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Microstructure characterization and mechanical properties of TiAl-based alloys prepared by mechanical milling and spark plasma sintering

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ABSTRACT

High Nb containing TiAl alloy powders with or without mechanical milling were consolidated by spark plasma sintering (SPS) technique. The effects of SPS temperature and mechanical-milling treatment on phase constitution and microstructure were studied, and the mechanical properties at room temperature were tested. The phases in the as-atomized powder are composed of major α_2 phases, a few γ phases and a trace of β phases. After milling, the diffraction peaks for α_2 phase are obviously broadened, and the intensities of diffraction peaks for both γ phase and β phase are decreased. Similar phase constitution including a large quantity of γ phases and a few α_2 phases are exhibited in the alloys sintered by either as-atomized powder or as-milled powder. At a sintering temperature of 1200 °C, the microstructure of the alloy sintered by using as-atomized powder consists of inhomogeneous γ and α_2 phases as well as a few α_2/γ lamellar colonies. By contrast, the densities increase and the microstructures are apparently refined for the alloys sintered by using as-milled powder. Fully dense alloys with uniformly distributed γ and α_2 grains can be obtained at extending milling time or increasing rotating speed. The nucleation of γ phase during SPS of the powders is dependent on recrystallization. The heterogeneity of deformation should be responsible for the formation of heterogeneous γ grains in the alloys sintered by using as-atomized powder. The heterogeneous grain size for the alloys sintered by using as-milled powders is mainly derived from the inhomogeneous nucleation of γ phase and the uneven distribution of α_2 grains. Good mechanical property accompanied by a fine grain size and a dense microstructure can be achieved by optimizing the process parameters.

1. Introduction

Intermetallic γ-TiAl based alloys are considered as promising materials for high-temperature aerospace and automobile applications [1–4]. As one of the most potential generations, high Nb containing γ-TiAl alloys own excellent mechanical strength as well as good oxidation resistance and creep properties at elevated temperatures [5,6]. However, due to high Nb content, the process of the alloys by ingot metallurgy (IM) [7] becomes very difficult. Compared with that processed by IM, high Nb containing γ-TiAl alloys with more homogenous microstructure and finer grain size can be obtained by powder metallurgy (PM) technique [8,9]. Conventional PM processing generally requires long and expensive procedures to achieve full densification. Spark plasma sintering (SPS) is a quick and efficient PM technique to sinter powder through the simultaneous application of a pulsed direct current of high intensity and uniaxial pressure [10]. By using SPS technique, full dense alloys with a refined microstructure can be obtained in a short time [11].

In the last decades, SPS technique has been demonstrated to be an effective route to consolidate high Nb containing γ-TiAl alloys, which exhibited high and remarkably reproducible room temperature strength as well as satisfactory ductility [12–14]. Studies about the effects of SPS parameters on microstructure and mechanical property, so as to obtain fully dense γ-TiAl alloys, have also been extensively reported [15–17]. However, the raw materials used in the previous studies focused on the pre-alloyed powder prepared by gas atomization process. It is known that mechanical milling is an effective way to produce pre-alloyed powder which could quicken solid diffusion and promote formation of compounds [18–20]. Up to now, the reports concerning the SPS behavior of mechanically-milled γ-TiAl alloy powder are still limited [21–24]. Chen et al. [22] investigated the microstructure and

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mechanical properties of a γ-TiAl alloy produced by SPS using high-energy mechanically-milled powder, and found that a low temperature and a short time are sufficient for reaching the phase equilibrium of the alloy. The optimized SPS temperature which can result in high-quality sintered alloys with ultrafine grain sizes and high strengths is also presented. Guyon et al. [23] studied the densification behaviors of as-atomized and as-milled γ-TiAl-based alloy powders consolidated by SPS, and proposed that the difference in initial structures would lead to different densification mechanisms. Shanmugasundaram et al. [24] utilized the cryo-milled powder to fabricate a fully dense fine-grained γ-TiAl based alloy by SPS technique, and investigated the effects of cryomilling on densification kinetics and final microstructure.

In the present work, high Nb containing TiAl alloys consolidated by SPS technique using the powders with and without mechanical milling were obtained. Phase transformation and microstructural evolution during SPS of the powders, as well as mechanical properties of the SPSed alloys at room temperature, were systematically investigated. Efforts were paid on discussing the effects of initial powder states on microstructure and mechanical property of the SPSed alloys.

2. Experimental

Gas-atomized powder with a nominal composition of Ti-45Al-7Nb-0.3W (at.%) were supplied by Xi'an Theron Metal Materials Co. Ltd. (Shanxi, China). The as-atomized powder with a particle size distribution ranging from 100 to 200 mesh were used. The contents of oxygen and nitrogen in the powder are 0.083% and 0.005%, respectively. A part of the powder was further mechanically milled in a planetary ball mill using a ceramic vial and ZrO_2 balls with different diameters. A ball-to-powder ratio of 5:1 was selected and about 60% of ethanol was put into the vial before milling for lubrication. The rotation speeds of the main disk are 150 rpm and 250 rpm, respectively. The milling times are 5 h and 15 h, respectively. The milling was stopped for 15 min at every 30 min. Then, the as-atomized and the as-milled powders were sintered in a FCT HP D 25/3 SPS device for 10 min under a pressure of 40 MPa at the temperatures of 1000 °C, 1100 °C and 1200 °C, respectively, with a heating rate of 100 °C/min. As-SPSed alloys with about 40 mm in diameter and 10 mm in thickness were obtained.

