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Effect of thermal stabilization on microstructure and mechanical property of directionally solidified Ti−46Al−0.5W−0.5Si alloy

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Abstract: Effect of thermal stabilization on the microstructure and mechanical property of directionally solidified Ti−46Al−0.5W−0.5Si (mole fraction, %) alloy was investigated. The specimens were thermal stabilized for different time (t) and directionally solidified at a constant growth rate of 30 μm/s and temperature gradient of 20 K/mm. Dependencies of the primary dendritic spacing (λ_1), secondary dendritic spacing (λ_2), interlamellar spacing (λ_L) and microhardness (HV) on holding time were determined. The values of the λ_1, λ_2 and λ_L increase with the increase of t, and the value of HV decreases with the increase of t. The increase of t is helpful to obtain a good directional solidification structure. However, it reduces the mechanical property of the directionally solidified TiAl alloy. The optimized value of t is about 30 min.

Key words: intermetallics; TiAl-based alloy; crystal growth; microstructure evolution; mechanical properties

1 Introduction

Intermetallic γ-TiAl based alloys have been known as attractive materials for high-temperature structure applications in aerospace and automotive industries. As reported in previous review [1], TiAl alloys present a good substitute for Ni-based superalloys because of their good elevated-temperature mechanical properties, high specific yield strength, good oxidation resistance and creep properties up to high temperature. LARSON et al [2] pointed out that plenty of researches had focused on refining the grain size by controlling microstructures and improving the mechanical properties by alloying with B, Si, W, Cr, Nb, V, Mn and Mo.

As reported by JOHNSON et al [3], TiAl-based alloys with aligned lamellar structures have a good combination of strength and ductility in a wide range of temperature. JOHNSON et al [4] indicated that the preferred orientation of lamellar structures can be obtained by directional solidification and seeding technique. In this respect, KIM et al [5] and LAPIN et al [6] investigated the directional solidification of TiAl-based alloys. Plentiful studies have focused on the control of lamellar orientation during directional solidification, such as LEE et al [7] for TiAl−(Mo, Si) alloys, JOHNSON et al [8] for TiAl−(Nb, Si) alloys, YAMANAKA et al [9] for TiAl−(Re, Si) alloys and YAMAGUCHI et al [10] for TiAl−(Si, Mo, B) alloys. Besides, FAN et al [11] studied the effect of solidification parameters on the microstructure and mechanical property of directionally solidified Ti−46Al−0.5W−0.5Si (mole fraction, %) alloy. FAN et al [12] also studied the dependency of microstructure and microhardness on the growth rate of directionally solidified Ti−43Al−3Si (mole fraction, %) alloy. Apart from that, LAPIN et al [6] investigated the effect of growth rate on the microstructure and mechanical properties of directionally solidified Ti−46Al−2W−0.5Si (mole fraction, %) alloy. However, the dependency of microstructure and mechanical property on solidification condition during directional solidification has rarely been studied.

As reported by BÖSENBERG et al [13] and BUCHMANN and RETTENMAYR [14], the parameters in directional solidification processing, such as thermal stabilization time, growth rate, temperature gradient and cooling rate, have an important influence on the microstructure and mechanical property. THI et al [15] and YOSHIOLKA et al [16] found that the initial

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solidification condition under which the growth starts is critical for directional solidification. After being heated to desired temperature, the specimens need to be held for a certain time to stabilize and homogenize the distributions of the temperature and composition to establish the initial solidification condition before directional solidification, e.g., solid−liquid interface morphology and solute distribution in the liquid phase. The holding time (t) of thermal stabilization is important to the microstructure and mechanical property of directionally solidified alloys. Previous works mostly focused on the preparation of the initial condition, but the effect of the initial conditions on the subsequence microstructure and mechanical property of the directionally solidified alloys has rarely been studied. For TiAl alloys, there are many values of t for thermal stabilization in different directional solidification experiments, such as 10 min used by LUO et al [17] for Ti−47Al (mole fraction, %) alloy, 15 min used by LAPIN et al [6] for Ti−46Al−2W−0.5Si (mole fraction, %) alloy, 15 min used by DING et al [18] for Ti−45Al−8Nb−(W, B, Y) (mole fraction, %) alloy and 30 min used by FAN et al [12] for Ti−43Al−3Si (mole fraction, %) alloy. However, few researches were found to focus on optimizing the value of t before directional solidification of TiAl alloys.

