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Micro and Nano-structure Development and Multiscale Physics at Sliding Metal Interfaces

PI: D. A. Rigney, The Ohio State University,
in collaboration with Los Alamos National Laboratory

Introduction:

This project is concerned with the response of ductile materials to extreme loading conditions such as those involved in impact loading with sliding friction. The research is performed by a multi-disciplinary team with backgrounds in physics, mechanical engineering and materials science and engineering. The work includes impact and sliding tests, materials characterization and computer simulation. The combined impact with sliding work is at Los Alamos National Laboratory (LANL), complementary sliding work is at The Ohio State University (OSU), materials characterization is primarily at OSU, and computer simulations are in progress at both sites.

The project grant announcement was made on August 30, 2002. The award papers were received at OSU on February 3, 2003, with an official project start date of Dec. 15, 2002. The OSU team was assembled during autumn, 2002, and planning and preliminary work began before the official start date. The four members of the Los Alamos (LANL) team visited OSU on November 15, 2002, for a full day of discussions, lab tours and project planning. The first test samples were sent by LANL to OSU in December, 2002. One member of the OSU team worked at LANL during the summer of 2003.

Project personnel:

At OSU: D. A. Rigney, PI; K. Subramanian (post-doc); A. Emge and H. J. Kim (PhD candidates)

At LANL: P. Rightley and K. Rainey (Mech. E.); J. Hammerberg (Physics); P. Crawford (MSE).

Coordination among team members has been accomplished via email, telephone, discussions at conferences, by a visit of the LANL team to OSU, and by an extended visit of one OSU student (Kim) at LANL.

Experimental Progress at LANL:

The most significant accomplishments involved improvements in the impact test system and determination of the most appropriate surface preparation for the test specimens. During the past year, several improvements and modifications were added to the Rotating Barrel Gas Gun (RBGG) facility. Most notable was the incorporation of a camera/flash system that enables us to take two consecutive images of the projectile just prior to impact. With careful markings on the projectile,

we will now be able to determine more accurately the longitudinal and rotational speeds of the projectile. To improve the automation and repeatability of each experimental test, a laser/photo detector trigger system was also added to the RBGG. This system allows precise timing of the data collection for both digital images and elastic wave impulses. All triggering and timing of images and lighting are now controlled via a custom Labview program. During July and August, H. J. Kim from the OSU group worked at LANL with P. Crawford and K. Rainey. Together, they were successful in implementing these latest and important modifications of the RBGG. Other modifications to the RBGG were performed to improve the safety of the electrical components, as well as to improve the ease of operation.

Repeated testing of the modified and updated RBGG has been performed during the last 6 months. All tests have used specimens of OFHC copper, 99.9% Cu. These tests have been successful in optimizing several aspects of the system, including the lighting conditions for the digital camera, the custom LabView program, alignment procedures for the target rod and the projectile and the acquisition of a square top elastic wave profile. Rainey and Crawford have also started plans for the building of a second generation RBGG. This new RBGG experimental facility will have the ability to reach rotational speeds of 100,000 rpm. A novel impulse-turbine-type design powered by pressurized air/gas will provide the rotational motion for the barrel, while helping to eliminating some of the electrical noise currently seen in the elastic wave signals. This new version will also have further modifications to improve both the operation of the RBGG and the data acquisition systems.

The holder for the target rod was also modified in an effort to improve the accuracy and precision of the alignment. The initial holder allowed for alignment of the target rod and the projectile using an alignment rod that was passed through the center of each. The new holder is composed of 4 optical stages capable of adjustments in the micron range. Numerous tests were performed using OFHC Cu target rods and projectiles to determine which set up is optimum for providing a clear elastic wave impulse signal and for repeatability/uncertainty estimates. These data are currently being analyzed.

We also investigated the effect of an epoxy joint on the stress seen by the sample. As mentioned in the conference paper cited below [1], there is some concern that the epoxy joint may cause some reflection of the impact wave in the sample, leading to plastic deformation. Figure 1 shows the stress data collected for a RBGG test using an epoxy joint. The residual stress seen in the axial direction indicates some residual strain in the sample, possibly caused by plastic deformation as a result of the impact. This effect has been repeated in specimens where the impact samples were attached to the target rod and projectile by a high strength epoxy. In addition, it was noted that the shear stresses reached at rotational speeds of greater than or equal to 3000 rpm (4m/s) were exceeding the shear strength of the epoxy. As a result, other methods of attaching the samples were considered. A mechanical joint was designed and tested. Tests indicated that this new joint was not transferring all of the shear stress seen by the sample to the strain gauges on the target rod. A decision was made to proceed with testing and data collection using projectiles and target rods without samples, eliminating any effects of a joint. Further consideration of this situation regarding the use of a joint is ongoing.

