

Supporting Information

Effects of protein species and surface physicochemical features on the deposition of nanoparticles onto protein-coated planar surfaces

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Figures and Tables

Table S1. Positive secondary ion peaks used to determine the amino acid chemistry of the surface-immobilized protein layers. Numbers indicate the unit m/z ratio of peaks. Relevant peaks were chosen based on Wagner and Castner (2001).⁵

| Amino acid source | Characteristic Ion |
|--------------------------------|---|
| isoleucine (Ile)/leucine (Leu) | 86: C ₅ H ₁₂ N ⁺ |
| methionine (Met) | 61: C ₂ H ₅ S ⁺ |
| phenylalanine (Phe) | 120: C ₈ H ₁₀ N ⁺ , 131: C ₉ H ₇ O ⁺ |
| valine (Val) | 72: C ₄ H ₁₀ N ⁺ , 83: C ₅ H ₇ O ⁺ |
| arginine (Arg) | 43: CH ₃ N ₂ ⁺ , 73: C ₂ H ₇ N ₃ ⁺ , 100: C ₄ H ₁₀ N ₃ ⁺ |
| asparagine (Asn) | 70: C ₃ H ₄ NO ⁺ |
| glutamine (Gln) | 84: C ₄ H ₆ NO ⁺ |
| glutamic acid (Glu) | 102: C ₄ H ₈ NO ₂ ⁺ |
| histidine (His) | 81: C ₄ H ₅ N ₂ ⁺ , 82: C ₄ H ₆ N ₂ ⁺ , 105: C ₅ H ₃ N ₃ ⁺ |

