Mat. Res. Soc. Symp. Vol. 634 © 2001 Materials Research Society

TEM Observation of Nanocrystalline Copper During Deformation

Carl J. Youngdahl, Northwestern University, Evanston, IL 60208 USA
Richard C. Hugo, Los Alamos National Laboratory, Los Alamos, NM 87545 USA
Harriet Kung, Los Alamos National Laboratory, Los Alamos, NM 87545 USA
Julia R. Weertman, Northwestern University, Evanston, IL 60208 USA

ABSTRACT

Nanocrystalline samples of copper were prepared using inert gas condensation and an optimized sequence of powder outgassing and compaction. TEM specimens were cut, electropolished, and mounted in a straining stage. *In situ* TEM observations including real-time video were captured during straining in the microscope. Areas of presumed increased stress concentration were identified near small cracks around the perimeter of the electropolished hole. Such locations were observed in the TEM while the specimen was pulled in tension. Several microstructural changes were captured during deformation including numerous sudden shifts in contrast of grains and parts of grains, occasional dislocation motion, opening and propagation of the crack. Relationships between grain size and deformation are described.

INTRODUCTION

The empirical Hall-Petch relation describes the dependence of several mechanical properties, including yield strength and hardness, on grain size. Various theories attempt to explain the dependence in terms of dislocation activity or its suppression. At very small grain sizes (below what is commonly used in structural applications), the relationship predicts strengths beyond the ranges of those attained at conventional grain sizes. As grain size decreases even lower, the relationship predicts values of yield stress that reach the theoretical limit. Possibly the mechanisms responsible for Hall-Petch behavior at conventional grain sizes give way to another mechanism at a certain low "threshold" size.

As methods to make materials with smaller and smaller grain sizes have increased in number and effectiveness, it is clear that the measured mechanical properties fall short of the values predicted by the Hall-Petch relation. It would be interesting to discover why, as doing so would lend insight into the microstructural workings of the Hall-Petch relation and could clarify how crystalline materials deform in general. *In situ* straining experiments carried out in a TEM offer the possibility of examining those deformation mechanisms that may be active [1,2]. Dislocation motion, if present, and displacement between grains may be witnessed and captured in real time. Minute changes in grain orientation (potentially on the order of seconds) can result in changes in contrast. The present paper describes such an *in situ* straining experiment of a nanocrystalline copper foil carried out at Los Alamos National Laboratory. It must be kept in mind that the deformation behavior observed in thin foils is not necessarily the same as that in the bulk material.

https://doi.org/10.1557/PROC-634-B1.2.1 Published online by Cambridge University Press

SAMPLE PREPARATION

Samples in this study were compacted from powders made via inert gas condensation [3,4] using a resistive evaporator at Argonne National Laboratory [5,6]. Fresh powder was dumped into a glass beaker while still in the evaporation chamber. Under continuous pumping and at around 10^{-7} Torr, the powder was moved to a compaction unit connected to the synthesis apparatus. Powders were gradually outgassed to prevent spikes in oxygen partial pressure by slowly moving the beaker closer to heat lamps and monitoring the pressure with an ion gauge. When no more pressure increases were seen after approaching the lamp, the powder was transferred to the compaction die and then compacted. Base pressures of both devices were on the order of 10^{-7} Torr. Compaction of the powders was performed at 1.4 GPa (10 tons).

The 9 mm diameter disk-shaped samples initially ranged between 500 and 1500 μm in thickness. To minimize cutting time and to remove surface layers, the discs were ground and polished using a sequence of polishing papers. To assist in the polishing, samples were glued to a steel cylinder 10 mm in diameter using tacky crystal bond (at ~90 °C) and quickly immersed in a beaker of cool distilled water. TEM foils 3 mm in diameter were cut from the thinned 9 mm discs. The TEM samples were then electropolished in a solution of 30% phosphoric acid (HPO_4) and 70% water using Struers Tenupol double-jet electropolisher.

