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Microstructure and Texture Evolution of Cryo Rolled and Annealed Ductile TaNbHfZrTi Refractory High Entropy Alloy

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***Abstract*—**The microstructure and texture evolution of cryo rolled and annealed ductile TaHfNbZrTi refractory high entropy alloy was investigated. To obtain that, the alloy is severely cryo rolled and subsequently annealed for the recrystallization process. The cryo rolled – 90% shows the presence of very fine grains and microstructural heterogeneity. The cryo rolled samples are annealed at a temperature ranging from 800°C to 1400°C, the partial recrystallization is observed at 800°C annealed condition, and at higher annealing temperatures the complete recrystallization process is noticed. The development of ND fiber texture is observed after the annealing.

***Keywords*—**Refractory high entropy alloy, cryo-rolling, annealing, microstructure, texture.

I. Introduction

THE high entropy alloys (HEAs) consist of a minimum of four metallic elements equiatomic or near equiatomic proportion. Therefore, it contributes higher configurational entropy to the alloy which can help in stabilizing simple solid solutions like BCC, FCC, and BCC+FCC phases [1]-[4]. HEAs show unique properties over conventional alloys. The uniqueness is because of high configurational entropy, severe lattice distortion, cocktail effect, and sluggish diffusion. The refractory high entropy alloys (RHEAs) are a new generation of potential high-temperature materials. It is a known fact that the softening of metallic materials generally occurs at temperatures above about 0.6 Tm of the melting point of an alloy. Then the increase in alloy melting temperature through suitable alloying may increase operating temperature, and thus such melting points of metals and alloys systems make them for application at elevated temperatures beyond Ni, Co, and steel-based superalloys. The high-temperature properties of these alloys are enhanced by selectively choosing the high melting point of individual refractory elements, i.e., group IV (Hf, Zr, Ti), V (Nb, V, Ta), and VI (Cr, Mo, W). The challenge of these RHEAs is poor ductility at room temperature (RT) [5]. The intrinsic design of a perfect crystal is incorporated to avoid brittle failure at room temperature and to improve ductility. The intrinsic ductility of RHEAs is achieved by utilizing electron theory by decreasing the number of valence electrons [4], [6], where the RHEAs show ductile failure mode if they contain VEC ≤ 4.4 and brittle failure mode for VEC ≥ 4.6, for example: (HfNbTiZr (VEC = 4.25) is ductile [7] and MoNbTaVW (VEC = 5.4) is brittle) [5].

Thermo-mechanical processing can affect the development of microstructure, texture, and mechanical properties. By incorporating this process route, one can expect to improve the properties of RHEAs.

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II. Experimental Procedure

A. Processing

The intrinsically ductile refractory high entropy alloy TaHfNbZrTi (VEC = 4.4) was prepared by argon arc melting, starting with high purity constituent elements. Then cold-rolled up to 50% and after that annealed at 1400°C for 10 min in a quartz tube under vacuum to break the as-cast microstructure and refine the grain size. This sample was cryo-rolled up to 90% reduction in thickness in several passes. The cryo-rolled samples were encapsulated in a quartz tube under vacuum and filled with small Ti chips to prevent oxidation during annealing treatment. The cryo-rolled samples were annealed at various temperatures (800°C, 1000°C, 1250°C, and 1400°C) for 1 h followed by water quenching, and these cryo-rolled and annealed samples were used for characterization.

B. Characterization

The cryo rolled and annealed TaNbHfZrTi samples were characterized using an electron backscatter diffraction (EBSD) system (Oxford Instruments, UK) attached to a scanning electron microscope (SEM) (Carl-Zeiss, Germany; Model: SUPRA 40). The specimens for EBSD experiments were prepared using mechanical polishing followed by electropolishing at 18.5 - 20 Volt and current 1.3 – 2.2 Amp (electrolyte: perchloric acid and methanol with 1:9 ratios by volume). For further analysis, the acquired EBSD dataset was exported to the TSL-OIM^{TM} software (EDAX Inc., USA).

C. Results and Discussions

A. Development of Cryo Rolled Microstructure and Texture

In Fig. 1(a), EBSD unique grain color map microstructure obtained after cryo rolling, 90% reduction in thickness reveals a very fine microstructure shown with a white arrow, and it has the average grain size of ~ 0.22 µm (see Table 1). The heavily deformed microstructure shows inhomogeneous microstructure containing regions of fine grains and elongate grains along the RD direction. And also, the formation of shear bands and deformation bands were observed. The evolution of microstructure resulted due to grain subdivision at different length scales. The development of deformation bands and shear bands which are common for BCC materials at high strain deformation. Therefore, with increasing deformation strain, the equiaxed grains transform into a needle-like structure and orient parallel to rolling direction (RD), and also a rotation of different parts of the grain to different end orientations, results in the development of local crystallographic texture. A large amount of strain is accumulated by dislocations and by the formation of new boundaries. The LAGBs convert into deformation-induced HAGBs due to the accumulation of dislocations. The grain subdivision process results in grain refinement and fine microstructure [8], [9].

