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# Development of Impressive Tensile Properties of Hybrid Rolled $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ Refractory High Entropy Alloy

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***Abstract*—**The microstructure, texture, phase stability, and tensile properties of annealed $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ alloy have been investigated in the present research. The alloy was severely hybrid-rolled up to 93.5% thickness reduction, subsequently rolled samples subjected to an annealing treatment at 800 °C and 1000 °C temperatures for 1 h. Consequently, the rolled condition and both annealed temperatures have a body-centered cubic (BCC) structure. Furthermore, quantitative texture measurements (orientation distribution function (ODF) analysis) and microstructural examinations (analytical electron backscatter diffraction (EBSD) maps) permitted to establish a good relationship between annealing texture and microstructure and universal testing machine (UTM) utilized for obtaining the mechanical properties. Impressive room temperature tensile properties combination with the tensile strength (1380 MPa) and (24.7%) elongation is achieved for the 800 °C heat-treated condition. The evolution of the coarse microstructure featured in the case of 1000 °C annealed temperature ascribed to the influence of high thermal energy.

***Keywords*—**Refractory high entropy alloys, hybrid-rolling, recrystallization, microstructure, tensile properties.

## I. Introduction

THE high entropy alloys (HEAs) consisted of 5 to 20 elements with equiatomic or near equiatomic proportion, because of the more number of alloying elements attribute to high configurational entropy. The high configurational entropy contributes to the decrease of Gibbs free energy and restricts the formation of intermetallics and stabilizes a single solid solution phase, such as BCC, FCC and HCP structures [1], [2]. The HEAs show better properties such as high strength, excellent softening, hardness, corrosion and wear resistance by contributing from four core effects [2]. Demand and pursuit of materials with higher strength, larger ductility, and high thermal stability (> 1300 °C) has never faded for aerospace applications and scientific curiosity. The new generation of the refractory high-entropy alloy (RHEA) came up with the concept of the high-entropy-alloys. Nevertheless, these refractory high entropy alloys promise to be new superalloys because of their substantial mechanical properties at high temperatures. From the literature, it was found that the RHEAs usually based on refractory elements like TaVWZrHfNbCrMo, have a stable microstructure and significant softening resistance at high temperatures even better than conventional Ni-based superalloys [2]. Simultaneously, so many refractory high entropy alloys (RHEAs) show extremely poor tensile ductility (less than 5%) at room temperature since over decade it has become a back-breaking problem for the processing and restraint their practical applications [2], [3]. Sincere efforts have been extended in this area, and a few techniques, such as decreasing the sample size and minimizing valence electron numbers, have been suggested. However, a little tensile ductility was achieved only in very few alloy systems. The $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ alloy has good ductility and density as well as an estimated (by rule of mixture) melting point is about 2055 °C; it could be a prominent alloy compared to some other RHEAs [4]. There were limited research studies available on deformation, microstructure, and mechanical properties of $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ alloy. Fascinatingly $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ alloy was severely hybrid rolled at room temperature and followed by cryo temperature (liquid $N_2$), and succeeding annealing effect on the phase stability, microstructure, texture, and mechanical properties development was investigated in the present research work.

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## II. Experimental Procedure

### A. Processing

The material used for the investigation $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ was produced by arc melting, starting with high purity individual constituent elements. The piece of as-cast material hybrid-rolled 93.5% reduction in thickness in multiple passes, which is equal to the strain of $\varepsilon = 3.2$ (initially cold rolled up to $\varepsilon = 1.6$ and then cryo rolled (in liquid $N_2$) $\varepsilon = 1.6$). The rolled specimens are then subjected to annealing at temperatures 800 °C and 1000 °C for 1 h, followed by water quenching. The hybrid rolled specimen encased in a quartz tube under vacuum to prohibit oxidation while annealing treatment. Annealed specimens mechanically polished to take off if any oxide surface is present after that used for further characterization.

