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Synthesis of bulk nanostructured Ni, Ti and Zr by repeated cold-rolling

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Abstract

Macroscopic nanocrystalline samples of Ti, Ni and Zr with average grain sizes well below 80 nm have been synthesized at ambient temperature by repeated cold-rolling. For pure Ni, macroscopic samples with an average grain size of 10 nm could be synthesized without simultaneous application of a high hydrostatic pressure.

Keywords: Accumulative roll bonding; Cold rolling; Severe plastic deformation; Nanostructured materials; Grain refinement

1. Introduction

In recent years, nanocrystalline bulk materials [1], i.e. dense and macroscopically continuous materials with a grain size smaller than 100 nm, have attracted considerable attention because of their modified and often improved properties. A wide variety of techniques are currently available to produce nanocrystalline materials including inert gas condensation [2], mechanical milling [3], electrodeposition [4] and controlled crystallization of amorphous materials [5]. Recently, severe plastic deformation (SPD) processes [6] have been studied intensively due to their potential to produce fine-grained structures with attractive and rather unusual mechanical properties [6,7]. However, SPD-processing of pure metals such as Cu, Ni and Ti typically yields ultrafine-grained (UFG) materials, i.e. materials with an average grain size in the 100–1000 nm range. Moreover, the production of bulk UFG materials by SPD techniques is still limited because of the demands that the ultra-high pressure poses on the selection of the machinery and the tooling material. Thus, alternate synthesis routes that do not involve a high hydrostatic pressure would be favorable. In order to obtain bulk nanocrystalline materials, we used a repeated rolling and folding process, which has the potential to produce large quantities of nanocrystalline materials. A similar procedure has previously been used to synthesize multilayer samples [8], UFG materials [9] or metallic glasses [10]. In the case of UFG-materials, the rolling and folding procedure with an additional intermediate ''brushing'' operation has been denoted as ''accumulative roll-bonding'' (ARB) [9]. In fact, cold-rolling is a well-known process that is frequently applied in industrial processing of sheet metal. Yet, the deformation process used here differs from classical cold-rolling as well as from ARB in the amount of strain (that is far larger than in both processes), in the absence of lubricating agents and in the absence of intermediate brushing. Thus, due to friction at the roll surfaces, a shear strain component adds to the rolling strain that is active in conventional cold-rolling processing [11]. Moreover, a rather small strain rate has been applied throughout the plastic deformation processing carried out in this work to minimize inhomogeneous coarsening

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due to friction-induced localized heating at the internal interfaces. Moreover, room temperature presents a rather low homologous temperature for the three metals under investigation. Thus, applying a low strain rate also minimizes fracture of the current samples during rolling.

