Strengthening and softening of nanocrystalline nickel during multistep nanoindentation

著者	Pan D., Nieh T. G., Chen M. W.
journal or publication title	Applied Physics Letters
volume	88
number	16
page range	161922
year	2006
URL	http://hdl.handle.net/10097/51849

doi: 10.1063/1.2197289

Strengthening and softening of nanocrystalline nickel during multistep nanoindentation

D. Pan
Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

T. G. Nieh
Department of Materials Science and Engineering, The University of Tennessee, Knoxville, Tennessee 37996 and Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831

M. W. Chen[a)]
Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

(Received 17 December 2005; accepted 6 March 2006; published online 21 April 2006)

Multistep load-unload nanoindentation was employed to address the effect of deformation-induced microstructural evolution on mechanical behavior of nanocrystalline Ni. Deformation discontinuity was deliberately introduced by unloading-reloading during nanoindentation testing, which allows us to examine the influence of microstructural evolution on the successive deformation. Strain strengthening/softening of nanocrystalline nickel, associated with the transition of deformation behavior from dislocation activity at high loading rates to a grain-boundary-mediated process at low loading rates, was uncovered by means of this experimental methodology. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2197289]

Nanoindentation has been widely used as a powerful tool for measuring Young's modulus and hardness of a variety of materials over the last two decades.[1–6] Moreover, precise resolutions in both displacement and force measurements promote nanoindentation as a robust technique in investigating novel material behavior beyond traditional mechanical properties, such as incipient plasticity and hardening/recovery of bulk metallic glasses[7,8] (BMGs) and pressure-induced phase transformations in inorganic materials,[9,10] which cannot be achieved by conventional mechanical tests. The mechanical behavior of nanocrystalline (nc) metals has been generally recognized to intrinsically differ from that of their microcrystalline (mc) counterparts.[11–15] Due to the metastable nature associated with small nanograins and high grain boundary volume fraction, deformation-induced microstructural evolution, for instance, deformation twins[16–18] and deformation-induced nanograin growth,[18–21] has been observed in a number of nc metals. Thus, the effect of microstructural evolution in successive deformation presents a key issue in developing a comprehensive understanding of mechanical behavior of nc metals. However, unlike discrete discontinuity in displacements such as pop-in events in BMGs (Ref. 7) and phase transitions in Si,[9,10] the length scale of structural evolution in nc metals during deformation is so small that even under depth-sensitive indentation the resultant changes in mechanical behavior appear to be "continuous." As a result, the effect of deformation-induced microstructural evolution on the consecutive deformation behavior of nanostructured materials has not been fully understood. In this letter, we report our effort to address this effect in nc Ni by employing a novel indentation method.

Fully dense nc nickel specimens were electrodeposited in a sheet form with a thickness of $\sim$200 μm and an average grain size of $\sim$15 nm. For comparison, mc Ni with an average grain size of $\sim$80 μm was also prepared by annealing commercial polycrystalline pure Ni at 1073 K for 1 h. Prior to indentation tests, the sample surfaces were polished to a mirror finish. A dynamic ultramicrohardness tester (Shimadzu W201S), equipped with a Berkovich indenter, was employed to perform the indentation tests. Intermittent load-unload indentation tests were conducted in ten load steps under a constant force rate during both the loading and unloading.

"Single-step" force-displacement (P-h) curves were obtained at four different loading rates (70.6, 35.3, 13.2, and 1.32 mN/s) from the mc [Fig. 1(a)] and nc [Fig. 1(b)] nickel to characterize their rate sensitivities. A nominal maximum force of 196 mN was selected for all the tests, yet small amount of overshoot can be observed at high loading rates. The nc Ni is apparently harder than the mc Ni, as shown by the fact that a shallower indentation depth is produced at the same maximum force. The P-h curves demonstrate no discernible effect of the loading rate on the mechanical response of mc Ni, whereas nc Ni exhibits increased resistance to deformation with increasing loading rates. Strain rate sensitivities m are estimated to be 0.005 and 0.023 for the mc and nc Ni, respectively [Fig. 1(c)], which are fairly consistent with the literature values.[21–23]