In the present work, the effect of the holding time (t) of thermal stabilization on the microstructure and mechanical property was experimentally investigated on directionally solidified Ti−46Al−0.5W−0.5Si (mole fraction, %) alloy. The relationships among the primary dendritic spacing (λ_1), secondary dendritic spacing (λ_2) and interlamellar spacing (λ_L), microhardness (HV) and holding time (t) during the directional solidification were also studied.

2 Experimental

Master ingot of Ti−46Al−0.5W−0.5Si (mole fraction, %) alloy was prepared in a vacuum induction melting furnace with a water cooled copper crucible by melting Ti (99.96%), Al (99.99%), W (99.98%), and Si (99.96%) of high-purity metals under an argon atmosphere. Chemical composition of the alloy was analyzed by spectral analysis, and the result is shown in Table 1. The specimens were machined to rods with 3 mm in diameter and 100 mm in length from the ingot with a spark machine. The fine diameter of sample was chosen to achieve a high temperature gradient during directional solidification. The experiments were performed in a Bridgman-type vacuum directional solidification furnace, consisting of heating coils, a water cooled liquid metal bath filled with a liquid Ga−In−Sn alloy, and an adiabatic zone which was located between the heater and the cooler. The details of the furnace have been described elsewhere [12].

Table 1 Chemical composition of Ti−46Al−0.5W−0.5Si (mole fraction, %) alloy

Al	W	Si	O	Ti	Other
45.27	0.62	0.52	0.05	Balance	<0.1

The specimen was placed into a 99.99% purity alumina crucible of 4 mm/5.5 mm in diameter (inside/outside diameter) and 150 mm in length. After 4 h heating to 1773 K, the specimens were thermal stabilized for different time (5−60 min) and pulled at a constant growth rate of 30 μm/s. After growing to about 40 mm, the specimens were dropped into the liquid Ga−In−Sn alloy to quench the solid−liquid interface.

The temperature gradient was measured by W/Re thermocouples that were placed near the outside surface of the alumina crucible. One thermocouple was placed 5 mm from the bottom of the sample where was near the solid−liquid interface. The other was placed 15 mm from the bottom of the sample where was the liquid region. The temperature gradient close to the solid−liquid interface was measured to be 20 K/mm. The temperature gradient can be changed by changing the temperature of the specimen. To keep the temperature gradient constant during directional solidification, the temperatures of the cooler and the hotter part of the furnace were kept constant by an automatic temperature controlling system.

The phase in the alloy was identified by a Rigaku D/max-RB X-ray diffractometer with monochromatic Cu K_α radiation. Both optical and scanning electron microscopy (SEM) analyses were used to characterize the microstructure of the specimens after polishing and etching by a solution of 10 mL HF+10 mL HON_3+180 mL H_2O. SEM backscattered electron imagining was used to help identify the phases.

The primary dendritic spacings were measured from the longitudinal sections of the specimens by using the linear intercept method. BÖYÜK et al [19] pointed out that measuring interlamellar spacing from transverse sections is more accurate than from longitudinal section. Thus, the interlamellar spacings were measured from the transverse sections of specimens on the BSE images.

Microhardness measurements were made on a standardized Vickers measuring test device using 0.98 N (100 g) load and a dwell time of 10 s on the surface of longitudinal sections with polished and slightly etched surfaces. The values of microhardness were average of at least 30 measurements.

3 Results

3.1 Microstructure

From the XRD diffraction pattern [20] (Fig. 1), three phases, namely, γ-TiAl, α_2-Ti_3Al and ξ-Ti_5Si_3 phases, can be identified for directionally solidified specimens. The typical microstructures of the directionally solidified specimens are shown in Fig. 2. According to the energy dispersive X-ray analysis (EDX) results (Fig. 3), the black phase is γ phase, the gray one is α_2 phase and the white one is ξ phase. In addition, some dark particles are found in the alloy after directional solidification (Fig. 2(b)). The chemical composition of the dark phase was analyzed by EDX, as shown in Table 2. The results indicate that the dark particles are Al_2O_3 phase. The Al_2O_3 particles were formed by the reaction between the crucible and the melting alloy during solidification.