### B. Characterization and Mechanical Testing

The phase analysis of the $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ samples was carried out using X-Ray Diffraction (XRD) with (Rigaku Ultima model). However, Cu source λ is 1.54 A° utilized for rolled samples and Co source λ is 1.78 A° utilized for annealed specimens. The microstructure and texture studies of annealed specimens were accomplished using a scanning electron microscope (SEM) (Carl-Zeiss, Germany; Model:

SUPRA 40) attached with an EBSD system (Oxford Instruments, UK). The samples for EBSD experiments prepared using mechanical polishing followed by electropolishing at room temperature (electrolyte: perchloric acid and methanol with 1:9 ratios by volume) (note: electropolishing is painful). AztecHKL software (Oxford Instruments, UK) used for acquiring the EBSD scans. The acquired EBSD dataset exported to the TSL-OIM™ software (EDAX Inc., USA). The harmonic series expansion method (series rank = 22) utilized to calculate ODFs of EBSD dataset. The volume fractions of various texture components implemented by a cut-off angle of 15°. The tensile test was carried out at $10^{\wedge -3}$ mm/sec strain rate using (Instron 5967, embedded with DIC) at room temperature.

## III. Results and Discussion

### A. Microstructure and Texture Development

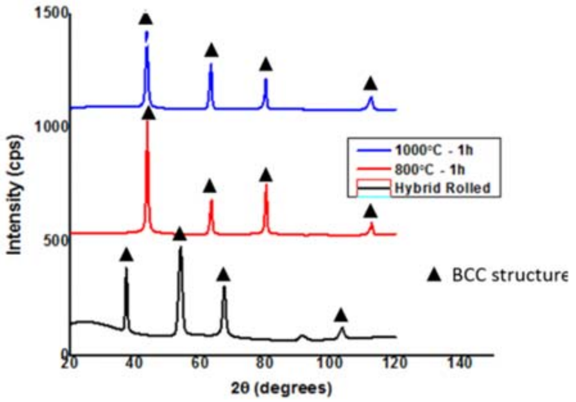


Fig. 1 XRD graph of $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ hybrid-rolled and annealed at 800 °C and 1000 °C temperatures for 1 h

The structure evolution and phase constitution of the $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ alloy were characterized by EBSD and X-ray diffraction (XRD) analysis, and the outcomes demonstrate the targeted change of phase stability as shown in (Fig. 1). A single BCC phase structure was acquired for all the conditions consisting only BCC diffraction patterns on its XRD trace. Furthermore, the XRD results reveal that there was no phase transformation occurred. In order to interpret the recrystallization and grain growth mechanisms that influence the development of the recrystallization texture from the deformation, one microstructural investigation was carried out utilizing electron microscopy. First, the annealing conditions accompany to complete recrystallization. The further grain growth process was also explored by hiking the annealing temperature. The examined annealing temperatures at 800 °C and 1000 °C, as seen in Figs. 2 (a) and (b), shows microstructures development during isochronal annealing for 1 h of hybrid rolled materials. The material becomes explicitly recrystallized (hybrid rolled 93.5%) after annealing at 800 °C. After recrystallization process accomplished, fine-grain and equiaxed microstructure is obtained as seen from the microstructure and composed of approximately equiaxed grains with a mean size 6.55 ± 4.2 µm (Table I) and distribution (Fig. 3). Therefore, heterogeneous microstructural features appeared as regions consisting of proportionally large recrystallized grains (marked by yellow arrow) in Fig. 2 (a) existing with regions incorporate of small grains (enclosed by yellow circles). Moreover, prevailing similar kind of morphology features in Fig. 2 (b) and relatively coarse microstructure evolution resulted for 1000 °C annealed temperature than 800 °C annealed condition has a mean grain size of 48.35 ± 29.9 µm (Table I) and distribution (Fig. 3). It is transparent that grain size found to increase with temperature, stipulates that the temperature actively contributed to grain growth. Heterogeneous microstructures development for both heat-treated conditions can be the existence of the inhomogeneous microstructure for the severely rolled material; recrystallization proceeds heterogeneously and starts in the highest stored energy regions. The nuclei of recrystallized grains originate from the pre-existed deformed state. In polycrystalline materials during the deformation process, the single grain inhomogeneously breaks up into regions of subgrains, cells, shear bands, and deformation bands, which are high energy regions and act as potential sites for recrystallization [5], [6]. A significant driving force provided for recrystallization is by the higher stored energy regions in the microstructure. Interestingly few recrystallized grains in both annealed microstructures are aligned along the 35°–65° direction (marked by white ellipse in Figs. 2 (a) and (b)) from the rolling direction. The new recrystallization grains nucleate favorably in the shear bands. Nucleation of recrystallization occurring favorably in shear bands has already been observed in many other alloys. Shear bands are the regions where deformation is enormously concentrated, are favorable sites for origin of recrystallization [5]. The microstructures evolution for the case of both annealed temperatures continues to show non-uniformity or inhomogeneity revealed by large preferentially grown recrystallized grains, and at the later stage of grain growth proceeds followed by the abnormal grain growth process. Thoroughly after completion of the recrystallization process grains involves abnormal grain growth [5], [6].

