Research Article

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Microstructure evolution of TC4 powder by spark plasma sintering after hot deformation

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Abstract: The cylindrical samples of TC4 titanium alloy prepared by spark plasma sintering (SPS) were compressed with hot deformation of 70% on the thermo-simulation machine of Gleeble-1500. The temperature of the processes ranged from 850°C to 1,050°C, and the strain rates varied between 0.001 and 5 s⁻¹. The relative density of the sintered and compressed samples was measured by the Archimedes principle. During hot deformation, the microstructure of the sample was observed. The results show that the average relative density of the samples was 90.2% after SPS. And the relative density was about 98% after the hot deformation of 70%. Under high temperature (>950°C), the sensitivity of flow stress to temperature was reduced. At low strain rate (0.001 s⁻¹), the increase in the deformation temperature promoted the growth of dynamic recrystallization (DRX). At the same temperature, the increase in strain rate slowed down the growth of DRX grains. And the variation tendency was shown from the basket-weave structure to the Widmanstätten structure at a low strain rate (<0.1 s⁻¹), with increase in the strain rate.

Keywords: spark plasma sintering, TC4 titanium alloy, microstructure, hot deformation, flow stress

1 Introduction

Titanium alloys are widely used in aerospace and marine applications due to its high specific strength, high-

temperature resistance and corrosion resistance [1]. However, the low-end titanium production capacity is excessive, and the high-end application level needs to be further improved [2–4]. Therefore, low cost and high performance have become the goals of the current development of titanium alloys. Powder metallurgy (PM) is one of the important methods to reduce the cost of titanium alloy parts. The main way is to shorten the production cycle and improve the utilization rate of metals. At present, the technology of PM has been widely used in the fields of machinery, electronics, aerospace, weapons and new energy and has become one of the most dynamic branches of new material science [5]. PM technology has a series of advantages such as remarkable energy saving, material saving, high product precision and good stability, which is suitable for mass production. In addition, some materials and complex parts which cannot be prepared by conventional casting methods and mechanical processing methods can also be fabricated by PM technology [6]. Therefore, it is highly valued by industries. The titanium alloy member having a tensile strength of 1,036 MPa and an elongation of 11.1% was obtained by Ng et al. while studying the formation of Ti-6242 titanium alloy powder [7]. Also, titanium PM parts comparable in performance to the forged titanium can be directly obtained by conventional, inexpensive uniaxial cold pressing and sintering. In order to obtain excellent high-temperature performance, Li et al. [8] studied Ti-3Al-4.5V-5Mo titanium alloy powder from the perspective of cross-rolling process.

Spark plasma sintering (SPS) has the characteristics of fast heating rate, short sintering time, controllable structure and uniform density [9,10] compared with pressureless sintering method, hot pressing method and hot isostatic pressing method. Fine and uniform grain samples can be obtained because SPS suppresses the growth of sintered grains [11]. Therefore, it is easy to obtain microstructures that can affect the mechanical properties. However, since the alloys produced by SPS are not fully dense, studies have been conducted to control the density of sintering by controlling the

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temperature [12,13]. In recent years, researchers have also conducted extensive research on the use of SPS technology to treat titanium alloys. For TC4 titanium alloys, more research should be done to understand cryomilling and SPS [14–16]. A few reports are available on the high-temperature hot deformation of TC4 titanium alloy prepared by SPS. The design of the related forging process also lacks the corresponding theoretical basis. Therefore, the main idea of the study was to realize the whole process by increasing the density of the powder by a forging process under conditions in which the powder was not completely densified during the SPS process. The purpose is to improve the process and make the TC4 titanium alloy more susceptible to deformation during the forging process (small resistance to deformation). In this article, the microstructural evolution of TC4 titanium alloy was investigated under the hot deformation condition. Therefore, the suitable density of TC4 titanium alloy was achieved by the SPS method. Then the isothermal compression test of TC4 titanium alloy was carried out. Changes in relative density, microstructure evolution and grain refinement behavior of TC4 titanium alloys were studied during high-temperature deformation. It provided the experimental basis for theoretical analysis and the formulation of hot deformation process parameters.

2 Experiment

The experimental material of TC4 titanium alloy powder was produced by Sino-Euro Materials Technologies of Xi'an Co., Ltd. The main components of the TC4 titanium alloy powder are shown in Table 1. The TC4 titanium alloy sample was prepared by SPS with SPS331-Lx. The pressure during sintering was maintained at 30 MPa and the sintering temperature at 850°C. After warming for 5 min, it was cooled in the furnace. Then the sample was processed into a cylinder of $\Phi 8 \text{ mm} \times 12 \text{ mm}$. The average value of the relative density of five cylinders was measured using the Archimedes principle. The thermal modeling test was performed on the Gleeble-1500. The amount of deformation was 70%. The

deformation temperatures were 850°C, 900°C, 950°C, 1,000°C and 1,050°C. And the strain rates were 0.001, 0.01, 0.1, 1 and 5 s^{-1} . Rapidly, the water cooled only after the deformation was completed.

