DOI: 10.1002/adma.200600397

From Micro- to Macroplasticity**

By *Stefan Brandstetter*, *Helena Van Swygenhoven*,* *Steven Van Petegem*, *Bernd Schmitt, Robert Maaß*, and *Peter M. Derlet*

Early results of experimental investigations on microplasticity have been gathered into collected works and treatises.[1–3] The plastic events taking place at lower strains are related to short-range dislocation motion[4] and are of different origin than the dislocation mechanism occurring in the macroplastic regime.[5] Therefore, macroplasticity theories based on a mechanism involving long-range dislocation motion cannot serve to predict micro-yielding. In terms of measured stress–strain behavior, the two regimes are poorly distinguished, especially when they are characterized by positive strain-hardening, i.e., an increase in strength when the microstructure is strained. To facilitate comparison of data, metallurgists have come up with the rather arbitrary 0.2 % definition of the macroscopic yield stress.[6,7] It is one of the most frequently used engineering materials parameters and very important for the precise design of functional components ranging from construction materials down to micrometer-sized microelectromechanical (MEMS) devices.

In polycrystalline materials, it is taken for granted that the majority of grains are plastically deforming at the macroscopic yield stress. However, there is no easy and suitable method to verify this assumption, and the real amount of strain that should be assigned to microplasticity is unknown. Experimental studies on granular materials have predominantly been performed by precise mechanical testing using load–unload cycles accompanied by surface-visualization techniques. The first experimental investigations on the effect of grain size on microplasticity[8,9] ascribed strain-hardening predominantly to the ability of grain boundaries (GBs) to act as barriers to dislocation motion, where the strain for a given stress varied as the cube of the grain size: large grains showed more strain than smaller grains. However, it was soon recognized that the relationship between microplastic strain and stress was not so simple. When a polycrystal is subjected to an external force, an inhomogeneous state of internal stress is developed resulting from elastic anisotropy and plastic incompatibilities arising from different resolved shear stresses within the different grains.[10] In addition to the fact that plastic yielding does not start at the same time in all grains, it was recognized that internal stresses arising from the elastic interactions of neighboring grains provide additional stresses that activate non-favored slip systems in grain interiors[11] and/or change the maximum shear direction in the vicinity of a GB. The latter may result in GBs emitting dislocations[12,13] on slip systems in the GB region that are different from the primary slip system of the grain interior.[14,15] Several plasticity models have been developed to predict the mechanical behavior in terms of stress–strain and hardening as a function of grain size. Some theories are based on the postulate that geometrically necessary dislocations account for the inherent difference in the accumulation of plastic strain between the GB region and the grain interior.[16,17] Others are based on models that use a two-phase approach, where the GB and grain interior are treated separately.[18,19] However, even when elastic anisotropy is included,[20] the initial strain-hardening regime is usually poorly described. To date, our knowledge on microplasticity in polycrystals is based on results obtained from coarse-grained structures. Very little attention has been paid to what happens in the microplastic region when grain sizes reach the nanometer scale, despite the tremendous amount of research on macroplastic behavior.[21,22] If one assumes that in a nanocrystalline metal all macroscopic plasticity is carried by dislocations emitted from, and subsequently absorbed in, GBs,[23,24] only 4 % of the grains would have deformed plastically at the 0.2 % yield stress.[25] In other words, a nanocrystalline structure deforms more heterogeneously than a coarse-grained polycrystal.

In the present work we show, through analysis of X-ray diffraction (XRD) spectra measured during in situ deformation of nanocrystalline and ultrafine-grained Ni, that the amount of strain assigned to microplasticity can significantly exceed the usual 0.2 % definition of the macroscopic yield stress when grain sizes reach the nanometer range.

XRD profile analysis is a well-known technique for microstructural analysis, where the broadening of the diffraction peaks result from limitations in the spatial extent of the coherent scattering volumes (in our case, the grain size) and the presence of inhomogeneous strain. The first type of broadening is diffraction-order independent, whereas the second is order dependent.[26] Possible sources of inhomogeneous strain may include lattice dislocations and extrinsic GB dislocations.

In situ XRD measurements were performed on Ni synthesized by electrodeposition (ED) and by high-pressure torsion

[*] Prof. H. Van Swygenhoven, S. Brandstetter, Dr. S. Van Petegem, Dr. B. Schmitt, R. Maaß, Dr. P. M. Derlet
Paul Scherrer Institute
5232 Villigen (Switzerland)
E-mail: helena.vanswygenhoven@psi.ch

[**] H. Van Swygenhoven thanks the Swiss National Science Foundation for their financial support.