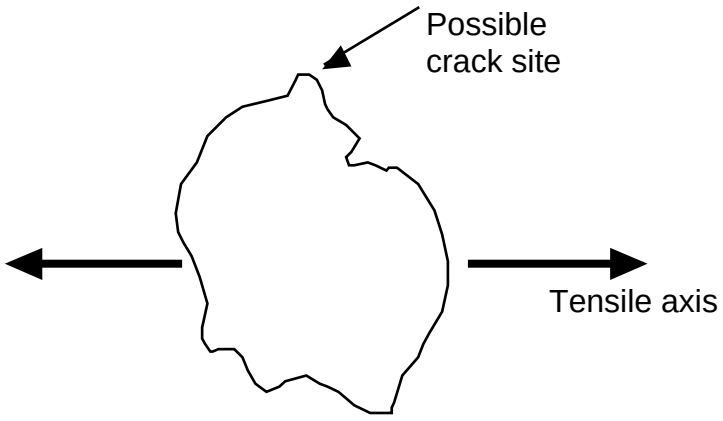

Figure 1. Orientation of TEM hole.

Before a given sample was affixed to the deformation fixture, a low-power microscope was used to find cracks or perforations around the perimeter of the hole. If a site somewhere on the perimeter was identified that appeared likely to produce a propagating crack under load, the sample was briefly examined under the TEM. If the sample looked promising (thin area near the potential crack site), a quick sketch was drawn of the hole and any notable or easily visible features. The sample was then removed from the microscope and affixed to a brass deformation fixture using very small drops of ethyl cyanoacrylate (nail glue) on each side of the hole. The sample was placed on the fixture so that the crack deemed most likely to propagate ran perpendicular to the straining axis, as shown in Fig. 1. The deformation fixture is shown in Fig. 2.

https://doi.org/10.1557/PROC-634-B1.2.1 Published online by Cambridge University Press

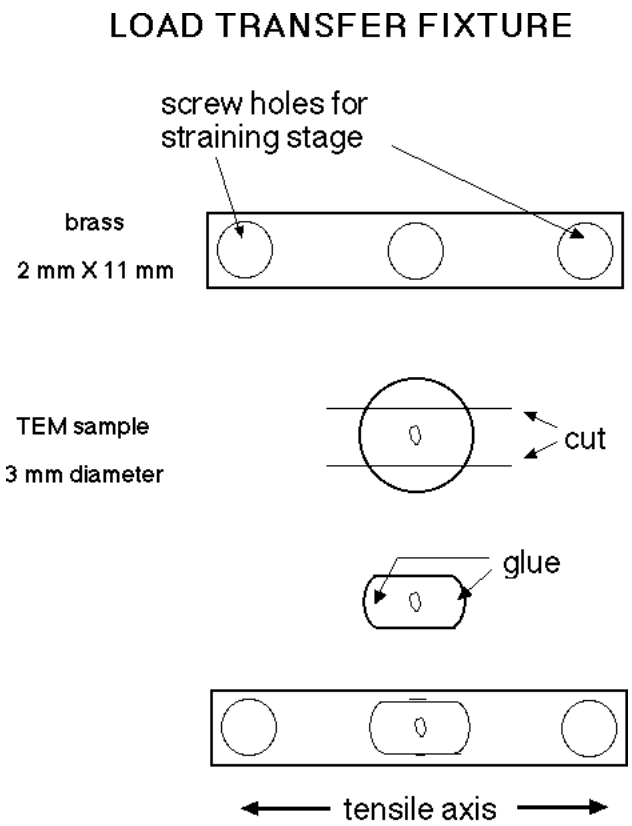

Figure 2. Fixture for supporting TEM samples during *in situ* deformation.

Straining Experiment

The *in situ* TEM tests were performed on a Philips CM30 with a LaB_6 filament and operating at 300 kV. Deformation was induced using a straining stage TEM specimen holder. Besides conventional micrographs, images were recorded digitally. Using a CCD camera running at 30 frames per second, real time "movies" were recorded onto half-inch digital beta videotapes. An attached Macintosh computer with a frame grabber was also used to record some digital images. When videotaping, the action was viewed on a 640 X 480 pixel monitor. The contents of the digital tapes were later transcribed onto consumer grade VHS videocassettes.

