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Grain refinement by means of phase transformation and recrystallization induced by electropulsing

SONG Hui[1], WANG Zhong-jin[2]

1. Research Station on Mechanics for Postdoctoral Fellows, Harbin Institute of Technology, Harbin 150001, China;
2. School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, China

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Abstract: The specimens cut from as-cast TiAl alloy and cold-rolled TA15 sheet were treated by high density electropulsing. The optical microscope (OM) and transmission electron microscope (TEM) were used to examine the changes of the microstructure before and after the electropulsing. It is found that fine and homogeneous microstructures with the grain size of 30−50 μm can be obtained from the as-cast TiAl based alloy with the initial lamellae of about 1 000 μm. The electropulsing treatment can induce the completed recrystallization in cold-rolled TA15 sheet. After electropulsing treatment, the microstructure of TA15 sheet transformed from primary α laths grain to smaller α equiaxed grain. The research results show that the electropulsing treatment is an effective method to refine grain of as-cast TiAl alloy and cold-rolled titanium alloys sheet. Because the conventional thermo-mechanical processing such as forging or extrusion or rolling as well as heating are not required, the refining process by electropulsing is simpler than conventional ones.

Key words: electropulsing; grain refinement; nucleation rate; TiAl alloy; TA15 alloy

1 Introduction

One of the most effective methods for reducing cost is through mass reduction of the aircraft and automobile structural materials. For this reason, lightweight titanium alloys and titanium aluminides are primary materials for aerospace, aeronautic and automobile applications.

On the one hand, titanium aluminides are difficult to be processed by conventional manufacturing routes such as forging, rolling and welding due to poor ductility at ambient temperature [1−2]. Therefore, casting and superplastic forming become the effective method for fabricating titanium aluminides. However, because the as-cast microstructure is fully lamellar structure, the properties of TiAl alloys are improved by the microstructural refinement [3−4]. Furthermore, fine grained superplastic materials are pre-treated for superplastic forming. Therefore, a considerable effort, such as thermomechanical treatment (TMT), has been expended to develop new approaches for grain refinement. But, thermo-mechanical treatment (TMT) in which the alloy is first forged or extruded, and then heat treated, is usually difficult due to its relatively high melting point and the extreme reactivity of Ti [5−6]. Therefore, significant difficulty in experiment and thus considerable cost in production are encountered with the conventional methods. To avoid the mechanical processing, heat treatment or cyclic heat treatment has been studied in recent years [7−8]. Although these studies are very fruitful, the time at temperature and the temperature control that is required have limited their general application [9]. On the other hand, as-rolled titanium alloys are heat treated in order to realize the restoration, to refine the grain and to produce an optimum combination of machinability. However, heat treatment for titanium alloy has its own limitation and weakness: the higher chemical activity of titanium in alloys makes metal surface form the oxidation layer and the gas absorbing layer during heating, which leads to decrease notably in plasticity, and tend to brittle failure.

So, it is valuable to find a new and more practical technique for grain refinement. The high density electropulsing treatment of materials has attracted more and more attention due to its special characters [10−11]. First, in a current-carrying metallic material, drift electrons can exert a force on dislocations, e.g. electron wind force. Second, the electropulsing can increase the

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Corresponding author: WANG Zhong-jin; Tel: +86-451-86413365; E-mail: wangzj@hit.edu.cn

nucleation rate by the decrease of thermodynamic barrier during phase transformation, consequently, can refine grain microstructure. In this work, a new method for grain refinement of titanium alloys and titanium aluminides is presented.

2 Experimental

Titanium aluminides used in this study had a nominal composition of Ti-48Al-2V (mole fraction, %). They were prepared by vacuum induction melting with a water-cooled copper crucible. Specimens with dimensions of 17.0 mm×4.0 mm×1.2 mm were cut from ingots using electro-discharge machining (EDM).

The titanium alloy specimens were cut from cold-rolled TA15 sheet (2.0 mm in thickness) along the rolling direction by electro-discharge machining (EDM). And the size of specimen's parallel portion was 15.0 mm in length, 3.0 mm in width and 2.0 mm in thickness.

