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Supplementary Materials for

Lowering coefficient of friction in Cu alloys with stable gradient nanostructures

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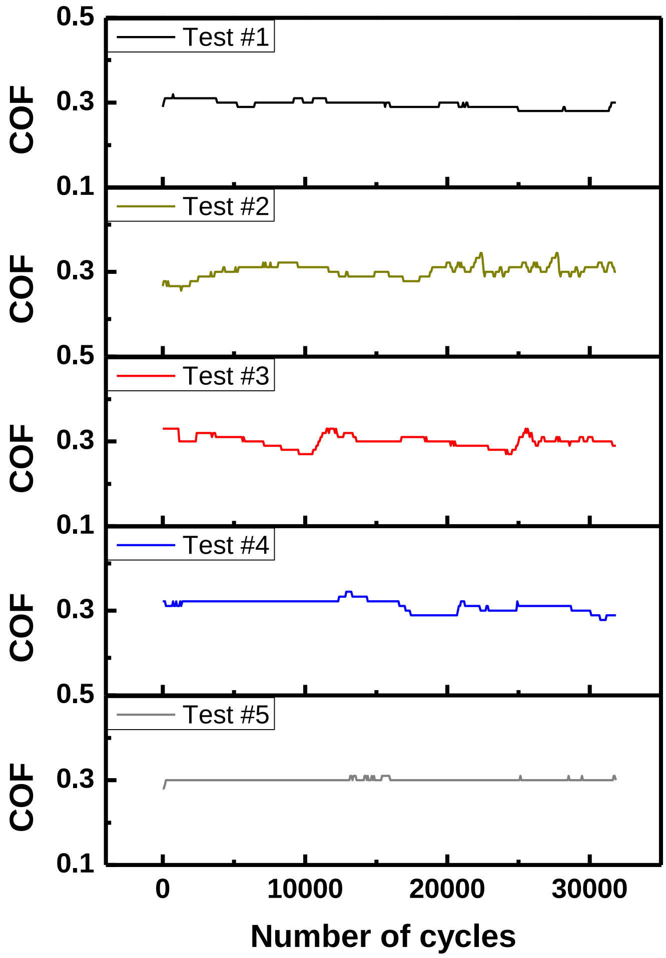

fig. S1. Measurement repeatability of COF. Measurement results of COFs with sliding cycles (more than 30000 cycles) in five tests on different GNG Cu samples using WC-Co balls under a load of 50 N, a slide stroke of 1 mm and a velocity of 10 mm/s.

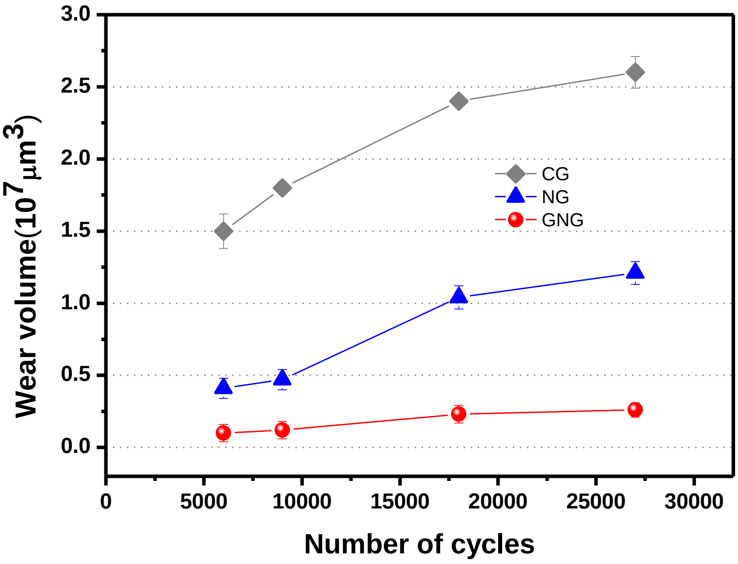

fig. S2. Measurement results of wear rates. Variation of wear volume with sliding cycles for the CG, NG, and GNG Cu-Ag samples sliding against WC-Co balls under a load of 50 N, a slide stroke of 1 mm and a velocity of 10 mm/s.

