Effect of growth rate on microstructure evolution in directionally solidified Ti–47Al alloy

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\textbf{A R T I C L E  I N F O}

Keywords: Directional solidification TiAl alloy Solidification path Lamellar structure

\textbf{A B S T R A C T}

The microstructures and morphologies of directionally solidified Ti–47Al alloys with different growth rates ranging from 1 to 200 \( \mu \)m/s were investigated using the Bridgman directionally solidified method. The results showed that numerous columnar grains were formed along the growth direction with the onset of directional solidification. With a variation in the growth rate, the solid/liquid interface changed from a flat to cellular and to dendritic interface. The flat-to-cellular interface transition rate of the Ti–47Al alloy varied from 1 to 3 \( \mu \)m/s. When the growth rate was higher than 10 \( \mu \)m/s, the solid/liquid interface showed typical dendritic growth. During the directional solidification process, the main phase of the directionally solidified Ti–47Al alloy was the \( \alpha \) phase, which can be attributed to the solute segregation, supercooling of the components, and contamination of the alloy melt by the \( \text{Y}_2\text{O}_3 \) ceramic shell. After reaching the steady growth state during the directional solidification process, the solidification path of the alloy was: \( L \rightarrow \alpha \rightarrow \alpha + \gamma \rightarrow (\alpha_2 + \gamma) + \gamma \). With an increase in the growth rate, the primary dendrite spacing (\( \lambda \)) and lamellar spacing (\( \lambda_s \)) of the alloy decreased gradually.

1. Introduction

Over the past few years, TiAl alloys have been extensively studied for high-temperature materials owing to their low density, high specific strength, good corrosion resistance, and excellent oxidation [1, 2, 3, 4, 5]. However, their low ambient temperature ductility restricts their large-scale applications. Directional solidification is used as a special casting technology to control the microstructure of TiAl-based alloys and produce near-net shape components. The lamellar structure of directionally solidified TiAl alloys shows a balanced combination of ductility and strength [6, 7, 8].

Four typical microstructures are observed in TiAl-based alloys. Among these, the fully lamellar microstructure shows the best comprehensive properties with excellent plasticity and strength. The fully lamellar microstructure mainly consists of TiAl (\( \gamma \)-phase) and TiAl\textsubscript{2} (\( \alpha_2 \)-phase). The relative contents of these phases (\( \gamma \)-phase) and (\( \alpha_2 \)-phase) can be controlled by adjusting the composition of the alloy [9]. In fully lamellar microstructure, the mechanical properties of TiAl-based alloys are strongly correlated to their lamellar orientation. When the lamellar orientation is parallel to the loading direction, the alloy shows the best comprehensive mechanical properties [10, 11, 12, 13, 14]. In this study, the microstructure evolution of a directionally solidified Ti–47Al (at.\%) alloy was investigated. The effect of the growth rate on the solid-liquid morphology and microstructure of the alloy was discussed. We studied the primary and solidification paths for the directional solidification of TiAl alloys to provide guidelines for controlling the microstructure of TiAl alloys during directional solidification.

2. Experimental procedure

In an argon atmosphere, commercial pure Ti (99.99\%) and Al (99.99\%) were used in a cold crucible induction furnace to produce a master bar with a nominal composition of Ti–47Al. The master batch was processed using an electric spark machine on a cylindrical rod with a diameter of 3 mm and a length of 120 mm. The directional solidification experiment was carried out in a Bridgman-type vacuum furnace. Yttrium was applied between the alumina tube and sample to prevent the TiAl melt from reacting with alumina. Before carrying out the directional solidification process, the sample was preheated to the solidification temperature. The solidification process was conducted in a Bridgman-type furnace. The growth rate was controlled by adjusting the pulling speed. The solidification path of the alloy was: \( L \rightarrow \alpha \rightarrow \alpha + \gamma \rightarrow (\alpha_2 + \gamma) + \gamma \). With an increase in the growth rate, the primary dendrite spacing (\( \lambda \)) and lamellar spacing (\( \lambda_s \)) of the alloy decreased gradually.

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https://doi.org/10.1016/j.heliyon.2021.e08704
Received 25 April 2021; Received in revised form 2 August 2021; Accepted 29 December 2021
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solidification of the alloy, the furnace cavity was evacuated to a level of $5 \times 10^{-3}$ Pa followed by the introduction of argon gas to prevent the evaporation of aluminum.

