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Microstructure of TiAl alloy prepared by intense plastic deformation and subsequent reaction sintering①

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[Abstract] TiAl alloy was prepared by intense plastic deformation and subsequent reaction sintering. The effect of plastic deformation on the microstructure of sintered TiAl alloy was investigated using energy dispersive X-ray spectroscopy (EDS), optical microscopy and transmission electron microscopy (TEM). The results show that the intense plastic deformation of reacting Ti and Al phases caused by high energy ball milling refines the as-sintered microstructure. The longer the milling time, the finer the grain size of γ and lamellar (α_2+ γ) phases. The finer grain size improves the properties of the TiAl alloy. It is also found that the volume fraction of lamellar (α_2+ γ) phases increases first, then decreases with increasing milling time. Based on the experimental results theoretical discussion was presented.

[Key words] TiAl alloy; high energy ball milling; reaction sintering; microstructure
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1 INTRODUCTION

TiAl alloys are suitable for high-temperature applications in aircraft gas turbine engines and other transportation vehicle, owing to their low densities, high melting points and good elevated-temperature strength[1, 2]. But their low-temperature brittleness and high strength at elevated-temperature make them difficult to be shaped by conventional machining or deforming methods. Recently, investigators studied the elemental powder metallurgy (PM) route for the fabrication of TiAl alloy. Not only PM can avoid grain coarsening and dendritic segregation associated with ingot metallurgy, but also make the fabrication of near-net shape components possible[3, 4].

In order to obtain TiAl alloy via elemental powder metallurgy route, reaction sintering is a necessity. Because of the significant difference between the diffusion coefficients of Ti and Al, pores tend to form due to Kirkendall effect during reaction sintering. Therefore, it is of great significance to refine the Ti and Al constituents prior to the reaction sintering so that porosity can be minimized and the microstructure of the sintered titanium aluminides can be refined. Indeed, it has been reported that the grain of the reactively sintered TiAl alloy was refined and the porosity was reduced by severe cold deformation, such as extrusion and rolling, of the mixed Ti and Al powder billet before reaction sintering[5, 6]. It is known that grain refinement provides a microstructural base for high performance material[7, 8].

High energy ball milling has a unique advantage of refining microstructure of powder materials[9, 10]. Compared with conventional deforming techniques, it is much more convenient and effective for the refining of powder materials. Thus, high energy ball milling and subsequent reaction sintering was employed to prepare the TiAl alloy. Recently, we developed and studied the preparation and consolidation of the ball milled fine grain Ti/Al composite powders with extremely fine alternative Ti and Al lamellar structure[11, 12]. The present paper reported the microstructure of the TiAl alloy via reaction sintering of the composite powders developed by our previous work.

2 EXPERIMENTAL

The composite powder was prepared from the mixture of commercially available titanium powder (> 99.9% in purity, 50 μm in mean diameter) and aluminum powder (> 99.5% in purity, 150 μm in mean diameter). The powders were mixed with an atomic ratio of 1∶1 in a stainless steel milling vial. High energy ball milling was carried out under the protection of pure argon gas. The milling vial was equipped with a water-cooling system in order to prevent possible alloying of Ti and Al during milling. Milling balls were 8 mm in diameter. The ball to powder mass ratio was 20∶1, and the rotational ve-

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locity was kept at 400 r/min.

The Ti/Al composite powders milled for various times were cold compacted under a specific pressure of 1 000 MPa, then degassed and encapsulated at 300 ℃ under a vacuum of less than 2.5×10^{-2} Pa. The encapsulated green compacts were further consolidated into fully-densified powder billets by hydrostatic extrusion with an extrusion ratio of 16∶1 at 250 ℃. For comparison, billets were prepared from commercial elemental Ti and Al powder mixture using the same consolidation route. The densified powder billets were reactively presintered at 630 ℃ for 2 h, then sintered at 1 250 ℃ for various times without pressure, so that their structure was transformed into titanium aluminides. The microstructure and microconstituent of as-sintered samples were studied by optical microscopy, TEM and characterized by EDS.

During presintering basically all Al phases reacted with Ti. Thus, after presintering, very little or no liquid Al remained in the billet and ensured stable billet shape after high-temperature sintering without pressure.

3 RESULTS AND DISCUSSION

Typical microstructures of TiAl alloys can be obtained by high temperature sintering of either mixed or milled powders, as shown in Table 1.

Table 1 Microstructures of sintered TiAl alloys

Sintering temperature/ ℃	Microstructure type
1 100	γ
1 250	Duplex
1 300	Full lamellae

The influence of high energy ball milling on the microstructure of the sintered sample is obvious. From Fig. 1, it is noticed that both TiAl alloy samples are composed of γ and (α_2 + γ) duplex microstructure after sintering at 1 250 ℃ for 8 h. As shown in Figs. 1 (a) and (b), the difference in the γ grain size is not large at all, but the differences in the grain size of (α_2 + γ) phase and γ to (α_2 + γ) volume ratio are remarkable.

