CONTINUOUS CASTING TECHNOLOGY OF SINGLE CRYSTAL COPPER ROD①

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ABSTRACT Single crystal copper rod, 8 mm in diameter, was successfully produced using self-designed horizontal continuous casting equipment. Experimental results showed that the single crystal copper rod with mirror-like surface can be obtained on condition that the temperature of the mould was 1373~ 1473K and the casting speed was 20~ 30mm/ min. The crystallization orientation was [100] and the crystallization face was (200). The mixed crystals (equiaxed and columnar grains) on the rod surface were formed when the mould temperature was lower than the melting temperature(T_m< 1356K) in the solidification front and the traction rate increased. Compared with those of unidirectionally solidified polycrystal copper rods, the tensile strength and the yield strength of the single crystal rod decreased by 8.82% and 91.62%, respectively, while the elongation and the reduction of cross-section area increased by 24.98% and 3.09%, respectively.

Key words single crystal copper rod continuous casting mechanical properties

1 INTRODUCTION

Recently, with the improvement in precision and miniature of electronic equipment, advancement in musical instrument, there was an urgent requirement for thin foils, fine wires, and single crystal rods, plates and cables[1]. For instance, the basic board material of integrated circuit and semiconductor element paste wire is desired to use grain boundary- and defects-free, high quality material. Single crystal cables have been used in sound equipment, which can improve fidelity of signal transmission[2]. The main producting processes of single crystal metal are Czochralski Process[3] and Brigman Process[4], but these processes are not suitable for the production of net-shaped single-crystal wires of substantial length and can not satisfy request of electronics industry. A new continuous casting method (OCC) was found by Ohno at the end of 1980s[5], in which the mould remained stationary within the heated zone from which a solidified crystal was withdrawn continuously, thereby it is possible to produce metallic single crystals of small diameter but unlimited length. This method is characterized by simple production technology, low cost, high efficiency. In Japan, USA and Canada[5-8], the technique of continuous casting of single crystal has been studied, and it has been preliminarilly studied in China. Although the single crystal copper wire has been successfully developed into a series of ideal audio cables PCOCC in Japan[6], the specific technical parameters, the crystallization orientation and mechanical properties have been studied very little. In this paper, the authors have studied above questions, which offers reliable technological parameters for the production of single crystal copper rods.

2 EXPERIMENTAL

Fig. 1 shows a schematic illustration of the self-designed horizontal continuous casting equipment, which consists of a melting furnace, a heating mould, a cooler, a withdrawal system

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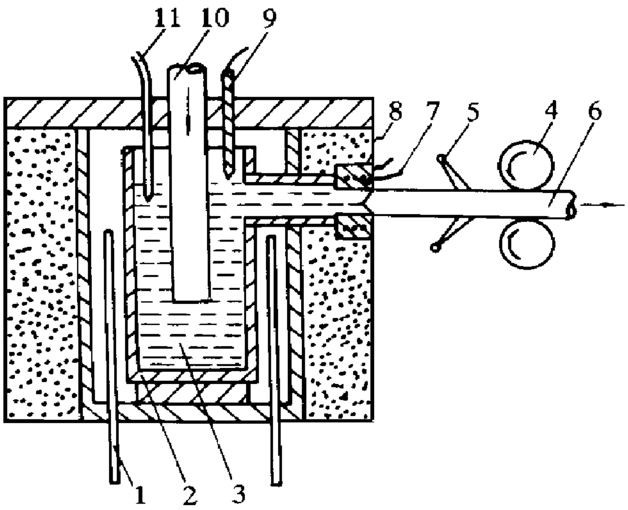

Fig. 1 Schematic illustration of horizontal crystal continuous casting equipment

1 —SiC heater; 2 —Crucible;
3 —Molten metal; 4 —Pinch rolls;
5 —Cooling water; 6 —Cast product;
7 —Thermocouple; 8 —Mould;
9 —Probe of the metal height;
10 —Control rod of metal height; 11 —Thermocouple

and a system for detecting and controlling the height of the liquid metal in the crucible. The various technological parameters for the continuous casting are as follows:

(1) Commercially available electrolytic copper was melted under a high-purity argon atmosphere. The furnace temperature was kept at 1 573 K;

(2) The mould temperature was controlled at 1 373~ 1 573 K;

(3) The amount of cooling water was approximately 60 mL/ min. The cooling device can move 5 cm to 30 cm away from the mould exit, in this work, this distance was kept at 7 cm.

