DOI: 10.24425/amm.2021.134766

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MICROSTRUCTURE EVOLUTION AND FORMATION MECHANISM OF COPPER-SILVER ALLOY FABRICATED BY INDUCTION HEATING DIRECTIONAL SOLIDIFICATION AT DIFFERENT MOLD TEMPERATURES

Cu-2wt%Ag alloy with diameter of 10 mm was fabricated by induction heating directional solidification (IHDS). The effect of different mold temperatures on microstructure of IHDS Cu-2wt%Ag alloy was investigated. The results show that IHDS Cu-2wt%Ag alloy is mainly composed of coarse columnar grains at mold temperature of 1075°C. While the mold temperature is at 1100°C, 1150°C and 1200°C, respectively, the IHDS Cu-2wt%Ag alloy is composed of columnar grains and equiaxed grains and the number of grains increases. Meanwhile, the growth direction of columnar grains in the edge of alloys deviates from the direction of continuous casting to form "V" shape. While the mold temperature is controlled at high temperature, the induced current increases, which leads to the enhancement of eddy current in the mold. Therefore, the dendrites fall off to form new grains under the effect of eddy stirring, resulting in an increasing in the number of grains.

Keywords: Induction heating directional solidification, Cu-Ag alloy, Mold temperature, Microstructure

1. Introduction

Copper and copper alloys are widely used in various fields due to their good conductivity and low price. With the development of modern industrial technology, copper alloys are required not only to have good electrical conductivity, but also to have high strength. The methods to improve the strength of copper alloy mainly include fine grain strengthening [1], precipitated phase strengthening [2] and alloying strengthening [3]. For example, adding Ag element to pure Cu is an effective means to fabricate high strength copper alloy [4]. At present, Cu-Ag alloy is widely used in fields such as high-intensity magnetic field equipment, leading framework of very large scale integrated circuits, transmission contacting line, etc. [5-7]. According to literature [8], the tensile strength of Cu-Ag alloy increases with the increasing of Ag content. However, the conductivity of Cu-Ag alloy is generally lower than that of pure Cu or pure Ag, and the conductivity of the alloy decreases gradually with the increasing of Ag content [9]. It can be seen that the high strength and high conductivity cannot be improved at the same time.

In order to improve the conductivity of copper alloys, it is a feasible method to change the microstructure of copper alloys under certain chemical composition. The columnar grain has higher conductivity because there is no large number of transverse grain boundaries in the axial direction. The formation of columnar grain is mainly relevant to the liquid temperature gradient (G_L) in front of solid-liquid interface and the growth velocity (R) of the solid-liquid interface. The angle between the growth direction of columnar grain and the axis is small when the ratio of G_L/R is greatly large [10]. Directional solidification can obtain columnar grains with good properties [11-13]. The principle is that the liquid metal in the mold is heated and the solidified metal at the exit of the mold is forced to cool. Then the directional heat flow is established. Therefore, the grains grow in an opposite direction of the heat flow [14]. During the process of directional solidification, the mold temperature is the main variable affecting the liquid temperature gradient (G_L) at the front of solid-liquid interface while the casting speed and cooling conditions are constant.

In this work, Cu-2wt%Ag alloy was used as raw material for fabrication of the directional solidification alloy. We use induction heating to heat the mold. The effect of different mold temperature on microstructure of induction heating directional solidification (IHDS) Cu-2wt%Ag alloy was investigated.

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2. Experimental

The raw material used in the experiment is Cu-2wt%Ag alloy, which the solidus temperature is 1050°C and the liquidus temperature is 1075°C. The samples were fabricated by IHDS technique. The schematic diagram of the IHDS equipment is shown in Fig. 1. It is mainly composed of graphite crucible, induction heating coil, temperature-controlling induction coil, graphite mold, cooling water and traction wheel. The IHDS equipment is similar to the industry equipment for continuous casting of copper and brass ingots [15]. The Cu-2wt%Ag alloy in the graphite crucible is melted by induction heating. The molten alloy flows into the graphite mold, which heated by the temperature-controlling induction coil. Cooling water is provided at the exit of the mold to cool of solidified alloy or dummy bar. A traction wheel is installed to continuously pull the alloy out of the graphite mold.