The electropulsing was performed under ambient conditions by capacitor banks discharge. The experimental arrangement for the electropulsing treatment is shown in Fig. 1(a). The waveform of electropulsing was detected in situ by a Rogowski coil and a TDS3012 digital storage oscilloscope (Tektronix Inc., Beaverton, OR, USA) and it was a damped oscillation wave (see Fig. 1 (b)). The maximum current densities J_{max} = 8.302×10^3 A/mm^2 and 5.4×10^3 A/mm^2 were used to titanium aluminides and cold-rolled TA15, respectively.

The microstructures were investigated by optical microscope (OM) and transmission electron microscope (TEM). For optical microcopy analysis, the samples were etched in the reagent of 2% HF+4%HNO_3+94% H_2O (volume fraction).

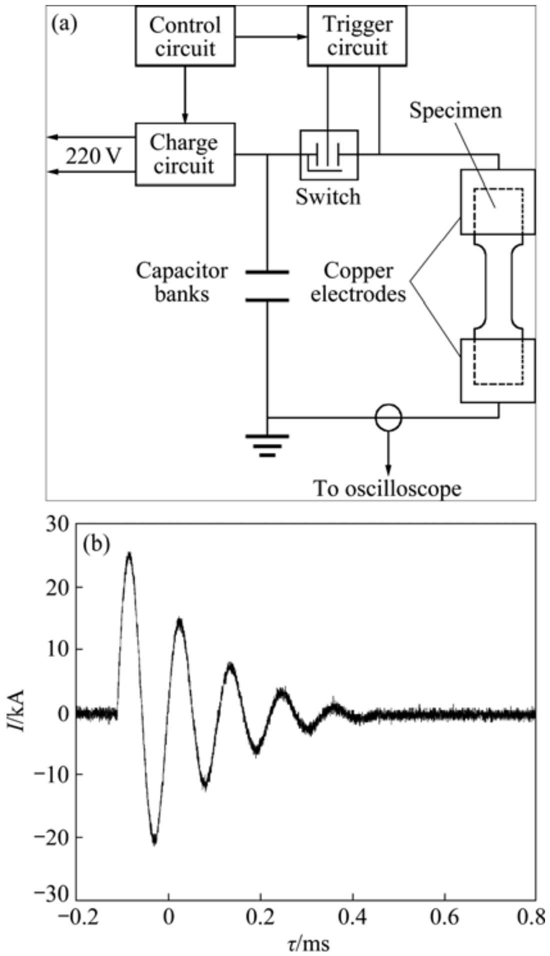

Fig. 1 Schematic of electropulsing experiment: (a) Experimental arrangement; (b) Typical electropulsing waveform

3 Results and discussion

3.1 Analysis of microstructure

The microstructures of the as-cast ingot and treated specimens are shown in Fig. 2. The micro- structure in the as-cast ingot consisted of columnar grains with a width of 1 000 μm and a length of about 2 000 μm. These columnar grains contained alternate γ and α2 plates, exhibiting a lamellar structure (Fig. 2(a)). The electropulsing treatment transformed the structure into a fine structure with an equiaxed grain size of 30−50 μm (Fig. 2(b)).

The optical microstructures of the TA15 before and after treatment are shown in Fig. 3. The microstructure of the as-rolled specimen is α laths grain with fine β-phase grain. Obvious recrystallization occurs in the sheets after electropulsing treatment. The recrystallized grains are very fine, and their distribution is uniform. The microstructure of TA15 sheet transformed from primary α laths grain to α equiaxed grain. It is shown that the completed recrystallization state is obtained at lower temperatures compared with conventional heat treatment.

The TEM images are shown in Fig. 4. It can be seen from Fig. 4 that most of the dislocation arrays became parallel with less dislocation nodes after the electropulsing treatment, and the dislocation density and the tangle degree of dislocation are decreased. It is demonstrated that electropulsing can promote the dislocation motion.