(4) The casting speed was kept at 10~ 40 mm/ min;

(5) The static pressure head was kept at 15 ~ 20 mm.

The crystallization face and crystallization orientation of continuous casting copper single crystal was investigated by XRD (D/ max- 3c).

3 RESULTS AND DISCUSSION

3.1 Microstructure evolution and crystallization orientation

Fig. 2(a) shows the microstructure evolution observed on longitudinal cross-section of the rod. Because of the effect of sharp cooling, the equiaxed grains are firstly formed at the start-up procedure. As the casting process continues, the unidirectional polycrystal structures grow on the equiaxed grains. The temperature on the inner wall of the mould is held higher than the melting point of the copper, so the nucleation on the wall of the mould can be avoided. Meanwhile, a temperature gradient is formed between the mould and the cooler, which makes the solidification process unidirectional. At the same time, because of the preferred growth on crystallization faces, the grains whose growth orientation is not in keeping with the thermal current are gradually eliminated. Finally, as the competitive growth of crystal, a single crystal rod is formed.

Fig. 2(b) shows XRD spectrum for the transverse cross-section of the rod. The gradually eliminated crystallization face are (311), (220) and (111). Finally, the single crystal growth crystallization face is (200), and the crystallization orientation is [100] which is perpendicular to (200). The growth speed of crystal is related to the crystallization orientation of crystal. The copper is a face centred cubic crystal, the preferred growth orientation is [100], the next is [001], and the growth speed of [111] is the slowest. The (200) is crowded with arrangement crystallization face. So, the crystallization orientation is [100] and the crystallization face is (200) at continuous casting.

3.2 Solid/ liquid interface position and morphology

By experimental observation and determination of the temperature field, the solid/ liquid interface was controlled at the inside of the mould (2~ 4 mm from the mould exit) during the continuous casting, so that a single crystal copper rod with mirror-smooth surface can be obtained. When the solid/ liquid interface existed outside the mould, the breakout occurred and the experiment was broken off. While the solid/ liquid interface is controlled at the outside (2.5 mm from the mould exit) of the mould at continuous casting of single crystal aluminum[9], this is because