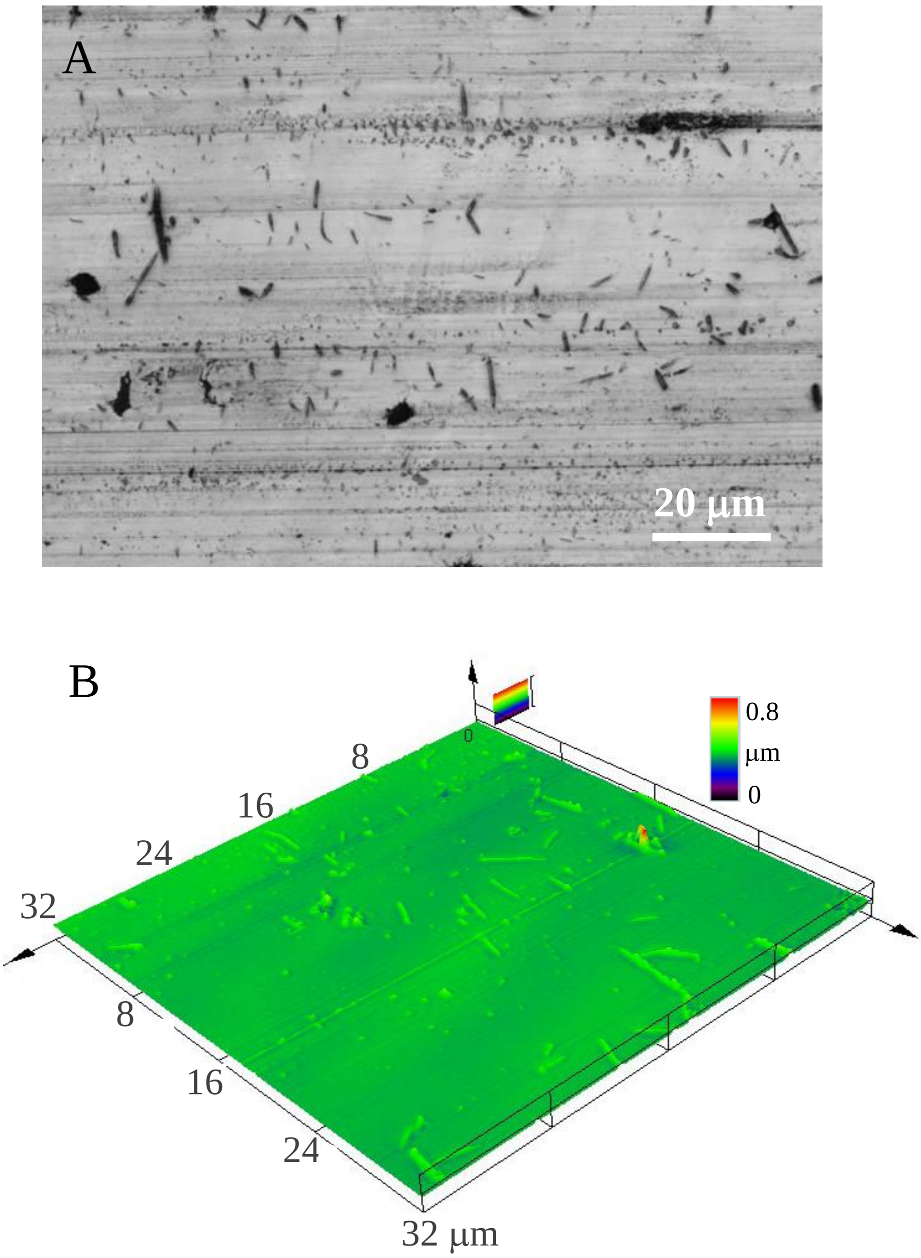

fig. S3. Surface profiles and morphology of the CG sample under low-load single sliding. Confocal laser microscopy image (**A**) and 3-D surface profiles (**B**) for surface morphology of a CG Cu-Ag sample after a single sliding at a load of 30 N, a slide stroke of 1 mm and a velocity of 10 mm/s.