The crystallographic texture is accountable for the directionality of properties; their origin and applications are a source of scientific curiosity. The recrystallization texture develops when deformed materials are subjected to the annealing process. Texture interpretation using ODF calculations was performed to determine the appropriate intensity and spread of each component. A typical deformation and recrystallization texture in BCC materials is represented by φ2 = 45° section of ODFs.

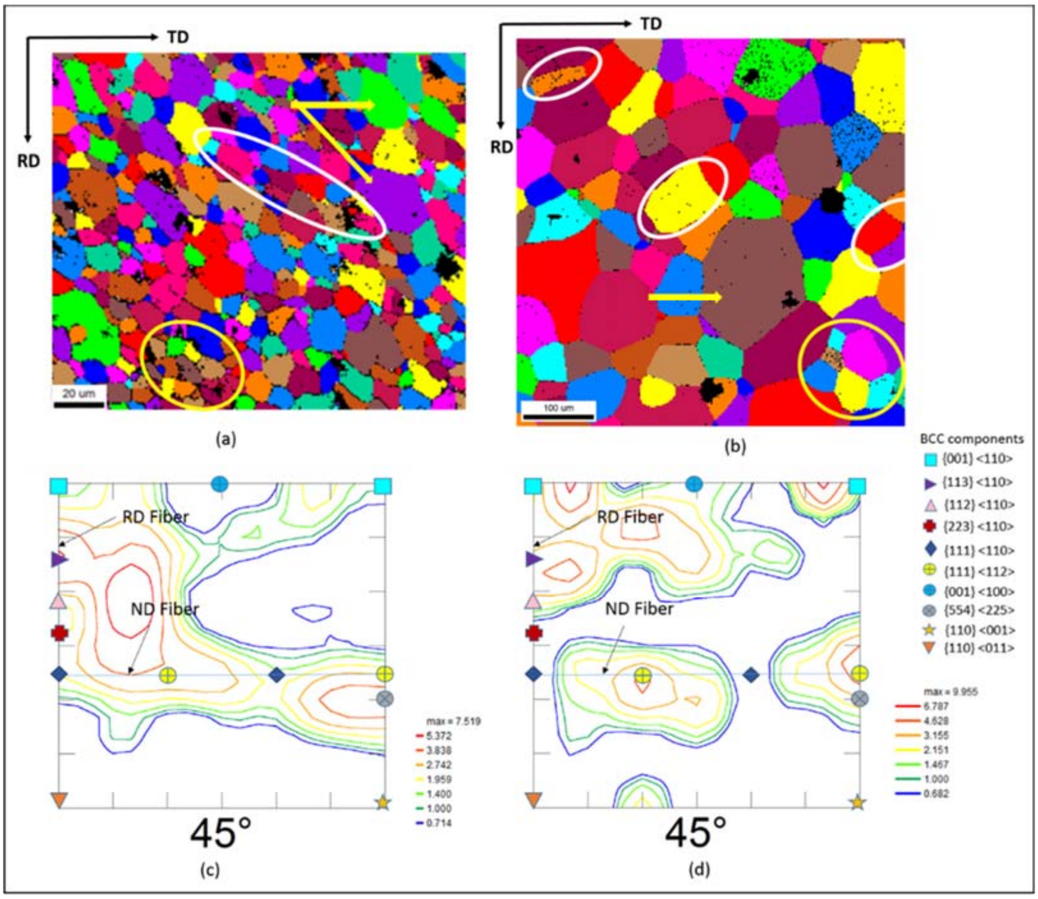


Fig. 2 (a), (b). EBSD unique grain color map and (c), (d) φ2 = 45° sections of ODFs of $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ annealed at 800°C and 1000°C temperatures for 1 h

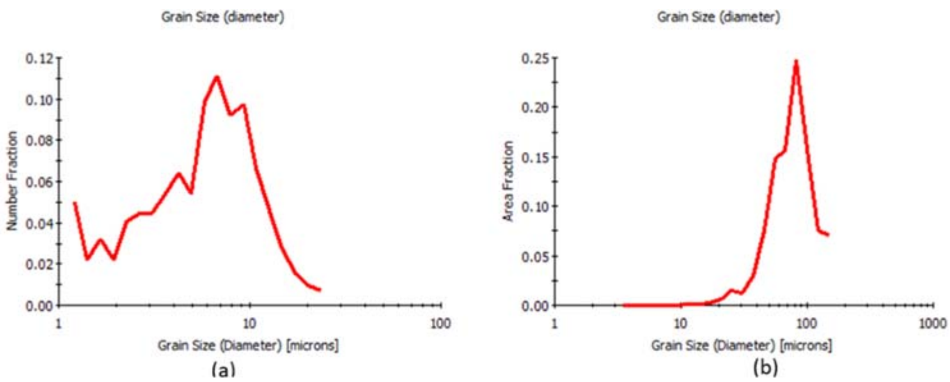


Fig. 3 The grain-size distribution of $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ annealed at (a) 800°C and (b) 1000°C temperatures for 1 h

TABLE I
THE AVERAGE GRAIN SIZE OF $TA_{0.5}NB_{0.5}HF_{0.5}ZRTI_{1.5}$ ANNEALED TEMPERATURES

| | 800 °C | 1000 °C |
|---|---|---|
| Average Grainsize (µm) | 6.55 | 48.35 |
| Standard Deviation | 4.20 | 29.90 |

The ODF results showed the ND and RD fiber texture development for the 800 °C annealed temperature in Fig. 2 (c), and eventually annealed at higher temperature 1000 °C showed the development of RD fiber and discontinued ND fiber texture in Fig. 2 (d). The recrystallization components conventionally follow the RD and ND fiber axis. The ND fiber texture with axis <111> is parallel to the sheet normal, the important components in ND fiber texture have <110> and <112> oriented with the rolling-direction and RD fiber texture with axis <110> is parallel to the rolling direction and maximum intensities are at {001}, {113}, {112}, {223} and {111} [6], [7]. The evolution of ND-fiber orientations after annealing favored by recrystallization and its orientations commonly undergo easy recrystallization because of their excessive stored energy, which is the usual behavior of annealing texture formation BCC materials [6], [7]. In the present investigation, it was clearly noticeable about both of these following annealing microstructure and texture development. On the one hand, it was revealed genuine complete recrystallization, inhomogeneous microstructures development featuring a difference in grain sizes, and preferential recrystallization. On the other hand, it was clearly shown that based on texture considerations, it could be put forward to explain the retention of the recrystallization texture upon annealing.