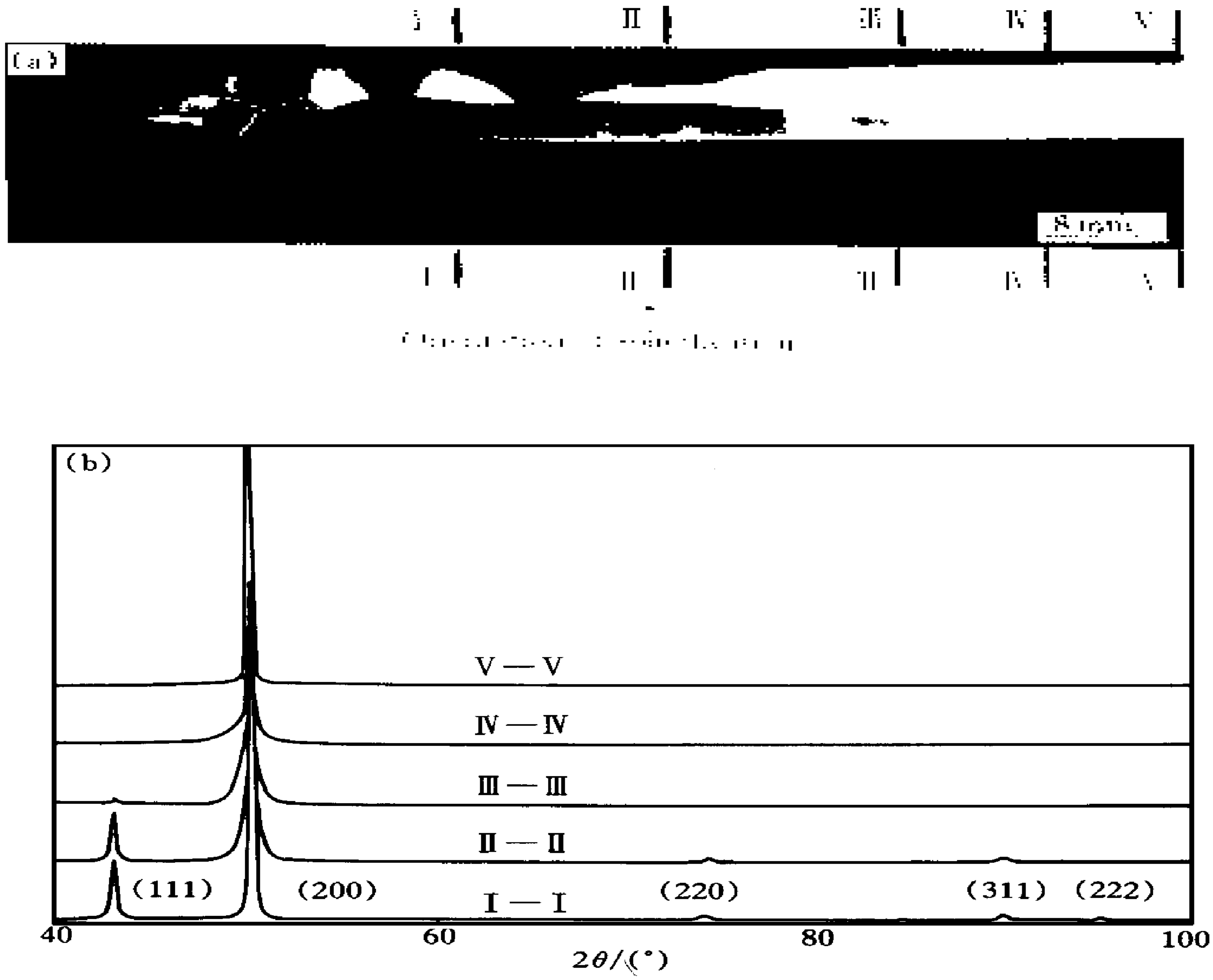

Fig. 2 Microstrucure evolution of copper rod

(a) —Longitudinal cross-section of rod;

(b) —XRD spectrum for microstructure evolution of copper rod

the static pressure head of copper is higher than that of aluminum during the continuous casting, and the density of copper($8.9\,g/cm^3$) is 3 times more than that of aluminum ($2.699\,g/cm^3$). Therefore, the additional pressure produced by the face tensile stress ($p_0 = 2\sigma/R$, R is the curvature radius) can not balance with the static pressure head and the weight of molten copper.

The solid/liquid interface morphology which has to do with the microstructure evolution and the internal quality are controlled by mould temperature, casting speed and cooling ability. Figs. 3(a) and(b) show the solid/liquid interface morphologies observed by means of the melt head being suddenly dropped and the melt head being not enough, respectively. It is shown that the solid/liquid interface morphology is a convex shape and it is favorable to the competitive growth of crystal. So, a single crystal rod can be easily gained. Because the central part of the rod solidifies before the surface part, the shrinkage porosity can be avoided and the gaseous and impurities can be easily moved to the liquid. For this reason, the solidified rod has no defect.

3.3 Formation of stray crystals

Fig. 4(a) shows that the stray crystals in the rod was not eliminated when the mould temperature was less than 1 356 ℃. A single crystal