The sample was heated to 1773 K over 2 h, and after 30 min of thermal stability, it was directionally solidified at a growth rate ($V$) of 3–200 μm/s under a constant temperature gradient of 18 K/mm. When the sample grew to 40 mm, the solid-liquid interface in the liquid Ga-In-Sn alloy was quenched. In the directional solidification zone, the directional solidified bar was cut longitudinally and transversely. After polishing and conditioning with 10 mL HF solution + 10 mL HNO₃ + 180 mL H₂O, the sample was subjected to microstructure analysis using optical microscopy (OM), scanning electron microscopy (SEM), and energy-dispersive X-ray spectroscopy (EDS). The distance between the primary dendrite arms and the interlayer distance were measured from the SEM electron backscatter images according to the method described in Ref. [15].

3. Results and discussion

3.1. Macrostructure of the directionally solidified Ti–47Al alloy

The macrostructures of the directionally solidified Ti–47Al binary alloys with different growth rates are shown in Figure 1. The macrostructure of the directionally solidified TiAl alloys could be divided into four regions: the as-cast, directional solidification, mushy zone, and quenching zones [16, 17]. At the bottom of the macrostructure, the sample did not melt and remained in the as-cast state during the heating process as it was in liquid Ga-In-Sn. The sample started melting when it was heated to the preset temperature. This temperature was held for 30 min so that the composition at the front edge of the interface was substantially formed. After the onset of the directional solidification, the grains nucleated and grew rapidly at the front edge of the interface of the mushy zone. The columnar crystals grew along a direction parallel to the longitudinal heat flow direction. As shown in Figure 1, columnar grains were formed along the growth direction at the bottom of the macrostructure. As the directional solidification continued, the grains began to compete for growth. In the steady growth stage of the directional solidification process, the macrostructure basically became single columnar as the grains replaced each other and occupied the entire region.

The lines in Figure 1 indicate the boundaries of the macroscopic grains of the directionally solidified Ti–47Al alloy. It can be observed that the orientation effect of the alloy was better at the relatively low growth rate of 1–50 μm/s. The directionally solidified grains could not reach a stable stage until after a period of competitive growth. During the competitive growth process, the growth of some grains was inhibited. It can be observed from Figure 1 that as the growth distance increased, the number of grains decreased gradually until they agglomerated into a single grain. At the same time, with an increase in the growth rate, the degree of supercooling increased, the number of initial nucleation sites increased, and the competitive growth distance also increased.

On the other hand, with an increase in the growth rate, the segregation of the solute elements in the directionally solidified Ti–47Al alloy increased. When the growth rate was higher than 10 μm/s, a white segregation zone appeared in the macrostructure. As the growth rate increased, the white segregation zone gradually increased. The segregation was caused mainly because the growth rate was too high and the time for solidification was less. It was too late to achieve a complete solid phase transition and have sufficient time for solute diffusion. Finally, the macrostructure transformed into a segregation zone at room temperature.

The morphologies of the quenched solid/liquid interfaces of the directionally solidified Ti–47Al alloys with different growth rates are shown in Figure 2. With a variation in the growth rate, the solid/liquid interface changed from planar to cellular and to dendritic. When the growth rate was 1 μm/s, the interface was a typical plane interface, as shown in Figure 2(a). When the growth rate was increased to 3 μm/s, the solid/liquid interface became unstable and showed cellular morphology, as shown in Figure 2(b). At the growth rate of 5 μm/s, the interface still had cellular morphology, but the cells became smaller, as shown in Figure 2(c). When the growth rate reached 10 μm/s, the cells elongated and a secondary arm appeared at their front ends, indicating the occurrence of an obvious cellular/dendritic growth. When the growth rate was increased beyond 10 μm/s, the solid/liquid interface transformed into a typical dendritic interface. Therefore, it can be concluded that the growth rate required for the planar-to-cellular transformation of the solid/liquid interface of the directionally solidified Ti–47Al alloy was 1–3 μm/s.

According to the supercooling theory of components, the criterion for the flat interface stability of binary alloys is [18]:

$$\frac{G}{V} \geq -\frac{mC_0(1-K_o)}{D_rK_o}$$

where $G$ represents the temperature gradient, $V$ represents the growth rate, $m$ represents the slope of the liquidus, $C_0$ represents the alloy composition, and $D_r$ represents the diffusion coefficient of the solute elements in the liquid phase. From the physical parameters of the Ti–47Al alloy listed in Table 1, it can be stated that the condition for the flat interface stability of the Ti–47Al alloy is: $G/V \geq 5.63 \times 10^{5}$ K s/m². In this study, the $G$ of the directionally solidified Ti–47Al alloy was 18 K/mm. Therefore, the critical stable flat interface growth rate of the Ti–47Al alloy was calculated to be approximately 2.5 μm/s. When the growth rate was lower than 2.5 μm/s, the solid/liquid interface of the directionally solidified alloy showed a planar growth. These observations are consistent with the experimental results. It can be seen from Figure 2 that the growth rate for the planar-to-cellular interface transition of the directionally solidified Ti–47Al alloy was 1–3 μm/s.
When the growth rate was higher than 5 μm/s, the solid/liquid interface became dendritic. With an increase in the growth rate, the primary dendrite spacing decreased gradually, as shown in Figure 3. The relationship between the primary dendrite spacing ($\lambda$) and growth rate ($V$) is as follows.

