Electronic Supplementary Material (ESI) for Analyst.

Supporting Information for:

Factors influencing polyelectrolyte-aptamer multilayered films with target-controlled permeability for sensing applications

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Supporting Figures

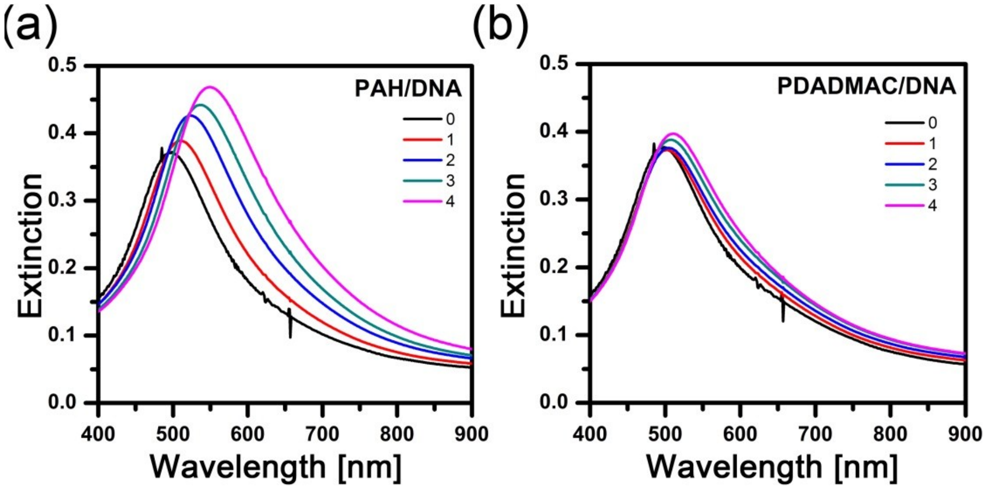

Fig. S1. Monitoring the layer-by-layer deposition via shifts in LSPR of the nanoparticles. Extinction spectra of immobilized nanoparticles with increasing number of bilayers of (a) PAH/DNA and (b) PDADMAC/DNA. The red shift in the LSPR indicates successful PAH/DNA multilayer deposition as the refractive index and film thickness increase. PDADMAC did not produce successive multilayer deposition with DNA as little shift in the LSPR was observed.