Fig. 1. Schematic diagram of IHDS equipment

At first, the Cu-2wt%Ag alloy in graphite crucible was melted with argon atmosphere and kept for 20 minutes. The graphite crucible temperature was set to 1200°C. The mold temperature was set to 1075°C, 1100°C, 1150°C and 1200°C, respectively. The temperature of the cooling water was 20°C, and the continuous casting speed was 2 mm/min. One end of the copper dummy bar with diameter of 10 mm was inserted into the mold, and the other end is clamped by traction wheel. An Ø10 mm IHDS Cu-2wt%Ag alloy can be continuously pulled out by the traction wheels.

Samples for microstructure observation were cut from the IHDS Cu-2wt%Ag alloys. After polishing, the samples were etched by corrosion solution of ferric chloride (5 g) and anhydrous ethanol (100 mL). The microstructure observation was performed by using optical microscope. XRD was used for the diffraction analysis of the IHDS Cu-2wt%Ag alloys.

3. Results and Discussion

Fig. 2 shows the microstructure of IHDS Cu-2wt%Ag alloy at different mold temperatures. The direction of continuous casting is indicated by CD (cast direction) arrow in the Fig. 2.

Fig. 2(a) is the microstructure of IHDS Cu-2wt%Ag alloy at mold temperature of 1075°C. According to the transverse section microstructure, it can be found that the number of grain is small. The average diameter of grains is about 2000 µm. It can be seen from the longitudinal section microstructure in Fig. 2(a) that the microstructure is mainly composed of coarse columnar grains. Meanwhile, the axial direction of columnar grains is basically parallel to CD. Fig. 2(b) shows the microstructure of IHDS Cu-2wt%Ag alloy at mold temperature of 1100°C. The number of grains begins to increase and the diameter of grains decreases. As can be seen from the longitudinal section microstructure of Fig. 2(b), the axial direction of columnar grains on the left side of the IHDS Cu-2wt%Ag alloy is parallel to CD. However, the columnar grains on the right side begin to deviate from CD, forming an angle of about 32°. Furthermore, the axial and radial dimensions of columnar grains begin to decrease. Fig. 2(c) shows the microstructure of IHDS Cu-2wt%Ag alloy at the mold temperature of 1150°C. The grain size has been measured and the average diameter of the grain is about 480 µm. From the longitudinal section microstructure in Fig. 2(c), it can be seen that the edge of the IHDS Cu-2wt%Ag alloy is columnar grains and the distribution is relatively symmetrical. The angle between axial direction of columnar grains and CD is about 26°. A small amount of equiaxed grains appear in the center of the alloy. Fig. 2(d) shows the microstructure of IHDS Cu-2wt%Ag alloy at the mold temperature of 1200°C. The average diameter of grains is 430 µm, thus indicates the grain size decreases with the increasing of mold temperature. The angle between the axial direction of columnar grains and the CD is about 25°. Compared with Fig. 2(c), the number of equiaxed grains in the center is increased. From the above results, it can be concluded that the axial direction of the columnar grains deviates from the continuous casting direction. The columnar grains become fine and the grain size begins to decrease as the mold temperature is controlled at a higher temperature.

As can be seen from Fig. 2, the columnar grains form "V" shape at the mold temperature of 1150°C and 1200°C. Thus indicates that the upward growth of columnar grains is restricted. The main reason is that the growth of columnar grains is limited by the high-temperature liquid due to the increase of mold temperature. When the mold temperature is maintained at 1075°C, the liquid temperature in front of the solid-liquid interface is low. The growth direction of the grain is almost parallel CD. The above results show that mold temperature has a great effect on the growth of columnar grain of IHDS Cu-2wt%Ag alloy. In the process of IHDS Cu-2wt%Ag alloy, although the liquid temperature gradient G_L at the front of solid-liquid interface can be increased by increasing the mold temperature, the columnar grains with uniform structure parallel to CD can not be obtained. It also can be seen from Fig. 2, at a certain continuous



Fig. 2. Microstructure of IHDS Cu-2wt%Ag alloy at different mold temperatures. (a) 1075°C; (b) 1100°C; (c)1150°C; (d) 1200°C

casting speed, the grain size of IHDS Cu-2wt%Ag alloy begins to decrease as the mold temperature increasing. While the mold temperature reaches 1200°C, the columnar grains becomes less obvious and the number of equiaxed grains begins to increase. Similar to literatures [16,17], the transformation of columnar grains into equiaxed grains occurred.

