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ORIGINAL ARTICLE

Effects of Solidification Conditions on the Crystal Selection Behavior of an Al Base Alloy During Directional Solidification

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Abstract Al base alloy can be used as model alloy of Ni base single crystal superalloy due to their similarity on microstructure, while its lower melt temperature can match the restricted temperature of furnace working in space. The crystal selection behavior Al base alloy during directional solidification is studied by Bridgman process. With rise of heating temperature and decrease of withdraw rate, the number of grains passed spiral selector reduces. At heating temperature 900 °C and withdraw rate 2mm/min, an Al base single crystal alloy can be produced. At higher heating temperature more Mg segregates to dendrite stem, which cause smaller liquid volume fraction. At lower withdraw rate less Cu segregate to interdendrite region, which cause reduced constitutional undercooling. These two factors lead to the shrinkage of secondary dendrite arm, thus the efficiency of spiral selector is improved.

Keywords Space materials science · Single crystal superalloy · Solidification defect · Dendrite · Crystal selection

Introduction

Bridgman process is an important method to produce single crystal materials, especially suitable to metallic materials (Elliott and Pollock 2007). Single crystal Al base alloy may be prepared by this method, the acquired single crystal can be used as the seed in continuous casting of Al single crystal line material to control its orientation (Zhou et al. 2006). It can also be used as a model alloy to study the solidification process under microgravity condition due to its relative low melt temperature. For instance, it can be used as model alloy of Ni base single crystal superalloy due to their similarity on microstructure, while its lower melt temperature can match the restricted temperature of furnace working in space. However, the crystal selection behavior Al base alloy during directional solidification is seldom studied despite of these latent applications. In present work, the crystal selection behavior of an Al-Zn-Cu-Mg alloy during Bridgman process is studied, the influence of solidification and its mechanism is also discussed.

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Experimental Procedure and Material

An alloy with nominal composition (wt %) Zn:9 %; Cu:2.5 %;Mg:2.82 %; Al:Bal is used to study the directional solidification behavior of Al base alloy and the influence of solidification parameters. The master alloy is melted by induction heating, then poured into an mould which is placed on a chill, the upper part of mould is surrounded by a cylinder shaped graphite heater, thereby a steady temperature gradient is setup, which can be modified by changing the heating temperature. When the mould is withdrawn from the heater with certain rate, the alloy melt in mould is solidified gradually along unique direction. A spiral channel is used to select single crystal alloy by shielding the growth of other grains with continuous change of direction. The chosen heating temperatures are 750 °C, 800 °C, 850 °C, 900 °C, while the withdraw rates are 2mm/min,6mm/min,10 mm/min respectively. The samples for optical microscope observation are cut from the solidified alloy bars (16mm in diameter) perpendicular to the axis by spark cutter. The samples are mechanically polished, then etched in a solution of 5 % HF in 95 % H_2O. Observation is carried out with a Leica optical microscope. The samples for EBSD(electron back scatter diffraction) testing is also cut perpendicular to the axis of alloys bars, which are mechanically ground, then electrochemically polished in a solution of 10 % $HClO_4$ in 90 % ethanol with 12V at room temperature for about 30s. EBSD testing in area scan mode with step 10μm were performed in Hitachi SEM equipped with Oxford EBSP camera. The orientation images are produced using HKL Channel 5 analysis package. Electron probe microanalysis (EPMA) (EPMA1610) is carried out to determine the composition at dendrite and corresponding interdendrite area of samples with different solidification conditions, the segregation of each element is then calculated according to the EPMA results.

Results and Discussion

The microstructures in transverse section of samples solidified under various conditions are shown in Fig. 1. It can be seen the alloy is solidified in dendrite morphologies at all experimental conditions, the dendrite become thinner with increase of withdraw rate at same heating temperature, moreover the dendrite shows irregular morphologies at lower temperature, while the amount of crossing-like dendrite increase with higher heating temperature. Especially, single crystal can be formed at 900 °C and 2mm/min, which exhibits dendrite microstructure similar to typical Ni base single crystal superalloys.