In the case of multistep load-unload indentation tests, both mc and nc nickel were tested under the same maximum load and loading rates as those used for the single-step tests. The resulting P-h curves are shown in the insets of Figs. 2 and 3 for the mc and nc Ni, respectively, and their corresponding hardness-displacement (H-h) curves have been calculated, following the algorithm in Ref. 6. All the H-h curves show the overall decrease in hardness with increasing indentation depth, which arises from the size effect of nanoindentation.[24] The dashed lines represent P-h curves from the single-step indentation while the solid lines are from the multistep load-unload indentation tests. It is noted that the multistep P-h curves for the mc nickel essentially

[a)]Author to whom correspondence should be addressed; electronic mail: mwchen@imr.tohoku.ac.jp

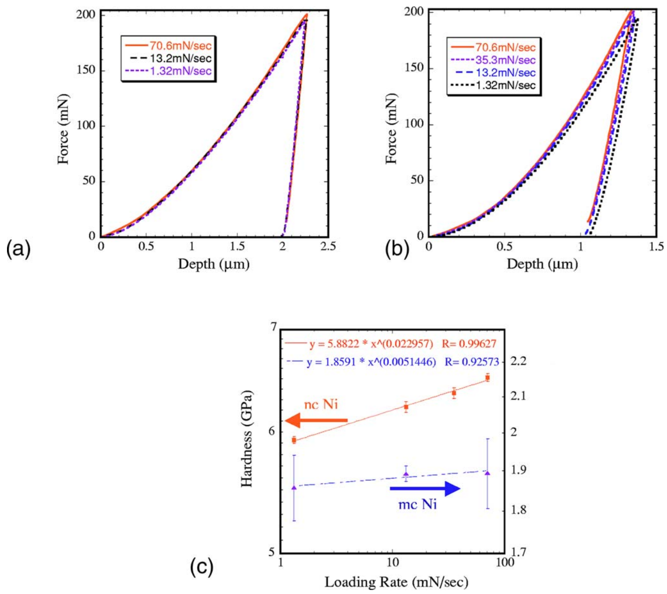

FIG. 1. (Color online) Strain rate sensitivities of (a) mc and (b) nc Ni characterized by nanoindentation. It can be seen that the strain rate dependence of stress is almost negligible for mc Ni, yet discernable for nc Ni. (c) Strain rate sensitivities *m* of 0.005 and 0.023 were estimated for mc and nc nickel, respectively.

overlap with the single-step curves at all the loading rates examined in this study, suggesting that the mechanical behavior of mc nickel is independent of the load-unload steps (Fig. 2). Additionally, the unloading and reloading curves in each load step follow exactly the same path, indicating a perfect elastic unloading/reloading process.

By contrast, a weak yet detectable hardening is observed in the nc nickel tested at the highest loading rate of 70.6 mN/s [Fig. 3(a)], whereas only a slight difference in material response is observed between the multistep and single-step profiles in the cases of medium loading rates of 13.2 and 35.3 mN/s [Fig. 3(b)]. At the slowest loading rate