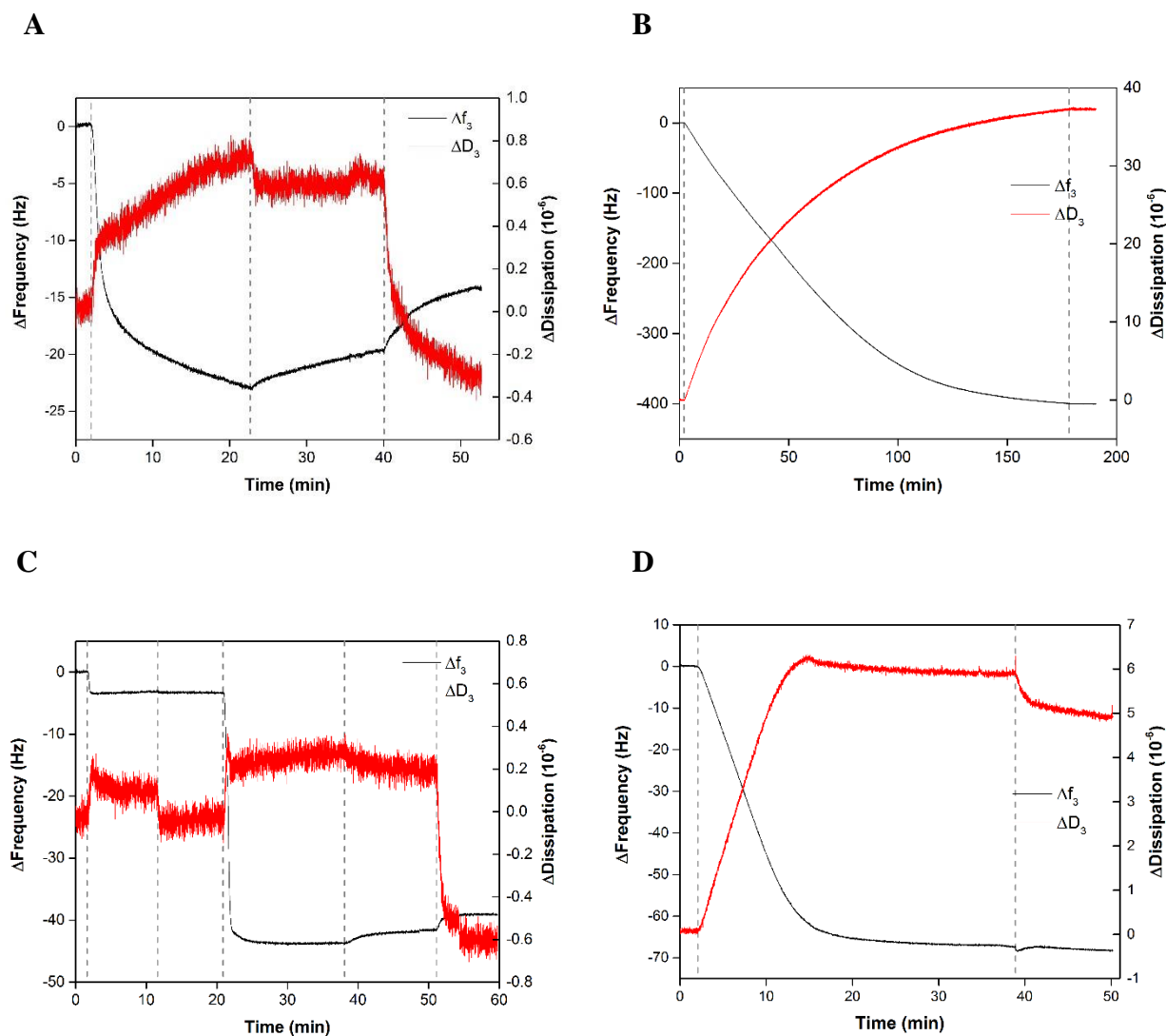


Figure S1. Representative third overtone QCM-D data for BSA adsorption followed by hematite NP deposition onto negatively-charged bare silica sensors (BSA adsorption shown in A and hematite NP deposition in B) and positively-charged PLL-precoated silica sensors (PLL and BSA adsorption shown in C and hematite NP deposition in D). The dotted gray lines indicate the time points at which the solutions being flowed through the QCM-D were changed according to the methods (A: 10 mM HEPES, BSA solution, 10 mM HEPES, 10 mM NaCl; B and D: 10 mM NaCl, hematite NP suspension, 10 mM NaCl; C: 10 mM HEPES, PLL solution, 10 mM HEPES, BSA solution, 10 mM HEPES, 10 mM NaCl).

