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Phase transformations in permanent-mould-cast aluminium bronze

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The phases obtained in aluminium bronze (Cu–10Al–4Fe) cast into a permanent mould were investigated. The parameters examined were the pre-heating temperature of the mould and the graphite coating thickness. The phases α and γ_2 were detected as well as the metastable phases β' and γ'. The intermetallics of the system Fe–Al were obtained in various stoichiometric compositions. The different cooling rates of the casting resulted in two mechanisms of transformation to α grains out of the unstable β phase, one being nucleation and growth producing needle-shaped α grains, the other exhibiting a massive transformation to spherical α grains. These two mechanisms determine the changes in the size of the α grains as a result of changes in the cooling rate in its various ranges.

1. Introduction

Cooling at equilibrium of the system Cu–10% Al from the liquid state results in the α and β phases. The eutectic reaction takes place at 1037° C and 8.5% Al. At 565° C and 11.8% Al the β phase decomposes to γ_2 phase by eutectoid decomposition [1].

During cooling in non-equilibrium conditions the martensitic phase β' replaces the β phase, and in addition α phase is obtained by solid-state transformation. There are phase diagrams that combine the metastable phases with the M_s temperature of the transition β–β' at about 520° C [2]. Adding 4% Fe causes only a small change in the phase diagram but contributes to the refinement of the structure [1].

High cooling rates produce the needle-like structure of the α phase. Nucleation starts at the β-phase grain boundaries and round the iron particles. Another possibility is the creation of a massive structure of α-phase, which takes place at high cooling rates without segregation during solidification and in very specific alloy structures [3]. Such a specific structure may, for example, be the alloy Cu–39% Zn or Cu–10% Al. It can be deduced from the phase diagram that with such compositions and at a temperature of about 500° C the α and β (or β') phases may exist side by side. If the β-phase is cooled sufficiently rapidly, so that the precipitation of the α-phase is suppressed, the β-phase transforms to the α-phase of the same composition. The only change is in the crystalline structure. The driving force is thermal activation, so that the massive α-phase quantity is seen to grow through a maximum as the cooling rate increases.

As yet higher cooling rates α-phase nucleation still takes place, but the mobility of the interface is insufficient for the growth of the α-phase, and no massive transformation ensues.

This paper reports on an investigation into the influence of the casting parameters on the phase transformation mechanisms as a function of the cooling rates characteristic of the process.

2. Experimental procedure

The experiments were carried out in a permanent mould made of heat resistant steel (AISI H-19 and coated with a layer of colloidal graphite, TRENEX CU PASTOS, made by the Gelger Co, Schmierstoff-Chemie GmbH (Fig. 1). The mould plate was 20 mm thick, and five Cromel–Alumel thermocouples were inserted in it at a distance of 2 mm from the casting-mould interface. Additional thermocouples were placed on the contact surface and at 6 and at 12 mm from the interface. The casting, in the form of a cylindrical disc diameter 100 mm and 15 mm thick was made of C-95200 aluminium bronze (Cu–10Al–4Fe). In a number of experiments the temperature of the casting was measured in the course of solidification, for which purpose a thermocouple had been introduced into the mould space before the cast, while its exact final location was found by cutting through the casting.

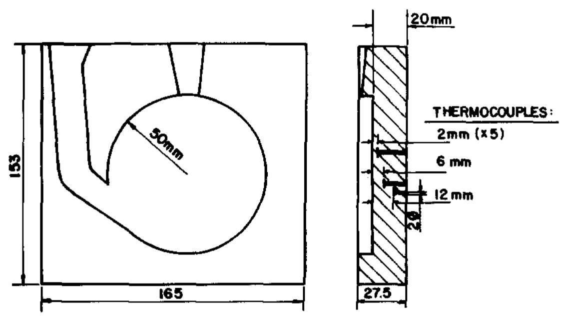

Figure 1 One half of the mould, showing the location of thermocouples.