Fig. 2(a) shows φ2 = 45° section of ODF of a 90% cryo-rolled sample. The ODF section of cryo rolled 90% shows the presence of continuous ND fiber and discontinues RD fiber texture. The deformation texture of single-phase BCC metals and alloys is characterized by the development of RD (RD//<110>) and ND (ND//<111>) fiber texture [9]. The strengthening of the RD-fiber components increases uniformly up to 70% deformation; the ND-fiber does not strengthen significantly up to a cold-rolling 80%. The further deformation of the ND-fiber component is preferentially strengthened [9].

B. Evolution of Annealed Microstructures and Texture

The microstructure of TaHfNbZrTi annealed at 800°C for 1 h (Fig. 1(b)) shows a partial recrystallization process. The presence of very fine grain clusters inside the deformed region indicates the nucleation of strain-free fine grains accompanied by its growth, which are pointed out with circles and arrows in the microstructure and having an average grain size of ~ 1.27 µm (Table 1). Subsequently, further annealed at 1000°C for 1 h microstructure (Fig. 1(c)) undergoes complete recrystallization process and grain growth than the previous temperature has an average grain size of ~ 29.21 µm (Table 1). Interestingly the microstructure has regions of small and large grains distribution marked with white circle and arrow, respectively, which reveals that larger grains consume smaller grains during the grain growth process and some regions in the microstructure show equiaxed grains. As the grains are becoming larger, the shape of the grain is changing and becomes equiaxed. A similar kind of trend is observed when a sample is annealed at 1250°C for 1 h (Fig. 1(d)), the microstructure shows almost complete consumption of small grains (indicated by arrow) by large surrounding grains in the process of grain growth mechanism, the average grain size at this annealing temperature is ~ 22.12 µm. The slight decrease in grain size featured than the previous annealed temperature, in general, the grain size increases with increasing annealing temperature. This reduction of grain size is anticipated due to the following two reasons: the first reason is grain pinning by texture and the second reason can be the shape of the grains. Both of these reasons could resist the grain growth and microstructures coarsening. Understanding the evolution of these kinds of microstructures in detail requires TEM studies. And a similar way of grain growth happened for annealing at 1400°C for 1 h (Fig. 1(e)), at this temperature, grains are continuing to grow extremely larger due to the effect of high annealing temperature, the average grain size is ~ 51.47 µm (Table 1). The partial recrystallization at 800°C for 1 h was observed since the annealed temperature might be lower than the recrystallization temperature. On increasing the annealing temperature to 1000°C for 1 h, the microstructure evidences the full recrystallization process, which indicates the annealing temperature is above the recrystallization temperature. The gradual increase in annealing temperature assists the grain growth, which can be explained by an increase in the average grain size and irregular grains turning into five and six-sided stable facets [9], [10]. The grain boundary energy act as a driving force for the grain growth process helps in coarsening of microstructure. The annealed at 1400°C temperature for 1 h showed the highest grain growth due to high diffusivity assisted by ultra-high annealing temperature.

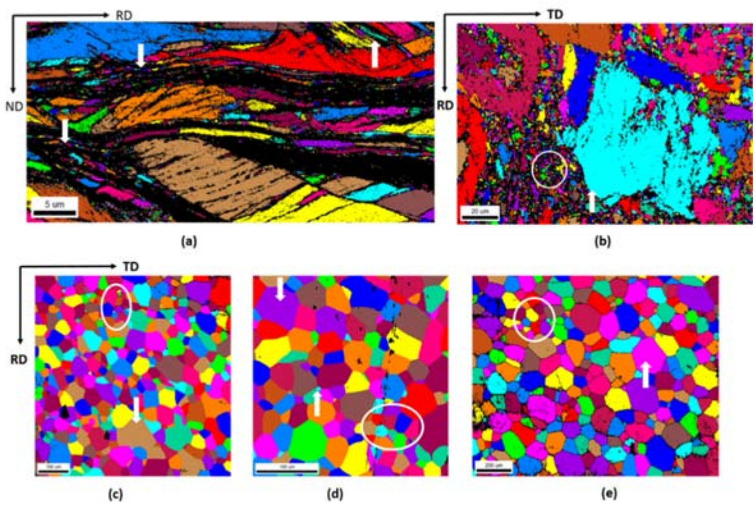

Fig. 1 EBSD unique grain color map of (a) cryo rolled 90% and annealed at (b) 800°C, (c) 1000°C, (d) 1250°C, and (e) 1400°C for 1 h, respectively

