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Microstructure and texture of heavily cold-rolled and annealed fcc equiatomic medium to high entropy alloys

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ABSTRACT

The evolution of microstructure and texture after heavy cold-rolling and annealing was investigated in FCC equiatomic medium to high entropy alloys. For this purpose medium entropy ternary CoFeNi and quaternary CoCrFeNi, and high entropy quinary CoCrFeMnNi alloys were cold-rolled to 90% reduction in thickness and annealed at different temperatures. The ternary alloy showed the development of lamellar microstructure and pure metal type texture after heavy cold-rolling. In contrast, the quaternary and quinary alloys showed heavily fragmented microstructure and brass dominated texture. After annealing, the ternary alloy showed abnormal grain growth at lower annealing temperatures and strengthening of the cube component ({001}<100>) with increasing annealing temperature. In contrast, the quinary and quaternary alloys showed more homogenous grain growth and only minor changes in the volume fraction of different texture components during annealing. The observed differences in the microstructure and texture formation during cold-rolling could be understood considering the differences in the stacking fault energy of the three alloys. On the other hand, the evolution of microstructure and texture of the quarternary and quinary alloys was affected by the sluggish diffusion behavior.

1. Introduction

High entropy alloys (HEAs) are multicomponent alloys based on the novel alloy design philosophy of mixing a large number of elements in equiatomic or near equiatomic proportions [1]. Surprisingly, despite having the presence of a large number of components, the HEAs may show rather simple crystal structures such as, FCC (e.g. equiatomic CoCrFeMnNi [2]), BCC or FCC + BCC [1]. This rather interesting behavior is explained on the basis of large configurational entropy of mixing of elements in equiatomic proportions [1]. However, it may be worth mentioning that recent investigations show that many multicomponent alloys can show the presence of ordered intermetallic phases [3].

Depending on the entropy of mixing, HEAs are defined as those having at least five elements, while alloys containing three to four elements in equiatomic proportion may be classified as medium entropy alloys [4]. Intensive investigations on HEAs are presently being carried out which have led to the discovery of many interesting and attractive properties [4–10]. The origin of these unique properties in HEAs is explained on the basis of the core effects of multicomponent solid solution formation, including distorted lattice structure [4], cocktail effect [4,5], sluggish diffusion [4,11,12] and extensive formation of deformation nano-twins [6,13].

An important area of research in HEAs is understanding the thermo-mechanical processing behavior of these alloys [14–21], including phase stability [2,22–28], microstructure and properties [13,29–35]. In order to fully unearth the effect of massive multicomponent solid solution formation in HEAs, it is important to understand and highlight the differences of HEAs with medium and low entropy alloys [32,36]. Wu et al. [32] have investigated the phase formation, recovery and recrystallization in various binary, ternary, quaternary and quinary alloys made from the constituent elements of the well-known equiatomic FCC single phase CoCrFeMnNi HEA system [2]. This particular study revealed phase stability in low to medium entropy alloys and thermo-mechanical processing behavior, particularly microstructure evolution during recovery, recrystallization and grain growth [32]. However, the evolution of texture which is an important aspect during thermomechanical processing of materials, has not been reported in that study. It is evident that understanding and control of microstructure and texture is important for controlling the properties of the

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HEAs. In order to bridge the gap, in the present work we systematically investigate the formation of microstructure and texture after cold-rolling and annealing in equiatomic FCC single phase medium to high entropy alloys, namely equiatomic ternary CoFeNi, quaternary CoCrFeNi and quinary CoCrFeMnNi having configurational entropy of mixing of 1.09R, 1.38R and 1.61R, respectively (R is the universal gas constant).

2. Experimental

2.1. Processing

The equiatomic FCC ternary, quaternary and quinary alloys used in the present investigation were prepared by vacuum arc melting and casting route. The as-cast alloys were subjected to homogenization treatment at 1100 °C for 6 h (hrs). The three homogenized alloys showed typical coarse microstructure of cast alloys. In order to break down the cast microstructures, samples with dimensions 25 mm (length)×8 mm (width)×5 mm (thickness) were obtained from the respective homogenized bars and cold-rolled to 50% reduction in thickness (~2.5 mm) using a laboratory scale rolling equipment with roll diameter 140 mm (SPX Precision instrument, Fenn Division, USA). These cold-rolled samples of the three alloys were annealed at 800 °C for 1 h (h) in a salt-bath furnace.

The fully annealed samples of the three alloys were used as the starting materials for further thermo-mechanical processing. These were subjected to cold-rolling up to ~90% reduction in thickness (final thickness ~ 250 μm). The deformation was carried out in steps of 20%, 40%, 60% and 80% reduction in thickness (with respect to the starting thickness ~2.5 mm). However, due to significant strain-hardening, 90% reduction in thickness was achieved using small incremental deformation in each pass. About 10 to 11 passes were required to achieve the total 90% reduction in thickness. Small rectangular samples were obtained from the 90% cold-rolled sheets of the three alloys and were isochronally annealed for 1 h at temperatures ranging from 700 °C to 1000 °C.