Fig. 2 shows the dependence of grain size and lamella volume fraction on high energy ball milling. The longer the milling time, the finer the grain size of γ and lamellar (α_2 + γ) phase. The TiAl alloy sintered from milled Ti/Al composite powders presents a much finer microstructure with an average grain size of 2~4 μm for γ phase and 5~8 μm for (α_2 + γ) phase, and a high volume fraction of up to 75% of (α_2 + γ) phase. This suggests a higher nucleation rate for both α and γ phases, and the acceleration of diffusion kinetics to phase equilibrium during reaction sintering. This can be attributed to the extremely fine Ti and Al lamellar structure, the fine-sized crystalline grains, the enhanced grain boundaries and lattice defects.

According to Johnson-Mehl formula, the relation between recrystallized grain size D and nucleation rate n is expressed as

$$D = K(G/n)^{\frac{1}{4}} \tag{1}$$

where G is the growth rate of grain, K is a constant. It was known from Eqn. (1) that α phase grain size D_α declines with increasing n at 1 250 ℃.

For an alloy system with dual phases, the growth equation of the new grain during heat treatment can be written as[13, 14]

$$\int_{D_0}^{D} \frac{\mathrm{d}D}{(1/D - 1/D_0)^{p-1}} = F(t - \tau) \tag{2}$$

$$F(T_1) = F(T_2)\exp[-\frac{Q}{R}(\frac{1}{T_1} - \frac{1}{T_2})] \tag{3}$$

where t is the duration of heat treatment, T_1 and T_2 are the temperatures of heat treatment, F is a constant related to the temperature of heat treatment and alloy composition, Q is the activation energy of nucleation, τ is the nucleation period, D_0 is the

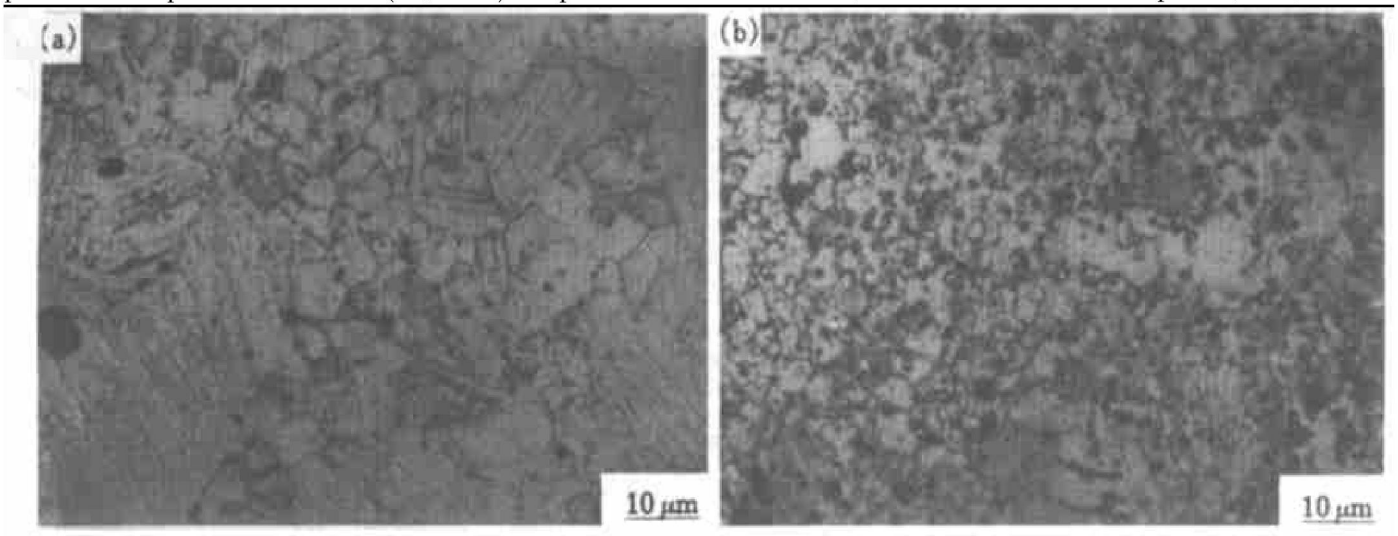

Fig. 1 Microstructure of TiAl alloy prepared by reaction sintering at 1 250 ℃ for 8 h
(a) —Without milling; (b) —Milled for 10 h