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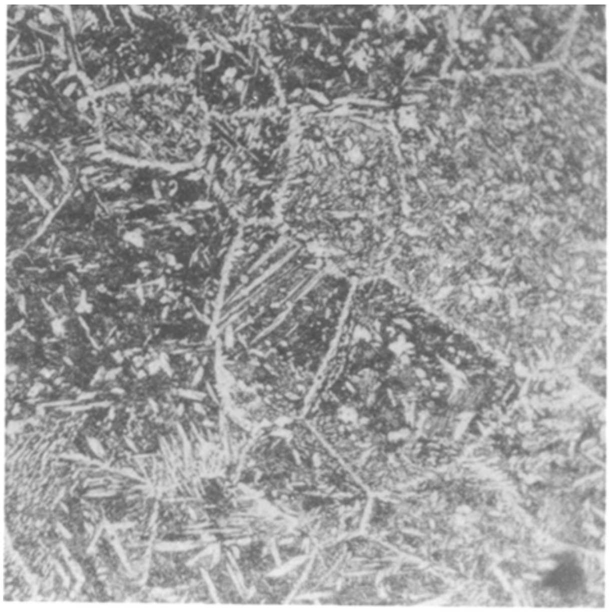

Figure 2 Aluminium bronze, water-cooled from the liquid-state. Nucleation of α phase (white) at the grain boundaries; at the grain centre-β phase. (× 460).

Figure 4 Needle-shaped α grains of a casting made in a mould pre-heated to 400° C. Graphite coating thickness 0.01 mm (× 360).

Temperatures were measured every second, the readings being fed into a computer for analysing the temperature field and calculating the heat flow to the mould.

The temperature field in the mould was calculated with the aid of the inverse solution of the heat conductance equation based on the measurement of the temperature close to the surface of the casting [4]. Knowing the temperature distribution in the mould at any time permits the calculation of the heat flow into the mould as a function of time. The source of the heat is the casting, so that it may be assumed that the heat flow into the mould equals the heat flow out of the casting. The temperature distribution in the casting is obtained by solving the one-dimensional heat conduction equation, account being taken of the emission of latent heat in the solidification range and of the dependence of the specific heat and the heat conduction on the temperature together with the boundary conditions determined before [4]. The temperature field in the casting was used for the calculation of the cooling rate in the solid-state [5].

All the metallographic specimens were taken from the centre of the casting, normal to the mould wall. The metallurgical tests and examinations comprised optical metallography, SEM, micro-analyses with the aid of EDS, X-ray diffraction and quantitative determination of the phases.

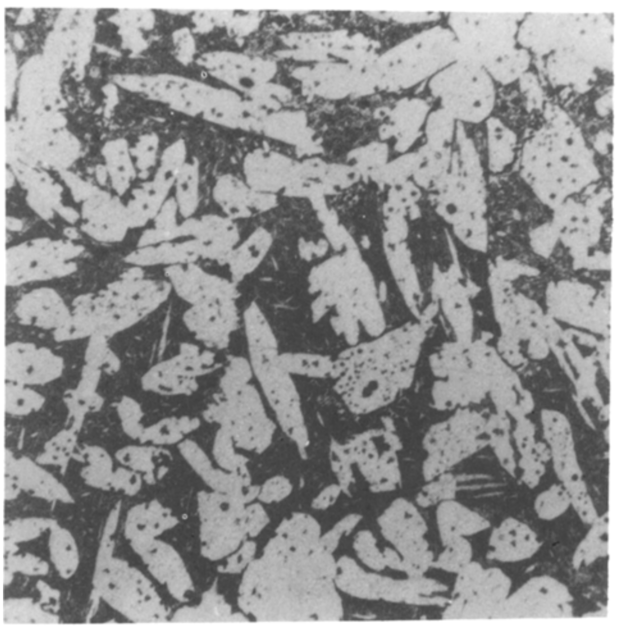

Figure 3 Needle-shaped α grains of a casting made in a mould pre-heated to 150° C. Graphite coating thickness 0.01 mm (× 360).