The initial focus of tests with the RBGG is on interfaces of copper/copper and aluminum/stainless steel. Subsequent testing will involve aluminum/tantalum as well. The following materials have been purchased and are being made into projectile and target rods for testing: Cu (OFHC-99.99% pure), Al (99.99% pure and 6061-T6), SS (21-6-9). To provide the OSU team with samples having optimum microstructures for examination, a detailed machining and heat treatment method has been developed for the OFHC Cu material. Similar methods are being developed for the remaining materials.

Currently, the RBGG facility is ready to begin testing with the Cu and Al/SS materials. These data

will be analyzed for the behavior of the kinetic coefficient of friction at varying impact loads and rotational speeds.

Experimental Work at OSU:

The OSU portion of this research project is focusing on the frictional behavior and structural changes in ductile FCC metals subjected to impact loading with sliding. The OSU group is examining the samples tested in LANL's novel RBGG and comparing them with samples produced at lower sliding speeds at OSU.

The experimental work at OSU during this reporting period has involved three tasks: (1.) designing and building an improved system for sliding tests at intermediate velocities, (2.) developing appropriate pre-testing surface preparation and (3.) developing post-test characterization techniques.

New Sliding Test System:

To complement the friction data provided by the Los Alamos, the wear group at OSU has designed and built a new pin/disk wear testing system. The older pin/disk system was capable of sliding speeds up to 5 cm/second. The new pin/disk wear testing system can achieve sliding speeds up to 1 m/s. The new set-up is also designed to create contact between the pin and disc for extremely short periods of time, as small as 0.1 s, and the system can be easily modified for longer tests. Coupled with the high acquisition rates, this enables the probing of dynamic friction behavior. It is possible to run these tests in a variety of environments including air, vacuum and nitrogen. Through a series of mechanical and turbo-molecular pumps, vacuum levels as low as 10^{-8} torr are expected in the testing chamber. The results of tests with this apparatus will bridge the high-speed friction data from Los Alamos and the lower speed data, obtained with the existing system. These are currently separated by nearly two orders of magnitude in sliding speed. The intermediate speed tests will also provide ample wear-tested material for characterization purposes. Results are expected soon from tests run on different tribo-pairs including stainless steel (SS) on Al, SS/Cu and Cu/Cu, at velocities up to 0.5 m/s at various loads.

A schematic of this apparatus is shown in Figure 2. In order to calculate the coefficient of friction, the tangential load (friction force), and the normal load must both be measured. A full bridge strain gauge configuration is used to measure the tangential load and a quarter bridge configuration is used to measure the normal load. The output from the strain gauges first goes through a strain conditioner and is then acquired by a computer for analysis.

The normal load is applied by lever arm action. A weight is suspended from a pulley on the end of the arm opposite the pin and disk as can be seen in Figure 2. The loading and unloading of the arm is done with the use of an electromagnet controlled by a timer. With the magnet switched on the arm is in an unloaded state. When the magnet is turned off the system is loaded. With the current timer, contact times as low as 0.1 seconds are possible. The rotational velocity is measured by a magnetic pick-up on the shaft of the motor. The entire apparatus is located inside a vacuum chamber, allowing control of environment.

The initial tests have used 25 mm diameter disks of Oxygen Free High Conductivity (OFHC) copper (99.99%Cu). The pin used for the tests is a 6 mm diameter ball. Both OFHC copper and 440C stainless steel balls will be used for the tests. The tests will be performed under various atmospheres such as air, nitrogen, and vacuum. The experiments will be performed at sliding velocities from 5-50 cm/s with loads ranging from 50-200 g.

The results from one of the first tests are shown in Figures 3 and 4. This test was performed with a

440C steel ball sliding on an OFHC copper disc. The sliding velocity was 5 cm/s, and the load was 50 g. The time of contact for the test was one hour. The resultant normal and tangential loads are shown in Figure 3 as a function of time. The coefficient of friction is calculated from the normal and tangential loads and is shown in Figure 4.

After the test the pin and disc are analyzed in the SEM. The analysis includes a study of the size and appearance of the wear track. The composition of wear debris and the presence of transfer material is checked with EDS. Once the pin and disc have been studied with the SEM they are prepared for TEM analysis.

Pre-Test Sample Preparation:

One of the primary concerns for this project was to determine the best sample preparation methods for the tests. A majority of the deformation from the tests is expected to be located relatively close to the sliding surface. This requires that before the tests are performed, the sample must have a minimal subsurface deformation from the machining processes. Early TEM analysis of a machined sample revealed extensive deformation just below the machined surface. Due to these factors, a study was conducted to find a machining method that would minimize subsurface deformation. The three machining methods analyzed were lathe turning, flycutting, and electrical discharge machining (EDM). Post-machining annealing was also considered as a method of removing deformation remaining from the machining processes.