ADVANCED MATERIALS

(HPT). Figure 1A and B shows the stress–strain curves and Figure 1C and D the stress as a function of time for ED-Ni and HPT-Ni, respectively. Each sample is sequentially submitted to two identical series of five load–unload cycles, starting with a cycle in the elastic regime (red loading and unloading curves) and ending with a cycle near the maximum flow stress (black loading and unloading curves). Care was taken to perform the cycles in both series at approximately the same percentage of the maximum flow stress of both materials: at about 25, 40, 75, and 95 %. After each unloading, the samples were kept at approximately zero stress for 20 min (a small and constant stress of 10 MPa was necessary to keep the sample in the grips), allowing the investigation of possible post-loading relaxation processes. For ED-Ni, the 0.2 % yield stress was 1050 MPa, whereas for HPT-Ni it was 920 MPa at respective total macroscopic strains of 0.95 % and 0.65 %.

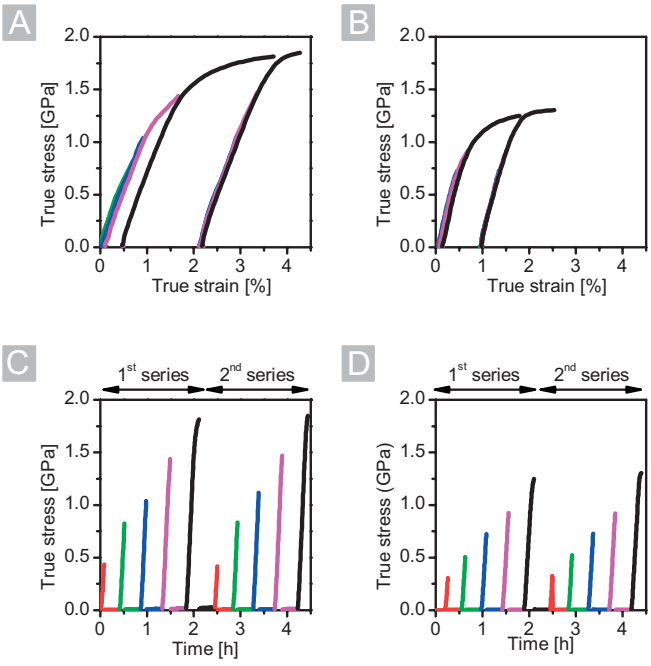

Figure 1. Mechanical data for a series of load–unload cycles. A,B) The true-stress/true-strain curves, and C,D) the true-stress/time curves for A,C) ED-Ni and B,D) HPT-Ni. The colors correspond to the different loading cycles. The unloadings were performed at 25 % (red), 40 % (green), 60 % (blue), 75 % (magenta), and 95 % (black) of the maximum flow stress.

Figure 2A and B shows the full-width at half-maximum (FWHM) values of the XRD peaks as a function of time for the (111), (200), and (311) peaks of ED-Ni and HPT-Ni, respectively. By comparing the stress–strain curves and the behavior of the peak broadening of ED-Ni, it becomes clear that the strain-hardening can be related to two clearly distinguishable behaviors of the peak width. Upon each unloading beyond the elastic regime and up to 1400 MPa (starting with the first green curve and ending with the first pink curve), extra amounts of recovery are observed in the FWHM. Once the maximum flow stress has been reached for the first time (first black curve), the recovery of the FWHM clearly becomes time dependent with an instantaneous part that is strongly diffraction-order dependent. In general, one can say that, with respect to the unloaded as-prepared state, the FWHM is fully or even more then fully recoverable, which agrees with a recent study on macroplasticity.[23,24] However, full recovery is not always observed with respect to the values of the FWHM after unloading at 1400 MPa (pink curve). In the most extreme case, the FWHM of the (311) peak is only fully recoverable with respect to the as-prepared state after a 10 min relaxation, and there is peak broadening with respect to the unloading at 1400 MPa even after relaxation for 2 h. However, the width of the (200) peak is instantaneously fully recoverable relative to the lowest value obtained after unloading at 1400 MPa, and continues to reduce gradually during relaxation. Additional load–unload cycles at stresses lower than 1400 MPa (second series of load–unload) do not result in extra recovery as was the case during the first series. In other words, the strain-hardening in the stress–strain curve up to 1400 MPa is related to a plastic deformation mechanism that results in a reduction in the FWHM of XRD peaks upon unloading. Above 1400 MPa, the plastic deformation mechanism is characterized by a time dependence in the recovery of the FWHM upon unloading, with respect to both the unloaded state and the lowest values reached upon unloading at 1400 MPa.