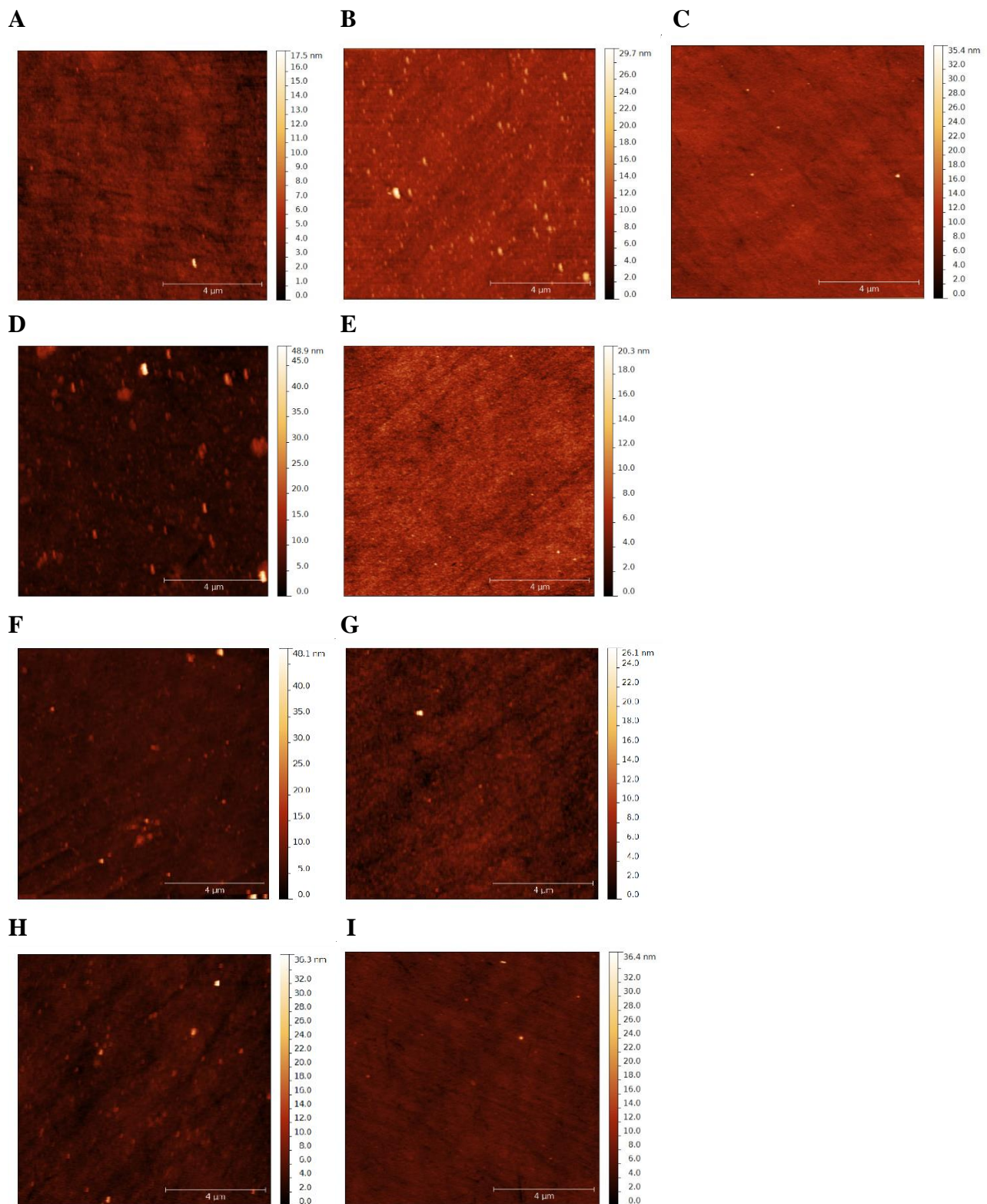


Figure S2. Representative AFM surface height images for BSA immobilized on a negatively-charged sensor surface (A) and positively-charged surface (B), lysozyme on a – surface (C), ubiquitin on a – surface (D) and + surface (E), *E. coli* protein extracts on a – surface (F) and + surface (G), and *P. fluorescens* protein extracts on a – surface (H) and + surface (I). The scale bars show 4 μ m lengths. AFM imaging was performed under dry ambient conditions.

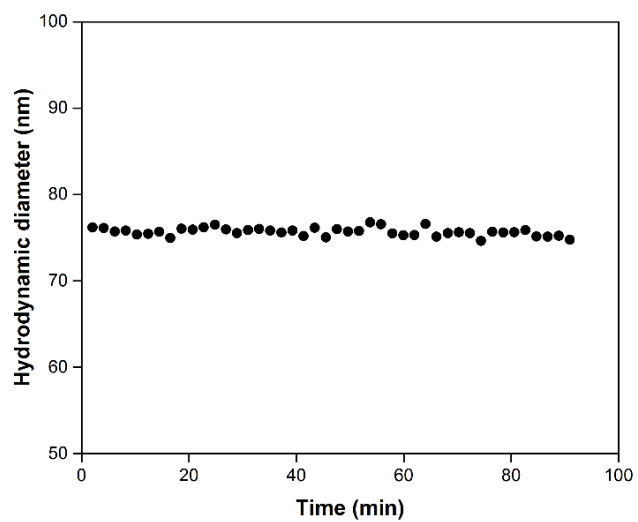


Figure S3. Representative hydrodynamic diameters of hematite NPs measured by DLS over the course of a QCM experiment. Hematite NPs were mixed in 10 mM NaCl (pH 5.7) immediately before starting the DLS measurement and QCM flow through.

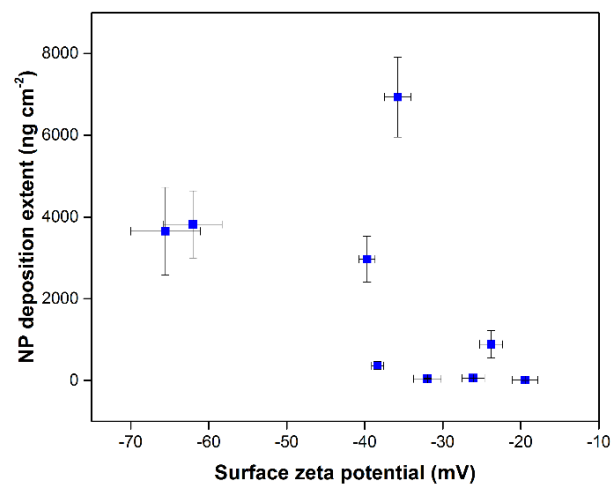


Figure S4. Correlations between surface zeta potentials of surface-immobilized protein layers and hematite NP deposition extents. Error bars indicate the standard deviations of at least triplicate measurements.