The surfaces of the machined samples were analyzed with optical microscopy and scanning electron microscopy (SEM). Microhardness tests were used to obtain hardness values as a function of distance from the machined surface. This was done for an unannealed lathe turned sample as well as for one sample from each machining method that was annealed. The annealing process was carried out at 275°C for one hour in a vacuum furnace. The results of the microhardness tests are indicated in Figure 5. The results from the microhardness tests show that the annealed EDM and flycut samples had similar hardness values that were considerably less than for the other machining methods. However, SEM surface analysis reveals scratches on the EDM surface, but not on the flycut surface. It is also known that the EDM process produces a heat-affected zone at the machined surface. From these results, flycutting along with the annealing was chosen as the preferred method for preparing specimens for RBGG testing at LANL.

Due to its very high ductility, the preparation of the aluminum samples needed to be done particularly carefully. Aluminum disks were cut by flycut machining to minimize machining effects on the subsurface microstructure. To remove the deformation history, all aluminum samples were annealed at 200°C for 1 hour.

Characterization:

In order to select suitable pre-testing sample preparation techniques, Transmission Electron Microscopy (TEM) was done on longitudinal and transverse sections of the as-machined annular OFHC copper samples. This revealed extensive subsurface deformation in the form of aligned cell structure with very high dislocation density. These features would complicate our efforts to study the changes produced by impact with sliding. The samples should have a minimal amount of subsurface deformation prior to testing, so the deformation due to sliding will not be obscured. Therefore, as discussed in the previous section, a study was conducted to find a test specimen preparation method that would minimize subsurface deformation. Three machining methods were analyzed: lathe turning, fly-cutting, and electrical discharge machining (EDM). Post-machining annealing at 275°C for one hour in a vacuum furnace was also performed to remove deformation remaining from the machining processes. Microhardness was measured as a function of the distance from the machined surface. This was a simple way to determine the extent of subsurface

deformation. The results show that annealed fly-cut samples are best for our purposes. Similar tests on pure aluminum samples suggest that annealing of fly-cut samples at 200°C for an hour is sufficient to remove subsurface deformation. Both Al and Cu samples are chemically polished before testing.

The material tested at OSU was characterized using optical microscopy, SEM and TEM. Wear tracks and wear debris were analyzed using SEM and energy dispersive spectroscopy (EDS). TEM samples were prepared using different techniques including dimpling, jet-polishing and chemical polishing. Innovative techniques involving a Focused Ion Beam (FIB) have also been explored.

TEM Sample Preparation Techniques:

A large part of the OSU work up to this point has involved optimizing TEM sample preparation techniques. The TEM sample preparation methods for the copper rings from the RBGG tests and the pin on disc tests are as follows. The sample is first sectioned on a wafer saw using a SiC blade. Next, the wear surface of one section is electroplated with copper. The electroplated layer is approximately 3 mm thick and is composed of two copper layers deposited with different solutions. The first layer is approximately 20 μm in thickness and is deposited from a solution of 500 ml distilled water, 17 g NaCN, 11 g CuCN, 7.5 g Na_2CO_3 , and 2.5 g NaOH. This first plating process uses a current density of $10\text{mA}/\text{cm}^2$ for one hour.

The second solution is composed of 500ml distilled water, 90 g $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$, 0.5 g phenol, and 12.2 ml H_2SO_4 . The current density for the second solution is $75\text{--}100\text{ mA}/\text{cm}^2$. The second plating process takes 5-6 days to plate the remainder of the 3mm layer. Nitrogen is slowly bubbled through the electroplating solutions during each step to ensure even plating.

Once the electroplating is completed a wire saw is used to further section the electroplated section. The wire saw uses a SiC slurry and produces sections approximately 1mm in thickness. Both transverse and longitudinal sections are made with the wire saw. After sectioning with the wire saw the specimen is manually polished to a final thickness of 100-125 μm with SiC polishing papers. The sample is then dipped in a chemical etchant solution, which reveals the interface between the electroplated layer and the bulk material.

The next step is to use a slurry drill to create a 3mm disk from the sample. The sample is drilled so that the electroplated/bulk interface is located at the center of the disc (i.e., half of the disc is the bulk material and the other half is the electroplated material). The disk is then jet-polished to produce a thin area at the center that is electron transparent for transmission electron microscopy. Polishing is performed on a Fischione model 110 submerged twin jet electropolisher. The jet polishing solution is 300 ml distilled water, 800 ml H_3PO_4 , and 100 g CrO_3 . The jet polishing is conducted with a voltage of 3.5 V at 25°C. The thin area is located at the bulk/electroplating interface, which means the sliding surface and the area just below it will be visible in the TEM. After jet polishing the disks are ready for TEM analysis.