The relative density of the sample was measured by the Archimedes principle after deformation and its formula was as follows:

$$\rho_r = \frac{m_0 \rho_{\text{water}}}{\rho_d (m_0 - m_1)}$$

where ρ_r was the relative density of the samples; m_0 and m_1 represented the quality of the samples in air and in water, respectively; the density of water $\rho_{\text{water}} \approx 0.9975 \text{ g/cm}^3$; ρ_d is the density of TC4 titanium alloy under full densification with the value of 4.51 g/cm^3 according to the measurement of the same batch of TC4 titanium alloy bars. The microstructure of the TC4 titanium alloy was observed under an optical microscope (OM). The microstructure of TC4 titanium alloy was also tested and analyzed by X-ray diffraction (XRD) scanning electron microscope and electron backscattered diffraction.

3 Results and discussion

3.1 Relative density

A high relative density and low porosity were required to ensure sintering and the subsequent forging. The relative density of the test piece was measured after the SPS, and the average value was 90.2%. The relative density of TC4 titanium alloy after thermal deformation was usually between 97% and 99% (as shown in Figure 1). The forming forces in the subsequent forging process were greatly affected by the relative density of the material. In the study by Sun *et al.* [13], when the SPS temperature was above 900°C, the density would reach 98%. By controlling the SPS temperature at 850°C, the same density can be achieved after performing the initial forging with the deformation of 70%.

3.2 Stress–strain behaviors

Figure 2 shows the effect of deformation temperature and strain rates on the peak stress of TC4 titanium alloy. It was observed that the stress decreased as the temperature increased and the strain rate decreased. At the same

Table 1: Main components of TC4 titanium alloy powder (wt%)

Al	V	O	Fe	C	H	N	Ti
6.1	4.2	0.18	0.04	0.015	0.02	0.021	Bal.

Figure 1: Relative density of TC4 titanium alloy at different temperatures and strain rates after hot deformation.

Figure 2: Curve of peak stress with temperature at different strain rates of TC4 titanium alloy during hot deformation.

time, the peak stress changed greatly at lower temperature ($<900^{\circ}\text{C}$), while the peak stress was closer at lower strain rates (0.001 s^{-1}) compared to that at high strain rates. Figure 3 shows the true stress–true strain curves of TC4 titanium alloy at different temperatures (0.001 and 0.01 s^{-1}). It can be more intuitively observed that as the temperature increased, the peak stress dropped significantly. Because of the increase in temperature, the softening mechanism was strengthened, and the stress was reduced at the time of dynamic equilibrium. At higher temperature ($>950^{\circ}\text{C}$), the change in flow stress decreased, indicating a reduced sensitivity of flow stress to temperature. The peak stress at 950°C was only about

37 MPa . On the other hand, at lower temperature ($<950^{\circ}\text{C}$), the flow stress gradually decreased with increase in strain, and the magnitude of the decrease was large, showing a characteristic curve of the dynamic recrystallization (DRX). At higher temperature ($>900^{\circ}\text{C}$), as the strain increased, the downward trend of flow stress slowed down.

Since the true stress–true strain curves were close at the same strain rate of more than 950°C , the two sets of true stress–true strain curves of TC4 titanium alloy were selected at 950°C and $1,050^{\circ}\text{C}$ (as shown in Figure 4). At low strain rates (0.001 and 0.01 s^{-1}), the stress decreased with the continuation of the deformation process, showing a low flow stress and a relatively steady flow curve. At a high strain rate (5 s^{-1}), the downward trend of stress was more significant. Because as the strain rate increased, the heat generated during deformation did not reach diffusion, leading to increase in material temperature. The increase in temperature was conducive for softening, and it showed a significant downward trend in the figure.

In addition, fluctuating flow stress can be observed by comparing Figure 3(a) $900^{\circ}\text{C}/0.001\text{ s}^{-1}$, Figure 4(a) $950^{\circ}\text{C}/0.1\text{ s}^{-1}$ and Figure 4(b) $1,050^{\circ}\text{C}/5\text{ s}^{-1}$. This type of curve was called discontinuous yielding, and what happens at this point was continuous DRX. In this variation, the increase in temperature was beneficial to softening, while the increase in strain rate was not conducive for softening. It was in this temperature-dependent variable, rate compensation, that the interaction exhibited a fluctuating flow stress curve between work hardening and dynamic softening.

