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Frictional performance and near-surface evolution of nanocrystalline Ni–Fe as governed by contact stress and sliding velocity

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ABSTRACT

While early reports on the wear performance of nanocrystalline alloys have suggested enhanced behavior consistent with their higher hardness compared to conventional microcrystalline alloys, there is still limited understanding of the mechanisms and limits of this enhanced behavior. In the present study, we examine the frictional response of a nanocrystalline Ni–20Fe alloy with 34-nm average grain size compared to the same film annealed to an average grain size of 500-nm. We examine the sliding friction performance of these films in contact with a 3.125 mm diameter Si_3N_4 spherical counterface under a range of normal forces (0.1–1.0 N) and sliding speeds (0.25–3.75 mm/s) in a non-oxidizing dry nitrogen environment. Under all conditions, the initial break-in coefficient of friction (COF) starts high, $\mu \approx 0.5$–0.8, typical of uncoated metallic friction. However, there is an evolution in the COF which depends on normal force and sliding speed. At low sliding speeds (or normal forces), the steady-state COF decreases to $\mu \approx 0.2$ whereas at higher sliding speeds and normal forces, the steady-state COF remains high at $\mu \approx 0.8$. Focused ion beam cross-sectioning and TEM imaging reveal that in all cases, a multilayer substructure is formed in the deforming film: a refined ultrananocrystalline layer at the top surface, over a region of coarsened grains, atop the parent nanocrystalline alloy. The key distinction between the high-friction and low-friction conditions appears to lie in the triggering of a delamination process: high-friction conditions are associated with a thickening of the UNC layer through repeated delamination, whereas low-friction conditions are associated with a thin UNC layer that does not delaminate. Finite element analysis is used to aid in the understanding of how the magnitude and location of stresses drive these two distinct regimes.

1. Introduction

Sliding contact on the surfaces of metallic materials often introduces plastic deformation underneath the wear surface that results in subsurface layers with microstructures that are significantly different from those of the bulk. The work of Kuhlman-Wilsdorf and co-workers [1–4] highlighted the similarities between the structures generated by rolling or fatigue, and those observed during frictional contact. Among others, Rigney and Hirth [5,6] extended the dislocation concept to friction and wear processes in metallic materials and postulated the relationships between the friction force and plastic flow. Suh and co-workers [7–10] advanced the theory of delamination wear, which again relies on plastic deformation of a surface layer, resulting in the creation of a soft subsurface layer and subsequent nucleation and growth of a plate-like debris. The formation of recrystallized structures and crystallographic texture has been confirmed by cross-sectional transmission electron microscopy (TEM) of wear surfaces. The advent of focused ion beam (FIB) microscopy has facilitated the preparation of TEM samples in site specific locations. By using a combination of FIB, TEM and electron backscatter diffraction (EBSD), Prasad and Michael [11], and Rainforth and co-workers [12–15], have characterized the friction-induced microstructural changes (e.g., recrystallization, formation of nanocrystalline structures, development of crystallographic textures, etc.) at specific locations underneath wear surfaces.

The dominant deformation mechanism in many FCC metals transitions as the grain size decreases: from collective dislocation structures such as tangles and pileups in coarse grained metals (grain size > 100's of nm) [16], to partial dislocation emission and absorption at grain boundaries in nanocrystalline metals (grain size < 100 nm) [17–20], to grain boundary mediated plasticity in very small NC grains (grain size < 10 nm) [21,22]. These transitions lead to emergent behavior such as Hall–Petch breakdown [23,24] and a shift to glass-like deformation as the grain size approaches the amorphous limit [25]. This cross-over from normal to inverse Hall–Petch effect occurs at a grain size of approximately 10 nm in most metals, where grain boundary

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processes dominate the plasticity. However, the grain structures of nanocrystalline metals can be unstable, often evolving due to the stresses generated during deformation. Grain coarsening can occur at room temperature under monotonic loading [26–31], fatigue loading [32–34]and sliding contact [35–37]. Sliding contact conditions may be particularly amenable to grain evolution, since shear stress is thought to be a significant driving force behind mechanical grain growth [29,38]. There is also evidence of wear-induced grain refinement in nanocrystalline metals [36,37,39] which must be distinguished from classical notions of wear-induced refinement due to cell formation [5,40–42], since the grain size is too small in nanocrystalline metals to support a classical dislocation cell structure.

The relatively high strength and hardness (according to Hall–Petch scaling) of nanocrystalline metals [24,31,43,44] makes them attractive candidate materials in a variety of applications including tribology. Confirmation of bulk strength measurement and improvements in processing methods facilitated friction and wear studies on these materials with the hope of finding enhanced wear resistance analogous to the strength improvements, since dislocation mechanisms are known to play a prominent role in both processes. Indeed, improved performance in this regard may be correlated with the increased hardness and refined grain size [45–49], although in general the effect of hardness on friction and wear is more complicated [42]. Rupert and Schuh have even shown that wear performance in nanocrystalline Ni-W exceeds that predicted by increased hardness alone [50].