3.2 Discussion

Usually associated with the dissipated motion of electrons, the energy provided by such an external resource (electropulsing) can be changed into heat. The course of temperature rise can be regarded as an adiabatic course due to a short time during the electropulsing treatment, the average temperature rise of the specimen by Joule heat is written as follows [11]:

$$\Delta T = (c\rho S^2)^{-1}\int_0^t \gamma I^2 \mathrm{d}t \qquad (1)$$

where I is the amplitude of pulse, t is the corresponding duration, S is the cross-sectional area of the specimen, γ, ρ, c are the electrical resistivity, the density and the specific heat capacity of the experimental material, respectively. For TiAl, $\gamma=1.2\times10^{-6}$ Ω·m, $\rho=4.0\times10^{3}$ kg/m^3, c=700 J/(kg·°C). According to this expressions, the average temperature rise for TiAl is ΔT=1 589 K (1 316 °C). According to the Ti-Al phase diagram [12], the phase transformation of γ to α takes place during heating in $\alpha+\gamma$ dual phase field of TiAl alloys, which is reason for grain refinement of titanium aluminides because the electropulsing itself can increase the nucleation rate by the decrease in thermodynamic barrier during phase transformation [10].

The physical properties of materials should be changed before and after the phase transformation due to the difference in the crystal structures of the new phase and the host medium. If electric conductivities of the nucleus of the new phase and the host medium are different, the configuration of the electric current will be redistributed after formation of the new phase [13]. For simplicity, the formation of a nucleus with radius a and conductivity σ_1 inside an old phase with radius b and conductivity σ_2, the energy change ΔW_e which arises due to additional work induced by electropulsing during formation of a nucleus of a new phase, is written as follows [10, 14−15]:

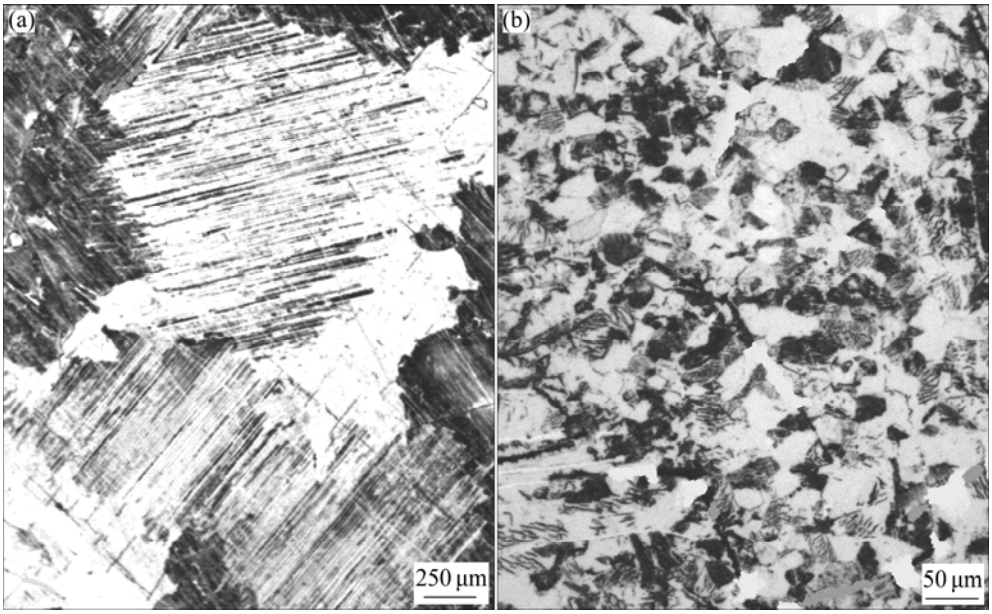

Fig. 2 Microstructures of titanium aluminides: (a) As-cast alloy; (b) Electropulsed alloy