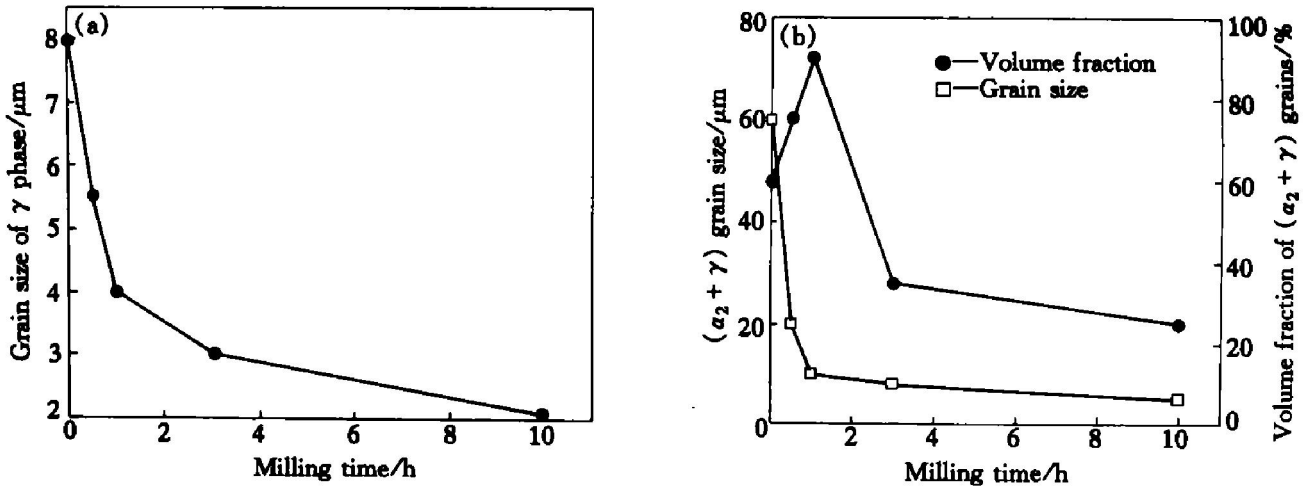

Fig. 2 Variation of grain size of γ(a), lamella grain size and volume fraction of lamella (b) in samples sintered at 1 250 ℃ for 8 h with milling time

initial grain size, and p is the grain growth factor that is usually equal to 1. When sintered at dual phase zone, the grain size of α phase can be further simplified as

$$D_\alpha \propto F(t-\tau) \tag{4}$$

After substituting Eqn. (4) into Eqn. (1), we obtain the volume fraction of α phase:

$$X_\alpha = 1-\exp[-\frac{\pi}{3}NnA(t-\tau)^4] \tag{5}$$

where A is a constant. From the above equation, it can be concluded that the volume fraction of α phase increases with the increase of its nucleation rate. Referred to the Ti-Al binary phase diagram, it can be found that for Ti-50Al alloy, ($\alpha_2+\gamma$) lamellae at room temperature originate from α phase at temperature above eutectic point such as 1 250 ℃. Therefore, under the same cooling condition, the volume fraction of ($\alpha_2+\gamma$) lamellae increases with increasing nucleation rate of α phase, that is, increases with increasing milling time.

The grain size of γ and α phases after sintering should obey Eqn. (1) in theory. Undoubtedly, the nucleation rate of γ and α phases increases with decreasing Ti/Al lamellar spacing of the composite powders. Therefore, their grains are refined with increasing milling time. Fine α phase grain brings up fine ($\alpha_2+\gamma$) lamellae.

From Fig. 2, it is clear that when powders are milled for more than 3 h, the volume fraction of ($\alpha_2+\gamma$) phase in sintered sample rapidly goes down. This doesn't mean the above theoretical analysis is mistaken. From the investigation on the influence of milling on powder's microstructure, it is known that 3 h of milling has induced amorphous phase among Ti/Al composite powders, and further milling can make all powders transform to amorphous phase. For sintering of amorphous powders prepared by long time milling, the nucleation periods τ of both α and γ phases should not be omitted. The period contains another nucleation period of transforming from amorphous to crystallite state. Hence, the crystallizing period takes up a part of the whole nominal sintering time. This leads to shortening of the time that spend on Ti and Al reaction and homogenization. So for powders prepared by long time milling, the volume fraction of α phase goes down at the beginning stage of the cooling following the sintering. Thus, the volume fraction of ($\alpha_2+\gamma$) lamella decreases. Compared with the unmilled powders or powders that are milled for short time, the existing of Ti and Al grains among the sintered amorphous powders under the same sintering process affirms the above analysis, as shown in Fig. 3.

