Scripta Materialia 153 (2018) 63–67

Contents lists available at ScienceDirect

Scripta Materialia

journal homepage: www.elsevier.com/locate/scriptamat

Regular article

The origin of surface microstructure evolution in sliding friction

Christian Greiner [a,*], Zhilong Liu [a], Reinhard Schneider [b], Lars Pastewka [a,c], Peter Gumbsch [a,c]

[a] Institute for Applied Materials (IAM), Karlsruhe Institute of Technology (KIT), Kaiserstrasse 12, 76131 Karlsruhe, Germany
[b] Laboratory for Electron Microscopy (LEM), Karlsruhe Institute of Technology (KIT), Engesserstrasse 7, 76131 Karlsruhe, Germany
[c] MicroTribology Center µTC, Fraunhofer IWM, Wöhlerstrasse 11, 79108 Freiburg, Germany

ARTICLE INFO

Article history:
Received 30 January 2018
Received in revised form 16 April 2018
Accepted 28 April 2018
Available online xxxx

Keywords:
Friction
Copper
Scanning/transmission electron microscopy (STEM)
Dislocation
Microstructure

ABSTRACT

A typical tribologically induced microstructure displays a discontinuity parallel to the surface separating the near-surface layer from the bulk. Despite its ubiquitous observation, the origin of this layer underneath the surface remains elusive. Here, we show that already after the first loading pass a localized dislocation structure is found at a depth of 100–150 nm. This structure is linked to the inhomogeneous stress field under the moving indenter, where dislocations are trapped when the stress drops below the material's yield stress. Control of the initial tribological loading could therefore be exploited to design surfaces and materials with superior tribological properties.

Tribology, the science and technology of interacting surfaces in relative motion, is of great importance for many aspects of modern life [1]. Friction and wear of metallic materials are crucial for energy efficiency and durability of many products from combustion engines [2] to artificial limbs and joints [3]. In the 1950s, Bowden and Tabor pointed out that frictional energy dissipation is mostly due to plastic deformation of the near-surface layer. The coefficient of friction largely depends on the yield strength of the softer material in the contact [4]. Plastic deformation induces changes in the material's microstructure [5–11]: Near-surface nanocrystalline regions consistently show grain growth [12–14], while applying load to an annealed, large-grained material leads to grain refinement near the surface [3,8,15–17]. In both cases, experiments regularly show a distinct discontinuity in the microstructure between a surface layer and the underlying bulk material [3,7,12,15–18]. The tribological surface therefore comprises not only the interfacial plane but also the region of the modified material underneath [19,20]. This modified surface materials have been given a variety of names in the tribological literature: Tribomaterial [21], tribomutation layer [22], Beilby layer [23], third body [24] and others. It is desirable to elucidate the origin of this discontinuity in order to significantly advance our understanding of friction and wear, because the surface layer is decisive in determining the tribological properties of the contact. The goal of the present study is to find the source of these discontinuities and to understand tribo-induced microstructures at early stages of sliding.

Our tribological model system is a hard and inert sapphire sphere (polished, with a diameter of 10 mm, from SWIP, Switzerland) sliding on annealed pure copper (OFHC copper with a purity >99.95%, from Goodfellow, Germany) as shown in Fig. 1. Copper samples were prepared following a procedure outlined in ref. [17]. Cu-5wt%Zn alloy (Wieland, Germany) and pure nickel with a purity >99.99% (Goodfellow, Germany) were chosen as additional materials to be tested. The sample preparation for both materials differed from that for copper as follows: The normalizing temperature for the Cu-5wt%Zn alloy was 550 °C and for pure nickel 750 °C. For nickel, a second normalizing step after mechanical polishing was followed by the final electro-polishing in A2 electrolyte (Struers, Germany). This allowed eliminating microstructural defects introduced during the mechanical polishing as well as removing any oxidation on the sample surface.