2. Experimental procedure

Nanocrystalline Ni, Zr and Ti samples were prepared by repeated cold rolling with intermediate folding and without the use of any lubricating agent. For each material, two foils (area: 80 mm × 80 mm) of each of the pure elements Ti (99.94%), Zr (99.8%) and Ni (99.95%) with initial thickness of 16 μm, 25 μm and 12 μm, respectively were stacked separately and then folded 4 times to yield a $20 \times 20\ mm^2$ multilayer sandwich. Thus, the number of layers in each starting multilayer amounted to 32. The total sample weight of the starting material was about 2 g. The folded samples were rolled in a motorized 2-high rolling mill at a strain rate of $0.5\ s^{-1}$ to a thickness of approximately 200 μm to obtain the starting multilayer. Thus, the equivalent strain during the preparation of the starting multilayers was −0.753 for Ni, −1.085 for Ti and −1.6 for Zr. Thereafter, each of the samples was folded and then rolled for 10 subsequent times with a continuously decreasing spacing between the rolls until the thickness was decreased to 200 μm. This procedure constitutes one folding and rolling (F&R) pass. Thereafter, the sample was cut (or broken) in half and the two halves were stacked and cold-rolled again, following the same procedure. This deformation procedure (1 F&R-pass) was repeated for up to 100 times for each specimen. The deformation time and the respective strain rate were calculated from the geometry of the rolls (diameter of the rolls 150 mm, length of the rolls 100 mm), the circumferential roll velocity (1 m/min) and the thickness of the samples (200–300 μm). In fact, the actual strain rate is smaller than the calculated upper bound, because of sliding at the internal interfaces (due to imperfect bonding) and due to certain areas of the sample that do not overlap after the folding. Consequently, a small area fraction is not deformed during rolling, as described by Bordeaux et al. [12]. Moreover, the bending of the rolls that occurs inevitably is not included in the simplified calculation. These issues also apply for the calculation of the amount of strain applied in the process of cold-rolling. Thus, assuming plain strain conditions leads to an overestimation of the amount of strain, which is the physical parameter that drives the microstructure modification. However, in order to allow a comparison between the current results and the results obtained by different SPD-techniques, the equivalent strain for rolling deformation has been calculated by assuming that the deformation obeys the von Mises criterion, $\varepsilon = -2/\sqrt{3} \cdot \ln(t/t_0)$, with t_0 and t the thickness of the material before and after rolling, respectively. Thus, the strain state is denoted by the number of folding and rolling passes in addition to the corresponding theoretical value for the equivalent strain for rolling, ε.

The samples were investigated by X-ray diffraction (XRD, Phillips X'Pert) in Bragg–Brentano geometry with Cu-Kα radiation using a fast, position-sensitive detector (Phillips X'Cellerator), followed by the characterization of their morphology in cross section by scanning electron microscopy (SEM, Leo 1530). For transmission electron microscopy/selected area electron diffraction investigations (TEM/SAED, Philips Tecnai F20 ST), 3 mm in diameter plan view samples were mechanically punched out. Thereafter, the Ni was thinned by twin-jet electropolishing (1/3 nitric acid and 2/3 methanol). Due to the detrimental impact of the electrolyte on Ti and Zr, these samples were thinned by ion milling with a low accelerating voltage (3.5 kV) to minimize the beam effects on the microstructure.

3. Results

The microstructure evolution at different stages of the deformation process was followed by XRD that averages over the sample volume. Fig. 1a shows the XRD scan of an as-rolled multilayer sample (Ni) after different F&R cycles. Initially the intensity of the Bragg peaks of the elements gradually decreased and the peaks broadened with an increasing number of rolling passes (increasing strain) due to the refinement of the crystallite size and the build-up of internal lattice strain introduced

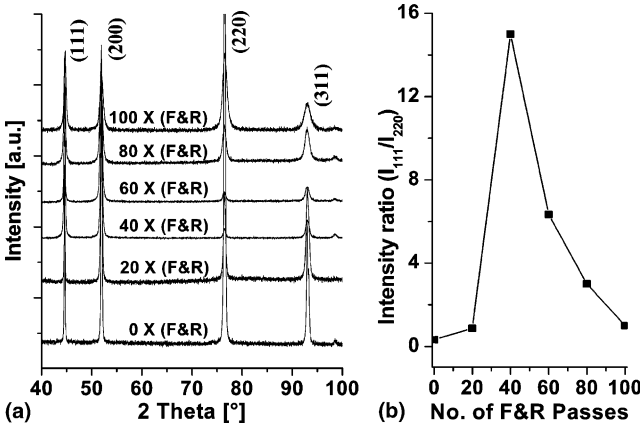

Fig. 1. (a) XRD scan of as-rolled multilayer samples (Ni) after different F&R cycles as indicated in the figure. The corresponding values for the equivalent strain for rolling, ε_{Ni}, are 0, −16, −32, −48, −64, −80, respectively. (b) Ratio of the intensities I of the (1 1 1) and the (2 2 0) Bragg reflection versus the number of F&R passes that scale linearly with ε. The line represents a guide for the eye.