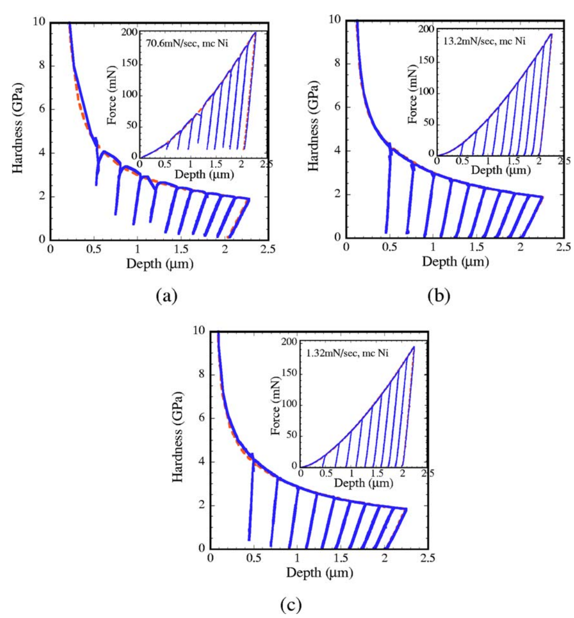

FIG. 2. (Color online) Hardness vs indentation depth curves of mc Ni under variable rates: (a) 70.6 mN/s, (b) 13.2 mN/s, and (c) 1.32 mN/s. Clearly no crucial difference is observed between the deformation behaviors of mc Ni by two methods. The insets are corresponding *P*-*h* curves.

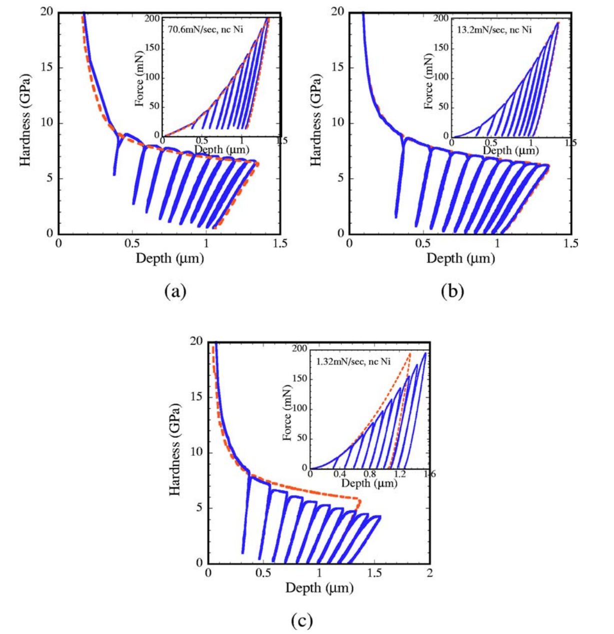

FIG. 3. (Color online) Hardness vs indentation depth curves of nc Ni under variable rates: (a) 70.6 mN/s, (b) 13.2 mN/s, and (c) 1.32 mN/s. The hardness of nc nickel is slightly enhanced at 70.6 mN/s while considerably decreased at 1.32 mN/s. However, at the intermediate loading rate such as 13.2 mN/s, the hardness is insensitive to load steps. The insets are corresponding *P*-*h* curves.

of 1.32 mN/s, considerable softening in the nc nickel takes place during the multistep test as compared to that during the single-step test [Fig. 3(c)]. The gain/loss of hardness at the maximum load by multistep nanoindentation in comparison with the single-step nanoindentation is summarized in Fig. 4. Again, mc Ni exhibits a nearly-rate-independent hardness. In contrast, a moderate gain in hardness is observed in nc Ni at high loading rates while significant loss in hardness at low loading rates.

The distinct difference in deformation behavior of nc Ni between single-step and multistep indentations, especially at low loading rates, may be attributed to three possible reasons: (1) geometric effect of indentation, (2) instrumental or methodological artifact, and (3) intrinsic behavior of nc Ni. At a certain load in a multistep indentation test, the complicated stress distribution in the deformation zone underneath the indenter may be essentially different from that at the same load in a single-step test, induced by contact geometry changes such as pileup and sinking-in during unloading/reloading cycles.[25] The overall mechanical response of the

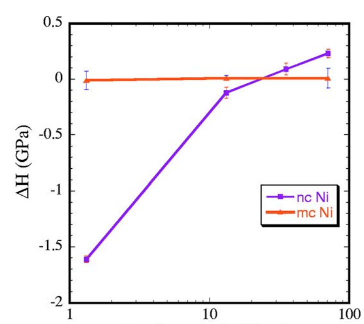

FIG. 4. (Color online) Hardness gain/loss of mc and nickel by multistep indentation vs loading rates. Zero value of ΔH indicates no hardness change caused by multistep indentation in mc Ni.