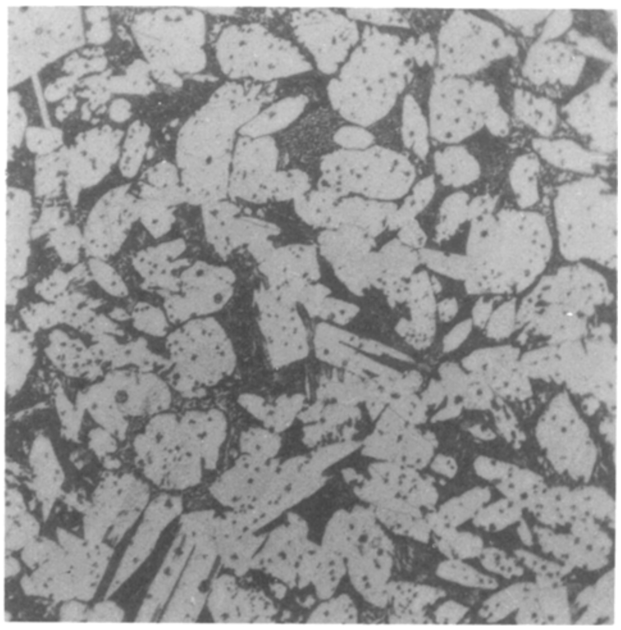

Figure 5 Highest cooling rate, spherical α grains. Mould pre-heated to 280° C. Graphite coating thickness 0.01 mm (× 360).

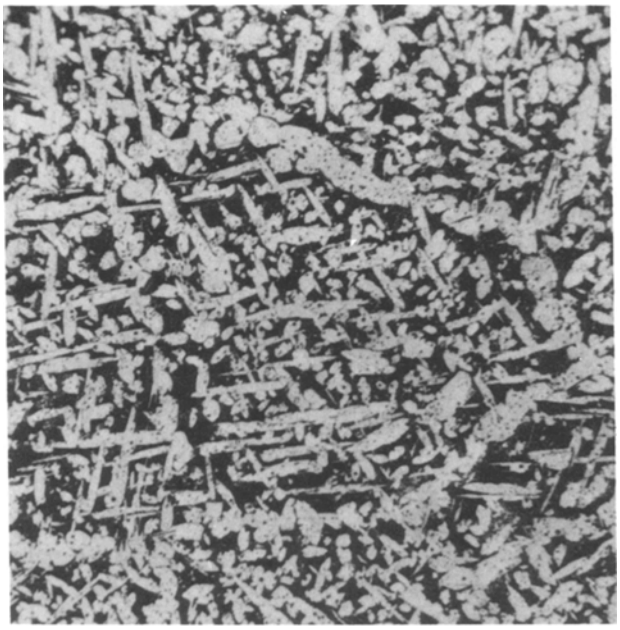

Figure 6 Needle-shaped α grains with emphasized boundaries , in a casting made in a mould coated with graphite 0.10 mm thick (× 230).

The relative amount of the α-phase (X_α) was determined by quantitative metallography, in addition the number of intersections (N_α) were counted along a line of a given length L. The grain size is thus equal to LX_α/N_α. This procedure was repeated at least five times and the mean value was used for the further analysis.

The phases of the Cu–Al system were identified with the aid of X-ray diffraction on a solid specimen and those of the Fe–Al system with the aid of a powder chemically prepared by the following procedures. Chips were dissolved in nitric acid, producing a sediment of the intermetallic phase Fe–Al. This was rinsed with water, the acid was neutralized with sodium carbonate, and the result dried, leaving the desired powder [6].

3. Results

The rates of solidification and cooling of the casting in the permanent mould are influenced by the temperature of the mould and by the thickness of the graphite coating on the mould walls [5]. It was found that these parameters also affect the microstructure of the casting made of Cu–10Al–4Fe in the as-cast state. In the course of the experiment the casting is allowed to cool in the mould for 2 min and is then exposed to air cooling. The characteristics of the microstructure and of the phases detected will be detailed below.

3.1. Metallography

From the phase diagram of the Cu–Al alloy it is seen [1] that at 10% Al a solid β-phase is formed first and that at about 930° C the α-phase begins to precipitate from the solid β-phase. The growth of the α-phase is thus dependent on the rate of heat extraction in the solid state.

Water quenching the alloy from the liquid state demonstrates the nucleation of the α-phase at the β-phase grain boundaries and round the iron particles and its inability to grow due to the rapid cooling (Fig. 2). A drop in the cooling rate in permanent-mould casting enables the α-phase to be obtained in two different morphologies.