$$\lambda = \frac{A}{C^{15}} \frac{V}{C^{0.334}}$$  \hspace{1cm} (2)

where $A$ represents a constant related to the material. The primary dendrite spacing and growth rate were obtained by nonlinear fitting to satisfy the following relationship.

$$\lambda = \frac{557.76}{C^{15}} \frac{V}{C^{0.334}}$$  \hspace{1cm} (3)

The exponent in Eq. (3), which defines the relationship between the primary dendrite spacing and growth rate, was 0.334, which is close to the theoretical value of 0.25 given by the Hunt [21] and Kurz-Fisher models [22]. This value is higher than that (0.24) reported by Lapin et al. for the Ti–46Al–2W–0.5Si alloy [23] and is close to that (0.31) reported by Jianglei for the Ti–49Al alloy [24]. This difference in the exponent values obtained in this study and those reported previously was mainly caused by the difference in the alloy composition and solidification conditions.

In the directional solidification process, the growth direction of the secondary dendrite arms was almost at an angle of 60° with the growth direction of the primary dendrite arms, as shown in Figure 4(a). Therefore, the main phase was the α phase according to the symmetry of the primary phase dendrites in TiAl alloys [25]. According to the binary equilibrium phase diagram of TiAl alloys, shown in Figure 5, it can be observed that the primary phase of the Ti–47Al alloy is the β phase. In this experiment, no primary β phase was observed. This is because, during the directional solidification of the Ti–47Al alloy, the solute (Al) element was continuously discharged into the solid/liquid interface,
leading to the concentration of Al in front of the solid/liquid interface. Figure 4(a) shows the dendritic morphology and EDS profile of the solid/liquid interface at the growth rate of 30 μm/s. It can be observed from the figure that some white Y₂O₃ particles existed between the dendrites. Because of the convection and thermal vibration effects, some of the Y₂O₃ particles were stuck in the alloy melt during the directional solidification. This resulted in the accumulation of Y₂O₃ particles at the quenching solid/liquid interface. Figure 4 shows the microstructure and EDS results of the directional solidification zone of the directionally solidified Ti–47Al alloy. The composition of the Ti–47Al alloy in the directional solidification zone was comparable to that of the as-cast alloy, but the Al content in the dendritic melt formed after quenching was 55.46%. According to the binary phase diagram of TiAl alloys, when the Al content is higher than 49%, the main phase is the α phase.

### 3.2. Microstructure evolution of the directionally solidified Ti–47Al alloy

The macroscopic and microscopic structures of the Ti–47Al alloys directionally solidified at different growth rates are shown in Figure 6. Figure 6(a) shows the macrostructure and microstructure of the Ti–47Al alloy solidified at the growth rate of 3 μm/s. It can be seen that in the directional solidification zone, the microstructure consisted of lamellar structures without significant segregation. The final structure was the lamellar structure, in which the orientation was parallel to the growth direction. Figure 6(b) shows the macrostructure and microstructure of the Ti–47Al alloy directionally solidified at the growth rate of 10 μm/s. As the lamellar orientation of the as-cast zone at the bottom of the sample was at an angle to the growth direction, the final lamellar orientation and...
Figure 6. Longitudinal macrostructures (a), (b), (c) and the corresponding microstructures (a₁) - (a₄), (b₁) - (b₄) and (c₁) - (c₄) for the local areas in (a), (b), and (c) of the directionally solidified Ti-47Al alloy at the growth rates of 3 μm/s, 10 μm/s and 20 μm/s, respectively.

Figure 7. Lamellar microstructures of the directional solidification zone in the directionally solidified Ti-47Al alloy at different growth rates, (a) 3 μm/s, (b) 30 μm/s, (c) 50 μm/s, (d) 100 μm/s.
growth direction were at an angle of approximately 45°. When the growth rate was increased to 20 μm/s, a white segregation zone appeared in the microstructure, as shown in Figure 6(c). The lamellar orientation of the Ti–47Al alloy in the directional solidification zone was parallel to the growth direction.