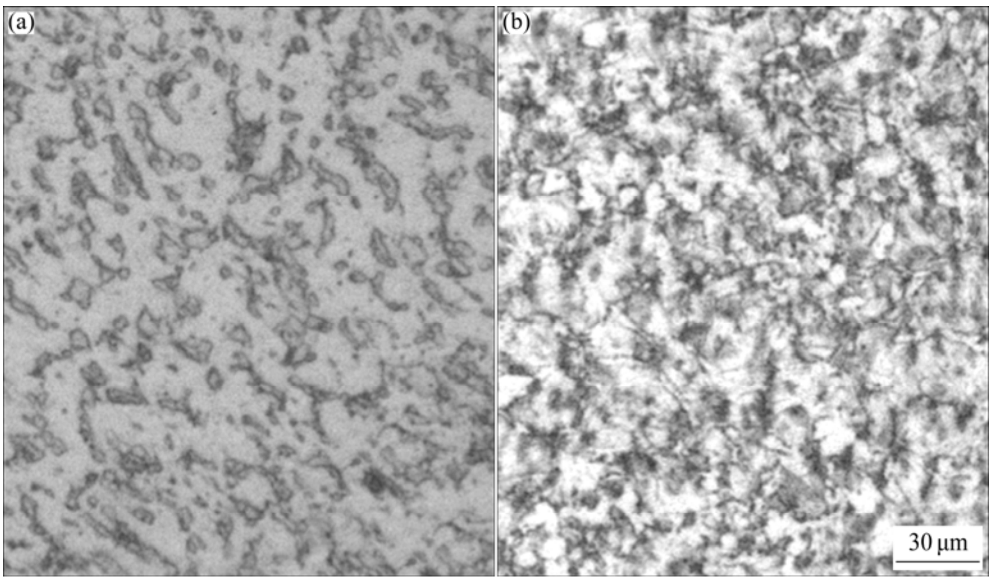

Fig. 3 Optical microstructures of TA15 sheets: (a) Cold-rolled sheet; (b) Electropulsed sheet

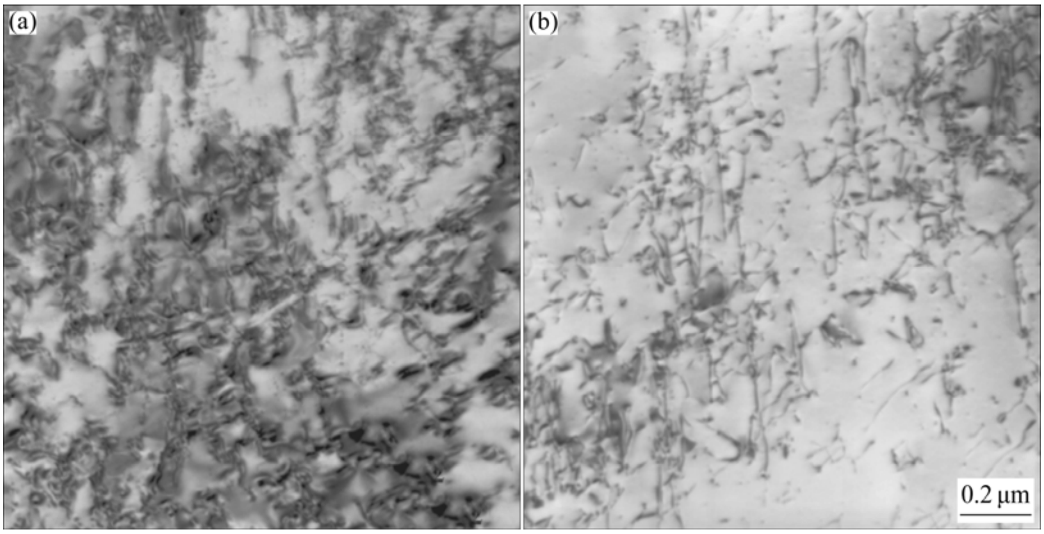

Fig. 4 SEM images showing dislocations of TA15 sheets: (a) Cold-rolled sheet; (b) Electropulsed sheet

$$\Delta W_e = \mu g(a,b)\xi(\sigma_1,\sigma_2)J^2\Delta V \tag{2}$$

where μ is the magnetic susceptibility; J is the current density;

$$g(a,b)=\left[\frac{3}{2}\ln\left(\frac{b}{a}\right)-\frac{65}{48}-\frac{5}{48}\xi\right]b^2$$

is a geometric factor that depends on the parameters of nucleus and medium;

$$\xi(\sigma_1,\sigma_2)=\frac{\sigma_2-\sigma_1}{\sigma_1+2\sigma_2}$$

is a factor that depends on the electrical properties of nucleus and medium; ΔV is the volume of a nucleus.