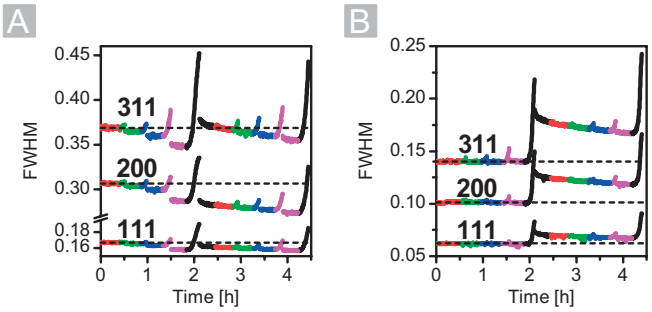

Figure 2. X-ray diffraction results. FWHM of the (111), (200), and (311) diffraction peaks as a function of time for A) ED-Ni and B) HPT-Ni. The color code corresponds to the one used in Figure 1.

For HPT-Ni, the FWHM behavior also indicated two distinct hardening regimes, the first characterized by a constant FWHM upon unloading, and the second by an increase relative to the as-prepared state, as expected from dislocation plasticity.[27] Similar to ED-Ni, the second regime demonstrated time dependence, albeit one less pronounced.

Changing the load–unload history before reaching the maximum flow stress by increasing the number of cycles or performing a continuous loading did not effect the FWHM values upon unloading at the maximum flow stress. Using methods

that allow the separation of broadening from grain size and inhomogeneous strains,[28] no increase in grain size was observed within the first regime for both HPT-Ni and ED-Ni. In other words, the extra recovery seen in ED-Ni can be entirely associated with a recovery in root–mean–square (RMS) strain, independent of deformation history.

The above results demonstrate the presence of two distinctly different hardening regimes that can be related to the microplastic and macroplastic regime. In particular, for nanocrystalline metal, strain-hardening in the microplastic regime results in a mechanism that upon unloading lowers the RMS strain. Such behavior is incompatible with the activation of multiple slip systems in the GB region and/or the increased presence of geometrical dislocations, since this would have the tendency to increase RMS strains or (when considering that the peak profile predominantly probes the grain interiors) at least keep the values constant, as observed for HPT-Ni. Here, as long as plastic events are scarce, incompatibility stresses are accommodated by the GB structure/network, reducing the RMS strain. Once the majority of the grains start deforming plastically, dislocations are still accommodated in the GBs but there is no room for network accommodations that allow extra relief of RMS strain.

Our results challenge the validity of the 0.2 % definition for macroscopic yield stress when grain sizes reach the nanometer range, as schematically represented in Figure 3. For ultrafine-grained Ni with a 300 nm grain size, the assumptions related

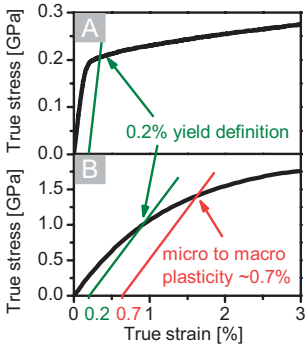

Figure 3. Illustration of the macroscopic yield stress for coarse-grained and nanocrystalline Ni. A) 0.2 % definition of the yield stress for coarse-grained Ni, and B) the 0.2 % definition (green) and the change from micro- to macroplasticity (red) for nanocrystalline Ni, as revealed by XRD measurements. The associated strain corresponds to a 0.7 % offset from Hooke's Law.

to the definition are still justified. For ED-Ni with a 30 nm grain size, the 0.2 % yield substantially underestimates the stress corresponding to the onset of macroscopic plasticity. The change from micro- to macroplasticity occurs in the nanocrystalline metal at 1400 MPa, i.e., at a strain that deviates 0.7 % from Hooke's Law, well beyond the 0.2 % definition of the yield stress. Using the correct transition from micro- to macroplasticity to study grain-size-dependent strengthening will not only reduce the scattering in experimental data but also provide a different view on the validity of current plasticity laws.[16,29]

In summary, by performing in situ X-ray peak profile analysis during mechanical testing of metals, it has been shown that different strain-hardening mechanisms in the micro- and macroplastic regimes are evident, and that the generally used 0.2 % yield criteria does not correspond to the onset of macroscopic plasticity in all materials.

Experimental

Materials: ED-Ni was purchased from Goodfellow and was the same batch as that used in previous studies [23,24,30]. The material exhibited a narrow grain-size distribution with a mean grain size of 30 nm. Cross-sectional analyses did not show columnar growth. HPT-Ni was obtained from Prof. M. Zehetbauer (University of Vienna, Austria). It was synthesized in the form of 8 mm disks with a thickness of 800 μm using a pressure of 8 GPa with two rotations leading to a van Mises strain of approximately 36 in peripheral sample areas. Transmission electron microscopy analysis showed large grains (200–400 nm) with a visible subgrain boundary structure resulting in a maximum coherent XRD domain size in the order of 80 nm. Mini-dog-bones, prepared as described previously [27], were used for mechanical testing.