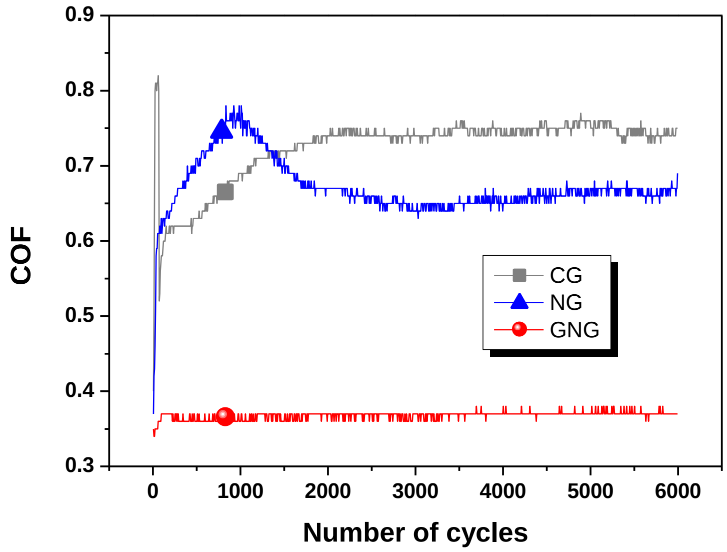

fig. S4. Effect of Ag addition on COF reduction—Measurement results in pure Cu samples. Variation of COFs with sliding cycles for the CG, NG, and the GNG pure Cu samples (purity better than 99.99%) sliding against WC-Co balls under a load of 30 N, a slide stroke of 1 mm and a velocity of 10 mm/s. The GNG and NG samples were prepared by using the same techniques as in the Cu-Ag samples. Analogous to the Cu-Ag samples, obvious COF reduction is obtained in the GNG Cu samples.

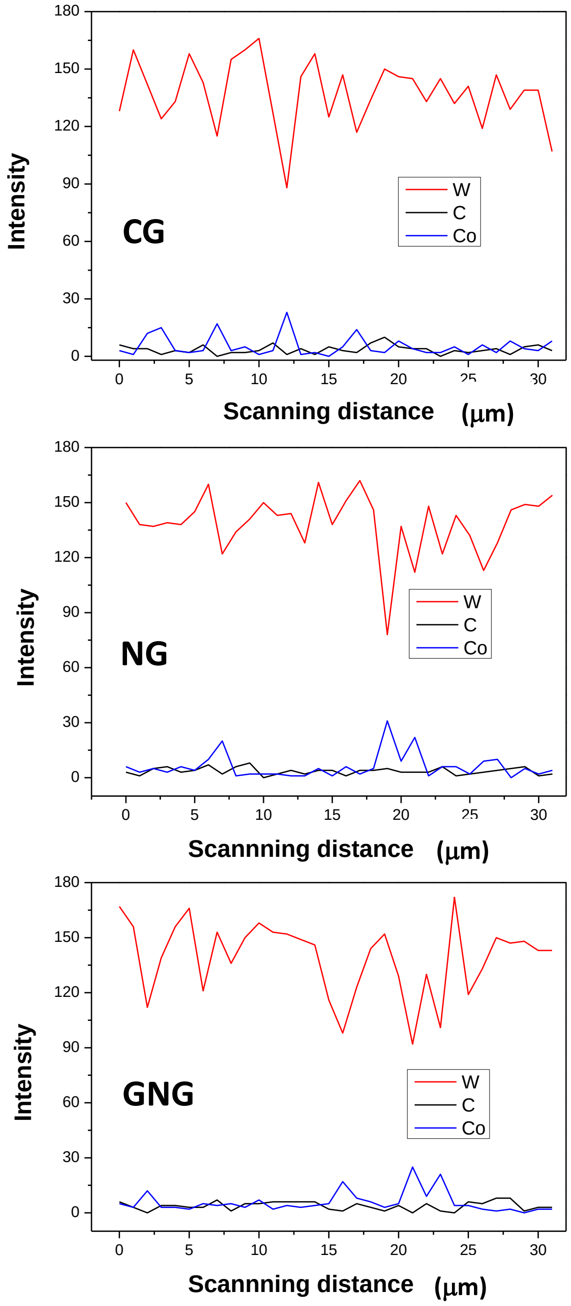

fig. S5. Counter surface analysis. Chemical composition analysis of the WC/Co ball counter surface after sliding for 18000 cycles on the CG, NG, and GNG samples under a load of 50 N, a slide stroke of 1 mm and a velocity of 10 mm/s. No obvious difference in the compositions of W, C, and Co is seen among the three samples. Elements of Cu and O have not been detected on the counter surfaces for the three tests, implying no transfer occurred (from and to the ball) during sliding.