Casting into a cold (about 150° C) or a hot (about 400° C) mould coated with graphite 0.01 mm thick led to the formation of mainly needle-shaped α grains (Figs 3 and 4). Casting into a mould of a median temperature (about 280° C) with the maximum cooling rate [5] resulted in the formation of spherical α particles (Fig. 5).

Casting conditions including a mould coated with a thick (0.10 mm) graphite coating led to the formation of exclusively needle-shaped α grains attended by emphasized β grain boundaries, especially in the zone close to the casting-mould interface (Fig. 6). With the graphite thickness stated above the needle-shaped α particles were obtained at any pre-heating

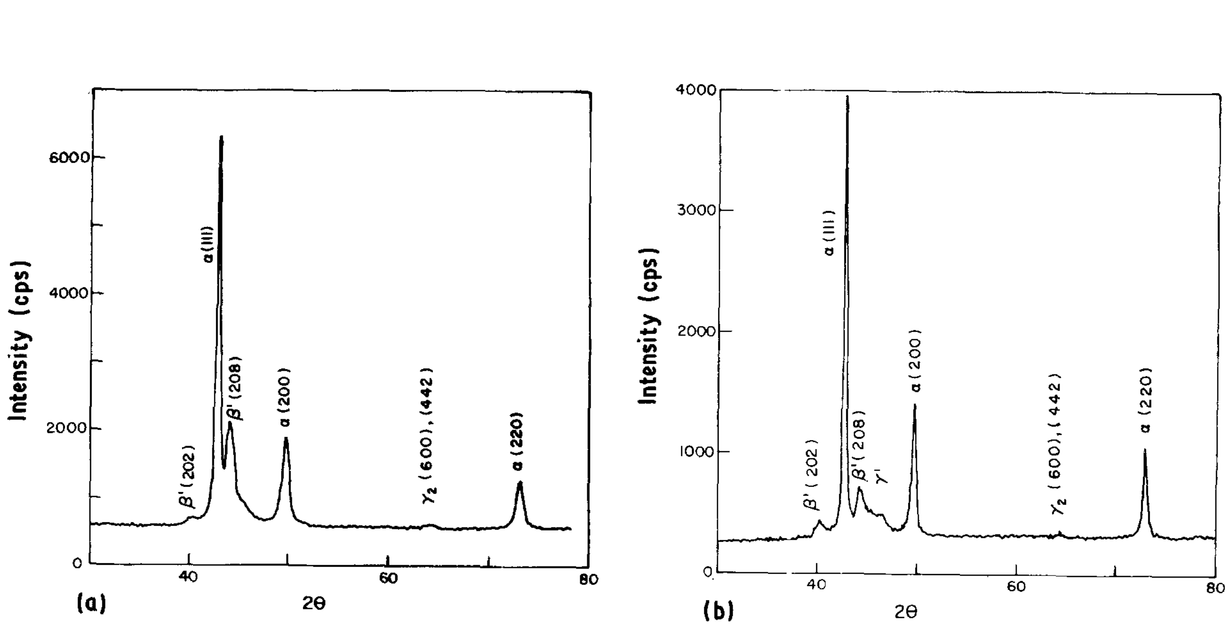

Figure 7 X-ray diffraction of aluminium bronze. (a) No formation of γ′ when casting into cold mould. (b) Segregation of γ′ when casting into hot mould followed by slow rate solidification.