In the previous study [21], the type of primary phase in TiAl alloys can be identified by the symmetry of

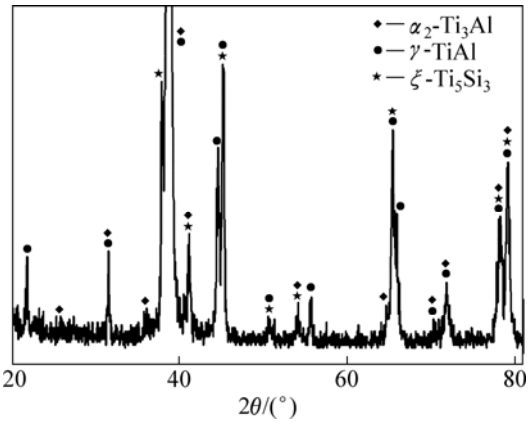

Fig. 1 XRD diffraction pattern of directionally solidified Ti−46Al−0.5W−0.5Si alloy [20]

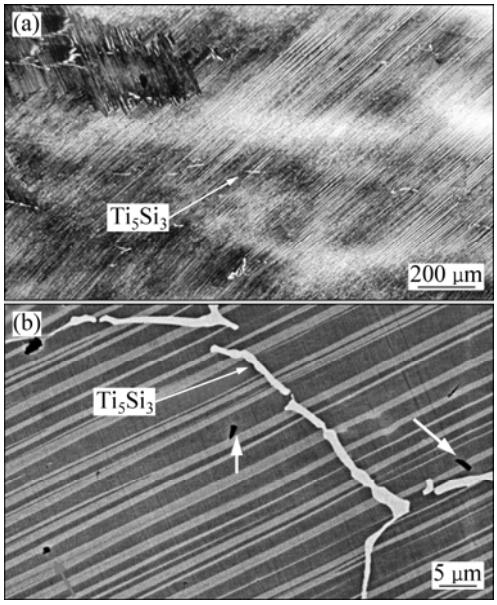

Fig. 2 Microstructures of directionally solidified Ti−46Al−0.5W−0.45Si alloy: (a) Optical image; (b) BSE image (White arrows indicate Al_2O_3 particles)

