The Interfacial Gradient and its Role in Ultralow Wear Sliding

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Wetting angles of three different probe liquids on the running films and transfer films are plotted against sliding cycles used to develop the tribofilms in Fig. S1. In general, wetting angle increased on the transfer film, decreased on the running film and the two converged after ~200k cycles. This wetting angle convergence is not surprising given that a high coverage polymer transfer film started to manifest after 200k cycles as shown in Fig. 2c. However, even after 200k cycles, wetting angle discrepancy persists between the running film and transfer film and the disparity is most for water (~22°) and least for formamide (~2°). When compared with the unworn bulk composite, wetting angles of water and formamide both decreased significantly while the wetting angle of glycerin remained largely unchanged (Fig.S1d).
Fig. S1. Wetting angles of the running film and transfer film against sliding cycles for (a) deionized water, (b) glycerin and (c) formamide as the probe liquid and (d) a summary of wetting angles on different surfaces. Early development of the wetting angles between 20 to 100k cycles is shown in linear-log scale in the insets. Error bars represent one standard deviation of measurement. Wetting angle of unworn bulk composite are shown in dashed lines with gray boxes representing one standard deviation.

Fig. S2 show representative wetting angle plots of the running film and transfer film at 20 and 500k sliding cycles for surface energy calculation using the prescribed method based on the Owen-Wendt theory. At 20 cycles, wetting angles on the transfer film were all smaller than on the running film. This is likely because the transfer film is mainly composed of large, patchy and discrete debris that only partially cover the counterface as seen in Fig. 2. At 500k cycles, wetting angles on the two tribofilms became comparable and the cosine of the wetting angle between water and tribofilms increase both in magnitude and uncertainty suggesting increasing contributions of surface energy from polar groups at the surfaces.
Fig. S2. Representative wetting angle plot of the (a) running film and (b) transfer film at 20 and 500k sliding cycles using the prescribed method. Solid line corresponds to the best fitted line using least square linear regression analysis. Uncertainties of the slope and intercept were calculated using the prescribed Monte-Carlo method.