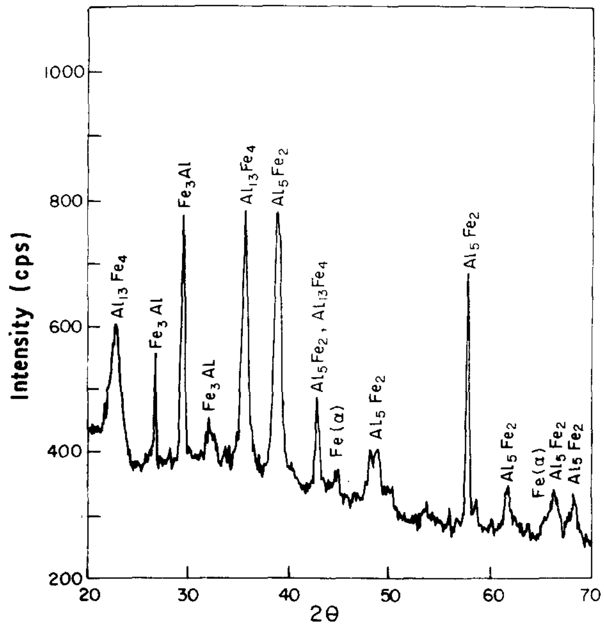

Figure 8 X-ray diffraction of the system Fe–Al after dissolution of the matrix and extraction of the powder.

temperature of the mould. It can be seen that the thick graphite coating – the dominant factor in determining a high cooling rate of the casting [5] – is also reflected in the microstructure.

3.2. Identifying the phases

The different phases were identified with the aid of X-ray diffraction (copper radiation). The principal phases of the Cu–Al system are the equilibrium α phase and small amounts of γ_2 and metastable β' phases. The metastable γ' phase appeared as a function of the cooling rates which created conditions of microsegregation during the solidification of the casting. A hot (about 400° C) mould which slows down the rate of solidification, causes the γ' phase to appear (Fig. 7a), whereas a colder (225° C) mould speeds up the rate of solidification, and the γ' phase does not form (Fig. 7b).

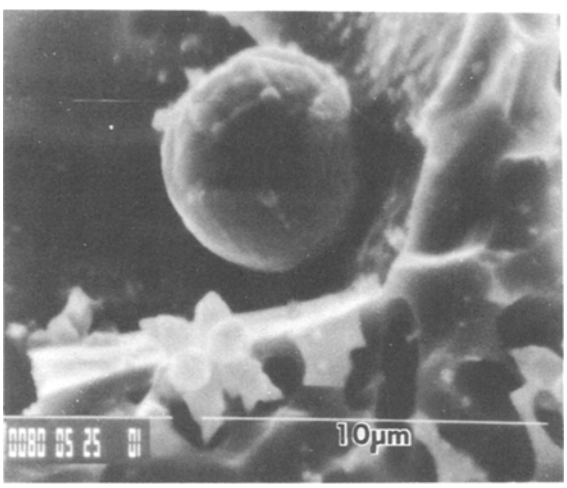

Figure 9 SEM image of a spherical iron particle (top) and a Fe_3Al precipitate in a grape-like structure (bottom).

The presence of iron in the alloy leads to the formation of intermetallics of the system Fe–Al. Owing to their low concentration (4% wt) no X-ray diffraction can be obtained when they are in the matrix, and they are identified after the dissolution of the matrix and the preparation of the powder. The results of the diffraction show that the precipitates are $Al_{13}Fe_4$, Fe_3Al, Al_5Fe_2, and Fe(α) (Fig. 8).

The iron particles that did not interact with the aluminium are spherical in shape, while the Fe_3Al precipitates are arranged in grape-like structures (Fig. 9). The precipitates in the SEM pictures were identified by determining their composition with the aid of EDS.

4. Discussion

The cooling rates ranged on an average between 238 and 404° C min^{-1} in different casting conditions, and the phases α, γ_2, β', and γ' were obtained. A connection was found between the cooling rate of the casting and the α grain size and shape.

Comparing Fig. 5 (high cooling rate) with Figs 3 and 4 (relatively low cooling rates) shows that, in Fig. 5, the majority of the α grains are spherical, whereas in Figs 3 and 4 the grains are needle-shaped. Needle-shaped α grains are formed, as a rule, by a nucleation-and-growth mechanism, while the spherical grains are created by one of massive transformation [3]. In addition, Fig. 10 presents the α grain size and the cooling rates as functions of the pre-heating temperature of the mould.

When casting into an uncoated mould or one with a coating up to 0.01 mm thick (Fig. 10a), the α grain grows as the cooling rate rises. After the cooling rate and the α grain have reached a maximum and the cooling rate begins to drop, the α grain shrinks to a minimum (Zone I, Fig. 10a). From this point onward the cooling rate continues to drop, but the α grain grows again (Zone II, Fig. 10a).