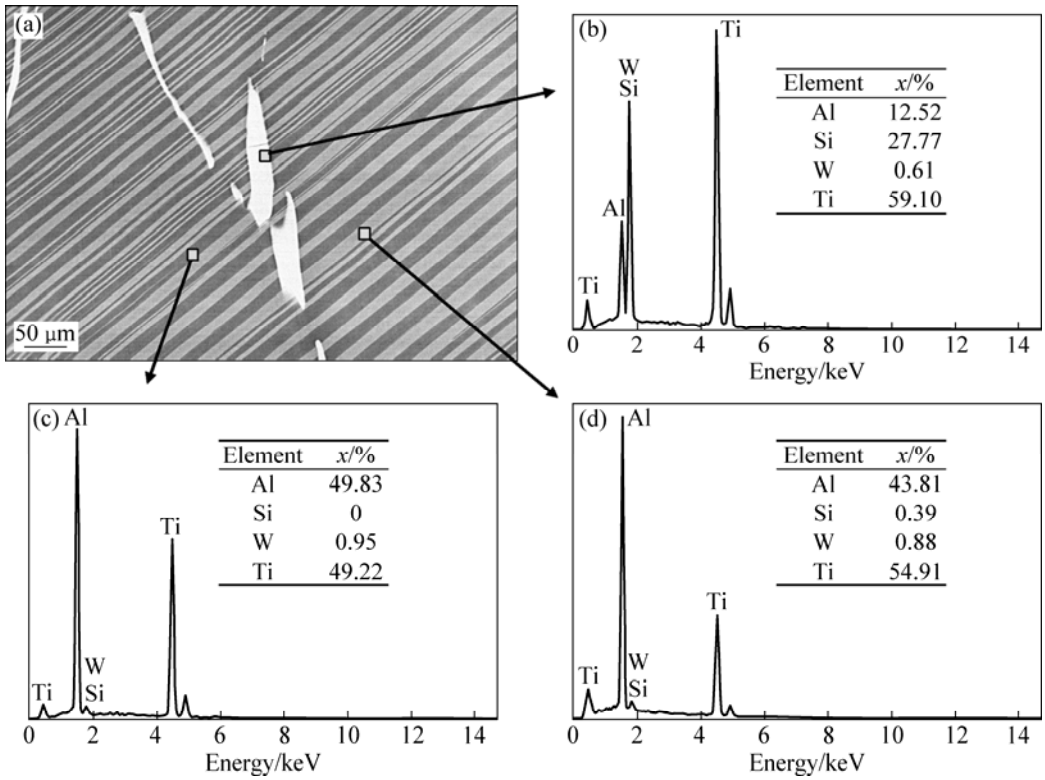

Fig. 3 SEM image (a) and EDX results (b, c, d) of Ti−46Al−0.5W−0.5Si alloy (Black phases are γ-TiAl phases, grey phases are α_2-Ti_3Al phases and white phases are ξ-Ti_5Si_3 phase)

dendritic shape, i.e., the angle between the secondary dendritic arms and the primary dendritic spines. According to the morphologies of the dendrites in transverse and longitudinal section, the dendrite presents a shape with six-fold symmetry (Fig. 4). Therefore, the α phase with hexagonal crystal structure is the primary phase of Ti−46Al−0.5W−0.5Si (mole fraction, %) alloy.

Table 2 Chemical composition of Al_2O_3 particle in Ti−46Al−0.5W−0.5Si alloy after directional solidification (mole fraction, %)

Ti	Al	W	Si	O
0.88	48.28	0.04	0.02	50.78

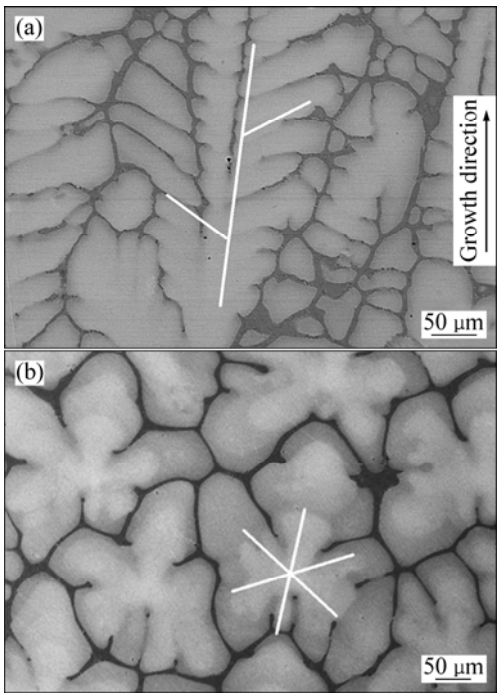

Fig. 4 BSE micrographs showing morphology of solid−liquid interface of directionally solidified sample (a) and transverse section near solid−liquid interface (b) at growth rate of 30 μm/s

3.2 Effect of holding time on solid−liquid interface

The morphologies of the solid−liquid interface of the alloy change with increasing t, as shown in Fig. 5. The solid−liquid interface is of dendritic growth at the growth rate of 30 μm/s under a constant temperature gradient (G=20 K/mm). The dendritic cores tilt with growth direction when t=5 min (Fig. 5(a)). With the increase of t, the dendritic cores align with growth direction gradually (Figs. 5(b)−(e)). The dendritic cores well align with growth direction when t=30 min (Fig. 5(c)). Therefore, it is helpful to increase t to obtain good directional solidification structure.

3.3 Effect of holding time on microstructure

The typical lamellar structures at different t are shown in Fig. 6. The lamellar structures coarsen with increasing t. The values of λ_1, λ_2 and λ_L increase with the increase of t at a constant V and G (Figs. 7(a)−(c)). The relationships among λ_1, λ_2, λ_L and t are obtained by using linear regression analysis, and the results are given as:

$$\lambda_1=288.9t^{0.22} \tag{1}$$

$$\lambda_2=32.36t^{0.10} \tag{2}$$

$$\lambda_L=1.05t^{0.33} \tag{3}$$

The regression coefficients of three relationships are r=0.976, 0.937 and 0.963, respectively.

