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TEM sample preparation by femtosecond laser machining and ion milling for high-rate TEM straining experiments

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ABSTRACT

To model mechanical properties of metals at high strain rates, it is important to visualize and understand their deformation at the nanoscale. Unlike *post mortem* Transmission Electron Microscopy (TEM), which allows one to analyze defects within samples before or after deformation, *in situ* TEM is a powerful tool that enables imaging and recording of deformation and the associated defect motion during mechanical loading. Unfortunately, all current *in situ* TEM mechanical testing techniques are limited to quasi-static strain rates. In this context, we are developing a new test technique that utilizes a rapid straining stage and the Dynamic TEM (DTEM) at the Lawrence Livermore National Laboratory (LLNL). The new straining stage can load samples in tension at strain rates as high as 4×10^3/s using two piezoelectric actuators operating in bending while the DTEM at LLNL can image in movie mode with a time resolution as short as 70 ns. Given the piezoelectric actuators are limited in force, speed, and displacement, we have developed a method for fabricating TEM samples with small cross-sectional areas to increase the applied stresses and short gage lengths to raise the applied strain rates and to limit the areas of deformation. In this paper, we present our effort to fabricate such samples from bulk materials. The new sample preparation procedure combines femtosecond laser machining and ion milling to obtain 300 μm wide samples with control of both the size and location of the electron transparent area, as well as the gage cross-section and length.

1. Introduction

Metallic alloys of aluminum, titanium, and magnesium are commonly used in the automotive, aerospace and military industries, and many studies have been conducted to optimize their resistance to deformation at quasi-static strain rates [1–6]. Similar efforts are also needed at high strain rates (up to 10^5s^{-1}) to design the next generation of armor materials. The nucleation and motion of dislocations, twins, and cracks must be characterized at high strain rates to effectively predict deformation during ballistic impacts, particularly since they can change as strain rate increases [7]. Compression experiments conducted on polycrystalline magnesium at strain rates ranging from 10^{-3} to 10^3s^{-1} show a change of the main deformation mechanism from twinning at low strain rates to a combination of twining and dynamic recrystallization at high strain rates [8]. Researchers have also shown that twinning and hardening are strain rate dependent in the Mg alloy AZ31B [9]. In general, a greater understanding of high rate deformation mechanisms is needed to accurately predict properties that are critical for armor applications such as dynamic strength and spall strength.

To date, microscopy studies of metals that have been deformed at high strain rate have been limited to *post mortem* analysis; *e.g.*, SEM, EBSD, or TEM studies after deformation. To capture the evolution of both microstructure and defects during loading at micro and nano-scales, several *in situ* TEM techniques have been developed [10,11], and much information can be extracted from *in situ* TEM experiments that cannot be gained from *post mortem* studies [11]. For example, *in situ* TEM tensile tests performed at room temperature on superalloys [12] and titanium aluminides [13] confirmed that the nonlinear motion of screw dislocations can be governed by lattice friction as dislocations jump between Peierls valleys in cubic phases. Another *in situ* study showed that lattice friction does not play a significant role in prismatic glide in zirconium [14], and a fourth, realized on pure magnesium single crystals at room temperature [15,16], showed that the jog-pair (or Friedel-Escaig) mechanism controls the gliding of screw dislocations in non-basal planes. The important findings in these studies could

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not have been obtained by *post mortem* studies. *In situ* observations were needed.

Unfortunately, current straining stages limit *in situ* TEM testing to quasi-static loading. Two reasons can be offered for this limitation. One is that TEM sample holders offer limited space in which to fit more rapid loading mechanisms and another is that conventional TEM electron detectors cannot image at rates greater than 30 frames per second [10]. Recently, a new direct detection camera has been developed by Gatan to capture TEM pictures at rates as high as 1600 frames per second [17,18]. However, such frame rates are still not fast enough for use in high strain rate experiments. Currently, the only *in situ* techniques capable of characterizing deformation at high strain rates are X-ray synchrotron methods, but these methods only provide statistics of the deformation at the macroscale [19]. To perform high strain rate mechanical testing within a TEM, unique microscopes, strain stages, and sample preparation methods are needed.

Rapid imaging within a transmission electron microscope can be accomplished using the Dynamic TEM (DTEM) at the Lawrence Livermore National Laboratory (LLNL). Unlike conventional TEM, imaging in the DTEM is accomplished using a series of nine electron pulses that are generated by a pulsed laser beam impacting a metal target. The electron pulses are deflected across the sample, and then are distributed onto different locations of a single detector. This allows one to record nine images, each one taken every 50–2500 ns with a temporal resolution of 20 ns and a spatial resolution of 20 nm [20]. A complete description of the instrument's functioning and applications can be found in reference [21].