The microstructures in longitudinal section of samples under part solidification conditions are shown in Fig. 2. At 750 °C and 10mm/min, even no directional solidification features are observed, the grains are almost equiaxial. While

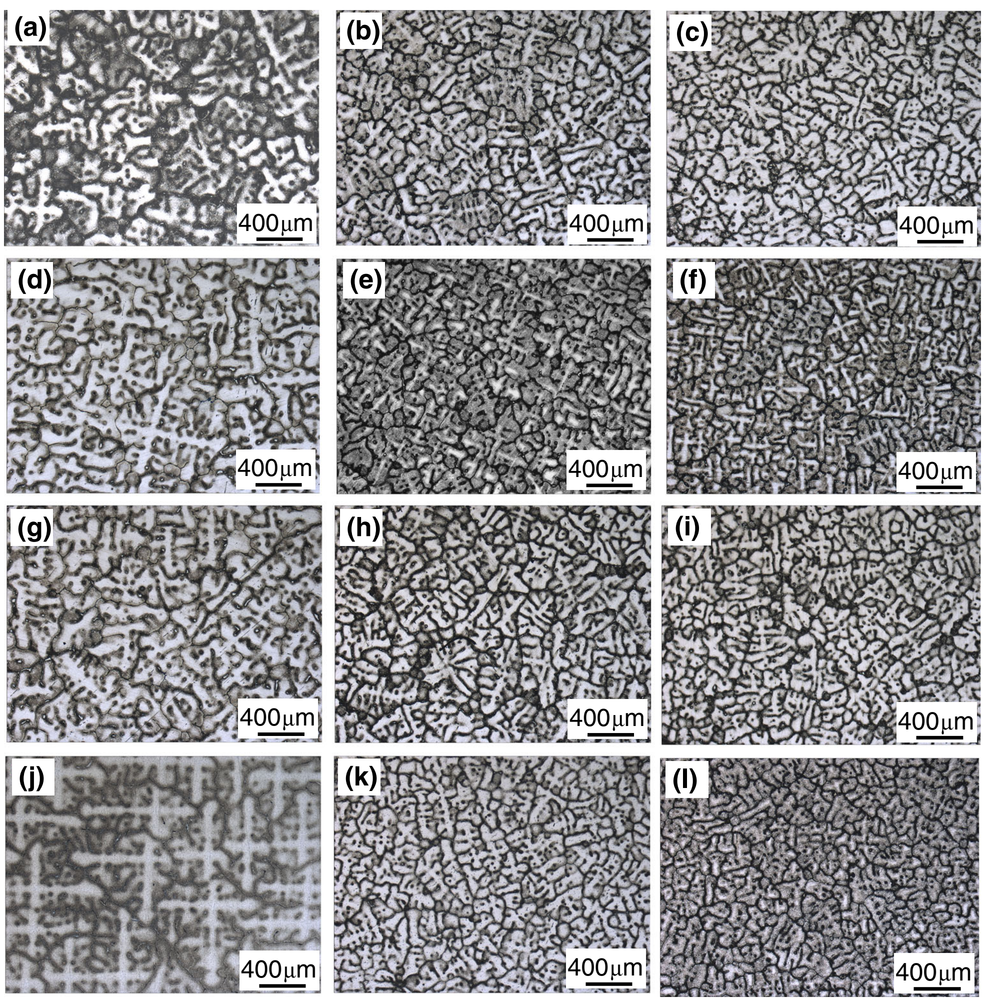

Fig. 1 microstructures in transverse section of samples solidified under various conditions (**a**)(**b**)(**c**) withdraw rate 2,6,10mm/min at 750 °C, (**d**)(**e**)(**f**) withdraw rate 2,6,10mm/min at 800 °C, (**g**)(**h**)(**i**) withdraw rate 2,6,10mm/min at 850 °C, (**j**)(**k**)(**l**) withdraw rate 2,6,10mm/min at 900 °C