3.4 Effect of holding time on microhardness

The values of HV decrease with increasing growth rate (Fig. 7(d)). The largest HV is obtained at t=5 min and the smallest HV is obtained at t=60 min. The relationship between HV and t is obtained by using linear regression analysis, the result is given as:

$$\mathrm{HV}=524.81t^{-0.09} \tag{4}$$

The regression coefficient of this fit is r=−0.969.

4 Discussion

There is no suitable refractory crucible that can absolutely avoid reaction with TiAl melts by now, as reported by GOMES et al [22] and CUI et al [23]. The Al_2O_3 crucible is common used for the directional solidification of TiAl alloys. LAPIN et al [24] pointed out that longtime interaction between the melt and Al_2O_3 crucible caused the contamination of the melt by Al_2O_3 particles, which would affect the microstructure and mechanical property of directionally solidified TiAl-based alloys. As reported by LAPIN et al [6], the volume fraction (f_c) of Al_2O_3 particles formed during directional solidification can be predicted from a kinetic equation in the form:

$$f_c = K_0 t_r^m \exp(-\frac{Q}{RT}) \tag{5}$$

where K_0 is a constant; t_r is the reaction time; Q is the activation energy for particle formation; R is the mole gas constant; T is the thermodynamic temperature. In this experiment, the reaction time can be expressed as $t_r = t + t_s$ where t is the holding time, and t_s is the solidification time. LAPIN et al [6] reported that the value of f_c during directional solidification of Ti−46Al−2W−0.5Si (mole fraction, %) alloy was expressed as:

$$f_c = 1.68\times10^8 t_r^{0.62} \exp\left(\frac{-37500}{RT}\right) \tag{6}$$

Figure 8 shows that the volume fraction increases with the increase of t. The measured results are agreed well with the predicted values by Eq. (6). LAPIN et al [6]