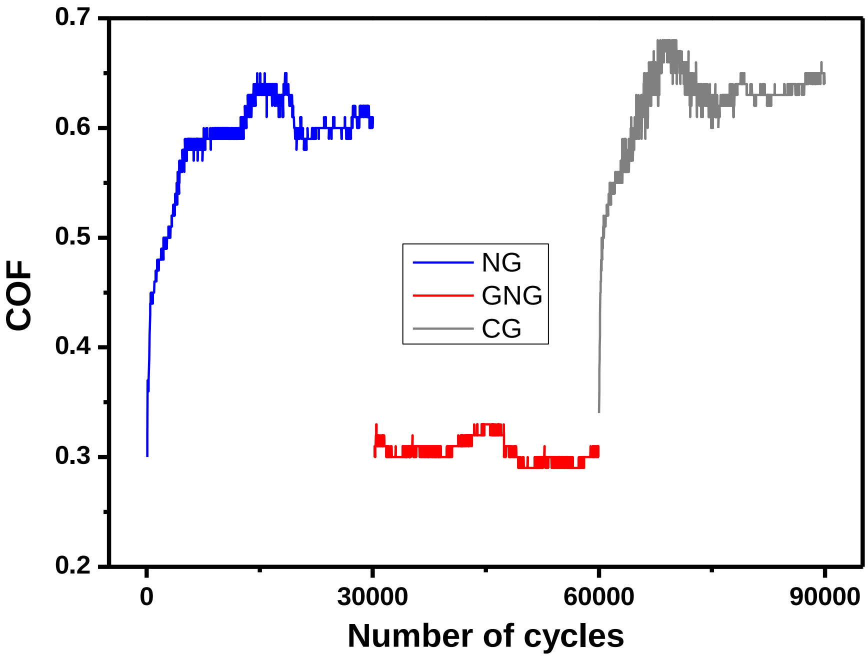

fig. S6. COF measurement on the NG, GNG, and CG samples subsequently using exactly the same contact surface of a WC-Co ball. Each sample was measured for 30000 cycles under a load of 50 N, a slide stroke of 1 mm and a velocity of 10 mm/s. Obviously, the effect of counter surface is negligible on the observed COF reduction in the GNG sample.

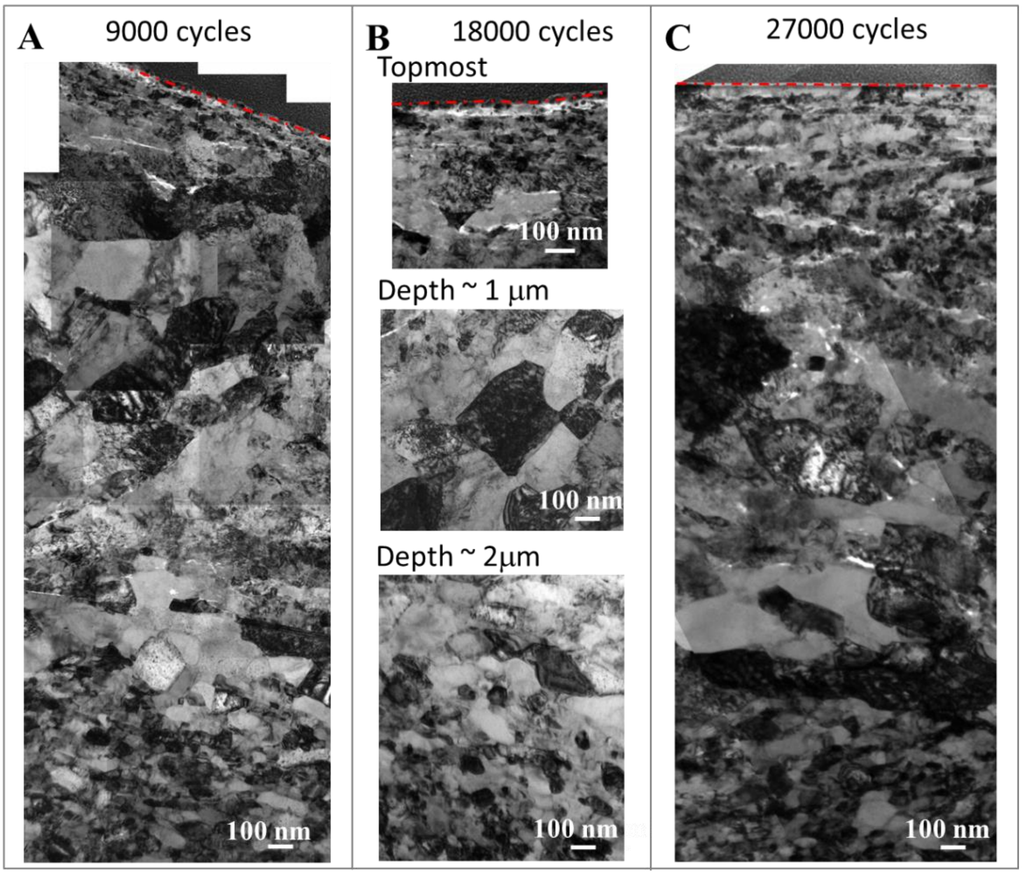

fig. S7. Stability of the subsurface microstructure in the GNG samples against sliding. (**A-C**) Typical TEM images of the subsurface microstructure in the GNG sample after sliding for 9000, 18000 and 27000 cycles at a load of 50 N, respectively. The worn surface is outlined by dash-dotted lines. It is clear that the subsurface microstructures remain basically unchanged against sliding in the steady-state stage.