Based on the present experimental observation and the above discussion, it can be concluded that the average grain size of the γ and ($\alpha_2+\gamma$) phases of the as-sintered TiAl alloy can significantly be reduced by using mechanically prepared Ti/Al composite powders as precursory material for reaction sintering. It can be expected that this microstructure is beneficial to mechanical properties of the as-sintered TiAl alloy such as strength and ductility, which has been experimentally tested. Because ($\alpha_2+\gamma$) lamellar microstructure possesses higher strength and lower ductility than γ phase, the comprehensive properties of the as-sintered TiAl alloy depend on the volume ratio of ($\alpha_2+\gamma$) lamella to γ phase to a certain extent. The study on the influence of high energy ball milling on mechanical properties of TiAl alloy prepared by the present method will be reported in another paper.

4 CONCLUSIONS

Compared to the samples prepared from elemental Ti and Al powder mixture, the as-sintered TiAl alloy prepared from milled Ti/Al composite powder possesses a much finer microstructure. This is due to the higher nucleation rate of titanium aluminides

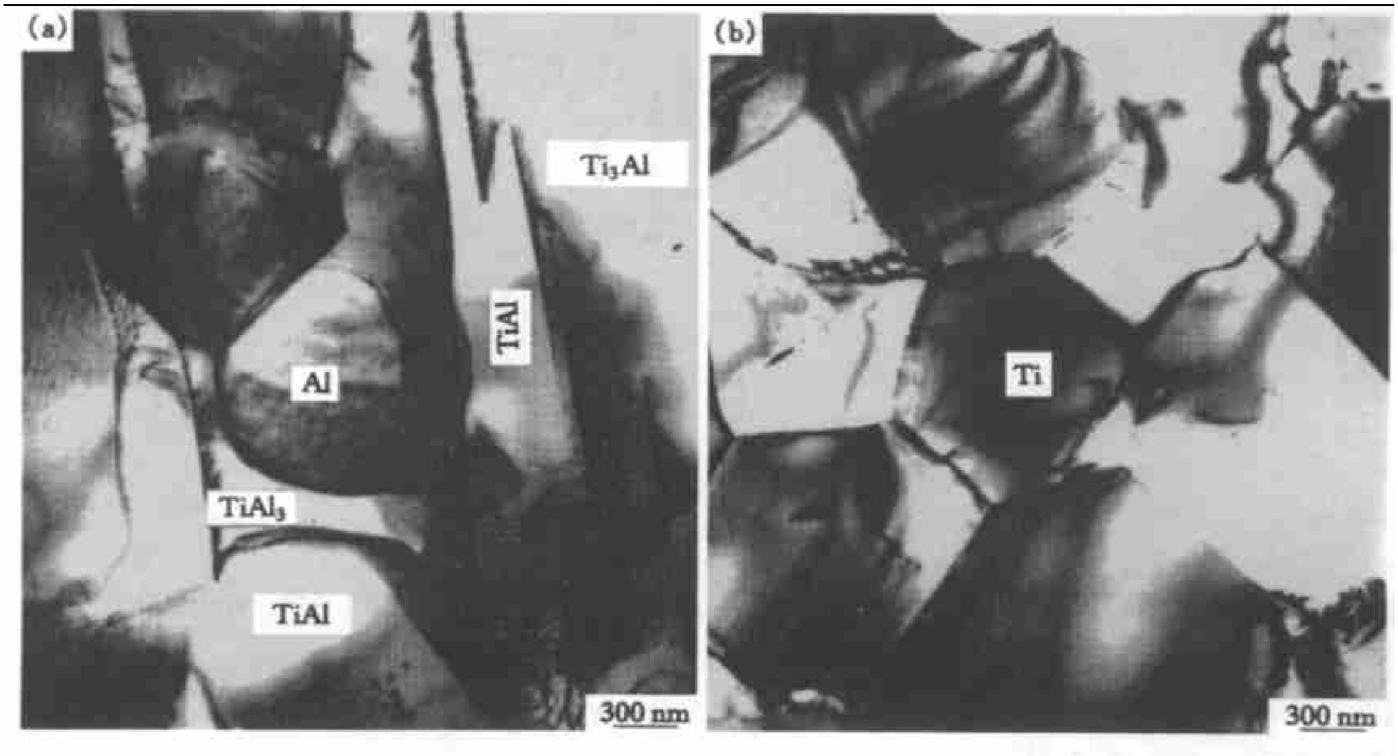

Fig. 3 Selected area TEM images of samples sintered at 1 250 ℃ for 1 h
(a) —Milled for 3 h; (b) —Milled for 10 h

during reaction sintering. The longer the milling time, the finer the grain size of γ and lamellar (α_2+γ) phases. Fine microstructure will be beneficial to the properties of the as-sintered TiAl alloy. It is also found and explained that the volume fraction of the lamellar (α_2+γ) phase first increases, then decreases with increasing milling time.

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