The electrical conductivity of an intermetallic compound is, in general, relatively small compared with that of metals. This is because the atoms combined with metallic bond give place to covalent one after an intermetallic compound is formed, and electron concentration is reduced [16]. Namely, $\sigma_\alpha=\sigma_1>\sigma_\gamma=\sigma_2$, $\xi(\sigma_1, \sigma_2)<0$. Thus, $g(a, b)>0$, $\Delta W_e<0$.

According to classical nucleation theory, the average number of stable spherical nuclei is given by [10]:

$$n \propto \exp[-W_c/(kT)] = \exp[-\max(\Delta W_0+\Delta W_e)/(kT)] \tag{3}$$

where k is the Boltzmann constant; T is the temperature; W_c is the thermodynamic barrier in forming a spherical nucleus with critical radius; and W_0 is the free energy of a current-free system. Because $\Delta W_e<0$, W_c in a current-carrying system is lower than that in a current-free system, the average number of stable nucle; n can be increased in a current-carrying system. Therefore, numerous fine microstructures (α phase) are formed in as-cast TiAl at high temperature. Because α phase grains are very fine, the volume fraction of grain boundaries is large. This can provide much more nucleation places for γ phase during subsequent cooling (α phase to γ phase). So, the nucleation rate of γ phase is high, and refined γ phase microstructure is formed.

For TA15 sheet, γ=1.63×10^{-6} (Ω·m), ρ=4.45×10^3 kg/m^3, c=755 J/(kg·°C), according to Eq. (1), the average temperature rise during electropulsing is 805 °C, which is lower than the static recrystallization temperature of TA15 (about 950 °C). The result means that electropulsing treatment is not a simple annealing treatment and electropulsing has the effect that a conventional annealing does not have.

It is also well-known that the gliding and climbing of dislocation and migration of atoms are important for the static recrystallization process. The drift electrons can exert a push on dislocations when high density electric pulses pass through the specimen, the force is named electron wind force, and it is proportional to current density [17]. The electron wind force can reduce the dislocation density and enhance the mobility of dislocation, thus can produce a more advanced stage of recrystallization and enhance the nucleation rate of recrystallization. In addition, electropulsing can enhance migration of atoms and reduce strength of the obstacles opposing dislocation motion. This is also a factor that recrystallization occurs in a metal by electropulsing. The TEM images shown in Fig. 4 are the microcosmic expression of these courses as well as. The lower residual dislocation density within the newly formed grains implies that the stored energy is decreased. Retardation of subsequent grain growth resulted from reduction in driving force for the growth of newly recrystallized grains, and then smaller recrystallized grains can be obtained finally.

4 Conclusions

1) The electropulsing treatment can induce the completed recrystallization in cold-rolled TA15sheet at lower temperatures compared with conventional heat treatment. After electropulsing treatment, the microstructure of TA15 sheet transform from primary α laths grain to smaller α equiaxed grain.

2) The as-cast titanium aluminides microstructures are significantly refined when they are treated by the electropulsing. The fine and homogeneous microstructures with the grain size of 30−50 μm can be obtained from the as-cast TiAl based alloy with the initial lamellae of about 1 000 μm.

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References

[1] ZHANG De-ming, CHEN Gui-qing, HAN Jie-cai, ZHANG Xing-hong, YAO Zhen-zhong. Macro-microscopic morphology and phase analysis of TiAl-based alloys sheet fabricated by EB-PVD method [J]. Transactions of Nonferrous Metals Society of China, 2007, 17: 777−782.

[2] QU H, WANG H M. Microstructure and mechanical properties of laser melting deposited TiAl intermetallic alloys [J]. Materials Science and Engineering A, 2007, 466: 187−194.

[3] PARKA H S, NAM S W, KIM N J. Refinement of the lamellar structure in TiAl-based intermetallic compound by addition of carbon [J]. Scripta Materialia, 1999, 41: 1197−1203.

[4] JIN Y G, WANG J N, YANG J, WANG Y. Microstructure refinement of cast TiAl alloys by b solidification [J]. Scripta Materialia, 2004, 51: 113−117.