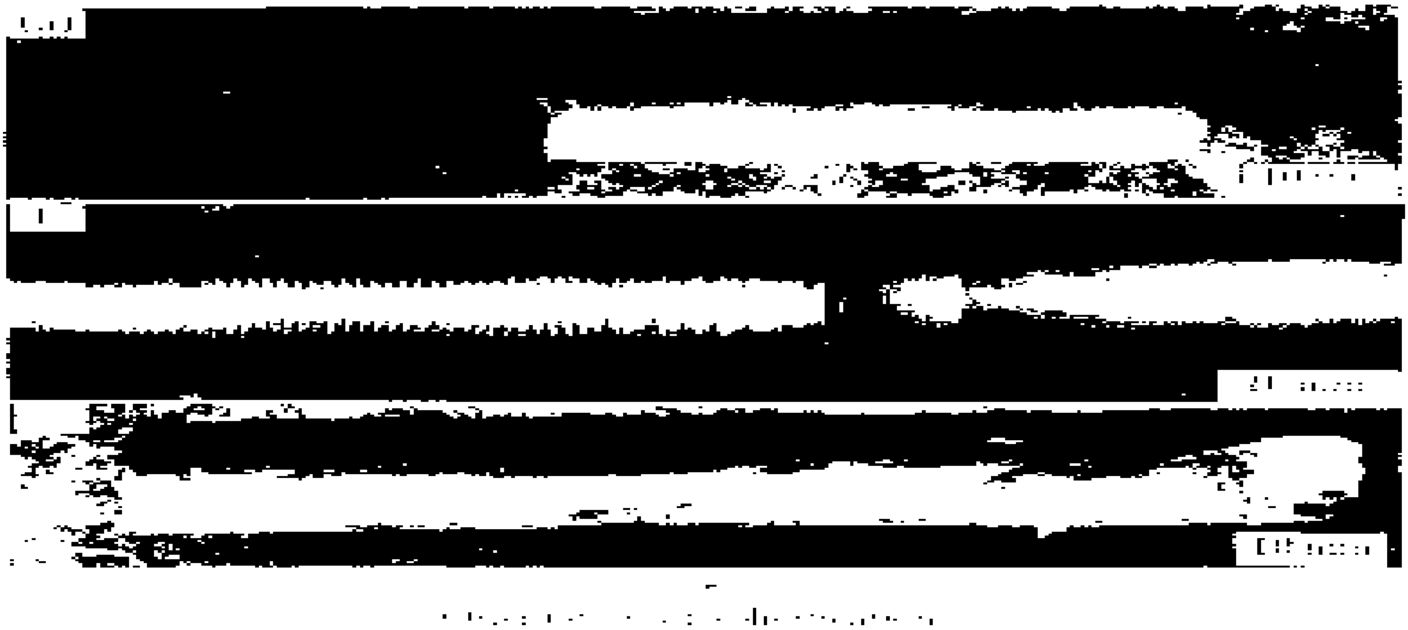

Fig. 3 Solid-liquid interface morphologies
(a) —Dropping melt head suddenly; (b) —Not enough melt head; (c) —Breaking-out

