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Synthesis of bulk nanostructured materials by repeated cold-rolling

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Abstract

Repeated cold-rolling with intermediate folding represents an alternative severe plastic deformation technique to produce extremely fine-grained bulk nanocrystalline materials at ambient temperature. In the present work, massive nanocrystalline samples of Ni with an average grain size less than 10 nm have been synthesized. Such small grain sizes have not been obtained for a pure fcc-metal by any other SPD technique. The main key to refine the microstructure via plastic deformation seems to be the requirement to apply a very large plastic strain but not necessarily the simultaneous application of a high pressure in the range of several GPa.

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

1. Introduction

The application of grain refinement is a powerful tool to design microstructures with improved properties. Recently, it has been found that intense straining of metallic materials can be used to produce ultrafine-grained (UFG)—materials with an average grain size in the 100–1000 nm range—or even nanocrystalline (nc) materials. In the last decade, various severe plastic deformation (SPD) processes have been proposed for ultra grain refinement [1,2]. However, the grain sizes achieved for pure metals, such as Cu, Ni and Ti are actually outside the nanocrystalline regime since the grain sizes obtained still remain in the 150–350 nm range.

On the other hand, it has recently been found that repeated cold-rolling and folding (F&R) can be used to produce nanocrystalline metals and alloys [3–5]. A similar procedure has previously been used to synthesize multilayer samples [6], UFG materials [7] or metallic glasses [8]. The present work was initiated to quantitatively investigate the grain refinement process during plastic deformation of pure Ni, including the limiting minimum grain size achievable by repeated cold-rolling.

2. Experimental procedure

Two foils (80 mm × 80 mm) of pure Ni (99.95%) with initial thickness of 12 μm were stacked and then folded four times to yield a 20 mm × 20 mm multilayer sandwich. The folded sample was rolled in a motorized two-high rolling mill (diameter of the rolls 150 mm, length of the rolls 100 mm) at a strain rate of 0.5 s^{-1} to a thickness of approximately 200 μm to obtain the starting material. Thereafter, the sample 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, stacked and cold-rolled again, following the same procedure. This deformation procedure (1 F&R pass) was repeated for up to 100 times.

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 and thinned by twin-jet electropolishing (one-third nitric acid and two-thirds methanol).

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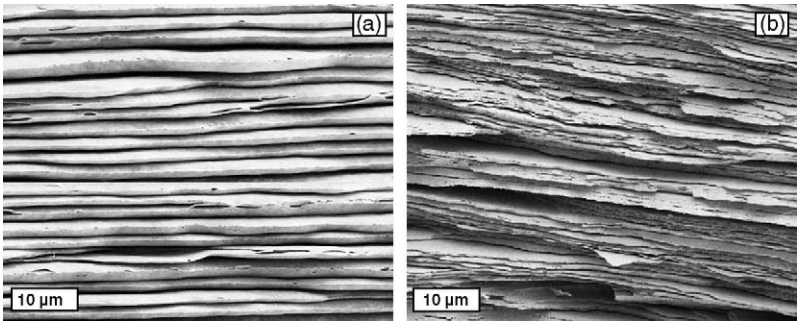

Fig. 1. SEM images showing overviews of the deformed Ni-multilayer samples after different F&R cycles: (a) 10 cycles and (b) 20 cycles.

3. Results

Investigating the cold-rolled Ni samples by SEM reveals a change in the layer thickness with increasing F&R cycles. Fig. 1 shows the cross-sectional SEM micrographs of the fracture surfaces of cold-rolled Ni at different deformation levels. The cross-sectional fracture surface was used for analyses, since most of the individual layers broke at different positions, which results in a higher resolution of the individual layers in the SEM images. It is clear that cold-rolling leads to a significant refinement of the microstructure. The average layer thickness is about 1 μm after 20 F&R cycles. The lamellar structure was highly refined with further rolling. Additional processing for up to 100 F&R cycles results in a refinement of the individual layers down to an average thickness of less than 50 nm. The von Mises equivalent strain corresponding to 100 F&R cycles amounts to −80, as obtained from the average layer thickness reduction.

The evolution of microstructure and texture at different stages of the deformation can be followed by X-ray diffraction methods. As expected for plastic deformation processes, the Bragg peaks of Ni gradually decreased and the peaks broadened with an increasing number of rolling passes due to the refinement of the crystallite size and the build-up of internal lattice strain. At the same time, it has been observed that after 40 F&R passes, the relative intensities of the {1 1 1} Bragg peak strongly increased and the {2 2 0} and {3 1 1} peaks almost vanished, indicating the formation of a strong rolling texture [5]. This result indicates a dominance of dislocation processes during the early stages of the deformation process [9]. Additionally, it was observed that further rolling up to 100 F&R passes gradually reduced the (1 1 1)/(2 2 0) intensity ratio down to a value close to 1. This later stage of the overall deformation process proceeds at decreased grain size. Thus, the local deformation mechanism may change and grain boundary sliding and grain rotation can become active [10], which would lead to a gradual decrease of the texture with continued plastic deformation.

It was seen from high resolution as well as dark field TEM images that a dense nanocrystalline structure with an average grain size of about 8–10 nm has been achieved after 80 F&R cycles. To further analyze the microstructure that has been obtained for pure Ni after 80 F&R cycles, annealing experiments with subsequent microstructure analyses have been conducted. In this case, a Ni sample was heated to 350 °C at a rate of 20 °C/min under a purified Argon atmosphere and cooled at about 150 °C/min directly after reaching 350 °C. The HR-TEM micrograph shown in Fig. 2a indicates that the microstructure shows very limited coarsening due to the annealing treatment. In fact, the average grain size increased only marginally from 8–10 nm previous of the annealing to about 10–12 nm after the thermal treatment. The corresponding dark field image (Fig. 2b) indicates clearly that the microstructure consists of a dense array of nanocrystalline grains of about 10–12 nm in diameter. Thus, a truly nanocrystalline structure has been obtained by repeated

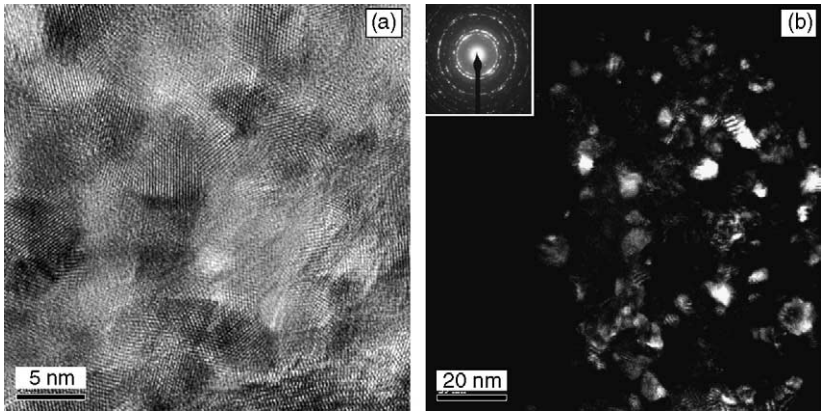

Fig. 2. (a) HR-TEM micrograph of Ni that has been rolled for 80 passes and that has additionally be heated at 20 °C/min to 350 °C followed by rapid cooling to room temperature. (b) Dark field TEM image corresponding to the HR-TEM shown in (a), indicating individual grains of about 10–12 nm in diameter.