A linear tribometer with unidirectional and reciprocating operation modes (Fig. S1) was employed for the dry sliding experiments [25]. The tribological tests were started with only one linear pass of the sapphire sphere on the copper plate, deliberately concentrating on the origin of an often observed sharp boundary in the subsurface microstructure between the tribologically deformed layers and the rest of the material. For investigating the effect of normal load on the stability of the dislocation trace line, we used normal loads of 0.5 N, 1 N, 2 N and 14 N, with corresponding maximum Hertzian contact pressures of 334 MPa, 420 MPa, 530 MPa and 1014 MPa. All other parameters, i.e. the sliding speed of 500 µm/s and the stroke length of 12 mm, were kept constant. The tests were conducted at room temperature and in air with 50% relative

* Corresponding author at: Karlsruhe Institute of Technology (KIT), Institute for Applied Materials (IAM), Kaiserstrasse 12, 76131 Karlsruhe, Germany.
E-mail address: greiner@kit.edu (C. Greiner).

https://doi.org/10.1016/j.scriptamat.2018.04.048

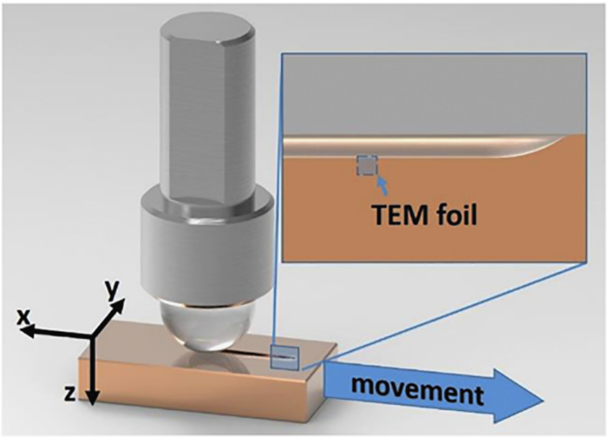

Fig. 1. Schematic diagram of the experimental setup. Perpendicular to the sample surface and in sliding direction, TEM foils are prepared by focused ion beam machining.

humidity. For testing the influence of multiple passes, we ran 2, 4, 6, 8, 10 and 20 unidirectional passes at a constant normal load of 2 N. Other experimental parameters are shown in Table S1. The profile of a wear track after 2 N single pass loading was examined by optical profilometry in phase shift interferometry mode (PLu Neox from Sensofar, Spain). The profile of the wear track is shown in Fig. S2. The friction forces are presented as a function of sliding time/distance in Fig. S3.

The microstructure was investigated using a focused ion beam/scanning electron dual beam microscope (Helios NanoLab™ 650 from FEI, USA). A state of the art focused ion beam lift-out technique was applied to prepare the TEM foils for this study [26]. The surface was protected by depositing two platinum layers, the first one employing the electron beam only. Scanning transmission electron microscope (STEM) images were taken with an acceleration voltage of 30 kV and beam currents of 50 pA or 100 pA. Conventional and high-resolution TEM as well as the energy dispersive X-ray spectroscopy (EDXS) were conducted at an acceleration voltage of 300 kV with an FEI Titan^3 80–300 microscope, which has a spherical-aberration corrector for the imaging ray path. X-ray photoelectron spectroscopy (XPS) measurements (PHI 5000 Versaprobe System with 15 keV monochromatic Al-Kα-X-ray excitation and an energy resolution of 0.2 eV) of an area of 1 mm × 1 mm on the contact area of a sapphire sphere after 1000 cycles of loading has been performed. The measurement was directly after the tribological test without cleaning the sphere tip. TEM foils were prepared for cross-sectional view at the center of the wear track, parallel to the sliding direction and perpendicular to the sliding surface. The depth of the trace line – the distance between the surface and the trace line – was measured at five different positions in each image; then the average value and the standard deviation were calculated.

Transmission Kikuchi diffraction (TKD), with a spatial resolution of 2–20 nm [27], was used to acquire crystallographic orientation information in cross-sectional areas. It is known that the step size plays an important role in the estimation of the misorientation measured as the density of geometrically necessary dislocations [28]. We chose a step size of 20 nm for our TKD measurements to guarantee reliable results with enough spatial resolution and a sufficiently low level of artefacts brought by contamination. TKD scans were taken above and below the trace line on cross-sectional areas of 2.2 × 3 μm^2. The misorientation was determined by comparing all pairs of points with the same horizontal position being 60 nm above and below the feature.