TABLE I
THE AVERAGE GRAIN SIZE OF TAHFNBZRTI CRYO ROLLED AND ANNEALED TEMPERATURES

	CRYO-90%	800ºC	1000ºC	1250ºC	1400ºC
Average Grainsize(µm)	0.22	1.27	29.21	22.12	51.37
Standard Deviation	0.12	1.97	18.60	15.02	41.58

The annealing texture at various temperatures obtained after cryo rolling 90% thickness reduction shown by $\phi 2 = 45^o$ constant ODF section (Fig. 2(b) to Fig. 2(e)). The TaHfNbZrTi refractory high entropy alloy annealed at 800°C for 1 h saw the development of strong ND fiber texture containing high intensity at {111} <112>. Further annealing of the alloy at 1000°C for 1 h shows continuous evolution of strong ND fiber texture and shifting maximum intensity about {111} <110> and also developing weak RD fiber texture.

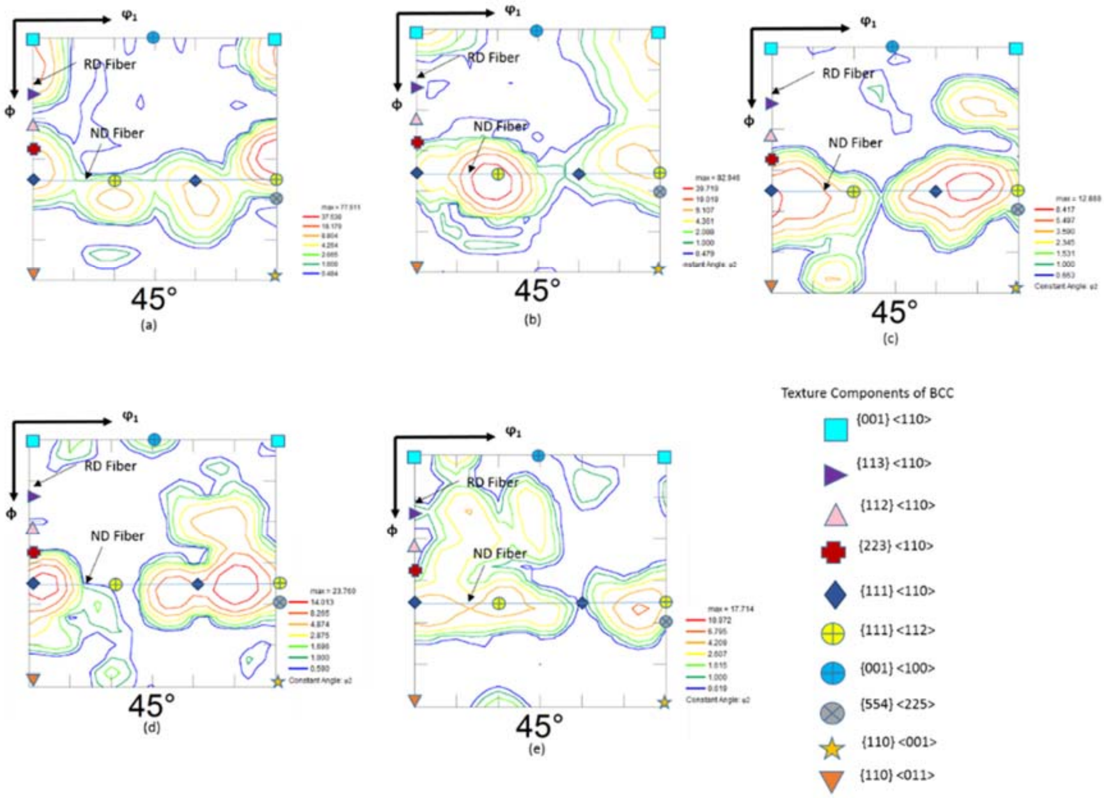

Fig. 2 $\varphi 2 = 45$ sections of ODFs of (a) cryo rolled 90% and annealed at various temperatures at (b) 800°C, (c) 1000°C, (d) 1250°C, and (e) 1400°C for 1 h, respectively

Increasing the annealing temperature to 1250°C and 1400°C for 1 h shows the development of discontinuous ND fiber texture as well as weak RD fiber texture, small intensity contours are present at various locations. The development of ND-fiber components on annealing is attributed to recrystallization [9], [11], which is the usual behavior of annealing texture formation in BCC materials. The deformed RD-fiber components show recovery behavior results in ND-fiber strengthening than the RD-fiber components after recrystallization.

D. CONCLUSION

The TaHfNbZrTi refractory HEA is successfully processed by cryo rolled up to a high strain level. The 90% cryo-rolled microstructure showed deformation heterogeneity and development of RD fiber texture. The partial recrystallization is observed on annealing at 800ºC. Annealing samples at higher temperatures showed a significant grain growth mechanism and strengthening of ND fiber texture.

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