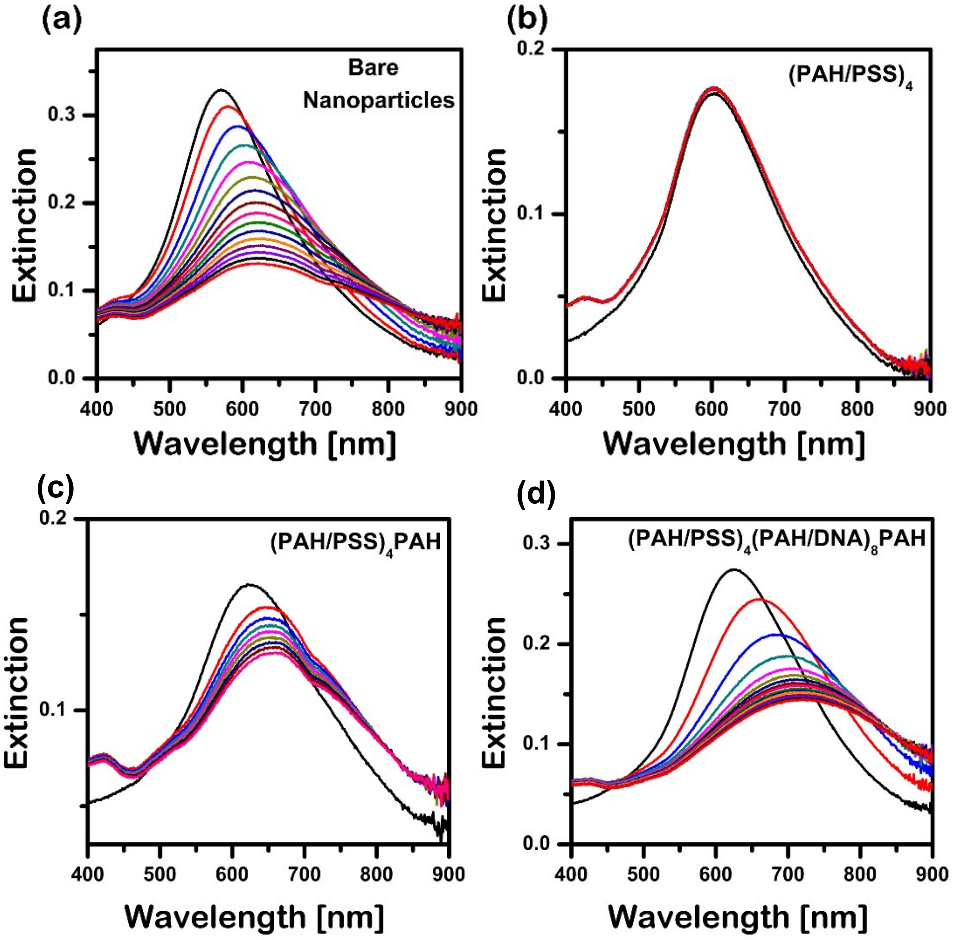

Fig. S2. UV-vis spectra of different samples during etching with 60 μM ferricyanide: (a) bare nanoparticles immobilized on cover slip; (b) nanoparticles covered with 4 bilayers of PAH/PSS; (c) nanoparticles covered with 4 bilayers of PAH/PSS and a PAH capping layer; and (d) nanoparticles covered with the optimized PE-aptamer film $(PAH/PSS)_4(PAH/DNA)_8PAH$. The diffusion of ferricyanide was impeded by the negative outermost layer (PSS); hence capping the film with the positive PAH is needed.

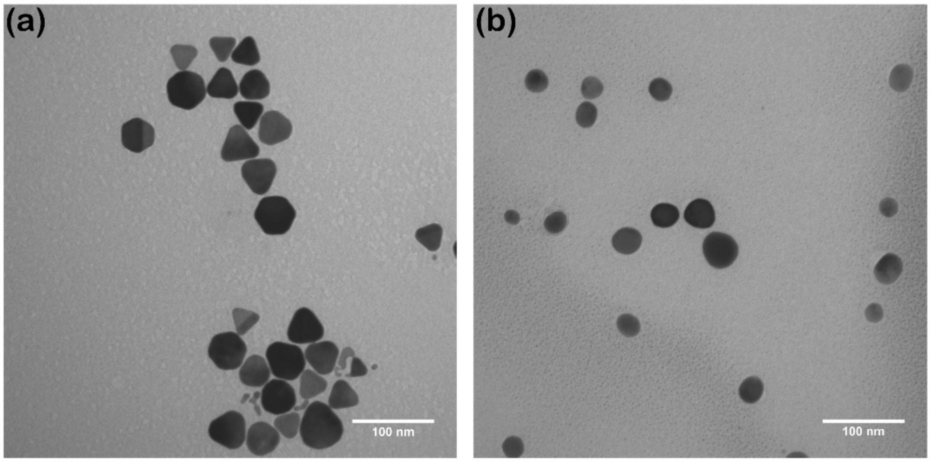

Fig. S3. Transmission electron microscopy images before (a) and after (b) etching of the nanoparticles with ferricyanide. The nanoparticles decrease in size and are more rounded after etching.

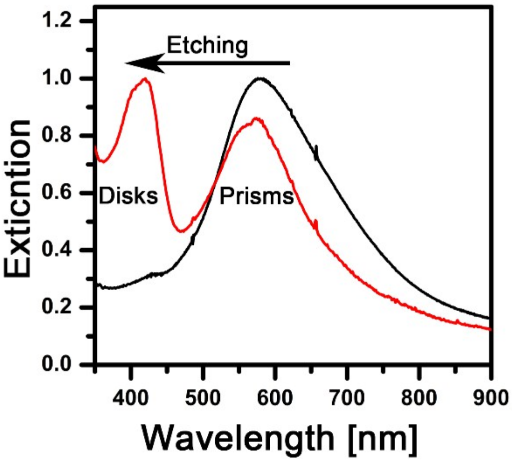

Fig. S4. Extinction spectra of gold-coated AgNP in solution before (black) and after (red) etching.