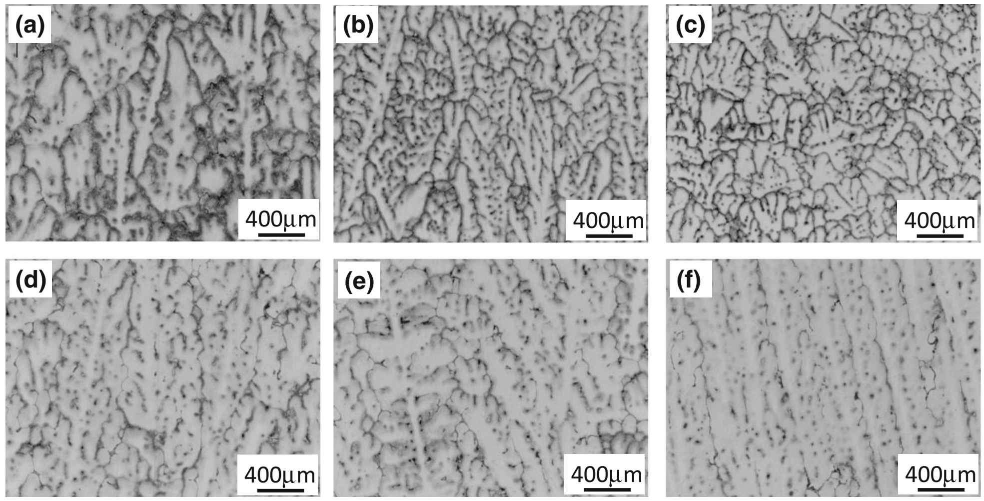

Fig. 2 microstructures in longitudinal section of samples of part solidification conditions (**a**) 750°/2mm/min, (**b**) 750°/6mm/min, (**c**) 750°/10mm/min, (**d**) 800°/2mm/min, (**e**) 850°/2mm/min, (**f**) 900°/2mm/min

the grains exhibit evident directional dendrite along the thermal gradient. At 2mm/min, the feature of directional growth become stronger with increase of heating temperatures, especially, parallel dendrite arm are observed in sample solidified at 900 °C, which demonstrates single crystal is formed. The results are agreed to that of transverse sections. Another remarkable feature is that the secondary dendrite arms are not perpendicular to dendrite stems in many grains, which is different to the case of Ni base superalloys.

The inverse pole figure of sample at part conditions are shown in Fig. 3, it can be seen the pole points appear as some sets, which correspond some grains in the visual area. Furthermore, the sets of pole points distribute uniformly over the inverse pole figure at low heating temperature