For aluminum specimens, several methods were tried. The best method involved the use of Torr Seal epoxy. It has a low vapor pressure and can be used in a vacuum chamber. The wear track was first covered by this epoxy to protect the surface. After curing, the mounted specimen was cut into slices. Mechanical polishing with 800 and 1200 grit SiC paper and slurry drilling followed. Finally, jet polishing created a small hole and a suitably thin area around the hole. The grain size was more than 500 μm , and some grains had mm-scale size. TEM showed that the sample contained only a small dislocation density, as desired for the starting material. Before sliding tests, all samples were mechanically polished with 800 grit and 1200 grit SiC paper. Subsequently, the samples were chemically polished. This removed $\sim 20\text{ }\mu\text{m}$ of material from the surface and ensured similar

surface conditions for all the tests. The normal load and sliding time were fixed at 50 g and 6 hours respectively. Sliding velocities ranged from ~1.3 cm/s to ~7.0 cm/s.

Preliminary Results of Sliding of Aluminum:

Preliminary tests used a pin of 440C stainless steel sliding on a disk of 99.99% aluminum. Figure 6 shows friction coefficient data vs. sliding time. Pure aluminum is much more ductile than stainless steel and the friction coefficient is high (above 1). All friction coefficient values are higher at the first stage than those observed at steady state. Sliding velocity and corresponding average friction coefficient results are summarized in Figure 7. Average friction coefficients decrease slightly with increasing sliding velocity. While the friction coefficient during early stages of sliding decreased with increasing velocity, the steady state values showed no such trend. After sliding, the wear track was characterized by SEM and EDS. Adjacent to the wear track, evidence of localized shear is observed (Figure 8). As sliding velocity increases, the amount of oxidized aluminum increased. This is due to the breaking of surface oxide and mechanical mixing of the oxide with the aluminum matrix.

The new pin-on-disc system provides more accurate data acquisition, higher velocity capabilities, and short time friction tests. Data acquired for a test in air using the new pin-on-disc system are shown in Figure 9. The initial normal load is 50 g, which is the same as the previous low speed tests and the sliding time is 5 minutes. The average friction coefficient is 0.194, which is much lower than the low speed and long time sliding tests.

For a closer comparison with LANL experiments, short time, extremely short time (less than 1 s) and high-speed friction tests will be performed. The new pin-on-disc system will allow this. Chemical analysis of surfaces, using XPS and/or SIMS, will help us to understand surface composition changes. Previous studies have indicated that environment has an effect on the friction behavior. Therefore, it is necessary to perform tests under different atmospheres, such as vacuum, nitrogen and air.

Training:

Students were trained in the use of OSU's XL-30 Environmental Scanning Electron Microscope (ESEM), Pad-V X-Ray diffractometer, pin/disk wear testing apparatus, transmission electron microscope (TEM) sample preparation techniques and other characterization tools. The post-doctoral researcher was trained in the use of our dual beam Focused Ion Beam (FIB) instrument, which enables the preparation of site-specific TEM samples.

Computer Simulations:

The theoretical aspects of the high-rate, large-compression behavior of the effective frictional force between two ductile metals have been pursued by Hammerberg. The theoretical work has focused on simulations (both large-scale NonEquilibrium Molecular Dynamics (NEMD) and continuum finite difference) and development of micro-, meso- and macro-scale models incorporating dynamically induced microstructure. The current Los Alamos experimental effort has concentrated on two ductile metal / ductile metal interfaces, Cu/Cu and Ta/Al. The Cu/Cu system is being modeled with NEMD using EAM potentials. The simple steady wave analysis for the effect of pressure for varying impact velocities in the rotating barrel gas gun experiments has been applied to the preliminary copper data. The preliminary analysis is consistent with an increase in the frictional force with velocities at low velocities. The Ta/Al system has been studied using NEMD and the results are the subject of a recent paper presented at the APS Conference of the Topical Group on Shock Compression of Condensed Matter [1]. These results have shown a power-law decrease in the frictional force at high velocities and an increase at low velocities with very localized shear

deformation in the Al at the interface. The current experiments in this program will access the lower velocity, lower pressure regime (velocities < 50 m/sec) and will potentially verify this predicted behavior. For these particular materials (Ta/Al) another set of high energy density (ATLAS and Proton Radiography) experiments will access the higher velocity regimes ($v > 100$ m/sec).

