Supporting Information

DNA-Embedded Au/Ag Core-Shell Nanoparticles

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Experimental details

Chemicals. Gold nanoparticles (15 nm) were purchased from Ted pella, Inc., Redding, CA, USA. Silver staining solutions (hydroquinone solution and $AgNO_3$ solution) were purchased from BBI international, cardiff, UK and used without further purification. Thiolated oligonucleotides were purchased from IDT Inc., Coralville, IA, USA.

Preparation of Oligonucleotide-Modified AuNPs. 3'-alkylthiol modified oligonucleotide (3'-HS-C_3-A_{10}-PEG_{18}-CTCCCTAATAACAAT-5') was used to functionalize 15-nm gold nanoparticles (Probe A), the disulfide was reduced using dithiolthretol (DTT, 0.1 M) in phosphate buffer for 2 hrs. The deprotected DNA solution was purified through a desalting NAP-5 column (Sephadex G-25 medium, DNA grade). The 5'-alkylthiol modified oligonucleotide (5'-HS-C_6-A_{10}-PEG_{18}-ACTCTTATCAATATT-3', Probe B) was deprotected with aqueous $AgNO_3$ (50 mM) for 20 min followed by treatment with DTT solution (10 mg/ml) for 5 min to remove excess $AgNO_3$. After centrifugation, the resulting precipitate was removed, and the supernatant was used for the next step. The reduced thiolated oligonucleotide solution (the supernatant) was purified by NAP-5 column. The 4 nmol of freshely reduced thiolated oligonucleotide was added to the 1 ml of 3.8 nM 15-nm AuNP solution. The mixed solution was wrapped in foil and placed on orbital shaker overnight at room temperature. The solution was then adjusted to obtain the final phosphate concentration of 9 mM and SDS concentration of ~0.1 % (wt/vol). After 30 min at orbital shaker, the colloids were brought to a final concentration of 0.3 M NaCl by six-time additions of one-sixth of the total salting buffer needed to reach a final concentration with 2 M NaCl. After the last salt addition and brief vortexing, the colloids were allowed to stand overnight at room temperature. The solution was centrifuged (14,000 rpm, 20 min), the supernatant was removed, and the precipitate was redispersed in 0.3 M PBS solution (this procedure was repeated twice). Finally, the solution was characterized by UV-Visible spectrophotometer.

In Situ Silver Staining-Based Preparation of DNA-em-Au/AgNPs. 100 µL of hydroquinone solution and 100 µL of $AgNO_3$ solution were diluted in NANOpure water (900 µL), respectively. The 12 µL of diluted hydroquinone solution was added to the 1 ml of 1 nM oligonucleotide-modified AuNPs. After gentle mixing, 12 µL of diluted $AgNO_3$ solution was rapidly added to the solutions. After vigorously mixed and incubated at room temperature, the reaction progress was monitored by UV-Vis spectrophotometry every 5 min. In general, the reactions started in a 2 minutes after $AgNO_3$ solutions added and it takes 30 min to reach the point of no substantial change in UV profile and to induce red wine to orange color change.

Next, the DNA-embedded Au/Ag core-shell nanoparticles were centrifuged (14,000 rpm, 15 min), the supernatant was removed, and the precipitated particles were redispersed in 0.3 M PBS solution. Finally, the solution was characterized by TEM and energy-dispersive X-ray (EDX) microanalysis.