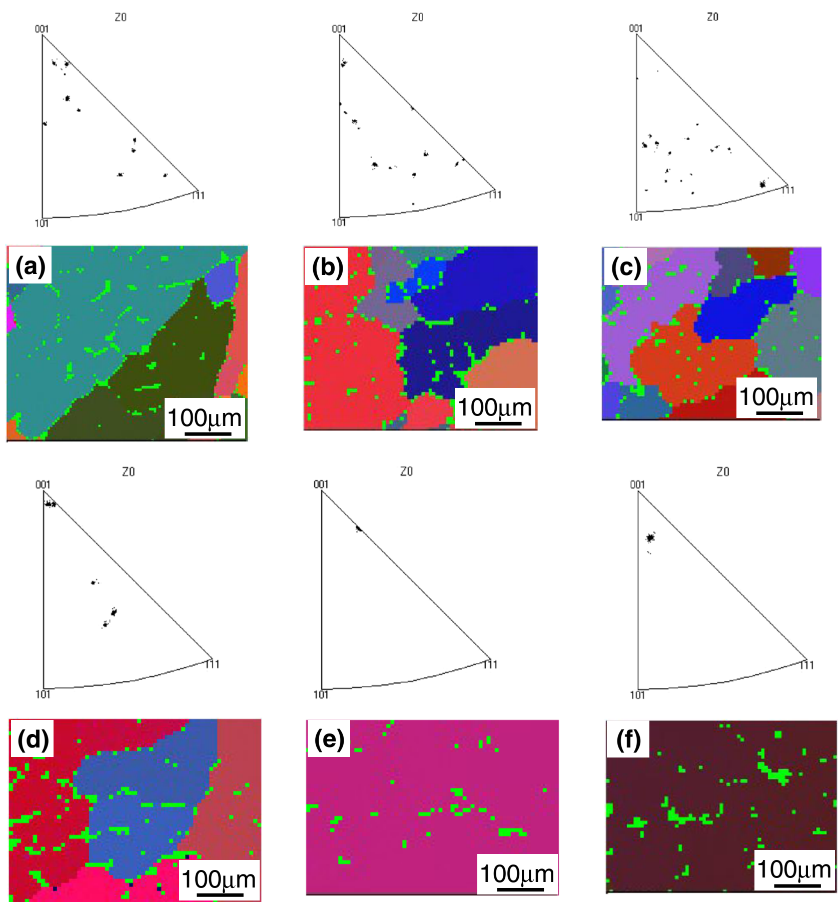

Fig. 3 The inverse pole figures and corresponding orientation maps of part solidification conditions (**a**) 750 °/2mm/min,(**b**) 750°/6mm/min, (**c**) 750°/10mm/min, (**d**) 800 °/2mm/min, (**e**) 850°/2mm/min, (**f**) 900°/2mm/min

(750 °C). In addition, the number of sets of pole points decreases with higher heating temperature, which corresponds to larger grain sizes. It can also be noted that the pole points become closer to the [001] point. Finally, only one grain appears in the visual area at 900 °C and 2mm/min.

Figure 4 shows the segregation ratio of elements in samples solidified under various experimental conditions. From Fig. 4a, it can be seen that the segregation ratios of all the three alloying elements increase with withdraw rate at heating temperature 750 °C. Further, segregation ratios of Cu changes more drastically, while segregation ratios of Mg increase slightly with high withdraw rate. From Fig. 4b, it can be seen, the higher the heating temperature become, the stronger Zn and Cu segregate to interdendrite area, while the stronger Mg segregate to dendrite.

According to microstructure observation, it can be seen that secondary dendrite arms of Al alloy are more developed than that in Ni base superalloys. Because the heat transfer rate of Al is much higher than that of Ni, so it facilitate the transport of latent heat and then the growth of secondary dendrite branch. If the secondary dendrite is well developed, the grains with deviated dendrite stem can get superiority in growth of secondary dendrite arm compared to those grains with parallel dendrite stem (D'Souza et al. 2002; Zhou et al. 2008). Moreover, the secondary dendrite arms are not perpendicular to dendrite stems, as enhances the co-growth of grains with different orientations. So the [001] oriented grains cannot shield the deviated grains by high order dendrite branches in starter block, then grains with various orientations with respect to thermal gradient can enter the spiral selector on the top of starter block. Moreover, the secondary dendrite arm belonging to different grains can intersect each other, so the spiral cannot shield the grains by gradual direction change. It is another case under low heating temperature and high withdraw rate, e.g. 750 °C and 10mm/min, the condition to directional solidification is not satisfied, therefore, nucleus of grains with random orientations are formed continuously accompanying withdraw of mould. Thus many grains with deviated [001] orientation can appear in the cast bar. This analysis can well explain the EBSD results, which show that the grains with [001] orientation paralleling to the thermal gradient do not prevail in the samples at low heating temperature. Furthermore, the higher the withdraw rate is, the pole points deviate from [001] more severely. Because elements with relative higher melting point, especially Cu, segregates to interdendrite region more intensively, so the secondary dendrite arm can acquire great constitutional undercooling, then grains with large deviation from preferential [001] orientation can pass the spiral selector.

When the heating temperature increases at low withdraw rate, the grains with preferential [001] prevail in the alloy bar gradually, also single crystal can be selected by spiral finally. This result can be attributed to two main causes. At first, steeper thermal gradient can be setup under higher heating temperature, so the mushy zone becomes short. Perhaps the anisotropy of surface energy of solid phase is enhanced due to change of segregation, the caused perpendicular relation between secondary dendrite arm and stem promotes the competition between grains. Then the grains deviated from preferential [001] direction are restrained rapidly by grains with preferential [001] orientation (Epishin and Nolze 2006; Zhao et al. 2011). So only grains with preferential [001] orientation paralleling to thermal gradient can enter spiral selector. The second, because Mg enrich in dendrite stem gradually, the volume fraction of liquid in mushy zone decreases, so the secondary dendrite arms shrink considerably, combined with the perpendicular relation between dendrite arms, then the efficiency of spiral in shielding grains is enhanced resultantly. Hence, only one grain can pass the spiral and a single crystal alloy bar is formed at 900 °C and 2mm/min eventually.