Considering the multitude of factors which seem to complicate even a basic understanding of how nanocrystalline metals behave during sliding contact, it is no surprise that there has not yet emerged a strong consensus on this issue. Although they are related concepts, friction and wear are not always synonymous [51]. The characterization of wear involves both fracture and deformation. In an effort to simplify the interpretation, this paper focuses predominantly on the friction behavior of a nanocrystalline Ni–20Fe under repeated unidirectional sliding contact, thereby building on previous studies of similar Ni–Fe systems [32,52–57]. Many previous measurements on the sliding contact of nanocrystalline alloys have been performed in an air environment leading to wear-induced surface oxidation, thereby complicating the interpretation of results. To avoid this issue, we performed cyclic, unidirectional, linear friction measurements in a dry nitrogen atmosphere while varying normal load and sliding speed. Our major objective is to investigate the formation and stability of friction-induced grain structures, and how these would in turn influence the friction behavior. Focused ion beam (FIB) cross-sections and transmission electron microscopy (TEM) were used to characterize the subsurface evolution in the wear tracks. Finite element analysis was used to interpret the role of the stress state on tribological evolution.

2. Experimental materials and method

The nanocrystalline (NC) Ni–20Fe materialused in this study was electroplated on a silicon wafer substrate with a 1000 Å copper seed layer. Films intended for friction experiments were deposited to a thickness of 50 μm. The bath contained nickel sulfate, iron sulfate, potassium sulfate, sodium citrate, saccharin, sodium salt and dodecyl sodium sulfate. The resulting alloy composition was measured by electron microprobe analysis (Table 1). The pulsed plating conditions employed in the present study utilized 3 s intervals with a current density of 20 mA/cm^2, resulting in an average grain size of 34 nm with a (1 0 0) deposition texture. For comparison to a larger grain size, some of the Ni–Fe coupons were annealed in a 10^{-6} Torr vacuum at 450 °C for 1 h, resulting in a 500 nm average grain size and a weak (1 1 1) out of plane texture. X-ray diffraction (XRD) pole figures and transmission electron microscopy (TEM) bright field images of the as deposited and annealed grain structures are shown in Figs. 1 and 2. The mechanical behavior of this alloy was characterized using microtensile tests on "dogbone" shaped samples with a nominal thickness of 10 μm. The 500 nm average grain size corresponded to a yield strength of approximately 300 MPa, while the 34 nm average grain size corresponded to a yield strength of approximately 1600 MPa. It is worth noting that even the 34 nm NC film possessed a grain size larger than that associated with any Hall–Petch breakdown, which is typically reported to occur below grain sizes of 10–15 nm [23–25]. Standard deviations for the average grain sizes in the 34 and 500 nm materials were 12 and 202 nm, respectively.

Friction measurements were made via unidirectional sliding with a custom built linear tribometer. More complete descriptions of the tribometer can be found elsewhere [58,59]. The tribometer was enclosed in an environmental chamber with precise control of oxygen content and dew point. Measurements were made in dry nitrogen (<10 ppm O_2 and <100 ppm H_2O) to minimize tribo-oxidation. This inert environment is in contrast to most other

Table 1
Element composition of Ni–Fe material in wt % as determined by electron microprobe.

Na	Si	S	K	Ti	Fe	Ni
0.007	0.019	0.026	0.001	0.004	18.648	81.132

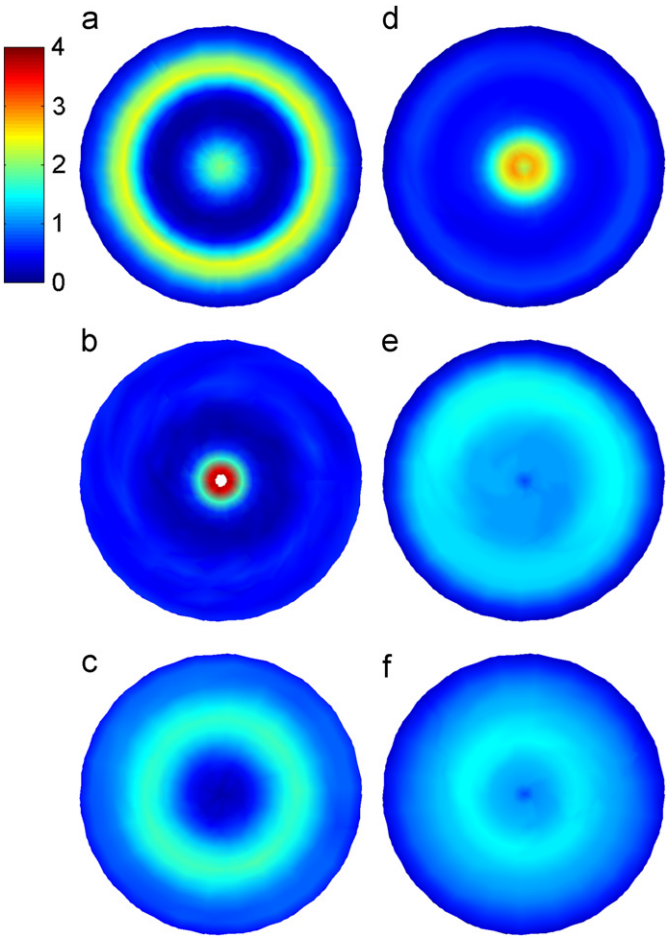

Fig. 1. XRD pole figures showing (1 0 0) texture in as-deposited Ni–20Fe (a)–(c) and a slight (1 1 1) texture in annealed Ni–20Fe (d)–(f). (a) (111) 34nm, (b) (200) 34nm, (c) (220) 34nm, (d) (200) 500nm, (e) (200) 500nm and (f) (220) 500nm.