Three-dimensional atomistic Non-Equilibrium Molecular Dynamics (NEMD) simulations with system sizes in the range of 10^6 atoms have been carried out for Ta/Al and Cu/Ag interfaces. The Ta/Al simulations have been the most extensive for both Ta(100)/Al(100) and Ta(110)/Al(111) interfaces. Both of these interfaces are incommensurate and the remarkable result for the velocity dependence of the frictional force was that both single crystal orientations showed an increase in the frictional force at velocities below 400 m/s and a power law decrease ($F_{\text{tangential}} \sim v^{-1/4}$) above this critical velocity with a common exponent of $-1/4$. Furthermore, the large deformation was confined to the weaker Al. Some of the details of the simulations are reported in [2]. Another set of simulations was carried out for the Cu/Ag tribo-pair where the differences in mechanical properties between the two metals are not so marked. Here Ag is the weaker material and both metals are fcc. A report on the initial Cu/Ag and Ta/Al simulations may be found in [3]. Cu/Cu interfaces are also being modeled using NEMD simulations and finite-difference continuum methods.

The connections among structural instability, large strains, heating and energy transport are being pursued for meso-scale based models of deformation. Some interesting aspects of the local processes leading to an increase in the frictional force at intermediate velocities have been treated in [4]. The experimental data anticipated in the near future for the mesoscopic changes in structure near the frictional interface will provide essential information for the theoretical modeling.

MD modeling at OSU has focused on simple amorphous materials [5,6]. This allows investigation of generic flow behavior and allows determination of similarities and differences when using different materials and structures. The results suggest that the flow of material close to the sliding interface is characterized by the formation of eddies, intimate mixing and gradual growth of the mixed layer. When the sliding speed is sufficiently high, the strain rate allows vorticity to develop. A comparison of eddy sizes with nanocrystal sizes in actual tribomaterial suggests that vorticity is directly responsible for the formation of such nanocrystal material. It is suggested that the flow of material near the interface is similar to turbulent flow in fluids. The resulting eddies affect frictional energy dissipation and mechanical mixing. A mixing-layer model of turbulent flow is qualitatively consistent with velocity profiles and friction behavior revealed by MD simulations. A continuum model is not sufficient for treating the vorticity at high sliding speeds because the length and time scales of the eddies are of the same order as the atomic length and time scales. The rate of energy dissipation cannot exceed that allowed by the smallest eddies, which are at the nanoscale.

References:

[1] P. J. Crawford, K. N. Rainey, P. M. Rightley, and J. E. Hammerberg, 2003, "A Novel Experimental Technique for the Study of High-Speed Friction Under Elastic Loading Conditions," presented at the 13th American Physical Society Topical Conference on Shock Compression of Condensed Matter in Portland, OR, July 20-25.

[2] J.E.Hammerberg, R.Ravelo, T.C.Germann, J.D.Kress and B.L.Holian, Sliding Friction at Ta/Al Interfaces, Proc. Conf. of the Topical Group on Shock Compression of Condensed Matter -2003, Portland, OR, July 21-25, 2003, to be published.

[3] J.E.Hammerberg, B.L.Holian, T.C.Germann, and R.Ravelo, NEMD Simulations of Metallic Friction at Ta/Al and Cu/Ag Interfaces, Metallurgical and Materials Transactions, 2004, to be published.

- [4] B.L.Holian and J.E.Hammerberg, Onset of Incommensurate Interfacial Instability in a Minimal Model of Dry Friction, Phys. Rev E, 68, 036101 (2003).
- [5] Invited as part of Viewpoint Set on driven systems: D. A. Rigney, Examples of Structural Evolution during Sliding and Shear of Ductile Materials, Scripta Mater. 49(10)(2003)977-983.
- [6] Invited plenary lecture at meeting of Wear Group of IRG-OECD, October 16-17, 2003: D. A. Rigney, Where the Action Is. This focused on changes that are found near sliding interfaces.