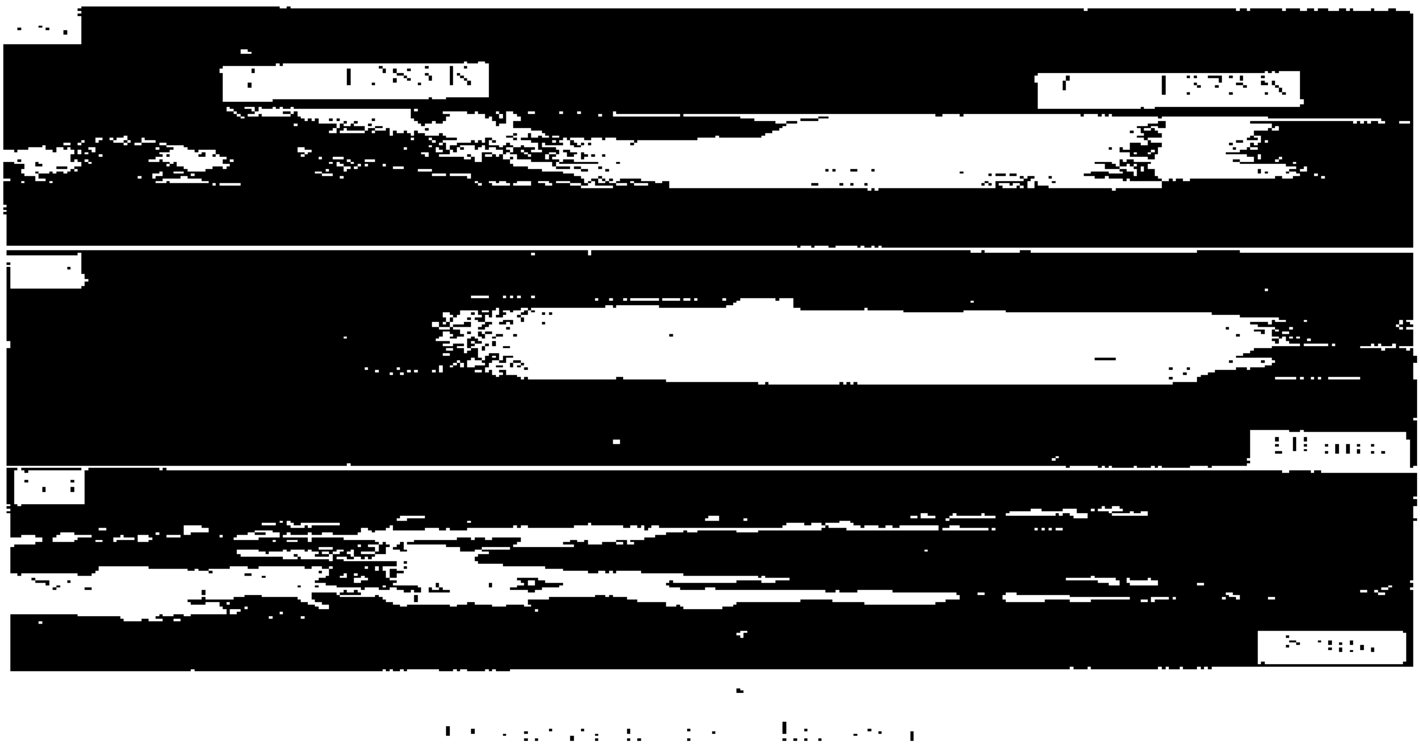

Fig. 4 Microstructures of longitudinal cross-section of rod
(a) —T_m= 1 283~ 1 373 K, R= 20 mm/min; (b) —T_m= 1 373 K, R= 30 mm/min;
(c) —T_m= 1 283 K, R= 35 mm/min

rod can be obtained only when the mould temperature is more than 1 356 K. With the increase of casting speed, the dispersed equiaxed grains and small columned grains (Fig. 4(b)) were formed. The unidirectional solidified structure was not formed (Fig. 4(c)) untill the casting speed was more than 35 mm/min.

When the mould temperature is less than 1356 K, the crystals nucleate on the wall of mould and grow. So, the stray crystals are formed in the rod interface. When the ratio between the temperature gradient of solid/liquid interface front (G_L) and casting speed (R) is gradually decreased, the crystal morphology turns from plane shape crystal to branched crystal. The increase of casting speed (R) results in decrease of G_L/R, which makes the equiaxed grains and columned grains formed on the rod surface, so that the unidirectional solidified structure are formed (Fig. 4(c)).

3.4 Surface quality of copper rod

The surface quality of the rod depends on the mould temperature, the casting speed and liquid copper pressure head when the finish of the mould surface is constant.

Fig. 5(a) shows that a single crystal rod with mirror-smooth surface can be obtained on condition that the temperature of mold was 1373 K and the casting speed was 20 mm/min. When the temperature of mold (T_m) was less than 1356 K, two types of cast surface (streaked and cracked) were identified under specific casting conditions. This is because the solid/liquid interface moves into the mould inner with the decrease of the mould temperature, which increases the friction of mould surface with the rod.

Fig. 5(c) shows that the pimple was formed on the surface of the rod when the R was suddenly increased. Because the solid/liquid interface moved to the mold exit, copper liquids had not enough time to solidify and were piled up in