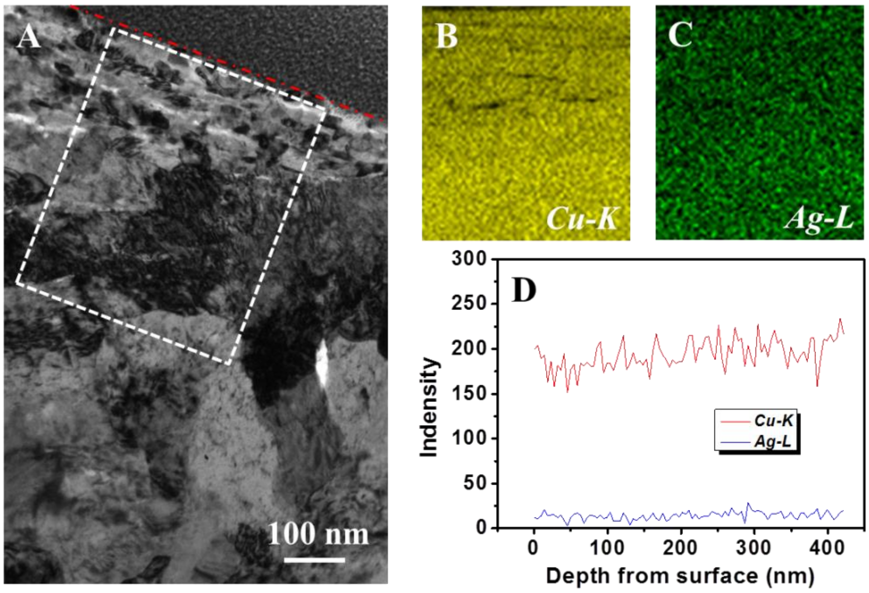

fig. S8. Chemical analysis of the topmost NG surface layer. (**A**) A cross-sectional TEM image below the sliding surface of the GNG sample after 9000 cycles. (**B** to **D**) Corresponding elemental maps analysis (B: Cu, C: Ag) and EDS line scanning along the depth (D) in the topmost region (as indicated in A) below the worn surface. The worn surface is outlined by dash-dotted lines. No detectable variation in Cu and Ag is observed across the topmost surface layer and the grain-coarsened structures. It means the sliding-induced contamination in the topmost surface layer with nano-sized grains is negligible.

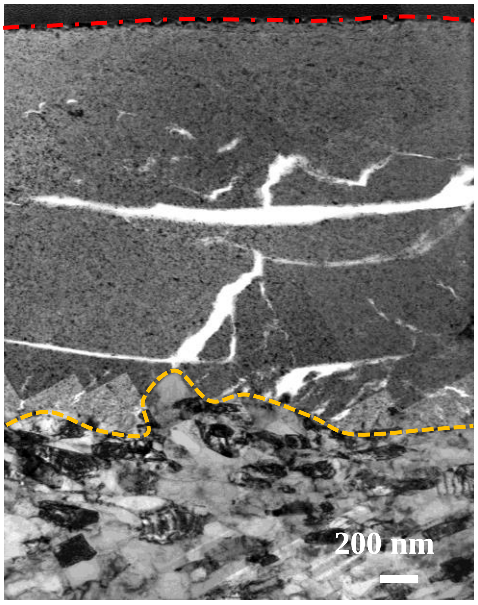

fig. S9. Subsurface microstructures in the CG Cu under sliding in the steady state. A cross-sectional TEM image below the sliding surface of the CG sample after 18000 cycles at load of 50 N, a slide stroke of 1 mm and a velocity of 10 mm/s. The sliding surfaces are outlined by dash-dotted lines and the tribolayer/recrystallization interfaces by dashed lines. Cracks (white lines) are seen in the top tribolayer.

table S1. Surface roughness change after dry sliding for 18,000 cycles. Measured surface roughness (Ra and Rz, in μm) for different Cu-Ag samples before sliding and after sliding for 18000 cycles.

	CG	NG	GNG
Original			
Ra (μm)	0.034±0.012	0.021±0.011	0.025±0.013
Rz (μm)	0.178±0.058	0.125±0.041	0.145±0.051
18000 cycles			
Ra (μm)	0.19±0.017	0.21±0.018	0.022 ±0.011
Rz (μm)	1.70±0.41	1.20±0.31	0.15±0.03