3.3 Microstructure analysis

The characteristic peaks of $\alpha\text{-Ti}$ and $\beta\text{-Ti}$ were shown in the XRD pattern (Figure 5(b)). And after hot deformation, the peak value rose significantly, indicating that hot deformation could improve the crystallization of the TC4 titanium alloy. Since no new characteristic peaks were observed and significant characteristic peaks were shifted, it indicated that no new phase appeared during the hot deformation process. Figure 5(a) and (c) shows the microstructure of the TC4 titanium alloy after SPS and after hot deformation, respectively. The microstructure in the figure was taken from the central part of the material. The effect of temperature on the microstructure evolution could be clearly observed in the figure. The non-fully dense black portion of the pores, the coarse

Figure 3: True stress–true strain curve of TC4 titanium alloy at the same strain rates and different temperatures: (a) 0.001 s^{-1} and (b) 0.01 s^{-1} .

Figure 4: True stress–true strain curve of TC4 titanium alloy at different strain rates (a) 950°C and (b) 1,050°C.

Figure 5: (a) OM before hot deformation; (b) XRD of the TC4 titanium alloy before and after the hot deformation and (c) OM after the hot deformation at $T = 900^\circ\text{C}$ at the strain rate of 0.001 s^{-1} .

lamellar grains and the clear original grain boundaries were exhibited in the TC4 after SPS sintering at 850°C. This appearance of the Widmanstätten structure was mainly due to the low degree of deformation during sintering under the pressure of 30 MPa. Thus, coarse original β grains and the lamellar α were gradually formed during the subsequent cooling. As shown in Figure 5(b), the β -transition and the equiaxed α phase were distributed on the substrate. The equiaxed α phase exhibited elongated, spherical and other irregular shapes at the temperature of 850°C. This was the characteristic of equiaxed tissue obtained by plastic deformation below the β transition temperature. At the same time, fine grains appeared at the dislocation slip formed at the grain boundary, which also indicated the occurrence of DRX.

At 900°C, as shown in Figure 6(a), the equiaxed α structure was also exhibited, which mainly composed of a primary α phase and a β -transformed structure containing acicular α . At the temperature of 950°C, the primary α phase was greatly reduced close to the β -transition temperature. It was characterized by the coexistence of the equiaxed α and the lamellar α . At this time, the lamellar α had the tendency to change to the basket-weave structure. Comparing Figure 6(c) and Figure 6(b), the organizational characteristics had basically changed into a more standard basket-weave structure in which the respective lamellar α structure was staggered. In addition to its characteristics of high durability and creep strength, the basket-weave structure is also known for its low plasticity and high thermal stability. In this regard, Wang et al. [17] believed that the

Figure 6: The OM and backscattered electrons of the samples after deformation at the strain rate of 0.001 s^{-1} (a) $T = 900^\circ\text{C}$; (b) $T = 950^\circ\text{C}$; (c) $T = 1,000^\circ\text{C}$ and (d) $T = 1,050^\circ\text{C}$.

Figure 7: Grain maps displayed by random colors at a strain rate of 0.001 s^{-1} : (a) 850°C and (b) 1,050°C.

fine basket-weave structure could be obtained by reducing the temperature of heating and shortening the heating time in the zone of β phase. The plasticity of the material does not decrease too much as long as the tissue contains 10–20% equiaxed α . Therefore, the near- β forging proposed by Yigang Zhou was aimed to control the content of equiaxed α in the structure and also to change the morphology and distribution of the lamellar α phase precipitated in the high-temperature β phase. Generally speaking, the larger the amount of deformation, the higher the deformation temperature, and the equiaxification was more favorable. However, as shown in Figure 6(d), when the temperature rose to 1,050°C, an elongated lamellar α phase and coarse grains of β

appeared, and the grain boundaries were also remarkable. As the deformation temperature increases, the migration and diffusion of atoms also increased while the nucleation and growth rate increased, which promoted the formation of DRX. When the temperature was too high (1,050°C), the DRX grains had grown completely. The lamellar α phase and the acicular α phase accumulated continuously, and the coarse Widmanstätten structure began to increase.

Figure 7 shows the grain maps displayed by random colors of the TC4 titanium alloy under hot deformation conditions under the strain rate of 0.001 s^{-1} at different temperatures. The characteristics of grain structure was confirmed using OM. The structure as shown in

Figure 8: Misorientation angles of the samples by hot deformation at strain rate of 0.001 s^{-1} : (a) 850°C and (b) 1,050°C.