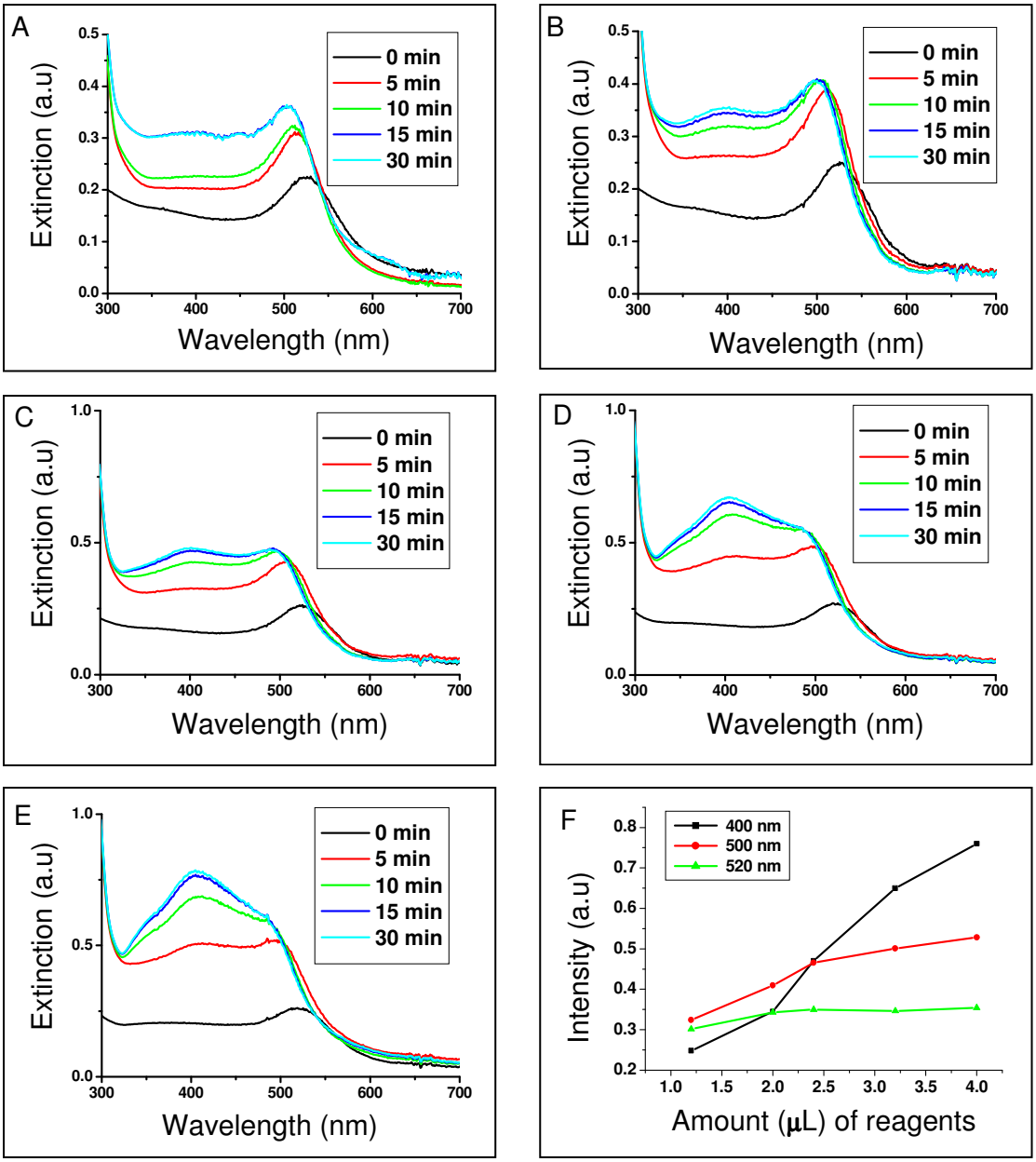

Figure S1. UV-visible absorption spectra from different amount of staining reagents and staining time (**A**. 1.2 µl of hydroquinone solution and 1.2 µl of $AgNO_3$ solution, **B**. 2.0 µl of hydroquinone solution and 2.0 µl of $AgNO_3$ solution **C**. 2.4 µl of hydroquinone solution and 2.4 µl of $AgNO_3$ solution. **D**. 3.2 µl of hydroquinone solution and 3.2 µl of $AgNO_3$ solution. **E.** 4.0 µl of hydroquinone solution and 4.0 µl of $AgNO_3$ solution. Absorption spectra of oligonucleotide-modified AuNPs (1.0 nM) are designated as 0 min in every profile (A-E). These results indicate that the final absorption spectra can be controlled by the amount of staining reagents (**F**), and the thickness of silver layer is controllable.