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Fabrication of pure copper rods containing continuous columnar crystals by continuous unidirectional solidification technology

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Abstract: Pure copper rods containing continuous columnar crystals were fabricated using the downward CUS (Continuous Unidirectional Solidification) equipment. When the technological parameters were set as the ranges of mould temperature 1100-1300°C, cooling distance (the distance from the exit of the cast mould to the start point of cooling) 10-20 mm, casting speed 0.2-2.5 mm/s, cooling water (20~25°C) volume 1000-1320 L/h, and when these parameters matched reasonably, the CUS process was performed stably, and pure copper rods containing continuous columnar crystals with bright and smooth surface were produced. The dendritic arm spacing of the crystals in copper rods decreased with increasing the casting speed. The results of the texture by X-ray diffraction analysis showed that the rods has strong ⟨100⟩ fiber texture.

Key words: copper; continuous unidirectional solidification; mould temperature; casting speed; microstructure; texture

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1 Introduction

In 1978 Atsumi Ohno invented the Ohno Continuous Casting technology (OCC process) on the basis of the theory of dissociative nucleation of grains. In this technology the water-cooled mould in the conventional continuous casting process is replaced by the heated mould and the heat is extracted from molten metal along the casting direction by using a cooling device near the mould. Thus nucleation of crystals on the mould wall is avoided and the conditions for unidirectional solidification are created, and cast rods with unidirectional crystals are fabricated [1,2]. There are three basic types for OCC equipment, the horizontal, the upward and the downward type. Among them the downward type produces high grade cast rods.

Based on the above principles a downward continuous casting equipment for unidirectional solidification was developed and the Continuous Unidirectional Solidification (CUS) technology for copper, copper alloy, aluminum alloy, iron and steel (including stainless steel, low-carbon steel, *etc.*) rods has been investigated [3,4]. T. Shimizu reported that it is possible to fabricate materials with good workability by this method [5]. The authors' results were in agreement with it. The cast pure copper and aluminum alloy rods produced by CUS method were worked down to ultra-fine wires at room temperature without any middle heat treatment. The cast copper rod was reduced from 17.28 mm to 19.7 μm in diameter with a total deformation of $7.0\times10^7\%$ at room temperature. The aluminum-silicon rods of 8 mm in diameter was processed to wires of 30 μm in diameter at room temperature. This process has a wide application for the fabrication of ultra-fine wires and the production of materials which are difficult to deform [5]. The objective of this work is to investigate the fabrication, microstructure and texture characteristics of the pure copper rod by CUS process.

2 Experimental

2.1 Equipment

Figure 1 presents the schematic illustration of a downward CUS equipment. It is beneficial to protect the copper from oxidation by conducting experiments under argon atmosphere. The equipment is mainly composed of mould, graphite crucible, induction coils, cooling device, *etc.* There are three technical characteristics with the equipment. First, the solid-liquid interface lies near the mould exit, which not only cast rods with bright and smooth surface are obtained due

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to the small friction between the mould and the rod surface, but also the break-out or tear-off phenomenon is avoided; second, it is easy for the cast rods to leave the mould attributed to a positive taper of the mould cavity; third, the quality of the cast rods can be enhanced because this equipment removes pores and impurities more easily.

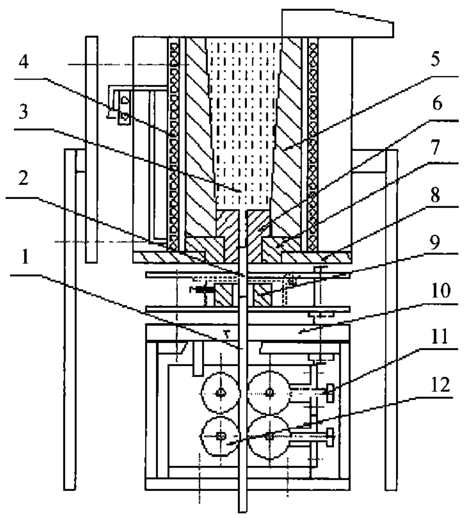

Figure 1 Schematic diagram of the downward CUS equipment: 1—dummy rod; 2—cast rod; 3—molten metal 4—induction coils; 5—crucible; 6—heated mould; 7—fireproof material; 8—aluminum board; 9—cooling device; 10—water tank; 11—press bolt; 12—drive wheel.

2.2 Materials and procedure

The electrolytic copper was used as melt stock in the experiments.