Particle size distribution and average size of the as-atomized and the as-milled powders were evaluated by a MICRO-PLUS laser particle size analyzer. X-ray diffraction (XRD) analyses were carried out on a Rigaku D/max 2550VB diffractometer using Cu-K_α radiation. Scanning Electron Microscopy (SEM) analyses were conducted on a Quanta FEG 250 microscope equipped with an energy dispersive spectroscopy (EDS) in order to observe the morphology of powders (secondary electron mode) and identify the microstructural characteristics of as-SPSed alloys (backscattered electron mode). Transmission Electron Microscopy (TEM) analyses were performed on a JEOL-2100F transmission electron microscope operated at 200 kV. TEM foils were thinned by a twin-jet electro-polishing at −30 °C in a solution of 20% perchloric acid, 30% n-butyl alcohol and 50% methanol under 25 V. Before electro-polishing, the foils with a thickness <80 μm were prepared by standard mechanical polishing with SiC sandpapers. The foils for TEM observations were further analyzed by electron backscattered diffraction (EBSD) technique. The data collection was conducted at an interval of 0.05 μm, and the collected EBSD data were processed by the TSL OIM analysis software.

The densities of the as-SPSed alloys were measured through Archimedes method with distilled water as the immersion medium. The concentrations of O and N in the powders were analyzed with an inert gas melting-IR absorption spectrometer. Tensile tests of the as-SPSed specimens with a dimension of 26 × 10 × 3 (mm^3) at room temperature were conducted at a constant strain rate of 10^{-4} s^{-1} using an Instron 3369 testing machine.

3. Results and Discussion

3.1. Characterization of As-atomized and As-milled Powder

The morphologies and the particle size distributions of TiAl powders with different initial states are shown in Figs. 1 and 2, respectively. It can be seen that the as-atomized powder appears spherical or near-spherical (Fig. 1(a)) with an average size of about 182.8 μm (Fig. 2). Some smaller particles adhere to the molten surfaces of larger ones, leading to the formation of satellite structure. Typical dendritic morphologies are exhibited on the surfaces of most powder (Fig. 1(b)). Individual smaller powder shows smooth surface, which could be single crystals without dendritic segregation [25]. The different sizes of the particles and its various morphologies are mainly derived from different cooling rates during processing [26].

The dendritic and smooth surfaces can hardly be found for the powder milled at 150 rpm for 5 h. Coarse surfaces with irregular or ellipsoidal cracks appear (Fig. 1(c)), due to the brittle rupture taking placing in particles (Fig. 1(d)). The average size of the powder is determined to be about 59.6 μm (Fig. 2). When the milling time increases to 15 h, severe deformation and further fragmentation are observed (Fig. 1(d)) and the average size is reduced to be about 21.0 μm (Fig. 2). It is found that increasing rotation speed of milling has a larger breaking effect on the powder than that of milling time (Fig. 1(e)). With the rotation speed of milling increasing from 150 to 250 rpm, the average size of the powder is reduced to be about 18.6 μm (Fig. 2), which can be mainly ascribed to the successive fragmentation/welding of the brittle powder during milling. However, the concentrations of O element in the as-milled powders are measured to be 0.18% (150 rpm, 5 h), 0.60% (150 rpm, 15 h) and 0.56% (250 rpm, 5 h), respectively, which indicates that a high content of oxygen is induced after milling, especially for the powder with increasing rotation speed or prolonging milling time.

Fig. 3 shows the XRD patterns of TiAl powders with and without milling. The phases in the as-atomized powder are composed of major α_2 phases, a few γ phases and a trace of β phases. Due to the high content of Nb, the TiAl alloy solidifies through a β path, and the metastable primary β phase would further transform to α phase and then α_2 phase. During the following solidification, γ phase can nucleate by $\alpha \rightarrow \gamma$ transition in the interdendritic regions where element Al is concentrated [27]. Because of the high cooling rate during gas atomization, a small number of β phases are still retained.

Compared with that of the as-atomized powder, the diffraction peaks for α_2 phases of the as-milled powders are obviously broadened (Fig. 3), which can be attributed to the lattice strain and the refinement of crystallite size during milling [24]. With the increase of milling time or rotation speed, the broadening phenomenon of diffraction peaks becomes apparent. It is suggested that the broadening of XRD peaks for the powder milled at 150 rpm for 5 h primarily originated from the accumulation of large amounts of lattice defects due to plastic deformation of fragmented particles. After being milled at prolonging milling time or increasing rotation speed, severe deformation would occur, leading to the increase of the amount of small flaky particles. The formation of extremely small crystallites may also contribute to the broadening. The intensity of diffraction peaks for γ phase decreases and the diffraction peaks for β phase almost disappear after milling, which imply that the occurrence of deformation during milling induced the phase transformation in the TiAl alloy powder. It has been reported that element O has a strong stabilizing effect on α (α_2) phase [28]. Apparent increasing content of O in the as-milled powders would be expected to promote the formation of α (α_2) phase in the sintered alloys.

3.2. Phase and Microstructure of As-SPSed Alloys

3.2.1. Effects of SPS Temperature on Phase and Microstructure

Fig. 4 shows XRD patterns of the alloys sintered by as-atomized TiAl alloy powder at temperatures of 1000, 1100 and 1200 °C for 10 min