Supporting Information: Surface–to–Surface Bridges Formed by Reversibly Assembled Polymers

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Experimental Details

Materials. Gold pellets (99.99%) were obtained form Kurt J. Lesker Co. (Clairton, PA) and chromium (99.995%) from Alfa Aesar (Ward Hill, MA). Absolute ethanol (Aaper Alcohol and Chemical Co., Shelbyville, KY) and 6-mercapto-1-hexanol, 97% (MH) (Aldrich Chemical Co., Milwaukee, WI) were used as received. Standard Si_3N_4 cantilevers (model AUHW) were obtained from Thermomicroscopes (Sunnyvale, CA). Deionized water was obtained from a Millipore filtering system.

Monolayer Substrate and Tip Preparation. Silicon wafers cut into 3 mm x 3 mm pieces were immersed in 70%:30% H_2SO_4/H_2O_2 (piranha solution) for 15 minutes, rinsed with water and ethanol, and dried with a stream of N_2. Au-coated substrates were prepared using an Edwards E306 high-vacuum evaporator (Wilmington, MA). Chromium layers (50 Å) were evaporated from a crucible followed by evaporation of gold (400-500 Å) at 4 x 10^{-6} Torr. Evaporation rates typically ranged from 1 to 3 Å/s. After evaporation of the gold, the substrates were immersed in solutions of 1 μM OM **1** for 4 hours under a humid atmosphere, rinsed with H_2O, and backfilled with 1 μM MH for 1 hour. After complete modification, tips and surfaces were rinsed with ethanol and deionized water before use. Substrates not modified directly after evaporation were stored in a Fluoroware container. Before being modified, the gold substrates were immersed in piranha solution for 24 hours, rinsed with ethanol and deionized water, and dried with N_2.

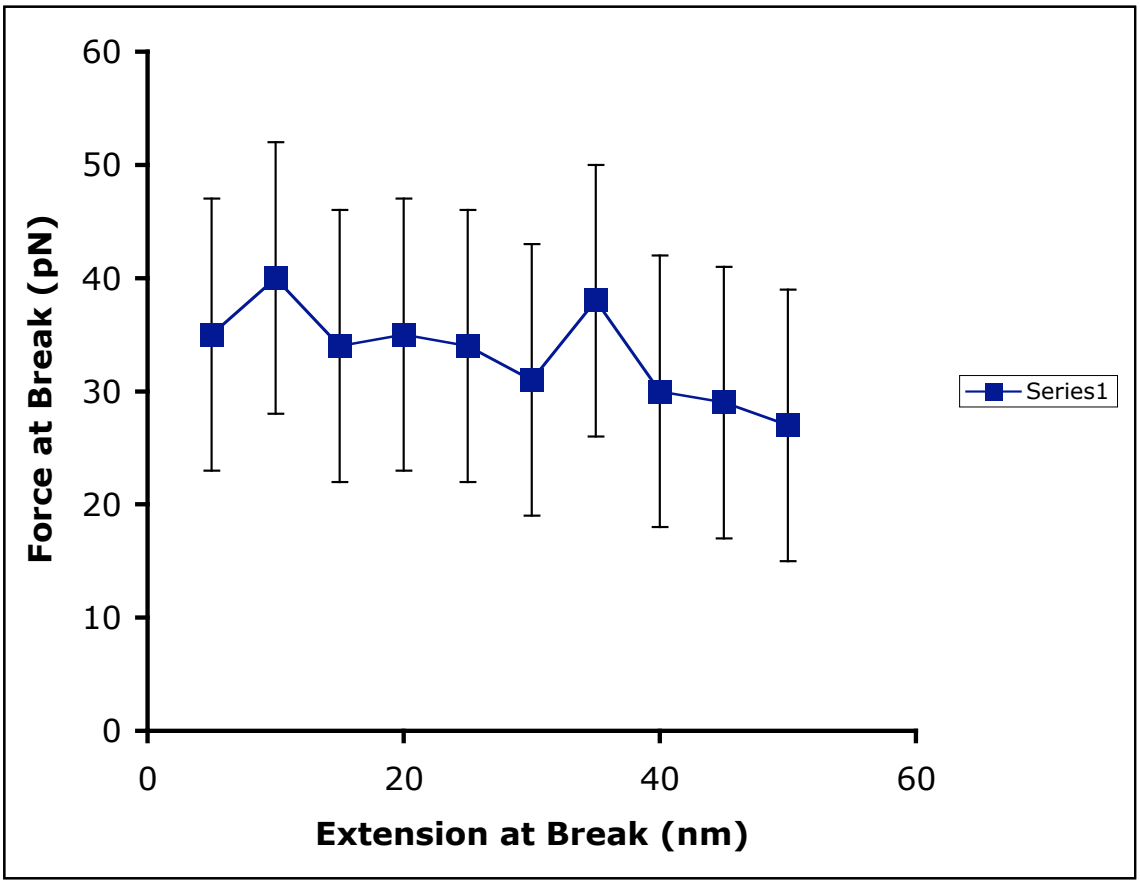
DNA Force Measurements. Pull-off forces were measured using a homemade AFM controller system incorporated with a Digital Instruments scanning head. Our AFM instrument and its mode of operation is similar to the one described in detail previously

(see Oberhauser, A.F., Marszalek, P.E., Erickson, H.P., and Fernandez, J.M. *Nature* **1998**, *393*, 181-185.) DNA-modified Au-coated Si_3N_4 cantilevers (rectangular-shaped, 200 μm x 20 μm, nominal tip radius ~ 40-50 nm, nominal spring constant k = 0.02 N/m, actual spring constant k = 0.017 N/m) were used. The spring constant of each AFM cantilever was calibrated in solution, using the thermal noise method, based on the energy equipartition theorem as described previously (see Florin, E.L., Rief, M., Lehmann, H., Ludwig, M., Dornmair, C., Moy, V.T., & Gaub, H.E. *Biosensors and Biolelectronics* **1995**, *10*, 895-901).

Measurements were carried out in a closed fluid cell with scanning set for a retract/approach cycle. Force curves were collected in LabView (National Instruments, Austin, TX) and analyzed using Igor Pro (Wavemetrics, Lake Oswego, OR). All data was filtered during acquisition at 500 Hz. After acquisition, the data was smoothed using Igor Pro, Box Algorithm, 15 points.

The responsiveness of the photodiode was calibrated against the piezo extension after the experiments by pressing the tip into a clean glass slide. The extension data represents the travel from rest of the actual AFM tip, corrected from the piezo displacement by the measured deflection of the tip. Thus, it represents the actual extension of the bridging assemblies. One consequence of the correction is that the noise in the force vs. extension graphs appears to be "tilted". The tilt is a direct consequence of the correction as follows. Thermal fluctuations downward reduce the tip-surface separation and appear as peaks that are "up" (apparent attractive force) and to the left (shorter separation). Fluctuations upward have the opposite effect. Hence, the noise "tilts" up and to the left in the corrected force curves. Without the correction there is no such tilt.

Rupture Force vs. Extension at Break (OM **2**, 5 mg/ml in 1 M NaCl phosphate buffer. Data points represent the average of measurements in a 5 nm range (e.g. 10–15 nm), and the error bars reflect an overall standard deviation of 12 pN taken from data sets of >10 measurements at short extensions.



Histogram of Extensions at Break (OM **2**, 5 mg/ml in 1 M NaCl phosphate buffer)