2.2. Characterization

The crystal structure analysis of the different alloys was carried out by X-ray diffraction (Make: PANalytical, The Netherlands; Model: X'Pert PRO) using Cu-K_{α} radiation (1.5406 Å). The microstructure and texture of the deformed and annealed materials were characterized using an electron backscatter diffraction (EBSD) system (Oxford Instruments, UK) attached to a scanning electron microscope (SEM) (Model: SUPRA 40, Make: Carl-Zeiss, Germany) equipped with field emission gun electron source (FEG). The samples for EBSD investigations were prepared using careful mechanical polishing followed by electropolishing using a mixture of methanol and perchloric acids (9:1 by volume).

The EBSD scans were acquired using the AztecHKL software (Oxford Instruments, UK). For deformed samples, scan step size of 0.04–0.05 μm was used. For annealed samples the step size varied between 0.20 μm and 4.0 μm. The acquired EBSD dataset were exported to the TSL-OIM^{TM} software (version 6.2) (EDAX Inc., USA) in the text file format for the analysis purpose. In order to determine the microstructural and textural parameters with a high degree of statistical accuracy, several EBSD scans were obtained from each deformed and annealed sample. The average grain size of different annealed samples was measured using the circle equivalent diameter method. For that the grain were defined with a tolerance angle of 15° and size of at least 5 scan points in a single grain. The annealing twins are excluded from the grain size calculation. The pole figures (PFs) and orientation distribution functions (ODFs) were calculated from the merged dataset (by stitching the edges of the maps using the merge function of the TSL-OIM^{TM} software) using the harmonic series expansion method (series rank = 30). Orthotropic sample symmetry was assumed for calculating the PFs and ODFs. The volume fraction of individual texture components was determined using a cutoff angle of 15°.

The melting behavior of the three alloys was characterized by a differential scanning calorimeter (Make: NETZSCH, Germany, Model: DSC 404). The measurements were carried out under a flowing high purity argon atmosphere using a heating rate of 10 °C min^{-1}.

3. Results

The XRD plots of the three homogenized starting materials are shown in Fig. 1 and compared with homogenized high purity nickel. The XRD patterns indicate that the three homogenized alloys form single phase FCC solid solution. However, systematic peak shifts to the left with respect to nickel indicate lattice expansion due to solid solution formation. The microstructures of three starting materials (obtained after 50% cold-rolling and annealing at 800 °C for 1 h) in the form of grain boundary (GB) maps are shown in Fig. 2. The microstructures of the three materials appear fully recrystallized as evidenced by the presence of recrystallized grains separated by high angle grain boundaries (HAGBs defined by misorientation angles (θ) ≥ 15° and highlighted in black in the GB maps in Fig. 2). The average grain sizes of the quaternary (Fig. 2(b)) and quinary alloys (Fig. 2(c)) are similar (~7 μm) but somewhat lower than that of the ternary alloy (~16 μm) (Fig. 2(a)). Therefore, the starting microstructures of the three alloys reveal quite refined and similar recrystallized microstructure. The grain size distributions of the ternary (Fig. 2(d)), quarternary (Fig. 2(e)) and quinary (Fig. 2(f)) are also shown. The grain size distribution of the ternary alloy (Fig. 2(d)) shows wider grain size distribution as compared to the other two alloys. Profuse annealing twin boundaries (TBs) (highlighted in red (in the web version) in the GB maps in Fig. 2) are clearly visible in the three starting materials.

The development of microstructure after heavy cold-rolling is shown in Fig. 3. The ternary alloy (Fig. 3(a)) shows lamellar deformation microstructure typical of heavily cold-rolled FCC materials. The microstructure also shows the presence of shear bands inclined to the RD and cutting across the lamellar structure (marked by arrow). The microstructures of the quaternary (Fig. 3(b)) and

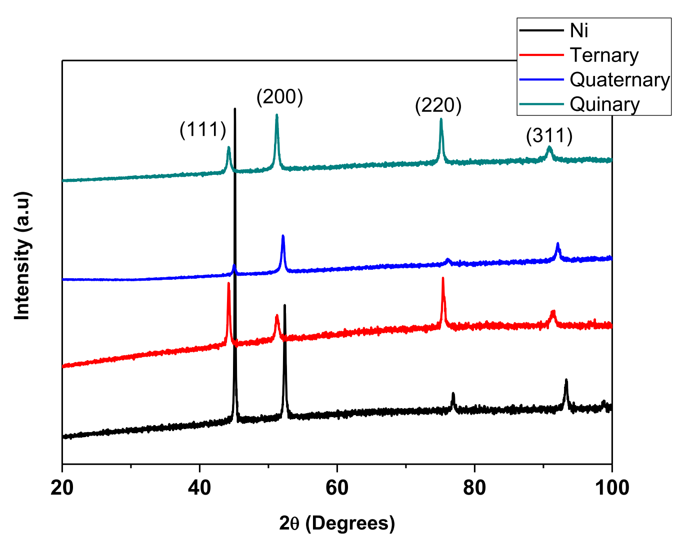

Fig. 1. XRD patterns of homogenized high purity Ni, ternary, quaternary and quinary alloys.