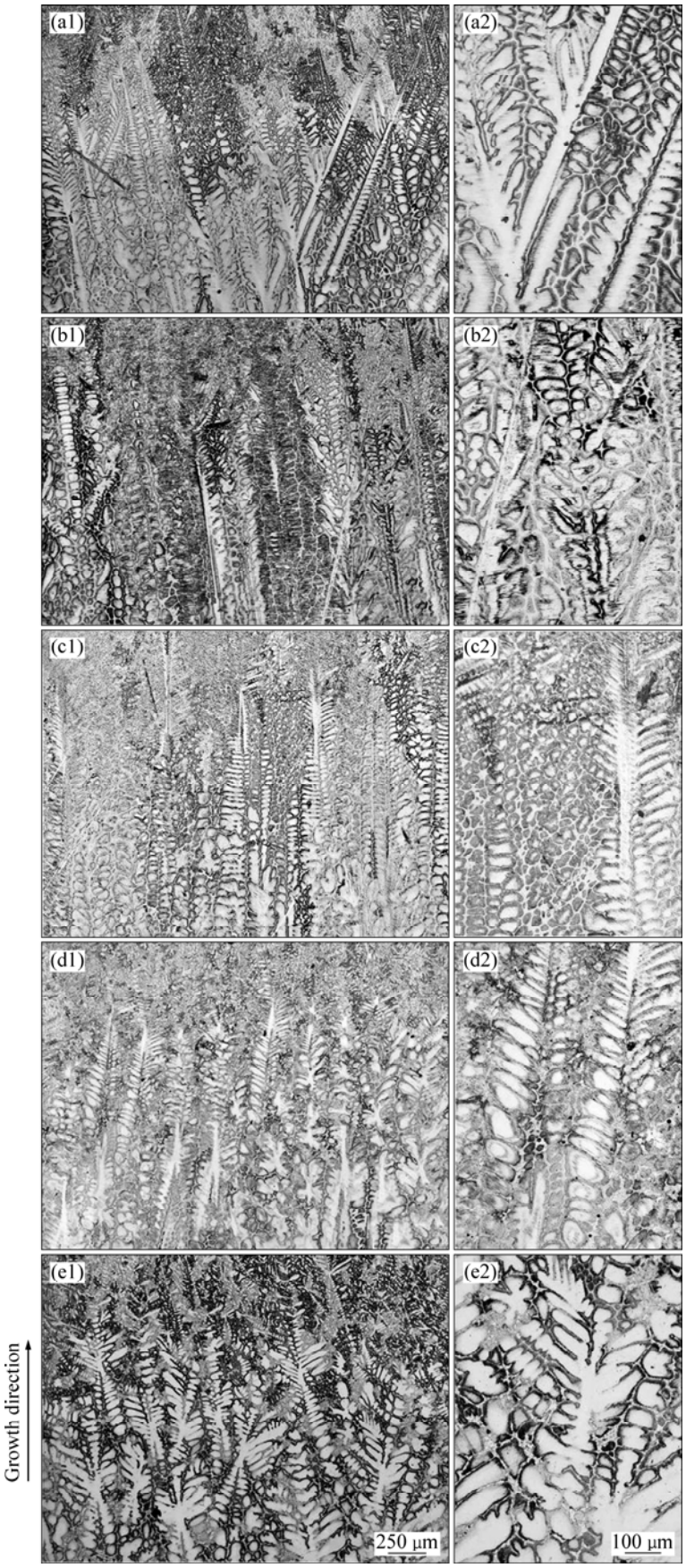

Fig. 5 Solid−liquid interfaces of directionally solidified Ti−46Al−0.5W−0.5Si alloys at growth rate of 30 μm/s for different holding time: (a1), (a2) 5 min; (b1), (b2) 15 min; (c1), (c2) 30 min; (d1), (d2) 45 min; (e1), (e2) 60 min

