatest. This is true probably because viscosity increases according to a suspension model, in which atom groups of Fe$_2$B type are formed inside the molten metal of hypereutectic composition. They behave similarly to suspended particles. The viscosity of the undercooled alloy liquid was estimated using Fulcher’s equation for alloys of hypereutectic and eutectic compositions. Figure 3 shows the estimated viscosity at the undercooled state of Fe$_75$Si$_{10}$B$_{15}$ (hypereutectic alloy) and Fe$_{79.5}$Si$_{8.5}$B$_{12}$ (eutectic alloy). Both plots were produced for temperatures used in this experiment using Fulcher’s equation for Fe$_75$Si$_{10}$B$_{15}$ (hypereutectic alloy) and Fe$_{79.5}$Si$_{8.5}$B$_{12}$ (eutectic alloy) obtained by Yamazaki et al. It was verified that viscosity falls as the melting temperature rises for both alloys. The plot of Fe$_{79.5}$Si$_{8.5}$B$_{12}$ lies under that of Fe$_75$Si$_{10}$B$_{15}$ overall; the viscosity change by melting temperature of the former is also smaller. Consequently, viscosity and its temperature dependence vary along with the alloy composition, even in an undercooled state in this alloy system. The viscosity is low; its variation by temperature is also low, especially in eutectic composition. The relationship between jetting temperature and ribbon thickness was examined to elucidate the effect of viscosity on ribbon thickness in greater detail, on a eutectic alloy Fe$_{79.5}$Si$_{8.5}$B$_{12}$, for roll speeds of 26 m/s, 31 m/s, and 36 m/s, at various ejection temperatures of 1573–1773 K. Figure 4 shows the relation between jetting temperature and average ribbon thickness of rapidly solidified Fe$_{79.5}$Si$_{8.5}$B$_{12}$ ribbons for roll speeds of 26 m/s, 31 m/s, and 36 m/s. The ribbon thickness decreased gradually with increased jetting temperature also at every roll speed, in spite of a slight difference in decrement in ribbon thickness at each roll speed. Results verified the jetting temperature dependence on the ribbon thickness of rapidly solidified Fe$_{79.5}$Si$_{8.5}$B$_{12}$. The reason is considered as follows: The lowest viscosity of rapidly solidified Fe$_{79.5}$Si$_{8.5}$B$_{12}$ at an undercooled state and viscosity drop with increased jetting temperature at every roll speed improved the contact conditions between the molten metal paddle and a roll; consequently, the cooling rate increased, and a decrease in ribbon thickness was promoted. To examine the contact state between a ribbon and a roll in detail, SEM observation of the ribbon surface will be conducted in a later study.

3.3 Hardness measurement and TEM observation of rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ and Fe$_{79.5}$Si$_{8.5}$B$_{12}$ ribbons

Figures 5(a) and 5(b) present results of Vickers hardness measurement of rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ and Fe$_{79.5}$-Si$_{8.5}$B$_{12}$ alloy ribbons, respectively, at various ejection temperatures at each roll speed. The figures show that Vickers hardness is almost constant for each alloy ribbon in spite of the increased jetting temperature. The hardness of the rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ alloy ribbon was about 1000 Hv, which agrees well with the hardness of this alloy ribbon reported by Masumoto et al. The hardness of the rapidly solidified Fe$_{79.5}$Si$_{8.5}$B$_{12}$ alloy ribbon was about...
The hardness of the rapidly solidified Fe$_{79.5}$Si$_{8.5}$B$_{12}$ alloy ribbon was slightly lower than that of the rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ alloy ribbon, which is presumed to be true because the less-metalloid elements such as Si and B lowered the partial covalent character. Figures 6(a) and 6(b) present the selected area diffraction (SAD) patterns obtained through TEM observation of rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ and Fe$_{79.5}$Si$_{8.5}$B$_{12}$ alloy ribbons respectively, spun at a roll speed of 36 m/s and ejection temperature of 1673 K. Both images portray only typical halo patterns of the broad ring shape commonly observed in rapidly solidified ribbons. They verify that an amorphous structure had formed. In addition, the diffraction patterns suggest no structural difference because of alloy composition differences. It is considered that detailed observation using high-resolution TEM is necessary to investigate this issue in future studies.

4. Conclusion

Rapidly solidified Fe-Si-B alloy ribbons were fabricated using single-roll melt spinning. Mainly, the change in ribbon thickness was investigated with different jetting temperatures. There by, the following knowledge was obtained.

1. In fabrication of rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ (hyper-eutectic alloy) ribbons, the ribbon thickness decreased concomitantly with increased roll speed. Ribbon thickness showed the minimum with increased jetting temperature at each roll speed.

2. In fabrication of rapidly solidified Fe$_{79.5}$Si$_{8.5}$B$_{12}$ (eutectic alloy) ribbons, the ribbon thickness decreased concomitantly with increased roll speed and jetting temperature.

3. The hardness of rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ and Fe$_{79.5}$Si$_{8.5}$B$_{12}$ alloy ribbons was almost constant over jetting temperatures. Hardness of the rapidly solidified Fe$_{79.5}$Si$_{8.5}$B$_{12}$ ribbon is lower than that of the rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ ribbon.

4. TEM observation of rapidly solidified Fe$_{75}$Si$_{10}$B$_{15}$ and Fe$_{79.5}$Si$_{8.5}$B$_{12}$ alloy ribbons showed a halo pattern, which verified their amorphous structure.

In conclusion, results suggest that the effect of jetting temperature on ribbon thickness varies with alloy composition, and that the viscosity of molten metal and undercooled alloy affects the ribbon thickness during fabrication of rapidly solidified Fe-Si-B alloy ribbons.
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