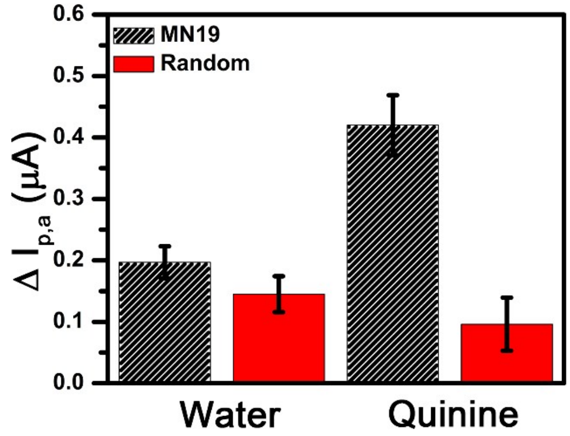

Fig. S5. A comparison of electrochemical responses of films containing quinine-binding aptamer vs random DNA. The random DNA sequence does not show different $\Delta I_{p,a}$when incubated with water vs quinine.

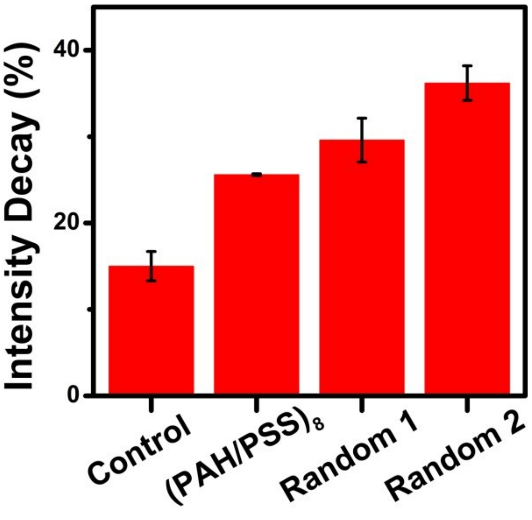

Fig. S6. Investigating the nature of sulfate interference. The PE-aptamer film in the colorimetric platform was replaced with regular polyelectrolytes (PAH/PSS) and those comprising random DNA sequences instead of MN19 aptamer. The films were incubated with sulfate for 1 h, and upon etching, an increase in the intensity decay of LSPR was observed for all cases as compared to the control film. It suggests that the interaction of sulfate is non-specific, most likely to the $-NH_3^+$ groups on the PAH.

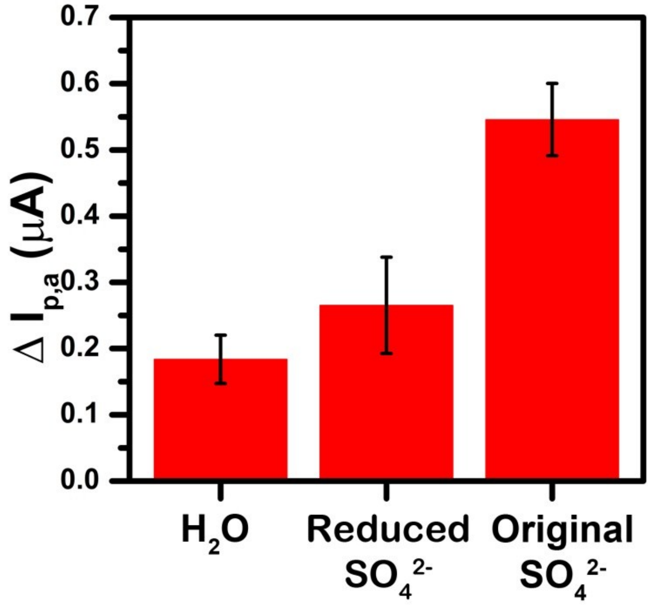

Fig. S7. Comparison of the electrochemical responses of PE-aptamer films exposed to water and solutions originally containing 1 mM sulfate with and without precipitation with $BaCl_2$. Pre-treatment of the solution reduces the false positive response and suggests possible remediation of the interference via chemical removal.

Experimental

TEM imaging

Morphological changes of the nanoparticles were examined using Phillips EM 201. The solutions of as-synthesized particles and after incubation with 1 mM of potassium ferricyanide for 10 minutes were concentrated by centrifugation. The samples were deposited via slow evaporation on copper grids.