Techniques: The samples were deformed at a constant strain rate of 6×10^{-5} s^{-1} under uniaxial tensile loading conditions, in situ at the Swiss Light Source. The in situ technique allows for a diffraction pattern to be taken continuously during deformation over a 2θ range of 60°. For more details on the technique and the fitting procedure of the XRD spectra we refer to previous publications [23,27,30].

Received: February 25, 2006
Final version: March 22, 2006

[1] C. W. Marschall, R. E. Maringer, *Dimensional Instability: An Introduction,* Pergamon, New York **1977**.
[2] *Microplasticity* (Ed: C. J. McMahon, Jr.), Interscience, New York **1968**.
[3] A. Puskar, in *Microplasticity and Failure of Metallic Materials* (Ed: I. Hrivnák), Elsevier, Amsterdam, The Netherlands **1989**.
[4] G. Fantozzi, C. Esnouf, W. Benoit, I. G. Ritchie, *Prog. Mater. Sci.* **1982**, *27*, 311.
[5] U. F. Kocks, H. Mecking, *Prog. Mater. Sci.* **2003**, *49*, 171.
[6] J. P. Hirth, J. Lothe, *Theory of Dislocations*, 2nd ed., Wiley, New York **1982**.
[7] In *Standard Test Methods for Tension Testing of Metallic Materials [Metric]*. Designation ASTM E 8M-04, American Society for Testing and Materials, West Conshohocken, PA **2004**, originally approved in 1924.
[8] D. A. Thomas, B. L. Averbach, *Acta Metall.* **1959**, *7*, 69.
[9] N. Brown, K. F. Lukens, *Acta Metall.* **1961**, *9*, 106.
[10] J. P. Hirth, *Metall. Trans.* **1972**, *3*, 3047.
[11] K. Hashimoto, H. Margolin, *Acta Metall.* **1983**, *31*, 773.
[12] R. E. Hook, J. P. Hirth, *Acta Metall.* **1967**, *15*, 535.
[13] J. C. M. Li, Y. T. Chou, *Metall. Trans.* **1970**, *1*, 1145.
[14] Z. Yao, R. H. Wagoner, *Acta Metall. Mater.* **1993**, *41*, 451.
[15] F. Delaire, J. L. Raphanel, C. Rey, *Acta Mater.* **2000**, *48*, 1075.
[16] M. F. Ashby, *Philos. Mag.* **1970**, *21*, 399.
[17] A. Needleman, J. G. Sevillano, *Scr. Mat.* **2003**, *48*, 109.
[18] M. A. Meyers, E. A. Ashworth, *Philos. Mag.* **1982**, *46*, 737.
[19] L. Capulongo, M. Cherkaoui, J. Qu, *Trans. ASME* **2005**, *127*, 400.

ADVANCED MATERIALS

[20] H. H. Fu, D. J. Benson, M. A. Meyers, *Acta Mater.* **2004**, *52*, 4413.
[21] J. R. Weertman, in *Nanostructured Materials: Processing, Properties, and Potential Applications* (Ed: C. C. Koch), William Andrews, Norwich, NY **2002**, pp. 397–421.
[22] K. S. Kumar, H. Van Swygenhoven, S. Suresh, *Acta Mater.* **2003**, *51*, 5743.
[23] Z. Budrovic, H. Van Swygenhoven, P. M. Derlet, S. Van Petegem, B. Schmitt, *Science* **2003**, *304*, 273.
[24] S. Brandstetter, Z. Budrovic, S. Van Petegem, B. Schmitt, E. Stergar, P. M. Derlet, H. Van Swygenhoven, *Appl. Phys. Lett.* **2005**, *87*, 231 910.
[25] G. Saada, *Mater. Sci. Eng., A* **2005**, *400*, 146.
[26] H. P. Klug, L. E. Alexander, *X-ray Diffraction Procedures for Polycrystalline and Amorphous Materials*, 2nd ed., Wiley, New York **1974**.
[27] H. Van Swygenhoven, B. Schmitt, P. M. Derlet, S. Van Petegem, A. Cervellino, Z. Budrovic, S. Brandstetter, A. Bollhalder, M. Schild, *Rev. Sci. Instrum.* **2005**, *77*, 013 902.
[28] T. Ungar, A. Revesz, A. Borbely, *J. Appl. Crystallogr.* **1998**, *31*, 554.
[29] P. G. Sanders, C. J. Youngdahl, J. R. Weertman, *Mater. Sci. Eng., A* **1997**, *234*, 77.
[30] Z. Budrovic, S. Van Petegem, P. M. Derlet, B. Schmitt, H. Van Swygenhoven, E. Schafler, M. Zehetbauer, *Appl. Phys. Lett.* **2005**, *86*, 231 910.

