Microstructure evolution in a CuZr alloy and CP Ti processed by a novel technique of free bending in rotating rollers

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Abstract. A novel technique referred to as free bending in rotating rollers was employed to refine the initial structure and to produce gradient fine-grained microstructures in the Cu0.5Zr alloy and commercially pure titanium (CP Ti). A newly designed die allowing a continuous and repetitive movement of a workpiece through a set of rollers in a channel bent by 90 degrees was employed to produce a series of billets after different numbers of passes resulting in different grains imposed on the billet. Microstructure evolution with increasing number of passes was characterized by precise measurements of microhardness (HV) distribution along the cross section of individual billets. The heterogeneous distribution of HV observed after a single pass was transformed to almost a uniform one in the whole cross section of the billets after 8 passes. EBSD analysis and TEM observations revealed a significant microstructure refinement in both materials after the final stage of processing. A homogeneous microstructure with an average grain size of about 300 nm was observed in CP Ti, whereas the microstructure of the Cu0.5Zr alloy was not fully refined and exhibited a bimodal grain size distribution. The experimental results were compared with the theoretical modelling of strain distribution by FEM.

1. Introduction
In the last decades, the grain size strengthening was widely used for increasing the mechanical performance of metallic materials. New methods of material processing and preparation were developed in order to obtain the refined microstructures in the ultrafine or even nanometer range [1,2]. Severe plastic deformation (SPD) techniques, which imposed a large plastic deformation of hundreds of percentages was found to be successful in this regard [3,4]. SPD-processed materials with FCC and BCC crystal structures usually exhibit improved strength at the expense of deterioration of ductility [5,6]. On the other hand, SPD methods were successfully employed to increase both the strength and ductility of magnesium alloys with a HCP crystal structure due to the grain refinement and the texture formation, which enables the easier activation of non-basal slip systems [7,8].

Preparation of a gradient structure was found to be another way of increasing both strength and ductility. Materials with a gradient structure, e.g. the grain size in the nanoscale level on the surface and a coarse-grained structure in the center of the billet, were found to be attractive for many practical applications. The combination of resistant surface layers with a plastic core provides enhanced strength and wear resistance simultaneously with increasing ductility and bending fatigue resistance [9,10].
The main aim of the present work is two-fold: i) to prepare a gradient structure by free bending in rotating rollers [11] in the Cu0.5Zr alloy and CP Ti, and ii) to investigate the possibility to produce a homogeneous, refined microstructure in the whole cross section of the billet by applying a higher number of passes.

2. Experimental material and procedures

The Cu-0.5 wt% Zr alloy and commercially pure Ti (grade 4) were supplied in the condition produced by hot rolling and subsequent mechanical treatment. Billets of the cross section of 10x10 mm were pressed through a die schematically shown in figure 1. This die contains a set of rollers forming a channel in which the sample is bent by 90 degrees during a single pass. A series of billets after 1 and 8 passes using rotation of the billet by 90 degrees after each pass (equivalent to route Bc used in the ECAP technique) was prepared. Cu0.5Zr and CP Ti specimens were processed at a temperature of 300 °C. Microstructure characterization of the initial state and samples after 8 passes was performed by transmission electron microscopy and EBSD on specimens taken from the central part of the cross section. TEM microscopy was performed on a Jeol 2000FX transmission electron microscope operating at 200 kV. EBSD maps were collected by a SEM FEI Quanta 200 electron microscope equipped with an EDAX EBSD camera. Microstructure evolution with increasing number of passes was investigated in detail by microhardness measurements along the plane perpendicular to the processing direction – the billet cross section. Vickers microhardness mapping under an applied load of 500 g and a dwell time of 10 s was measured using an automatic microhardness tester Qness Q10a. A regular network of indenters consisting of an orthogonal square net with the lateral distances of 300 µm (1 and 8 passes) and 500 µm or 1000 µm (initial states of CP Ti and Cu0.5Zr) where used. Prior to all measurements, the surface of the Cu0.5Zr alloy was mechanically polished successively using 1200, 2400 and 4000-grit SiC papers followed by polishing on diamond suspensions with a particle size of 3 µm and 1 µm. The final treatment consisted of polishing by 0.04 µm colloidal silica suspension. CP Ti samples were first ground using 1200, 2400, 4000-grit SiC papers and then polished on the colloidal alumina suspension with decreasing particle size down to 0.02 µm for over 16 hours on a Vibromet machine. Mechanically thinned TEM foils to a thickness about 120 µm were prepared by twin jet electropolishing in a solution of 6% HClO₄ and 33% butyl alcohol in methanol at a temperature of -20 °C.