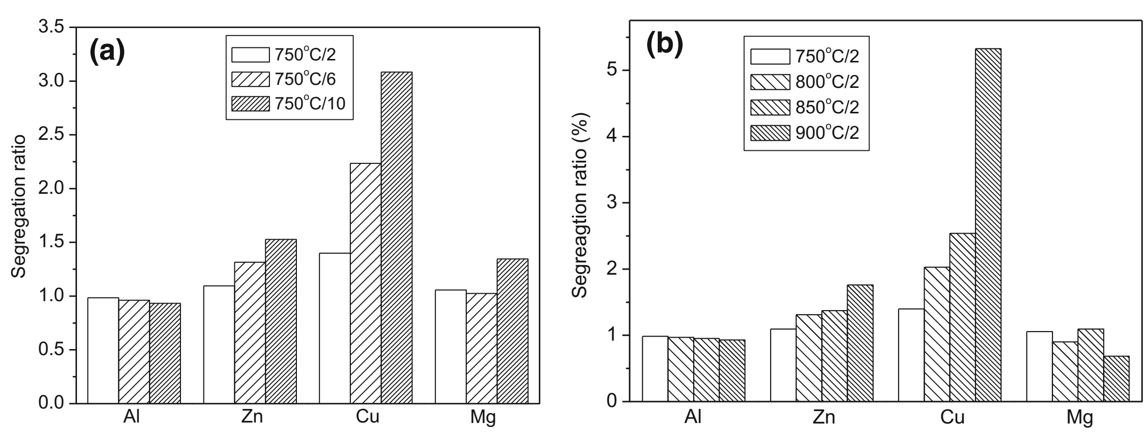

Fig. 4 Segregation ratios of elements in samples solidified under different conditions (**a**) with different withdraw rate at 750 °C, (**b**) with different heating temperature at 2mm/min

The segregation of Cu to interdendrite region can suppress the selection of single crystal due to great constitutional undercooling, while Mg can promote formation of freckle chain related to its low density, and Zn can facilitate selection of single crystal for enhancement to anisotropy of surface energy of solid phase but it is evaporated easily. Therefore Al alloys with more Mg and less Cu and less Zn are more suitable to used as a model alloy to study phenomenon related to gravity in Ni base single crystal superalloys, such as freckle chain. Under microgravity condition, the convection driven by density difference due to segregation of Cu and Mg is almost eliminated. The diffusion is the only mass transport process inside the mushy zone, so the resulting solute distribution must have significant difference with the case under normal gravity condition (Ruiz 2007; Ratke et al. 2009; Hu et al. 2014). The related dendrite morphology and solidification defect under microgravity condition need further experimental study in space combined with simulation approach.

Conclusions

(1) Within the range of experimental condition in present study, with rise of heating temperature and decrease of withdraw rate, the number of grains passed spiral selector reduces. At heating temperature 900 °C and withdraw rate 2mm/min, an Al base single crystal alloy can be produced by Bridgman method.

(2) The Al base alloy exhibits rather developed secondary dendrite arm than that of Ni base single crystal superalloys, so it's difficult to select single crystal. At higher heating temperature more Mg segregates to dendrite stem, which cause smaller liquid volume fraction. At lower withdraw rate less Cu segregate to interdendrite region, which cause reduced constitutional undercooling. These two factors lead to the shrinkage of secondary dendrite arm, thus the efficiency of spiral selector is improved.

(3) An Al base alloy can be used as model alloy of Ni base single crystal to study its solidification behavior due to similarity of microstructure and lower melting temperature.

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