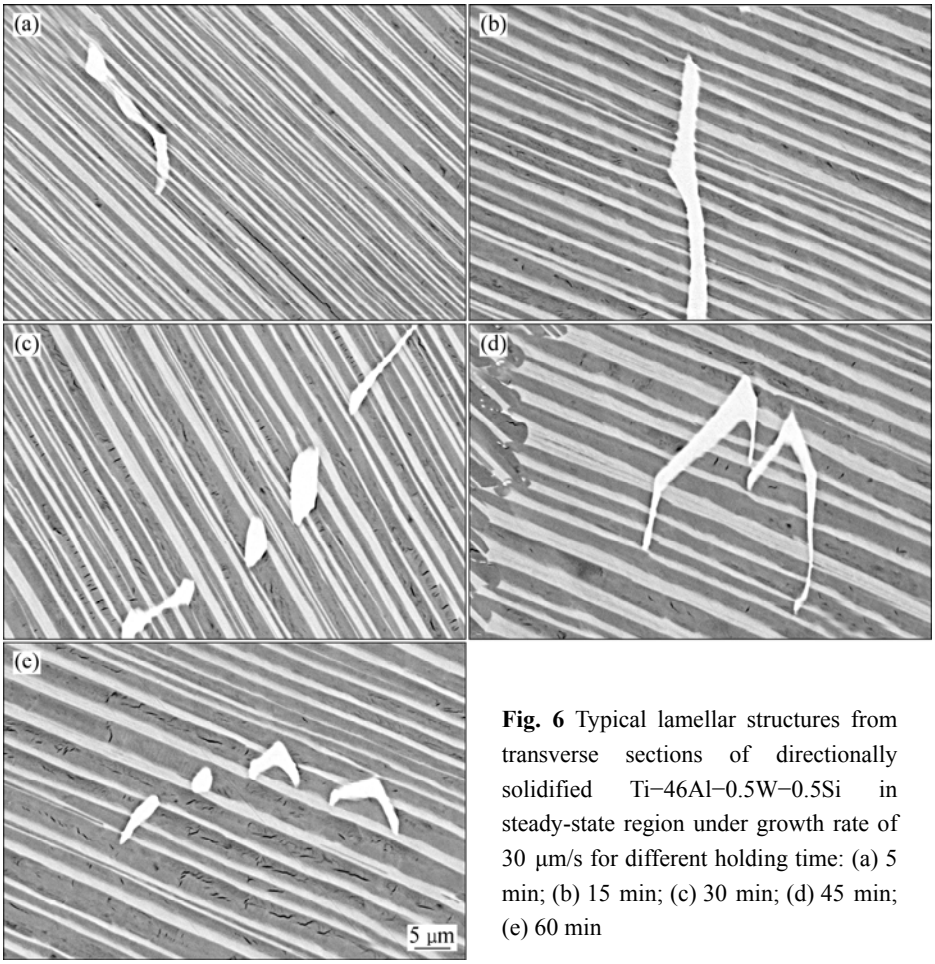

Fig. 6 Typical lamellar structures from transverse sections of directionally solidified Ti−46Al−0.5W−0.5Si in steady-state region under growth rate of 30 μm/s for different holding time: (a) 5 min; (b) 15 min; (c) 30 min; (d) 45 min; (e) 60 min

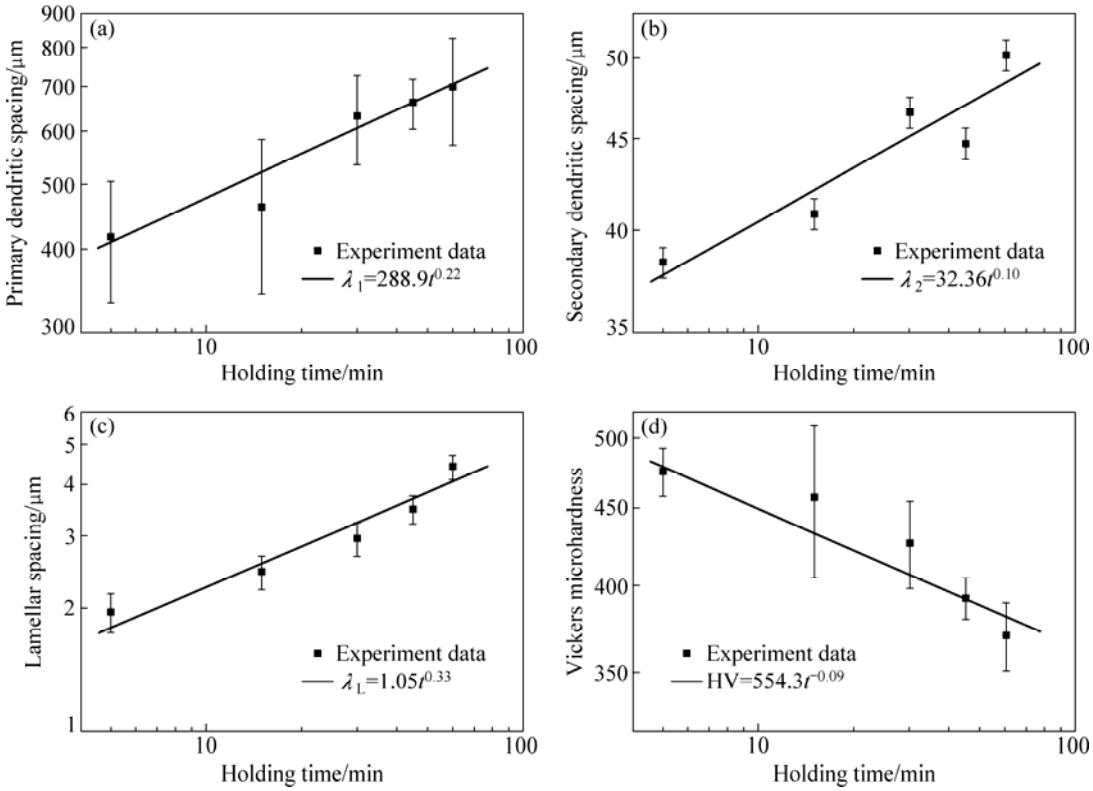

Fig. 7 Effect of holding time t on primary dendritic spacing (a), secondary dendritic spacing (b), interlamellar spacing (c) and microhardness (d)

considered that these particles were captured by moving solid−liquid interface and distributed in interdendritic region, which would hinder the growth of dendrites and cause the increase of dendritic spacing in directionally solidified specimens. It increases the difficulty to control the lamellar orientation of directionally solidified TiAl alloys. Therefore, it needs to reduce t to decrease the volume fraction of Al_2O_3 particles.