Fig. 3 is the transverse section microstructure of IHDS Cu-2wt%Ag alloy at different mold temperatures. While the mold temperature is 1075° C (Fig. 3 (a)), dendritic structure can be clearly found in the grains and the average distance between dendrites is about 130 µm. While the mold temperature is 1100° C, 1150° C and 1200° C (Fig. 3(b) ~ 3(d)), it can be seen from Fig. 3(b) ~ 3(d) that there is no obvious dendritic structure in the grains. This indicates that at a lower mold temperature (1075° C), the grain growth is mainly carried out in the form of dendrite. On the other hand, the liquid temperature in front of the solid-liquid interface increasing is not conducive to the increase of undercooling at the front of dendrite, which leads to the dendrites growth being restricted.

Fig. 4 is the XRD results of IHDS Cu-2wt%Ag alloys. From Fig. 4, it can be found that there exists a peak value at 2θ of 50.43° as the mold temperature is 1075°C, and the corresponding grain plane is (200). There is no obvious peak value of other diffraction angles. The results suggest that the grain grows along a single direction and the growth of other grain planes is inhibited at mold temperature of 1075°C. When the mold temperature is increased to 1100°C, 1150°C and 1200°C, respectively, the peak values of (111), (220) and (311) grain planes begin to appear in the IHDS Cu-2wt%Ag alloy, which indicates that the grain orientation begins to diversify and no longer appears a single growth direction.

Directional solidification is a kind of technique based on the theory of mold wall nucleation. When the mold temperature begins to be heated above the liquidus of the alloy, the alloy in the mold maintains the liquid phase and avoids the grain nucleation on the mold wall. Simultaneously, forced cooling at the exit of the mold leads to forming downward heat flow. Near the exit of the mold, the grain begins to grow in the opposite direction of the heat flow, and finally forms columnar grains.

During columnar grain growth, only under the condition of high G_L/R value, the actual growth direction of columnar grain is closer to the theoretical growth direction. Otherwise, the



Fig. 3. Transverse section microstructure of IHDS Cu-2wt%Ag alloys at different mold temperatures. (a) 1075°C; (b) 1100; (c) 1150°C; (d) 1200°C



Fig. 4. XRD results of IHDS Cu-2wt%Ag alloy

grain growth will deviate from the axial direction. According to undercooling theory [18]

$$\frac{G_L}{R} \ge \frac{m_L C_0 \left(1 - k_0\right)}{D_L k_0}$$
(1)

Where m_L is the liquidus slope, C_0 is the original composition of the alloy, k_0 is the partition coefficient and D_L is the diffusion coefficient of solute in liquid. The dendritic structure is formed because of the instability of the solid-liquid interface caused by the composition undercooling, which is produced by the accumulation of solutes in the front of the solid-liquid interface. If the solidification condition satisfies Eq. (1), the interface is a stable flat, whereas the interface is unstable to form cell or dendrite. In front of the solid-liquid interface, the undercooled zone appears due to the liquid temperature gradient decreasing, which destroys the stability of the flat interface. At this time, any protruding at the flat interface will lead to greater fluctuations. Furthermore, composition undercooling in front of the interface gradually widens as G_L/R decreases, resulting in protruding quickly growth.

In general, the growth speed of grain is related to grain orientation. In face-centered cubic metals, [100] is the preferred direction for grain growth, followed by [110], and [111] is the slowest [19,20]. The grains growing faster will gradually eliminate those growing slower and finally form coarse columnar grains during competitive growth. From the Fig. 4, at mold temperature of 1075°C, the preferred orientation of grains in α -phase is (200) plane. The normal direction perpendicular to the (200) plane is [100]. Comprehensively analyzing Fig. 2(a) and Fig. 4, it can be found that the growth speed of dendritic grain is faster at [100] direction. The other orientations are gradually eliminated during the growth of columnar grains with dendritic structure.