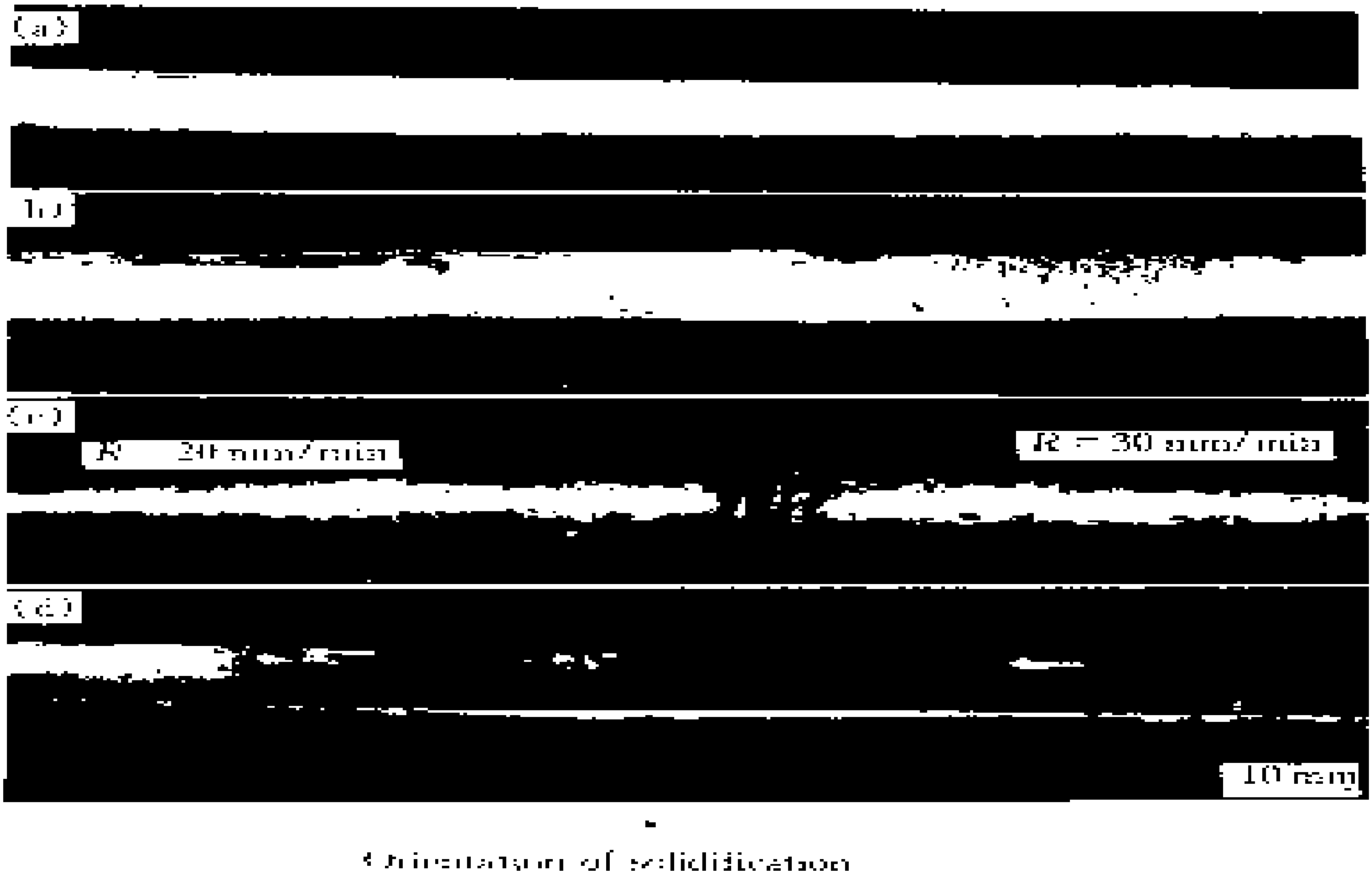

Fig. 5 Appearance morphologies of single crystal copper rod

(the melt head was not enough)

(a) — T_m = 1373 K, R = 20 mm/min; (b) — T_m = 1283 K, R = 20 mm/min; (c) — T_m = 1373 K, R = 20~30 mm/min; (d) — T_m = 1373 K, R = 20 mm/min mould exit. Fig. 5(d) shows that the ditches were formed on the surface when the melt head was not enough.

Overall, when the variable technology parameters rationally match, the single crystal copper rod with mirror-smooth surface can be obtained (Fig. 6).

Fig. 6 Rod of single crystal copper by continuous casting

3.5 Properties of single crystal copper

Table 1 shows the mechanical properties of the industrial-purity single crystal copper rods and unidirectional solidified polycrystal copper rods by continuous casting, respectively. It shows that, compared with those of the unidirectional polycrystal copper rods, the tensile strength and the yield strength of the single crystal rod decreased by 8.82% and 91.62%, respectively, while the elongation and the reduction of cross-section area increased by 24.98% and 3.09%, respectively. All these indicates that the single crystal copper by continuous casting has excellent capacity of cold working.

Table 1 Mechanical properties of single crystal copper and unidirectional crystal copper by continuous casting

Sample	σ_b/ MPa	σ_s/ MPa	δ_{10}/ %	ψ/ %
Single crystal rod	148.23	6.96	48.33	90.2
Unidirectional crystal rod	162.56	83.01*	38.67	87.3

* —value of $\sigma_{0.2}$

4 CONCLUSIONS

(1) Single crystal copper rod, 8 mm in diameter, was successfully produced using self-designed horizontal continuous casting equipment. Compared with those of unidirectionally solidified polycrystal copper rods, the tensile strength and the yield strength of the single crystal rod decreased by 8.82% and 91.62%, respectively, while the elongation and the reduction of cross-section area increased by 24.98% and 3.09%, respectively.

(2) The crystallization orientation is [100] and the crystallization face is (200). The mixed crystals (equiaxed and columnar grains) on the rod surface were formed when the mould temperature was lower than the melting temperature (T_m< 1356 K) in the front of solidification and the traction rate increased. When the casting speed (R) was more than 30 mm/min, the directional solidified rod was formed.

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