Misorientation is interpreted as the density of geometrically necessary dislocations (GND) [29], ρ_{GND}:

$$\rho_{GND} = \frac{2\vartheta}{ub}, \tag{1}$$

where u is the distance between the two points used to calculate the misorientation and b the length of the Burgers vector of a $\frac{1}{2}\langle 110\rangle$ dislocation (0.255 nm for copper), which is the most common perfect dislocation in fcc crystals. This method assumes a cube with an edge length of u and each edge type GND in this cube will stretch one edge with the length of one Burgers vector. The GND density can then be correlated with a misorientation tilt angle, regardless of slip system [30]. This method gives results very close to a more detailed analysis [29]. As the angular resolution limit of our electron backscatter diffraction setup is 0.08°, the lowest GND density which could be detected with our method is 5×10^{13} m^{-2} [31]. This limit also describes the minimum error of the GND analysis [31].

We observe the formation of a distinct microstructural discontinuity already after a single (unidirectional) pass of the sapphire sphere. To elucidate the character of this discontinuity, we measure crystallographic orientation differences by TKD on focused ion beam cuts into the surface in the middle of the wear track parallel to the sliding direction. The result for a single pass with a 2 N load is presented in Fig. 2A, which displays the misorientation of any pixel with the neighboring four pixels [27]. This misorientation is the signature of the dislocation density that is geometrically necessary to generate a corresponding tilt of the crystal structure. Fig. 2A shows a line-like feature with significantly increased dislocation density roughly 140 nm under the surface. TKD scans over this line reveal an average misorientation of 4.6° between the region above and below. This indicates that the line-like feature effectively has the character of a small angle grain boundary.

To further clarify the nature of the line, high-resolution transmission electron microscopy (HRTEM) is performed after 2 N single pass loading on another specimen (Fig. 2B, C). The line-like feature is at a depth of 114 ± 8 nm. The brighter appearance of the layer above the line in Fig. 2B indicates a dramatically reduced dislocation density. The zone axis (ZA) is a [103] direction, as determined from the fast Fourier transform image of the area beneath the line. The {200} atomic planes visible in Fig. 2C are rotated by 7.9° (marked as α in Fig. 2C). The high-resolution TEM image in Fig. 2C shows that the line itself consists of many dislocation-like contrast features and has a width of 5–8 nm. Hence, it clearly is not an ideal small angle tilt or twin boundary but rather a complex self-organized dislocation network composed of many dislocations with different Burgers vectors. Energy dispersive X-ray spectroscopy (EDXS) spectra from the areas above and beneath this line (Fig. S4) show no obvious difference in oxygen concentration, excluding the influence of oxidation on its formation. XPS spectra (Fig. S5) indicate that there is no material transfer during our experiments. The line therefore is referred to as a dislocation trace line in the following.

We hypothesize that the motion of dislocations within the strongly inhomogeneous stress field below a sliding spherical indenter determines the formation of the trace line. The main contribution to the force on dislocations moving in the cross-sectional area underneath the indenter is the in-plane shear component σ_{xz} of the stress field [32]. Fig. 3A shows σ_{xz} obtained from a superposition of Hertz's solution and frictional surface traction [33] (see Supplementary materials). The friction coefficient between copper and the sapphire spheres in our experiments was around 0.25, as measured in several independent experiments (Fig. S3A). Simplifying the discussion to four perpendicular slip systems underneath the indenter, dislocations with a Burgers vector parallel to the surface either follow the motion of the slider or are left behind. Dislocations with a Burgers vector perpendicular to the surface are either pushed into the bulk or they disappear at the surface. This generates an area of reduced dislocation density near the surface, which explains the brighter contrast above the trace line in the TEM image in Fig. 2B. As the sphere continues to move, the sign of σ_{xz} changes for a material point below the indenter (the solid line in Fig. 3 shows the location of the sign change), pulling dislocations that were previously pushed into the bulk back up towards the surface again (Fig. 3B). In the near-surface region at the trailing edge of the indenter, σ_{xz} steeply goes to zero underneath the surface. The exact location of zero stress depends on the friction coefficient and load (see