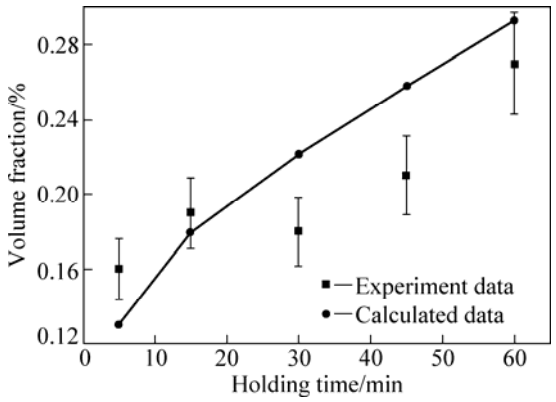

Fig. 8 Variation of volume fraction of Al_2O_3 particles with holding time (t)

LAPIN et al [6] indicated that the mechanical properties of fully lamellar TiAl alloys depended on several microstructural parameters, such as interlamellar α_2−α_2 spacing, thickness of γ lamellae and thickness of α_2 lamellae. As reported by LAPIN et al [6], the strengthening effect of lamellae can be described by Hall-Petch type relationship in the form:

$$\sigma_y = \sigma_0 + k\frac{1}{\sqrt{\lambda_L}} \quad (7)$$

where σ_0 is the stress corresponding to the lattice resistance to dislocation slip in the γ phase; k is a parameter which is related to the critical stress necessary to generate a dislocation in the next α_2 lamellae. According to Eq. (7), the yield stress decreases with the increasing of λ_L. Increasing holding time (t), which causes the coarsening of lamellar structures in directionally solidified specimens, is bad to the mechanical property of TiAl alloys.

DIMIDUK et al [25] found that there is a linear relationship between yield stress and microhardness, which allows mechanical properties of directionally solidified TiAl ingots to be predicted from the values of Vickers microhardness. According to the relationship, increasing t will decrease HV and reduce the yield stress of directionally solidified TiAl alloys.

In this work, a good combination of good directional solidification structures and high HV can be achieved when t=30 min. Therefore, the optimized value of t is 30 min for the directional solidification of this TiAl alloy.

5 Conclusions

Effect of thermal stabilization on the microstructure and mechanical property of directionally solidified Ti−46Al−0.5W−0.5Si (mole fraction, %) alloy was studied. The values of λ_1, λ_2 and λ_L increase with the increasing t. However, the values of HV decrease with increasing t. Increasing t is helpful to obtain good directional solidification structure, but it reduces the mechanical property of directionally solidified TiAl alloys. Under the present experiment condition, the optimized value of t is about 30 min, which will provide a reference to optimize the value of t for the directional solidification of TiAl alloys.

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熔体热稳定处理对定向凝固 Ti−46Al−0.5W−0.5Si 合金组织和性能的影响

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摘　要：通过改变定向凝固前的保温时间，研究热稳定处理对定向凝固 Ti−46Al−0.5W−0.5Si(摩尔分数，%) 合金组织和性能的影响。在稳定的温度梯度下(G=20 K/mm)，定向凝固启动前试样分别保温 5、15、30、45 和 60 min，然后试样以恒定的速度(v=30 μm/s)进行定向凝固实验。测量定向凝固组织的一次枝晶间距(λ_1)、二次枝晶间距(λ_2)、层片间距(λ_L)和显微硬度(HV)，并分析这些参数与热稳定处理时间(t)的关系。λ_1、λ_2 和 λ_L 的值随着 t 的延长而增大，而 HV 则随着 t 的延长而减小。在定向凝固启动前，延长热稳定时间有助于获得良好的定向凝固组织，然而，过长的保温时间则会降低合金的力学性能。因此，需要优化定向凝固前的热稳定处理时间。根据实验结果，在当前实验条件下，定向凝固前热稳定处理时间选择 30 min，既能够获得良好的定向凝固组织，又能保持合金较高的力学性能。

关键词：金属间化合物；TiAl 基合金；晶体生长；组织演化；力学性能

(Edited by YANG Hua)