It follows from this that the growth of the α phase in Zone I does not obey the nucleation-and-growth mechanism but rather that of massive transformation, characterized by large and spherical α grains, as can be seen in Fig. 5.

In Zone II solidification is slower because of the hot mould, and resulted in segregation of (γ' phase-Fig. 7). This in turn, caused the formation of α phase by nucleation-and-growth mechanism and not by the massive transformation mechanism. Therefore, a drop in the cooling rate of the solid causes an increase in the α grain size and the α phase has the needle-like shape as well (Fig. 4). This process has also been observed in the Cu–39 Zn alloy [3].

At even higher cooling rates, as encountered in moulds with a graphite coating 0.10 mm thick, no massive transformation takes place, the α grains are needle-shaped, and with rising cooling rate the α grain shrinks, as is seen in Fig. 10b. The reason for the absence of massive transformation (in Figs 2 and 6) at such high cooling rates is that the alloy rapidly cools to such low temperature that the mobility of the interface α–β is insufficient to permit the growth of α into the unstable β.

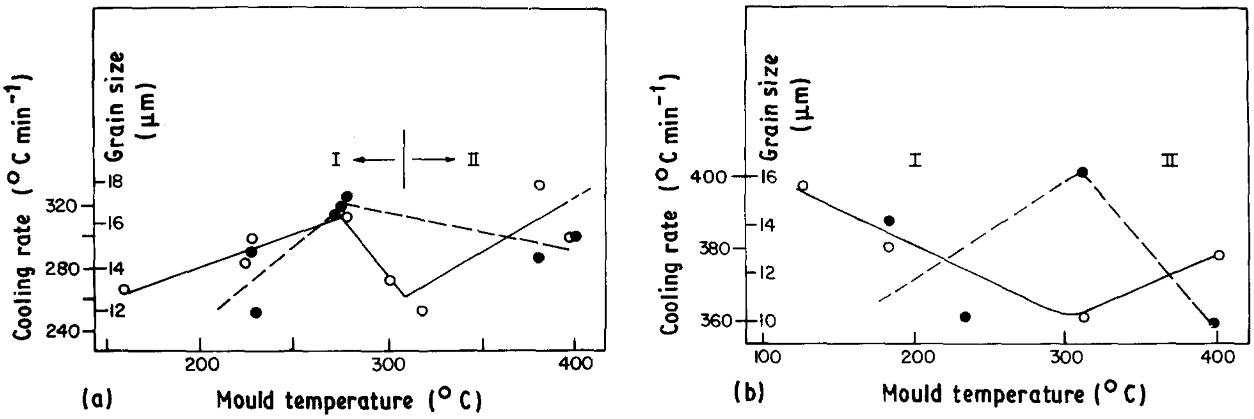

Figure 10 The interdependence of the cooling rate (●) and α grain size (○) as a function of the pre-heating temperature of the mould. (a) Graphite coating thickness – 0.01 mm; (b) Graphite coating thickness – 0.10 mm.

5. Conclusions

The conclusions are as follows.

(1) The phases obtained by casting aluminium bronze into a permanent mould are α and γ_2 and the metastable phases β' and γ'. The phases γ_2 and γ' are created locally because of microsegregation in the casting.

(2) At high or low cooling rates the α phase precipitates by a process of nucleation and growth to needle-like grains. At median cooling rates phase α is obtained by a process of massive transformation, the grains being spherical in shape and of large sizes. The size increases with rising cooling rates. Segregation of the γ' phase impedes the massive transformation, so that the nucleation-and-growth mechanism prevails in the creation of the α phase although, in the absence of the γ' phase, massive transformation would occur.

(3) Coating the mould with a thick layer of graphite increases the cooling rate and leads to a needle-like structure that emphasizes the grain boundaries of the β phase. In these conditions there is not massive transformation.

(4) The iron and aluminium intermetallics formed are of the compositions, $Al_{13}Fe_4$, Fe_3Al, and Al_5Fe_2. In addition there are spherical α iron particles that did not interact with the aluminium.

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