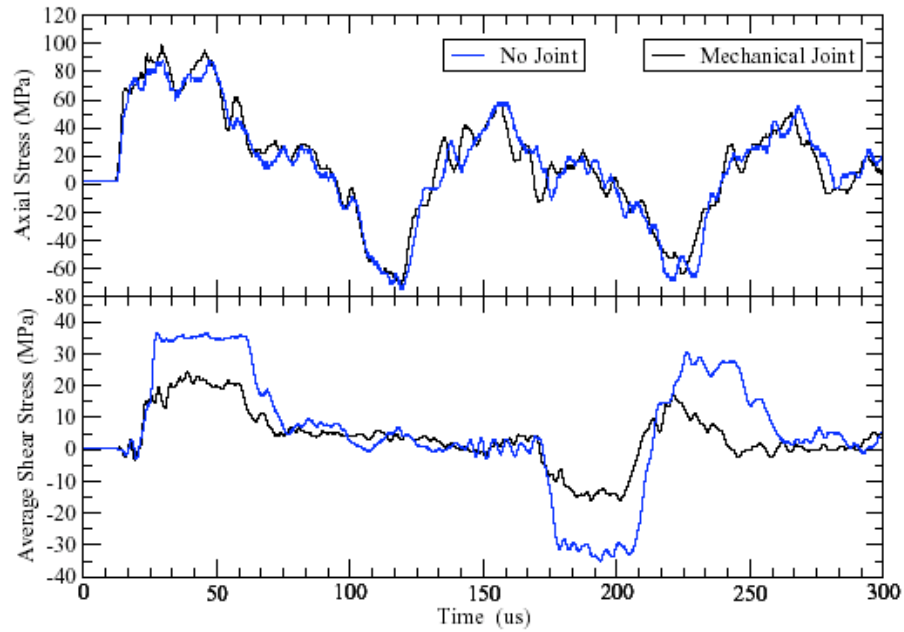


Figure 1. Comparison of mechanical joint and no joint for RBGG tests conducted on OFHC copper at 20psi and 3000 rpm.

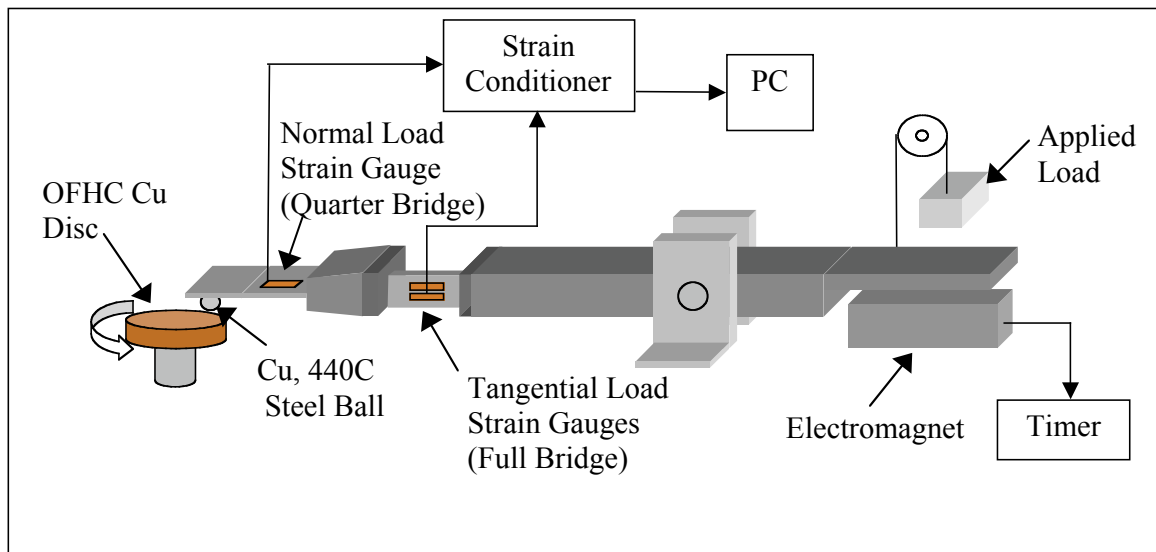


Figure 2. Schematic of the new friction testing system.

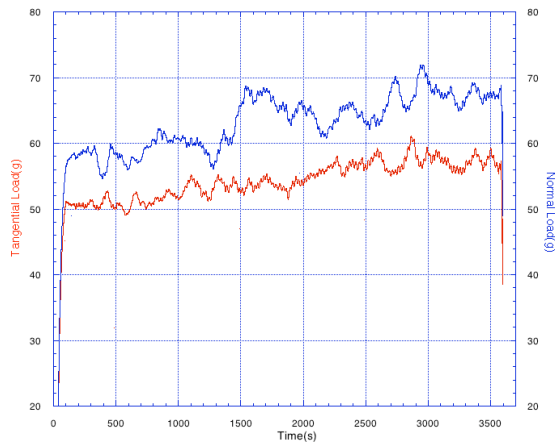


Figure 3. The normal and tangential loads for a friction test conducted on OFHC copper (sliding against a 440C steel pin). Sliding velocity was 5 cm/s

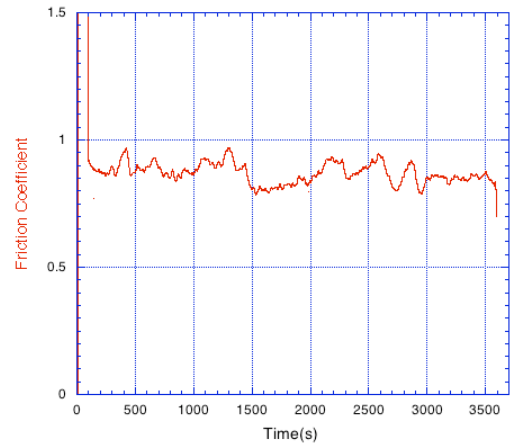


Figure 4. The evolution of the friction coefficient with time for a friction test involving a 440C steel pin on OFHC copper disc, sliding at 5cm/s

