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The effects of adhesive strength and load on material transfer in nanoscale wear

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ABSTRACT

Molecular dynamics simulation is used to study the effects of adhesive strength and load on material transfer during wear of nanoscale sliding contacts. Material transfer is quantified in terms of the number of atoms transferred from a copper substrate to a silicon dioxide tip where the interaction strength between the two materials is varied to modulate the work of adhesion. Material transfer is quantified before and after sliding, providing a means of isolating adhesive and abrasive wear mechanisms. Results reveal that both adhesion and abrasion contribute to material transfer during sliding, but that their relative contributions depend on the applied load and adhesive strength.

1. Introduction

An atomic force microscope (AFM) tip is often used to measure, deposit, modify, or manipulate material at the nanoscale. During these processes, contact between the tip and the sample can result in wear of the sample, tip or both. Thus, a fundamental understanding of the nanoscale material evolution of contacting surfaces is critical for reliable and precise measurements at the nanoscale as well as tip-based nanomanufacturing methods. The two primary mechanisms through which nanoscale wear occurs are abrasion and adhesion [1–8]. The key features of abrasive wear are cutting or plowing of the softer surface by the harder counterface. Typical observations of abrasive wear at the nanoscale are broken bonds and displaced material. Adhesive wear evolves through the formation of adhesive junctions, their growth, and fracture. The transfer of material, specifically atoms transferred due to bonds breaking and reforming at the nanoscale, between the contacting solid surfaces, is regarded as the characteristic feature of this mechanism of wear [9]. However,material transfer has been used as a means of quantifying wear in general [10–18]. It is likely that both adhesion and abrasion contribute to material transfer, with one or the other mechanism being dominant, depending on condition such as load [18].

Previous AFM-based studies have shown that load and adhesive strength significantly affect nanoscale wear [6,19–26]. However, it is difficult to quantify this relationship since adhesive strength cannot be easily controlled during the wear process. Further, AFM cannot be used to investigate wear in terms of material transfer. To address the latter issue, atom probe tomography (APT) has been used to characterize the composition of the AFM tip before and after sliding [18,27,28]. APT, however, does not provide a means of characterizing adhesive strength. Thus, previous experimental efforts have not directly shown how the adhesive strength of an interface affects material transfer.

Experimental studies of nanoscale wear have been complemented by Molecular Dynamics (MD) simulations of the near-contact region of an AFM tip [29–34]. There are fewer MD simulations specifically focused on the role of adhesive strength on nanoscale wear. Simulations of tip loading and unloading showed that large adhesive strength led to substantial plastic deformation on separation and some material transfer, while small adhesive strength resulted in separation of the materials without material transfer or plastic deformation [35,36]. Simulations of sliding predicted that wear increased with adhesive strength. In one case, a simulation of sliding on graphene showed that the rate of graphene bonds broken and the shear experienced by the tip increased as the adhesive strength between graphene and substrate was increased [3]. Also, simulations of two contacting, sliding asperities revealed that increasing adhesive strength resulted in more bonding between the asperities along with more heat generation and an increase of temperature in the contact area [37].

Material transfer wear has also been studied theoretically [10,12,13,16]. It was found that the transfer tendency can be predicted by comparing the cohesive strengths of the sliding components [10]. Also a lump growth model was developed based on

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transfer mechanisms observed at the macroscale and it was predicted that lump growth can be decreased by higher surface hardness of the workpiece, lower roughness of the tool surface, lower nominal contact pressure, or lower shear strength of the interface [12,13,16].

Here, building on previous research focused on understanding the mechanisms underlying wear at the nanoscale, we quantitatively characterize the effect of adhesive strength on material transfer, and the dependence of that effect on load and sliding. We use MD simulations of a model SiO_2 AFM tip sliding on a Cu substrate with artificially modulated adhesive interactions between the two. Predicted work of adhesion is correlated to material transfer before and after sliding at normal loads between 100 and 1000 nN. The results indicate that there is a work of adhesion- and load-dependent threshold below which no material transfer will occur. Above that threshold, the severity of the material transfer wear increases with adhesive strength and load. In addition, the relative contributions of adhesion and abrasion to observed wear are analyzed in terms of the ratio of the number of atoms transferred at 0 nm to that at 30 nm. The findings shed light on the complex processes by which adhesive and abrasive mechanisms determine the load-dependent wear of nanoscale sliding contacts.

2. Methods

The model consists of a SiO_2 tip, the α-quartz crystal structure of which is created in Materials Studio, and a face-centered-cubic Cu (100) surface as shown in Fig. 1. The substrate lattice orientations along the x, y and z directions are [100], [010] and [001], respectively. The tip radius is 30 nm and the height of the spherical cap is 2 nm. The dimensions of the Cu substrate are $60 \times 40 \times 2.2$ nm in the x-, y- and z-directions, respectively. The total number of atoms in the model is 475,415. We find that, at the largest work of adhesion and applied load, the maximum penetration depth of the tip is much less than the thickness of the substrate and the average displacement of the substrate atoms near the fixed bottom layer is smaller than one tenth of the substrate lattice constant. Both indicate that the substrate thickness is sufficient in our simulation. Periodic boundary conditions are applied in the x- and y- directions. The atoms in the top three layers of the tip and the bottom two layers of the substrate are fixed. The inter-atomic interactions within the tip are described by the Tersoff potential [38] with previously reported parameters for SiO_2 [39]. The Cu–Cu interaction is modeled using the Embedded Atom Method [40] with parameters reported in [41].