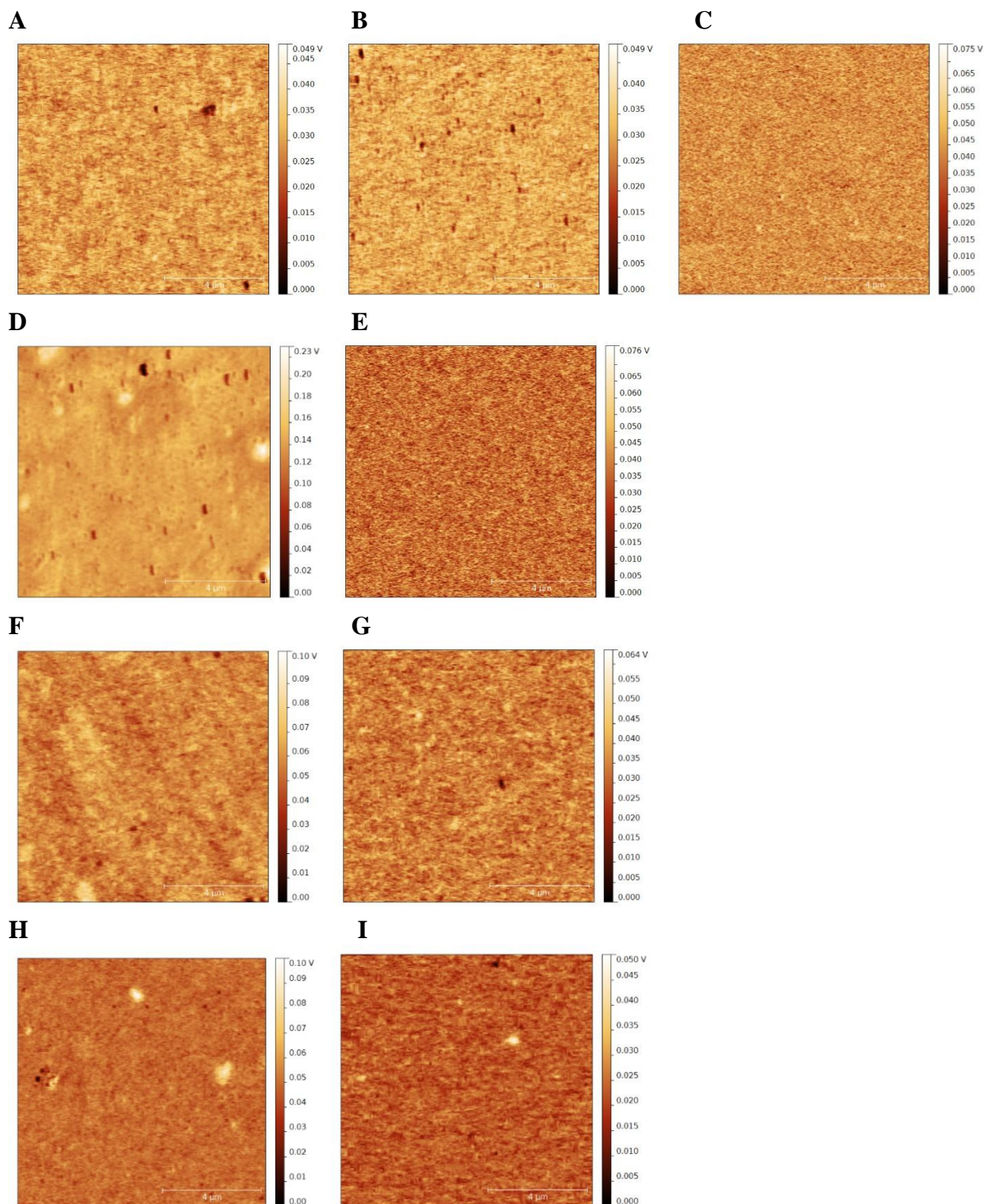


Figure S5. Representative KPFM surface potential images for BSA immobilized on a negatively-charged sensor surface (A) and positively-charged surface (B), lysozyme on a – surface (C), ubiquitin on a – surface (D) and + surface (E), *E. coli* protein extracts on a – surface (F) and + surface (G), and *P. fluorescens* protein extracts on a – surface (H) and + surface (I). The potential values corresponding to the color gradient are relative values with the zero threshold set individually for each image by the analysis software. The scale bars show 4 μm lengths. KPFM imaging was conducted under dry ambient conditions.