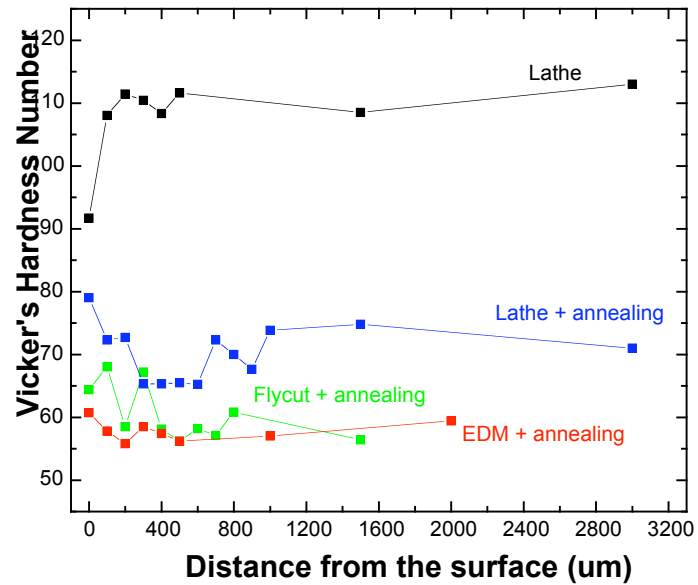


Figure 5. Microhardness test results indicate that the annealing helps in the recovery of subsurface deformation.

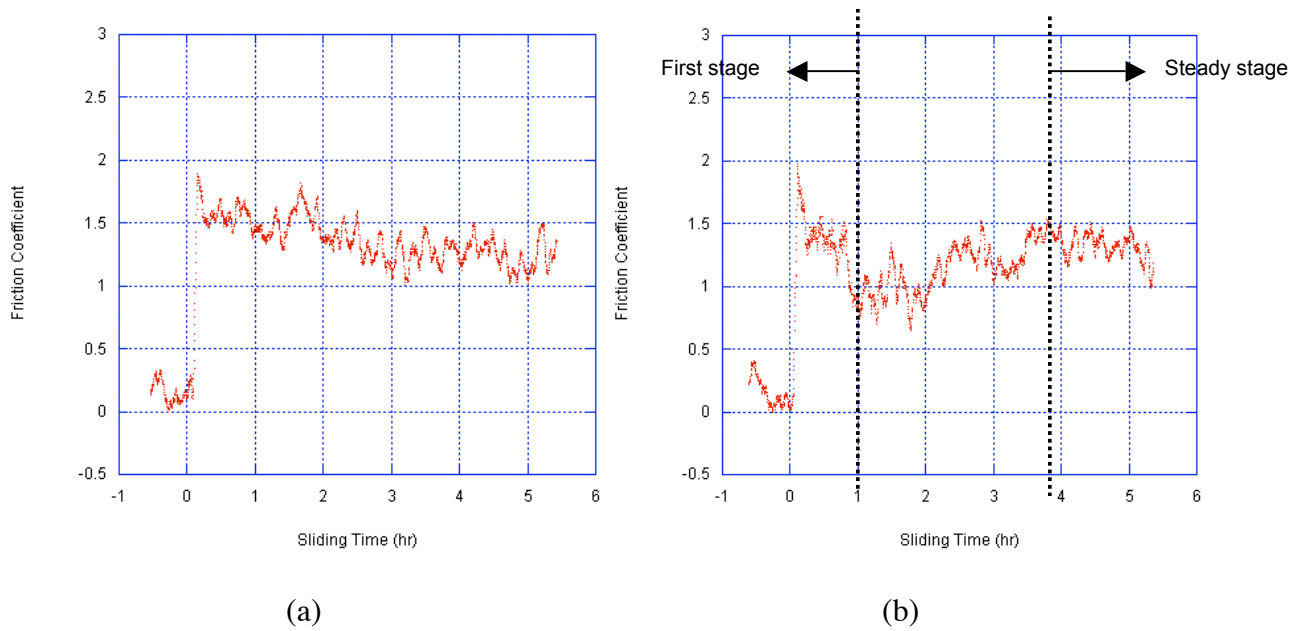


Figure 6. Friction coefficient data for two sliding velocities. (a) 1.34 cm/sec (b) 7.04 cm/sec