The Lennard-Jones (LJ) potential is used to model the interactions between tip and substrate atoms:

$$E_{ij} = 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right] \quad (1)$$

where ε is the LJ interaction strength parameter with units of energy, σ is the zero-crossing distance with units of length, and r_{ij} is the distance between a given substrate atom i and a given tip atom j. The LJ parameters for Cu–Si ($\varepsilon_{Cu-Si} = 0.942$ eV and $\sigma_{Cu-Si} = 0.2217$ nm) are the same as those reported in recent work [42]. The LJ parameters for Cu–O ($\varepsilon_{Cu-O} = 0.043$ eV and $\sigma_{Cu-O} = 0.2644$ nm) are obtained via the combination rule [42] using previously-reported LJ parameters for Cu–Cu ($\varepsilon_{Cu-Cu} = 0.415$ eV and $\sigma_{Cu-Cu} = 0.2277$ nm) [43] and O–O ($\varepsilon_{O-O} = 0.0045$ eV and $\sigma_{O-O} = 0.3010$ nm) [44]. To simulate different adhesive strengths, the magnitudes of ε_{Cu-Si} and ε_{Cu-O} are artificially changed, while the parameters σ_{Cu-Si} and σ_{Cu-O} are held constant as 0.2217 nm and 0.2644 nm, respectively; the tip-substrate interaction parameters studied are summarized in Table 1. The ratio of ε_{Cu-Si} to ε_{Cu-O} is approximately constant across the four groups of LJ potential parameters. Changing the interaction strength parameter of the LJ potential has been successfully employed in the previous MD simulations to modulate the adhesive strength between two surfaces [3,37,45]. The adhesive strength between the tip and substrate could be affected by the tip termination, size or shape. However, to exclude these effects, the same tip is employed in all simulations and adhesive strength is changed using LJ parameters only. All simulations are performed using LAMMPS simulation software [46,47] with a time step of 1 fs.

To assess the adhesive strength of the interfaces with different LJ interaction strengths, we calculate the ideal work of adhesion W as:

$$W = \frac{E_1 + E_2 - E_{12}}{A_c} \quad (2)$$

where A_c is the contact area, E_{12} is the total energy of the interface (consisting of the tip and substrate) at equilibrium, and E_1 and E_2 are the total energies of the tip and the substrate at equilibrium, respectively [48–50]. E_1 and E_2 are calculated from energy minimization after the tip and substrate are relaxed separately. To obtain E_{12}, the tip is placed 0.3 nm above the substrate surface and then the system is relaxed without applied load for 0.05 ns to allow the system to reach a stable potential energy. Then the system energy is minimized to calculate E_{12}. Note that the distance between the tip and substrate is allowed to change freely during the relaxation process, so the initial distance does not affect the calculated energy. The area A_c is calculated from the positions of contacting atoms, where tip contact atoms are identified as those within 0.3 nm of a substrate atom; 0.3 nm is chosen here because it is slightly larger than the equilibrium distance between tip atom and substrate atom. Two different cross sections are used to estimate the diameter of the circle formed by the contacting atoms from which we can calculate contact area. These calculations are performed with the tip-substrate geometry since that is the configuration used in the sliding wear simulations. However, we find that calculations performed with a slab-on-slab geometry yield qualitatively and quantitatively comparable results.

After determining the work of adhesion for each case, MD simulations are conducted with each model to investigate the wear behavior at various loads and adhesive strengths. A constant normal load (100, 400, 700 or 1000 nN) is maintained on the rigid top layers of the tip and the system is allowed to relax for 0.01 ns. This method of applying load directly to the atoms in the rigid top layers of the tip makes it possible to isolate the effect of load from that of adhesive strength. Then the tip slides along the x-direction at a constant speed of 10 m/s through a distance of 30 nm. This speed, although fast compared to an AFM experiment, is necessitated by the timescale limitation of MD simulation, particularly for the relatively large number of atoms in this model. The sliding distance is determined based on the observation that

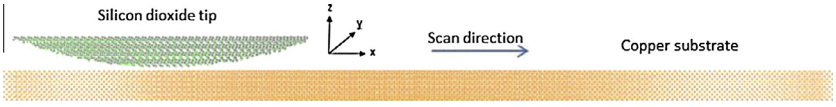

Fig. 1. Snapshot of the initial configuration of the simulation. Copper atoms are shown in orange, silicon atoms in green and oxygen atoms in gray. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)