specimen may show a primary discrepancy in single-step and multistep indentations. However, a closer investigation of residual indentation by scanning electron microscopy and atomic force microscopy, does not uncover a clear evidence of pileup or sinking-in for both samples. One may argue that the observed softening effect during multistep indentation at low loading rates could be induced by thermal drift of the nanoindentation instrument. This argument is unlikely because of the fact that under the same test condition the softening is only observed in nc Ni [Fig. 3(c)], but not in mc Ni [Fig. 2(c)]. We also ruled out the possible artifact effect caused by the instrument inertia, in particular, at high loading rates. In this study, the indentation displacement overshoot, even at the highest loading rate, is estimated to be less than 3 nm, which is insignificant to produce the distinct difference in mechanical behavior. Therefore, the appreciable mechanical response of nc nickel under multistep indentation is most likely associated with the microstructural change in the nc metal during deformation.

In our recent transmission electron microscope observations, we have found that deformation behavior of nc Ni is not only a function of grain size, as intensely discussed before, but also strongly depends on loading rate.[26] At high loading rates, plastic deformation is mainly controlled by intragranular processes, such as normal dislocations in large nanograins and stacking faults and twins in small nanograins. This dislocation-mediated plasticity could lead to work hardening as observed in the multistep nanoindentation tests with a high loading rate. At low loading rates, significant grain growth occurs and the deformation takes place through a stress-assisted grain boundary process. Based on the Hall-Petch relationship for Ni, the effective grain size after multistep load deformation is estimated to be ~20–38 nm (Refs. 27 and 28) from the measured hardness. Indeed, postmortem TEM observation of deformed nc Ni revealed abnormal grain growth, and a number of large grains with a size up to 200 nm, mixing with small nanograins, were observed around the residual indenters. These coarsened grains are expected to deform preferentially at a stress level that is lower than that for a sample with the original grain size. Incorporated with a large number of grains with an original grain size, the coarsened grains are expected to show a moderate increase in the "effective" grain size in Hall-Petch relationship and result in significant work softening at low loading rates.

In this study, the main difference between single-step and multistep indentation tests lies in the introduction of the deformation discontinuity in the multiple unloading/reloading steps, which greatly enhances the effect of deformation-induced microstructural evolution on the successive deformation behavior. Upon unloading or the partial removal of the stresses at high loading rates, some dislocation-dislocation interactions are expected to occur in nanograins. The microscale stress redistribution underneath indenter during reloading may considerably alter the slip systems of some nanograins and result in the interaction between the newly generated dislocations, stacking faults, and deformation twins with the preexisted ones. This could lead to work hardening of the nc Ni upon reloading. At low loading rates, nanograin growth is induced by deformation, either through stress-assisted grain boundary migration or nanograin rotation. The growth strongly depends on the local stress states applied to individual nanograins and can only occur in certain nanograins with favorable stress states.[29,30] The redistribution of local stresses as a result of slow loading/unloading definitely promotes the coarsening of more nanograins with various orientations and results in the strength decrease in nc Ni during multistep testing. For the coarse-grained Ni, the grain size (~80 μm) is much larger than the size of impression (~15 μm). In such a case, dislocations produced by deformation can readily run away without being impeded by barriers such as grain boundaries. Although unloading and reloading may alter the stress distribution underneath the indenter, the overall crystal orientation of the indented grain is essentially unchanged and deformation mainly occurs by the same slip system as that in the single-step loading. Thus, the gain/loss of strength between the multistep and single-step nanoindentation tests is virtually zero, as seen in Fig. 4.

This work was sponsored by Grant-in-Aid, Japan Society for Promotion of Science (JSPS) and partly supported by Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy under Contract No. DE-AC05-00OR-22725 with UT-Battelle.