### *B. Mechanical Properties*

Fig. 4 shows the engineering stress-strain curves of the $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ alloy of hybrid rolled 93.5% and as well annealed at 800 °C and 1000 °C temperatures for 1 h. Distinct variation can be seen in the tensile graph of the alloy with different processed conditions.

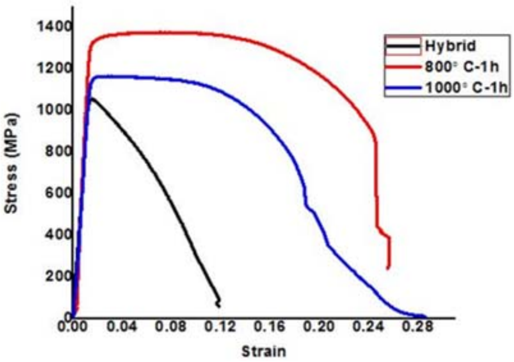


Fig. 4 Tensile graph of $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ alloy hybrid rolled 93.5% and annealed at 800 °C and 1000 °C temperatures for 1 h

Severely hybrid rolled condition has surprising tensile properties, shows yield and tensile strengths of 852 MPa and 1060 MPa, respectively, along with significant elongation of 11.8%. The most striking tensile results obtained in the present research are at 800 °C and 1000 °C annealed temperatures. The annealed specimen at 800 °C temperature reveals simultaneous enhancement of both tensile strength and elongation. Incredible yield strength, tensile strength, and elongation are 842 MPa, 1380 MPa, and 24.7%, respectively (in Fig. 3), which are fascinating tensile properties compared to previously developed RHEAs. Further annealed at 1000 °C temperature also shows attractive tensile properties compare to other refractory HEAs has 834 MPa yield strength, tensile strength (1170 MPa), and extensive elongation (26.2%) shown in Fig. 4. It is clearly noticeable that the tensile strength reduced than 800 °C annealed temperature, but elongation increased. Both of the annealed 800 °C and 1000 °C temperatures show more uniform elongation and contains superior tensile properties than severely rolled condition having higher strength and elongation. The high yield strength for the hybrid rolled condition may be considering small grain size induced during heavy rolling, and decreasing yield strength with annealing temperature can be due to the coarsening of recrystallized microstructure well-known phenomena observed by many researchers. The tensile strength can be influenced by strain hardening; in the past, most of the experimental studies carried out on different metallic materials found that the enhancement of tensile strength with an increase in strain hardening. The amount of uniform elongation is influenced by deformation mode in polycrystalline materials, based on whether homogeneous or inhomogeneous deformation mode prevails in the tensile loading [8], [9]. However, homogenous deformation suspends early necking and fracture. In the case of 800 °C and 1000 °C annealed temperature featuring uniform elongation predicted by manifest of homogeneous deformation, whereas non-uniformed elongation for hybrid rolled condition maybe because of the assistance of heterogeneous deformation. Overall, the exciting tensile properties of $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ RHEA of hybrid rolled 90% and annealed at 800 °C, and 1000 °C temperatures prodigious containing a better combination of strength and ductility compared to other brittle RHEAs

## IV. Conclusion

The $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ RHEA was able to be severely hybrid-rolled. Both annealed temperatures have a single-phase BCC structure. Moreover, annealed conditions contain recrystallized inhomogeneity in the microstructure, regions containing relatively large recrystallized grains and small grains. A coarse microstructure prevailed for 1000 °C annealed temperature because of a higher thermal energy effect. The recrystallized microstructures showed the development of ND fiber and RD fiber textures for both annealed temperatures. Remarkable tensile properties development confirmed the Ta0.5Nb0.5Hf0.5ZrTi1.5 alloy. The excellent combination of tensile strength and elongation and also easy manufacturing process of the $Ta_{0.5}Nb_{0.5}Hf_{0.5}ZrTi_{1.5}$ alloy is promising as a metallic superalloy for high-temperature applications.

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