In this study, the mobile end of the straining stage was set to move at 100 nm/s. The motor can be set to push or pull the specimen. Tiny screws hold the specimen in the straining stage. The far end is fixed, and the near end moves. Since only 1-3 μm were usually in the field of view depending on the magnification, the motion from the straining stage caused the image to move steadily off the screen during the tests. To keep a particular area in view, the specimen translators were continuously adjusted during specimen extension.

When a sample was ready for straining, an appropriate location to watch was selected. Magnification was usually set to 46 kX, a compromise between obtaining sufficient detail and having a reasonable field of view. After a few pictures were taken straining was begun. The sample was too unstable to take reasonable static pictures during straining. Thus documentation of the microstructural behavior during deformation relied on the video images. The stage pulled on much more than the copper TEM sample within the field of view (brass deformation fixture, glue, and sample) so that the great majority of the displacement was accommodated outside the viewing area. While the overall displacement rate of the straining stage was known, the heterogeneity of the deformation made it impossible to determine the straining rate of the sample.

The first four samples appeared to exhibit some changes in relative positions of grains. When

https://doi.org/10.1557/PROC-634-B1.2.1 Published online by Cambridge University Press

successive pictures were compared via computer, no grain translation was found. It is likely the perceived changes were caused by minor contrast changes from small tilts experienced by the whole specimen during straining. A video image of a typical sample area examined during straining is shown in Fig. 3.

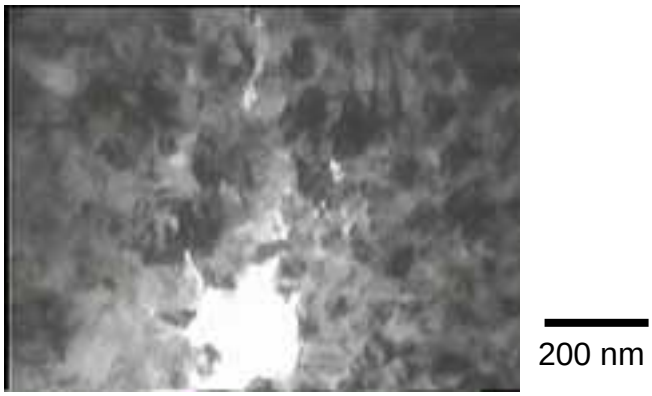

Figure 3. A video image of the crack front during *in situ* tension.

RESULTS AND DISCUSSION

Activity during straining in the form of sudden contrast changes and dislocation motion was seen primarily in the intense stress fields around cracks. The contrast changes took place rapidly, were usually confined to one or a few contiguous grains, and lasted for some tens of seconds. The action then shifted to another grain or grains. The recording VCR runs at 30 frames/second. A frame-by-frame examination failed to unambiguously catch any contrast in the process of changing. Sometimes the changes were clearly confined to a single grain; in other cases it appeared that the changes took place over different regions of a large grain. However, because of grain overlap in the foil, it often was difficult to determine the positions of the grain boundaries and the "different regions" of a large grain may actually have been several small grains. Grains showing contrast changes averaged 60 nm in size as measured from video images. (However, some contrast changes were observed in regions as small as 10 nm.) The average grain size in the foil was measured to be 50 nm, though both this value and that of the average size of grains undergoing contrast changes may be overestimates for the reason just mentioned. A representative picture of the microstructure of a nanocrystalline copper sample is shown in Fig. 4.

Static dislocations were observed in grains as small as, or smaller than, 40 nm. Dislocations were seen moving in several grains, but there was no evidence of pile ups (except in very large grains of $d > 100$ nm) or transmission of dislocation arrays across grain boundaries. In one case, dislocations appeared at the edge of a hole and moved inward, where they seemed to disappear into a dislocation sink. After about 30 seconds the movement of the dislocation array abruptly stopped.