Figure 7(a) retained the β phase which was formed during slow cooling of the $\alpha + \beta$ processing temperature, and the spherical α phase can be obtained. And Figure 7(b) shows the phase structure was interlaced with the lath α phase and acicular α phase. Figure 8 reveals the reason for the change from the angle grain boundary. The effect of dislocations was demonstrated at different temperatures on the grain structure. The amount of deformation has an effect on the microstructure [18]. Long et al. [19] found that the average grain size was $15\mu\text{m}$ which exhibited a microstructure consisting of equiaxed α and transformed β phases by hot forging method. Relatively speaking, the average grain size could reach $3.8\mu\text{m}$ when the amount of deformation reached 70%. At the temperature of $1,050^\circ\text{C}$, the average grain size reached $14.9\mu\text{m}$.

At 950°C , the OM of the TC4 titanium alloy at different strain rates is shown in Figure 9. As shown in

Figure 9(a), in addition to the equiaxed α phase, the discontinuous grain boundary α and a small amount of large α appeared, showing as the basket-weave structure. As the strain rate increased, the equiaxed α phase disappeared, as shown in Figure 9(b). Instead, the microstructure included coarse colonies of the discontinuous grain α and the relatively coarse lath α structure also appeared. Typical lamellar α and large β grains appeared as the strain rate (1s^{-1}) increased. When the strain rate was further increased, this characteristic of the Widmanstätten structure became more pronounced at the grain boundary, which was shown as the elongated lamellar α phase parallel to each other. Therefore, when the four images in Figure 9 are compared, it was not difficult to find that the increasing of the strain rate would change the tendency from the original basket-weave to the Widmanstätten structure. Under this trend, a significant decrease in high-

Figure 9: The OM at 950°C : (a) 0.01s^{-1} ; (b) 0.1s^{-1} ; (c) 1s^{-1} and (d) 5s^{-1} .

Figure 10: Grain maps displayed by random colors at the temperature of 950°C: (a) 0.1 s^{-1} and (b) 5 s^{-1} .

temperature strength and plasticity was observed, although the creep resistance and fracture toughness of the material had been improved. At high strain rates (5 s^{-1}), the hot deformation time shortened; and the dynamic recovery (DRV) process occurred first after the elongation of the grains during the hot deformation process; however, DRX occurred only in a small portion. Referring to Figure 6(a) and (b), it could be found that when the strain rate lowered to 0.001 s^{-1} , the grains grew again, which involved a process in which the recrystallized grains were grown and refined.

Figure 10 illustrates the feature changes in the OM map by comparing different strain rates. At a strain rate of 5 s^{-1} , a large number of fine DRX grains were exhibited. At the

same time, the relationship between DRV and DRX during hot deformation is also shown in the figure. As shown in Figure 10(a), the recrystallized grain grew compared with the strain rate of 5 s^{-1} . Because at lower strain rates, the hot deformation time was found to be longer, and the fine grains formed at the high dislocation entanglement had sufficient time to further grow at a high temperature. So more complete growth of DRX grain structure could be obtained. On the contrary, DRX grains were more likely to remain in a finer form as the strain rate increased. Figure 11 shows the presence of a higher proportion of low-angle grain boundaries at a strain rate of 5 s^{-1} . As the strain rate decreased, the proportion of high-angle grain boundaries increased, and the DRX grain size increased.

Figure 11: Misorientation angles of the samples by hot deformation at temperature of 950°C: (a) 0.1 s^{-1} and (b) 5 s^{-1} .

4 Conclusion

- (1) The relative density of TC4 titanium alloy was measured after SPS, and the average value was 90.2%. And the relative density of TC4 titanium alloy was about 98% after the hot deformation of 70%.
- (2) Under high temperature ($>950^{\circ}\text{C}$), the sensitivity of flow stress to temperature was reduced.
- (3) At low strain rate (0.001 s^{-1}), the increase in the deformation temperature promoted the growth of DRX. The effect of temperature on the evolution of the α phase was shown, which successively changed from the equiaxed α phase to the lath α phase and lamellar α phase, with an increase in temperature. At the same time, the lamellar α phase increased, which showed as the Widmanstätten structure at high temperatures ($1,050^{\circ}\text{C}$).
- (4) At the same temperature, the increase in strain rate promoted the refinement of DRX grains. The equiaxed α phase gradually disappeared and the lamellar α phase gradually increased. The overall variation tendency was shown from the basket-weave structure to the Widmanstätten structure with an increase in the strain rate.

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