[1]J. B. Pethica, R. Hutchings, and W. C. Oliver, Philos. Mag. A **48**, 593 (1983).
[2]M. F. Doener and W. D. Nix, J. Mater. Res. **1**, 601 (1986).
[3]W. C. Oliver and G. M. Pharr, J. Mater. Res. **7**, 1564 (1992).
[4]Y. T. Cheng and C. M. Cheng, Appl. Phys. Lett. **73**, 614 (1998).
[5]M. Dao, N. Chollacoop, K. J. Van Vliet, T. A. Venkatesh, and S. Suresh, Acta Mater. **49**, 3899 (2001).
[6]A. C. Fischer-Cripps, *Nanoindentation*, Mechanical Engineering Series (Springer, Berlin, 2002).
[7]C. A. Schuh and T. G. Nie, Acta Mater. **51**, 87 (2003).
[8]B. Yang, L. Riester, and T. G. Nieh, Scr. Mater. **54**, 1277 (2006).
[9]X. Q. Yan, X. M. Huang, S. Uda, and M. W. Chen, Appl. Phys. Lett. **87**, 191911 (2005).
[10]G. M. Pharr, W. C. Oliver, and D. S. Harding, J. Mater. Res. **6**, 1129 (1991).
[11]T. G. Nieh and J. Wadsworth, Scr. Metall. Mater. **22**, 955 (1991).
[12]K. S. Kumar, H. Van Swygenhoven, and S. Suresh, Acta Mater. **51**, 5743 (2003).
[13]M. W. Chen, E. Ma, and K. J. Hemker, in *Nanomaterials Handbook*, edited by Yury Gogotsi (Taylor & Francis, Boca Raton, FL, 2006).
[14]Z. W. Shan, E. A. Stach, J. M. K. Wiezorek, J. A. Knapp, D. M. Follstaedt, and S. X. Mao, Science **305**, 654 (2004).
[15]M. W. Chen, E. Ma, K. J. Hemker, H. W. Sheng, Y. M. Wang, and X. M. Cheng, Science **300**, 1275 (2003).
[16]X. Z. Liao, F. Zhou, E. J. Laverinia, S. G. Srinivasan, M. I. Baskes, D. W. He, and Y. T. Zhu, Appl. Phys. Lett. **83**, 632 (2003).
[17]H. Rosner, J. Markmann, and J. Weissmuller, Philos. Mag. Lett. **84**, 321 (2004).
[18]M. Jin, A. M. Minor, E. A. Stach, and J. W. Morris, Jr., Acta Mater. **52**, 5381 (2004).
[19]M. W. Chen and X. Q. Yan, Science **308**, 356c (2005).
[20]K. Zhang, J. R. Weertman, and J. A. Eastman, Appl. Phys. Lett. **87**, 061921 (2005).
[21]R. Schwaiger, B. Moser, M. Dao, N. Chollacoop, and S. Suresh, Acta Mater. **51**, 5159 (2003).
[22]Q. Wei, S. Cheng, K. T. Ramesh, and E. Ma, Mater. Sci. Eng., A **381**, 71 (2004).
[23]F. Dalla Torre, H. Van Swygenhoven, and M. Victoria, Acta Mater. **50**, 3957 (2002).
[24]W. D. Nix and H. J. Gao, J. Mech. Phys. Solids **46**, 411 (1998).
[25]O. Casals and J. Alcala, Acta Mater. **53**, 3545 (2005).
[26]M. W. Chen, MRS Fall Meeting, Boston, MA, 2004 (unpublished).
[27]C. A. Schuh, T. G. Nieh, and T. Yamasaki, Scr. Mater. **46**, 735 (2002).
[28]G. D. Hughes, S. D. Smith, C. S. Pande, H. R. Johnson, and R. W. Armstrong, Scr. Metall. **20**, 93 (1986).
[29]D. Moldovan, V. Yamakov, D. Wolf, and S. R. Phillpot, Phys. Rev. Lett. **89**, 206101 (2002).
[30]A. J. Haslam, D. Moldovan, V. Yamako, D. Wolf, S. R. Phillpot, and H. Gleiter, Acta Mater. **51**, 2097 (2003).