On the other hand, induction heating was used for mold heating. At lower continuous casting speed (2 mm/min), the solid-liquid interface of the alloy is always inside the mold due to the effect of cooling water at the exit of the mold, despite the mold temperature is above liquidus temperature. The induced current increase as the mold temperature is increasing. Thus results to the eddy current intensity in the mold increasing, which has function of stirring for the liquid at the front of the solid-liquid interface. Therefore, secondary or primary dendrites will fall off under the increasing stirring. Those falling dendrites become new nuclei in the liquid and grow to form the equiaxed grains at the center of IHDS Cu-2wt%Ag alloys.

5. Conclusion

In this work, the Cu-2wt%Ag alloys were fabricated by IHDS technique. Under certain casting speed and cooling conditions, an effect of different mold temperatures on microstructure of IHDS Cu-2wt%Ag alloys was investigated. The following conclusions, which are valid for the sample with diameter of 10 mm, are obtained:

- When the mold temperature is low (1075°C), the columnar grains of IHDS Cu-2wt%Ag alloy grows faster in [100] direction, showing obvious directional growth.
- (2) The columnar grains deviated from the direction of continuous casting and formed "V" shape as the mold temperature increased. There exists equiaxed grains in IHDS Cu-2wt%Ag alloy.

REFERENCES

- P. Zhang, A. Ma, S. Lu, G. Liu, P. Lin, J. Jiang, C. Chu, Materials and Design 32, 348-352 (2011).
- [2] G. Xie, Q. Wang, X. Mi, B. Xiong, L. Peng, Materials Science and Engineering: A 558, 326-330 (2012).

- [3] J. Zhang, N. Gao, M.J. Starink, Materials Science and Engineering A 527, 3472-3479 (2010).
- [4] Y. Sakai, K. Inoue, T. Asano, H. Wada, H. Maeda, Applied Physics Letters 59, 2965-2967 (1991).
- [5] L. Zhang, L. Meng, Scripta Materialia 52, 1187-1191 (2005).
- [6] T. Hirota, A. Imai, T. Kumano, M. Ichihara, Y. Sakai, K. Inoue, H. Maeda, IEEE Transactions on Magnetic 4, 1891-1894 (1994).
- [7] D.W. Yao, L. Meng, Physica B 403, 3384-3388 (2008).
- [8] L. Meng, Y.W. Zeng, Materials Science and Engineering A 435-436, 237-244 (2006).
- [9] A. Benghalem, D.G. Morris, Acta Materialia 45 (1), 397-406 (1997).
- [10] M.C. Flemings, Metallurgical Transactions 5, 2121-2134 (1974).
- [11] M. Okayasu, S. Yoshie, Materials Science and Engineering A 527, 3120-3126 (2010).
- [12] Y.A. Kwon, Z.A. Daya, H. Soda, Z. Wang, A. McLean, Materials Science and Engineering A, 368, 323-331 (2004).
- [13] M. Okayasu, S. Takasu, S. Yoshie, Journal of Materials Processing Technology 210, 1529-1535 (2010).
- [14] A. Ohno, Journal of Metals 38 (1), 14-16 (1986).
- [15] W. Wołczyński, Z. Lipnicki, A.W. Bydałek, A.A. Ivanova, Achieves of Foundry Engineering 16 (3), 141-146 (2016).
- [16] W. Wołczyński, A.A. Ivanova, P. Kwapisiński, Scientific Journals of the Maritime University of Szczecin, 56 (128), 47-54, (2018)
- [17] W. Wołczyński, A.A. Ivanova, P. Kwapisiński, Proceedia Manufacturing 30, 459-466, (2019).
- [18] W.A. Tiller, K.A. Jackson, J.W. Rutter, B. Chalmers, Acta Metallurgica 1, 428-437 (1953).
- [19] P.R. Harowell, D.W. Oxtoby, The Journal of Chemical Physics 86 (5), 2932-2942 (1987).
- [20] R.J. Braun, J.W. Cahn, G.B. McFadden, H.E. Rushmeier, A.A. Wheeler, Acta Materialia 46 (1), 1-12 (1998).