Current TEM sample holders dedicated to *in situ* mechanical testing typically load samples in tension or by indentation using an electric motor and a gear box located outside the TEM [10]. This combination permits one to apply large forces and strains to samples. However, the displacement rates are limited to a maximum of 10 μm/s. A new emerging technique uses MicroElectroMechanical Systems (MEMS) and allows one to deform nanoscale samples with very accurate measurements of force and the displacement [22,23]. Unfortunately, MEMS-based systems typically limit studies to a few nano-grains, which is incompatible for most structural materials. More importantly, this technique cannot enable high-rate straining experiments. To reach strain rates of $10^3 s^{-1}$ or higher, we designed a new TEM holder using two bending piezoelectric actuators mounted in parallel to pull samples in tension [24]. The maximum relative displacement speed of the piezo actuators is 200 mm/s, which allows one to achieve strain rates ranging from quasi-static to $10^4 s^{-1}$. However, the maximum force that can be applied on samples is limited to 200 mN. This force restriction, and the desire to reach very high strain rates, required us to design and develop novel TEM specimens for rapid *in situ* testing in tension.

In designing novel TEM specimens, several restrictions were considered. First, the specimens must be fabricated from bulk samples with micro-scale microstructures if conventional structural materials are to be studied. Second, the samples need to be small enough in cross-sectional area (limited width and thickness in the gauge section) to deform in tension at strain rates as high as $10^4 s^{-1}$ when the maximum applied force is only 200 mN. However, the cross-sectional area must be large enough to allow handling of the samples in their free-standing form. Third, specimens also need to be shorter than 2 mm in length to fit into the TEM holder [24]. Fourth, the gauge geometry and thus, the size and location of the electron transparent area, must offer relatively consistent and repeatable straining from one specimen to another. Finally, the sample preparation technique must allow the fabrication of numerous samples in a relative short time as many *in situ* experiments will be necessary to ensure a perfect synchronization between loading, defect motion and imaging.

This paper presents two attempts to address these challenges using two model materials. In the first, unsuccessful attempt, specimens were prepared from rolled Al and Cu foils using electropolishing and in the second, successful attempt, specimens were prepared from rolled Cu foils by combining femtosecond laser machining and ion milling. We also consider the effect of laser machining on the microstructure of the Cu foils and how best to optimize the geometry of the specimens.

2. Experimental

99% pure 15 μm thick aluminum foils and 10 μm thick 99% pure OFHC copper foils were used for the conventional preparation of samples *via* electropolishing, and 10 μm and 25 μm thick 99% pure OFHC copper foils were used for the laser machining and ion milling of specimens. Copper was chosen over aluminum for laser machining because more extensive literature is available for this manufacturing process [25–30]. All foils were annealed prior to specimen preparation to generate a coarse grained microstructure with a low dislocation density.

The electropolishing was realized with a Tenupol-5 double-jet electropolisher developed by Struers. Aluminum samples were electropolished with an electrolyte made of 25% Perchloric acid, 25% methanol and 50% distilled water at −10 °C and 20 V. Copper samples were electropolished with an electroplyte made of 10% Phosphoric acid and 90% distilled water at −30 °C and 20 V. Ion milling on copper samples was performed at liquid nitrogen temperature (close to −180 °C) with argon ions in a Precision Ion Polishing System (PIPS II) produced by Gatan using the following recipe:

1. 3 V/20 min with a top beam angle of 10° and a bottom beam angle of 5°
2. 2 V/10 min with a top beam angle of 5° and a bottom beam angle of 5°
3. 1 V/10 min with a top beam angle of 3° and a bottom beam angle of 3°

unless specified otherwise. A Clark MXR CPA Femtosecond laser was used to machine small copper samples with a 25 μm spot size at energies varying from 1 to 10 mJ.

Several microscopes were used to characterize the samples that were prepared during this study: A conventional optical microscope with magnification ranging from 1.4x to 18.0x, a Tescan MIRA-3 FEG Scanning Electron Microscope (SEM) coupled with an Electron Back Scattered Detector (EBSD) operating at 20 keV, and a Transmission Electron Microscope (TEM) CM300FEG developed by Philips operating at 300 keV. A Memrecam HX6 high speed camera was used to record *ex situ* experiments at 15,000fps.

2.1. Electropolishing

Two conventional TEM sample preparation methods were considered initially – Focus Ion Beam (FIB) and electropolishing. FIB fabrication can position electron transparent areas very accurately within small samples [31,32], but the method was considered too slow and expensive for this study given that many samples are required. In addition, FIB fabrication can introduce damage in the samples [33–35], such as ion implantation and dislocations loops, both of which can influence the deformation mechanisms that are active during loading. Thus, we chose electropolishing as the preferred conventional method for fabricating multiple samples at a reasonable cost [36]. All samples, either aluminum or copper, were cut from the 15 or 10 μm thick foils, respectively, and were glued onto TEM aperture grids with silver paint as shown in Fig. 1. The grids were used so that the samples can be handled during processing without damage. In addition, we varied the positioning of samples relative to the 0.3 mm hole at the middle of the aperture grids to identify the best location for insuring that the final specimen has both electron transparent regions that can be deformed on loading and unpolished regions that provide structural support and prevent unwanted deformation during handling (Fig. 1(a) & (b)). As