The experiments were performed as the following steps: first, heated the electrolytic copper, the cooling distance was preset at 10-20 mm, the volume of cooling water (20-25°C) was fixed at 1000-1320 L/h; then, when the temperature of the mould reached 1200-1300°C, kept the copper at this temperature for about half an hour, so that the copper fully melted; finally, adjusted the mould temperature to 1100-1300°C and started up the withdrawing device. At the beginning of the withdrawing, the casting speed was set at a reasonable slow speed, when the equilibrium of the CUS system was obtained the casting speed was raised. In the process of the experiments the technological parameters should be stable and be properly adjusted to make them matched to each other.

3 Results and discussion

Figure 2 shows the cast copper rod produced in the experiments. Its chemical composition (mass fraction) is 99.98% Cu, Ag<0.005%, 0.0038% Fe, Si<0.0005% and other elements in balance. The rod is so flexible that it is easy to bend it in a circle by hand (figure 2(a)). Figure 2(b) is a higher magnification view of the copper rod in figure 2(a), the image of the word "copper" in the rod surface is clear, reflecting the mirror-like surface of the rod.

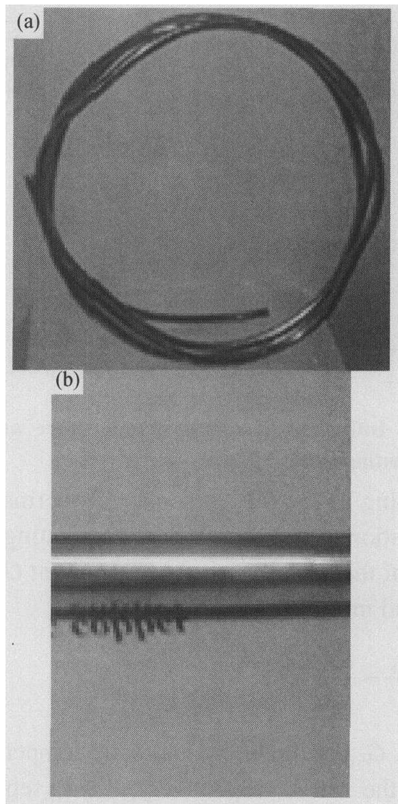

Figure 2 Copper rod produced by CUS process: (a) low magnification; (b) high magnification.

3.1 Technological parameters

The temperature gradient G_L of the molten metal near the solid-liquid interface and the growing speed R of the crystals are two important technological parameters of CUS process. The improvement of the temperature gradient of the molten metal near the solid-liquid interface is one of the objectives of CUS process [6]. The crystal morphology is controlled by above mentioned two parameters. In the CUS process, the comprehensive effects of the major technological parameters determine either the position or the shape of the solidification interface. These parameters are mould temperature, casting speed, cooling distance and cooling water volume. Since the parameters of cooling distance and cooling water volume are seldom changed once they are determined, the principal adjustable processing parameters are mould temperature and casting speed.

Figure 3 presents the influence of mould temperature and casting speed on the casting state. The area I, II, III are steady state, break-out state and tear-off state, respectively for a cooling distance of 10 mm and a cooling water of 1320 L/h. It is seen that the mould temperature range becomes narrower and narrower with increasing the casting speed. The influence of the mould temperature and casting speed on the solidification process and the microstructure is analyzed as follows.

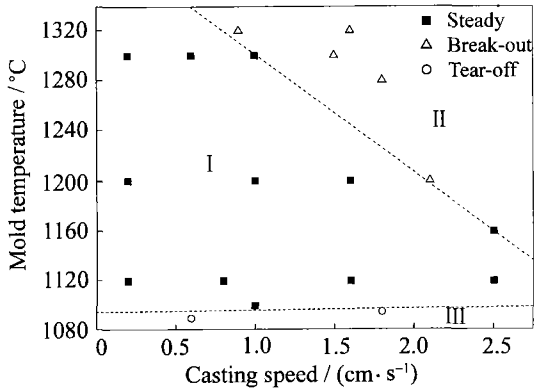

Figure 3 Influence of mould temperature and casting speed on casting state.

According to the one-dimensional thermal equilibrium equation and Fourier heat-conducting law, the equation of the liquid temperature gradient G_L near the solid-liquid interface was derived [7]:

$$G_L = \frac{\lambda_S G_S}{\lambda_L} - \frac{LR\rho_m}{\lambda_L} \quad (1)$$

where G_L, G_S are the liquid and solid temperature gradients of the metal, respectively; λ_S, λ_L represent the heat-conducting coefficient of solid and liquid metal, respectively; L denotes the crystallization latent heat for crystal growth for unit mass; R is the solidification speed; ρ_m is the density of molten metal near the melting point.