It can be observed from Figure 6(b1) and 6(c1) that the primary phase of the Ti–47Al alloy during the directional solidification was the α phase. However, the orientations of the lamellar structure formed in the final microstructure were different, as shown in Figure 6(a2), 6(b2), and 6(c2). The blue arrows in the figure represent the final lamellar orientations of the directionally solidified Ti–47Al alloy. According to the Burgers relationship, the lamellar orientation formed in the final microstructure is perpendicular to the growth direction when the primary phase is the α phase [26]. The black arrows represent the preferred orientation of the primary α phase. The results indicate that the growth orientations of the primary α phase were different at different growth rates. Both the initial directional solidification initiation interface and growth rate had a great effect on the growth orientations of the primary α phase, resulting in different orientations of the final lamellar structure. There was a single α-phase region below the solid/liquid interface. In this case, it was too late to achieve the α → α + γ → α2 + γ solid phase transition. Therefore, no lamellar structure was formed. As shown in Figure 6(a2), 6(b2), and 6(c2), the boundaries between the single α-phase region and lamellar structure, in which the solid phase transition was completed, gradually became blurred with an increase in the growth rate.

At relatively higher growth rates, there were still many white α phase segregation regions in the lamellar structure region, which were formed because of the segregation of the solute elements. During the dendritic growth, the dendrite arms were rich in Ti and the dendrites were rich in Al. After the liquid-to-solid phase transition, it was too late to achieve a complete solid phase transition because the growth rate was too high. Therefore, it resulted in the segregation of the solute elements, which had an adverse effect on the properties of the alloy. Hence, it was more appropriate to carry out the directional solidification at a relatively lower growth rate or to sufficiently diffuse the solute elements by a subsequent heat treatment to reduce or avoid the segregation. From the microstructure evolution of the alloy during the directional solidification, it can be inferred that the solidification path of the alloy under this condition was $L \rightarrow α \rightarrow α + γ \rightarrow (α2 + γ) + γ$.

The lamellar structures of the directionally solidified Ti–47Al alloy at different growth rates are shown in Figure 7. An increase in the growth rate resulted in the refinement of the lamellar structure of the directionally solidified Ti–47Al alloy. For TiAl alloys with a full lamellar structure, the mechanical properties are affected by many factors, such as the grain size, lamellar orientation, and lamellar spacing. It is believed that small lamellar spacings are conducive to improving the properties of an alloy. As shown in Figure 7, an increase in the growth rate had a significant refinement effect on the lamellar structure of the directionally solidified alloy.

Linear-regression analysis was used to analyze the relationship between the lamellar spacing ($λ$) and growth rate ($V$), as shown in Figure 8. The following relationship was obtained.

$$λ = 14.79V^{-0.55}$$

The results showed that under the experimental conditions used in this study, the growth rate was an important factor affecting the lamellar spacing. The lamellar spacing decreased with an increase in the drawing rate.

4. Conclusions

1. The hexagonal α phase was the primary phase in the Ti–47Al alloy. The solid/liquid interface of the alloy changed from planar to cellular and to dendritic with the growth rate. The growth rate for the planar-to-cellular transformation of the interface was 1–3 μm/s.
2. From the microstructure evolution of the directionally solidified Ti–47Al alloy, it can be inferred that the solidification path of the Ti–47Al alloy was $L \rightarrow α \rightarrow α + γ \rightarrow (α2 + γ) + γ$.
3. An increase in the growth rate had a significant refinement effect on the lamellar structure of the directionally solidified Ti–47Al alloy. The primary dendrite spacing gradually decreased with an increase in the growth rate ($V$) according to the relationship $λ = 557.76 \cdot V^{-0.254}$. The interlamellar spacing ($λ_s$) decreased with an increase in the growth rate ($V$) according to the following relationship $λ_s = 14.79V^{-0.55}$.

Declarations

Author contribution statement

Tong Liu: conceived and designed the experiments; performed the experiments; analyzed and interpreted the data; wrote the paper.

Jing Tao, Xiaoye Cai, Dayong Chen: conceived and designed the experiments; contributed reagents, materials, analysis tools or data.

Jiansheng Li, Liangshun Luo, Zonghui Cheng: analyzed and interpreted the data; contributed reagents, materials, analysis tools or data.

Yanqing Su: conceived and designed the experiments; analyzed and interpreted the data; wrote the paper.

Funding statement

This research was supported by National Natural Science Foundation of China (grant no. 52001001), Scientific Research Foundation of AnHui Polytechnic University (2020YQQ005) and Industrial Collaborative Innovation Foundation of AnHui Polytechnic University-Fanchang District (2021FCGYXTA2)

Data availability statement

Data included in article/supplementary material/referenced in article.

Declaration of interests statement

The authors declare no conflict of interest.


**Additional information**

No additional information is available for this paper.

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