3. Experimental results and discussion

3.1 Microhardness

Figure 2 shows the Vickers microhardness maps of the initial samples and those processed by free bending in rotating rollers. The variations of microhardness across the cross section of the billets are clearly displayed by the color code at these maps. The variation of the HV values after a single pass is similar for both materials. The microhardness increases preferably at the top part of the billet (deformed in compression). Further, a gradual decrease of HV from the edge to the central part is apparent. On the other hand, the bottom part of the billet (deformed in tension) exhibits a comparable increase of HV only in a very thin layer (thickness less than 1 mm) and outside this layer the HV rapidly decreases towards the central part. This dominant strengthening in the peripheral parts of the cross section is consistent with the results of numerical simulations performed for Cu-0.18 wt% Zr alloy, where the maximal values of strain were observed at the peripheral areas of the cross section [12]. After a total of 8 passes both materials exhibit almost a uniform distribution of HV in the whole cross section of the billet. The microhardness of CP Ti increases from 240 (initial state) to about 290 (8 passes). Such an
increase in HV is comparable with the CP Ti processed via the ECAP technique [13]. On the other hand, a much higher increase in HV of about 200% was observed in the Cu0.5Zr alloy.

![Figure 2](image_url) Microhardness maps measured on the cross section of (a) CP Ti and (b) Cu0.5Zr alloy. Black dots inside the maps represent the individual measured points.

### 3.2 Microstructure

Microstructure observation revealed a significant grain refinement in both materials processed by 8 passes via free bending in rotating rollers, cf. figure 3 and figure 4. As it is apparent from figure 3b, a fully refined, homogenous microstructure was observed in the central part of CP Ti. The microstructure consists of equiaxed grains separated by HAGBs with a size below 400 nm. A typical grain size obtained in CP Ti processed by ECAP is usually about 300 nm [14]. Moreover, numerous heavily deformed grains, containing a high dislocation density, are clearly visible in the micrograph.

![Figure 3](image_url) Bright-field TEM micrographs of CP Ti (a) in the initial state and (b) after 8 passes.

On the other hand, the microstructure of the Cu0.5Zr alloy after 8 passes exhibits a bimodal grain size distribution, cf. figure 4b. The microstructure contains a high fraction of low-angle grain boundaries (misorientation lower than 15 degrees) highlighted by the white color, indicating the dislocations arranged into dislocations cells or walls. Such a microstructural feature is common for FCC materials processed via methods of SPD [15]. The average grain size in the Cu0.5Zr alloy (taking into account
grains separated by HAGBs) decreased from about 150 μm in the initial state to 4 μm after 8 passes of free bending in rotating rollers.

![Figure 4](image.png)

**Figure 4.** Inverse pole figure maps of the Cu0.5Zr alloy (a) in the initial state and (b) after 8 passes.

### 4. Conclusions

The present paper investigated the microhardness and microstructure evolution in the Cu-0.5 wt% Zr alloy and commercially pure Ti processed by free bending in rotating rollers. The following conclusions can be drawn from this experimental investigation:

- The gradual decrease of HV from the upper edge to the central part of the billet after a single pass of free bending in rotating rollers indicates the presence of a gradient type microstructure in both materials.
- The formation of a gradient fine-grained microstructure during one pass is consistent with the theoretical modelling of strain distribution by FEM.
- The application of 8 passes leads to an almost homogeneous distribution of microhardness values in the whole cross sections of the samples.
- A UFG microstructure with grains having an average size of 200-500 nm was achieved in CP Ti processed by 8 passes.

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