The equation indicates that G_L can be improved by improving G_S, but the usual method is improving the temperature of molten metal near the solidification front. In the experiments the mould temperature is used to reflect the temperature of the molten metal. Too low or too high mould temperature will lead to some adverse consequences. Although low mould temperature is helpful for high casting speed, it will make the solid-liquid interface move to the intake of the mould if the mould temperature is too low under the condition of other parameters maintaining constant. Thus three problems will be caused: (1) the temperature gradient reduces; (2) the friction increases because of a rise of the contact area between the cast rod and the mould wall, cracks are easy to appear on the surface of the rod and even tear-off phenomenon will occur; (3) the crystal growth directions may deviate from the axis of the rod. High mould temperature assists the solid-liquid interface to protrude in the molten metal. Such convex solid-liquid interface is conducive to eliminate the crystal growth directions deviating from the axis through contest. But very high mould temperature makes the interface descend or even causes break-out phenomenon. So it is necessary to control the mould temperature in an appropriate range so as to control the solid-liquid interface near the mould exit, hence the liquid temperature gradient is increased and the friction between the rod surface and the mould wall is decreased, which aids to obtain cast rods with high surface quality. In the experiments, the reasonable mould temperature is in the range of 1100-1300°C.

In the stage of stable CUS process, the solidification speed and the casting speed are consistent. When the cooling intensity is constant, the lower the mould temperature, the greater the casting speed; when the mould temperature remains constant, the greater the cooling intensity, the greater the maximal casting speed. High casting speed is advantageous to the improvement of production efficiency and the fining of the crystals. But there is a limit for the casting speed, when it exceeds this limit, crackles will occur on the surface of the rod or the break-out phenomenon will happen. So the casting speed must also be controlled properly. Known from the figure 3 that when the casting speed is in the range of 0.2-2.5 mm/s the CUS process is performed normally.

3.2 Dimensional stability

In the CUS experiments, the solid-liquid interface lies above the mould exit. Because the mould cavity is a positive taper, the diameter of the cross-section along the length is different. When the solid-liquid interface moves up or down as a result of the fluctuation of technological parameters, the diameter of the rod will vary accordingly. **Figure 4** shows that the variation in diameter along the copper rod approximately 4500 mm in length is less than 0.19 mm and the average diameter is 17.28 mm with a standard deviation of 0.05 mm. To achieve better dimensional stability, the processing parameters, especially mould temperature and casting speed, should be maintained more stably.

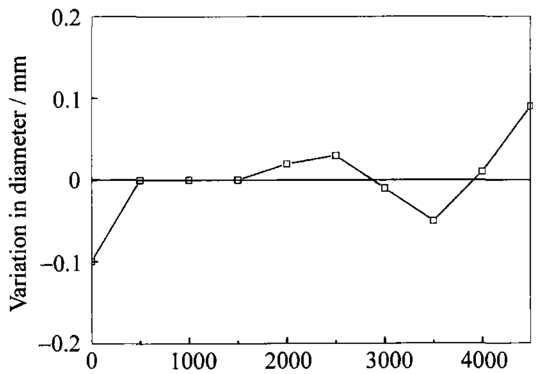

Figure 4 Variation in diameter along the length of the copper rod, the average rod diameter is 17.28 mm.

3.3 Microstructure

The morphology of the solid-liquid interface and the solidified microstructure are mainly influenced by the value of G_L/R. For a given temperature gradient G_L, when R is very small, the shape of the interface is planar; the value of G_L/R decreases with increasing R, and the planar shape changes into cellular one. In the experiments when the mould temperature was remained at 1100-1200°C and the casting speed ranged from 0.2 mm/s to 2.5 mm/s, the crystals of the cast rods are columnar. The characteristic dimension of this microstructure is dendritic arm spacing. The classical theoretical model on the dendritic arm spacing is Jackson-Hunt model [8].

$$\lambda = AG^{-1/2}R^{-1/4} \qquad (2)$$

where λ denotes the dendritic arm spacing; A represents a coefficient; G is the temperature gradient; R is the casting speed.

The results of Young *et al.* supported this conclusion [9]. A. Ohno *et al.* also investigated the influence of casting speed on the dendritic arm spacing of solidified microstructure. They came to the conclusion that fastening casting speed shortened the dendritic arm spacing [10].