It is not clear if the sudden contrast changes observed in the present *in situ* straining experiment result from dislocation activity or from grain sliding and rotation. In one instance, contrast changes clearly seem to be from dislocation motion. A long grain was observed to be twinned into three parts, the twin boundaries running parallel to the long axis of the grain. The two sections of the "parent" grain underwent extensive contrast changes while the twin in the grain interior remained unchanged. It is unlikely that sliding would take place on a low energy twin boundary. If dislocations are indeed causing these contrast changes, it can be concluded that twin boundaries are effective barriers to dislocation motion.

All crack propagation took place in an intergranular fashion. The cracking appeared somewhat ductile. For example, formerly adjacent grains were later separated by a crack spanning 80 nm. Such positional changes happened gradually rather than via sudden brittle fracture and were not associated with an increase in cracks or porosity. Also, it was noted that throughout the test there were no sudden jolts or skips such as might be expected from an instance of brittle fracture.

https://doi.org/10.1557/PROC-634-B1.2.1 Published online by Cambridge University Press

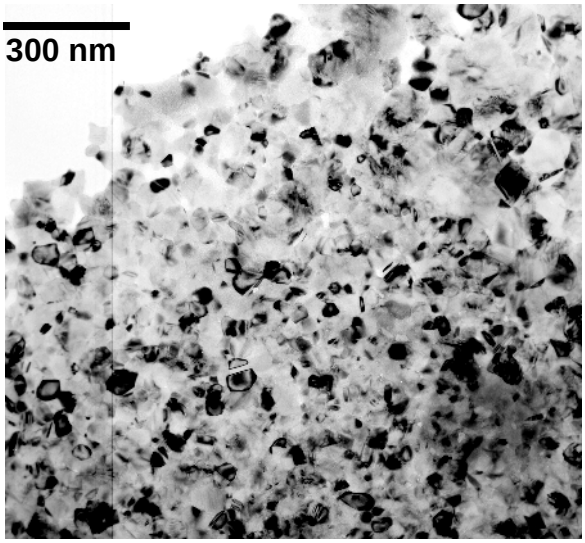

Figure 4. Microstructure of area typical for *in situ* tension tests.

https://doi.org/10.1557/PROC-634-B1.2.1 Published online by Cambridge University Press

CONCLUSIONS

An *in situ* straining experiment was carried out in the TEM on nanocrystalline copper with average grain size of approximately 50 nm.

• Sudden contrast changes were seen in individual grains in the stress field of cracks.

• Generally it could not be determined whether the contrast changes are caused by dislocation activity or by grain sliding and rotation. However in at least one case dislocation motion seems to be responsible.

• Dislocations are observed in grains down to at least 40 nm in size, probably lower.

• Crack propagation in the foil is intergranular.

ACKNOWLEDGEMENTS

This research was partially supported by the Department of Energy's Office of Basic Energy Science and by the LDRD program at the Los Alamos National Laboratory.

REFERENCES

1. W.W. Milligan, S.A. Hackney, M. Ke, and E.C. Aifantis, Nanostructured Materials, **2**, 267-276 (1993).
2. J.E Carsley, A. Fisher, W.W. Milligan, and E.C. Aifantis, Metallurgical and Materials Transactions A. **29A**, 2261-2271 (1998).
3. C.G. Granqvist and R.A. Buhrman, Journal of Applied Physics, **47**, 2200-2219 (1976).
4 R. Birringer, H. Gleiter, H.-P. Klein, and P. Marquardt, Physics Letters, **102A**, 365-369 (1984).
5 J.A. Eastman, L.J. Thompson, and D.J. Marshall, Nanostructured Materials, **2**, 377-382 (1993).
6 P.G. Sanders, G.E. Fougere, L.J. Thompson, J.A. Eastman, and J.R. Weertman, Nanostructured Materials, **8**, 243-252 (1997).