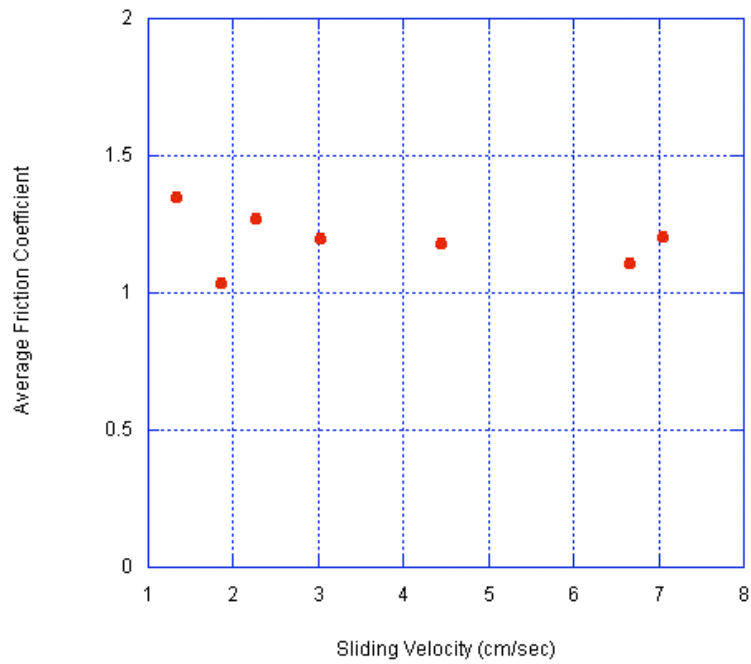


Figure 7. Average friction coefficient as a function of sliding velocity

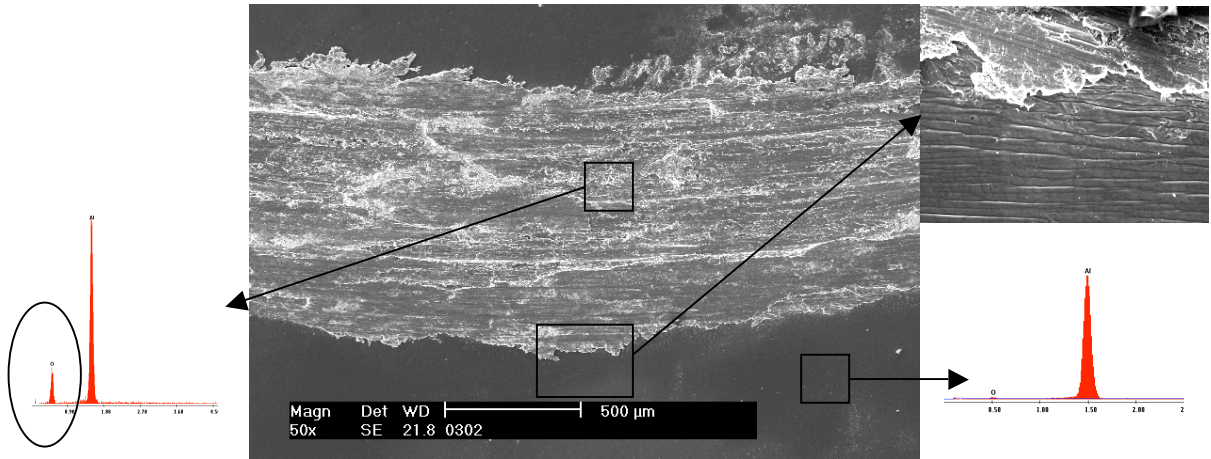


Figure 8. SEM and EDS analysis of wear track. EDS reveals an additional oxygen peak along the wear track suggesting the breakage of the surface oxide layer and its mechanical mixing with the aluminum matrix.

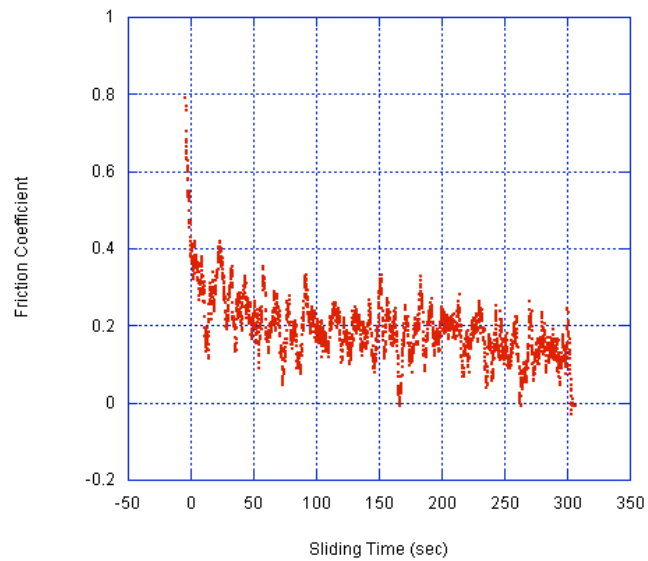


Figure 9. The evolution of the friction coefficient with time for a friction test involving a 440C steel pin on aluminum disc, sliding at 12.5cm/s. This test was conducted on the new friction testing setup.