In this work, the dendritic arm spacing of the pure copper rod also decreased with increasing the casting speed. **Figure 5** is the longitudinal microstructure of the cast copper rod at different casting speeds. Figure 5(a) and (b) were obtained at casting speeds of 0.2 mm/s and 2.0 mm/s, respectively. The corresponding dendritic arm spacings are approximately 230 μm and 100 μm respectively.

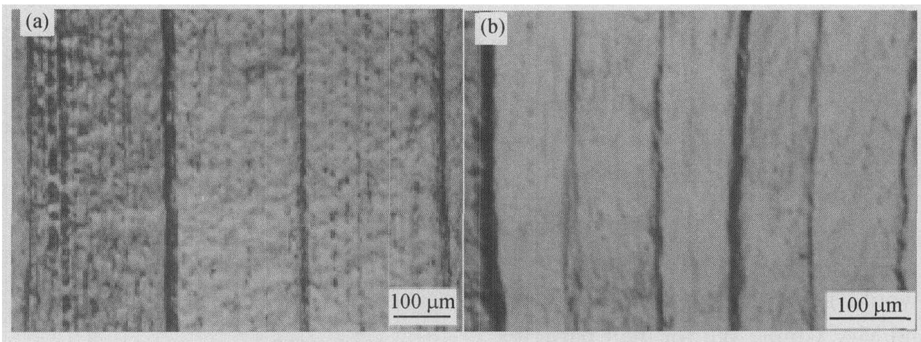

Figure 5 Micrographs of the copper cast rod at different casting speeds: (a) *R*=0.2 mm/s; (b) *R*=2.0 mm/s.

3.4 Texture

The micrographs of the copper rod shows that it has unidirectional crystals. In order to investigate the details of crystal orientation, the texture of the copper rod was observed. The {100} and {111} pole figures of the cast copper rod were determined by General Area Detector Diffraction System/Solution, which are shown in **figure 6**. From the pole figures, it is found that the high density clusters correspond to texture variants of {110}<001> and {001}<$\bar{1}$00>. Thus the copper rod has strong <001> fiber texture. Zhenming Xu *et al.* [11] reported a preferred growth orientation of [100] in copper single crystals produced by continuous casting. H. Soda *et al.* [12] suggested that the preferred orientation in copper crystals may be due to an impurity segregation effect at the interface caused by a faster casting speed, which in turn, changes the mode of solidification. The factors of impurity and casting speed may also influence the textures in CUS copper. The cast copper rod by CUS process exhibited strong <001> fiber texture. This characteristic is probably one of the reasons why the cast copper rod by CUS process possesses excellent cold workability. There are perhaps also other reasons for its outstanding workability, such as the working method, deformation conditions, and so on, which need further study.

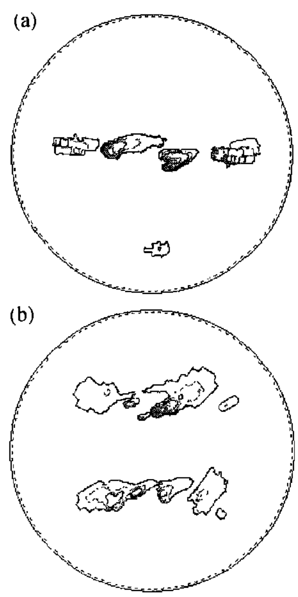

Figure 6 {100} (a) and {111} (b) pole figures of the copper cast rod.

4 Conclusions

(1) The copper cast rod by CUS process exhibits excellent workability, which was worked to ultra-fine wires at room temperature with a total deformation of

7.0×10^{7}%.

(2) When the technological parameters varied in the following ranges: mould temperature 1100~1300°C, cooling distance 10-20 mm, casting speed 0.2-2.5mm/s, cooling water volume 1000-1320 L/h, and when these parameters matched appropriately with each other, the CUS process could be conducted stably and copper rods with bright and smooth surface could be fabricated.

(3) The copper rod with an average diameter of 17.28 mm had a standard deviation of 0.05 mm in diameter. More stable technological parameters can further improve the dimensional stability.

(4) When the mould temperature ranged from 1100°C to 1200°C and the casting speed varied between 0.2-2.5 mm/s, the crystals of the cast rods were columnar. And when the casting speed rose the size of the crystals lessened.

(5) There is strong <100> fiber texture along the longitudinal direction of the cast copper rod, which is the major reason for the excellent cold workability.

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