[5] CHEN Y Y, LI B H, KONG F T. Microstructural refinement and mechanical properties of Y-bearing TiAl alloys [J]. Journal of Alloys and Compounds, 2008, 457: 265−269.

[6] WANG, Y H, LIN J P, HE Y H, WANG Y L, CHEN G L. Microstructure and mechanical properties of as-cast Ti-45Al-8.5Nb-(W, B, Y) alloy with industrial scale [J]. Materials Science and Engineering A, 2007, 471: 82−87.

[7] YANG J, WANG J N, WANG Y, XIA Q F. Refining grain size of a TiAl alloy by cyclic heat treatment through discontinuous coarsening [J]. Intermetallics, 2003, 11: 971−974.

[8] WANG J N, YANG J, WANG Y. Grain refinement of a Ti-47Al-8Nb-2Cr alloy through heat treatments [J]. Scripta Materialia, 2005, 52: 329−334.

[9] WU X H, HU D. Microstructural refinement in cast TiAl alloys by solid state transformations [J]. Scripta Materialia, 2005, 52: 731−734.

[10] ZHOU Y Z, ZHANG W, WANG B Q, GUO J D. Ultrafine-grained microstructure in a Cu-Zn alloy produced by electropulsing treatment [J]. J Mater Res, 2003, 18(8): 1991−1997.

[11] ZHANG W, ZHAO W S, LI D X, SUI M L. Martensitic transformation from α-Ti to β-Ti on rapid heating [J]. Applied Physics Letters, 2004, 84(24): 4872−4874.

[12] ZHU H L, MARUYAMA K, MATSUDA J. Microstructural refinement mechanism by controlling heating process in multiphase materials with particular reference to γ-TiAl [J]. Applied Physics Letters, 2006, 88: 131908−131910.

[13] DOILINSKY Y, ELPERIN T. Peculiarities of coexistence of phases with different electric conductivities under the influence of electric current [J]. Materials Science and Engineering A, 2000, 287: 219−226.

[14] DOILINSKY Y, ELPERIN T. Thermodynamics of phase transitions in current-carrying conductors [J]. Phys Rev B, 1993, 47: 14778−14785.

[15] WANG X L, GUO J D, WANG Y M, WU X Y WANG B Q. Segregation of lead in Cu-Zn alloy under electric current pulses [J]. Applied Physics Letters, 2006, 89: 061910−061913.

[16] ZHANG Yong-yang, HAN Ya-fang, CHEN Guo-liang, GUO Jian-ting, WAN Xiao-jing, FENG Ti. Intermetallic compounds for structural application [M]. Beijing: National Defence Industrial Press, 2001: 486. (in Chinese)

[17] ZHOU Y Z, XIAO S H, GUO J D. Recrystallized microstructure in cold worked brass produced by electropulsing treatment [J]. Materials Letters, 2004, 58: 1948−1951.

脉冲电流处理过程中的相变和再结晶细化晶粒

宋 辉[1]，王忠金[2]

1．哈尔滨工业大学 航天学院力学博士后科研流动站，哈尔滨 150001；
2．哈尔滨工业大学 材料科学与工程学院，哈尔滨 150001

摘 要：在室温下对铸态 TiAl 合金和冷轧 TA15 合金进行高密度脉冲电流处理。应用光学金相显微镜和透射电子显微镜研究脉冲电流处理前、后试样的显微组织。实验结果表明：通过脉冲电流处理可以细化铸态 TiAl 基合金的晶粒，从约 1 000 μm 的原始粗大层片组织，经脉冲电流处理后可以得到尺寸为 30−50 μm 细小、均匀的晶粒。对于冷轧 TA15 合金，脉冲电流处理后发生了完全的再结晶，晶粒组织由原始的 α 板条晶粒转变为细小的 α 等轴晶粒。研究结果表明，脉冲电流处理是一种有效的细化晶粒方法；由于不需要热机处理所要求的挤压等变形工序和高温加热、真空保护等条件，简化了工艺过程。

关键词：脉冲电流处理；晶粒细化；形核率；TiAl 合金；TA15 钛合金

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