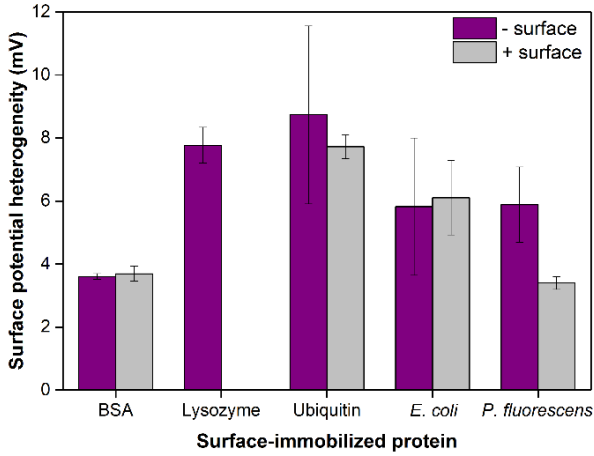


Figure S6. Surface potential variability of model and extracted proteins adsorbed on negatively- or positively-charged sensor surfaces. The variability was calculated as the RMS value of KPFM potential images. Lysozyme only adsorbed onto negatively-charged sensor surfaces; as such, no data were collected for positively-charged surfaces. Error bars indicate the standard deviations of the RMS values from at least five KPFM images.

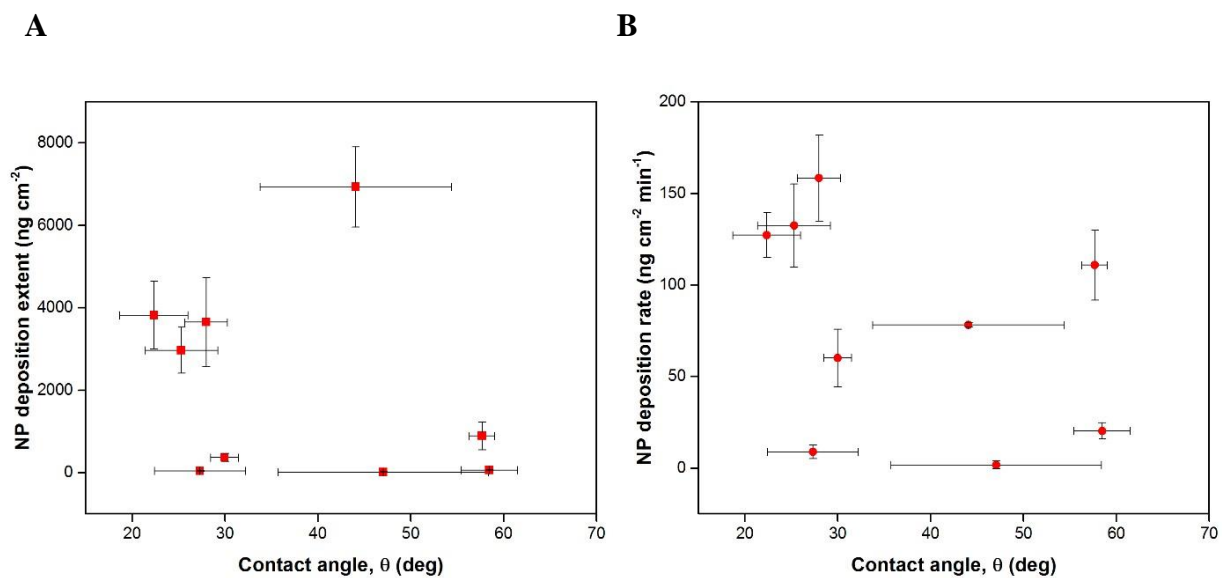


Figure S7. Correlations between contact angles of surface-immobilized protein layers and hematite NP deposition extents (A) and rates (B). Error bars indicate the standard deviations of at least triplicate measurements.