

## **Supporting Information**

### **Experimental procedures**

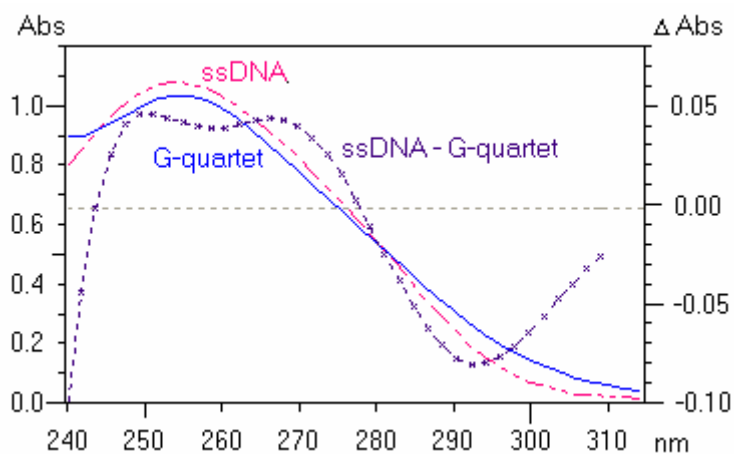
AuNPs (~13 nm) were prepared according to the published protocol.<sup>[1]</sup> Briefly, A sodium citrate solution (1 %, 3.5 mL) was rapidly added to a boiled HAuCl<sub>4</sub> solution under vigorous stirring. The mixed solution was boiled for 10 min and further stirred for 15 min. The resulting solution was cooled to room temperature and filtered, which was stored in the refrigerator and ready for use. A DNA aptamer for potassium ions (K<sup>+</sup>)<sup>[2]</sup> was employed as the model system in this work, which has a sequence of GGG TTA GGG TTA GGG TTA GGG. Two DNA oligos with random sequences (AGC AAC CTC AAA CAG ACA CCA TGG; TAG CTA TGG AAT TCC TCG TAG GCA) were used as the control.

A typical colorimetric test includes the following steps. First, ten microliters of DNA solutions (10 μM) was mixed with 2 μL K<sup>+</sup> with an appropriate concentration. This solution was heated at 85 °C for 5 min and slowly cooled to the room temperature. Second, 4 μL of such prepared solution was added to the solution of gold nanoparticles (200 μL). The solutions were allowed to react for 4 min at room temperature and then 35 μL of buffered salt (10 mM phosphate, 0.5 M NaCl, pH 7.4) was added to produce color change.

### **UV-vis studies on the formation of G-quartets:<sup>[3]</sup>**

The formation of G-quartets of K<sup>+</sup> aptamers in the presence of K<sup>+</sup> can be studied by

UV-vis absorption spectroscopy. Briefly, 5  $\mu\text{L}$  of aptamer (75  $\mu\text{M}$ ) was first diluted with 60  $\mu\text{L}$  of water and then mixed with 10  $\mu\text{L}$  of  $\text{K}^+$  (2 mM). The mixed solution was measured with UV-vis spectroscopy (Hitachi, U-3010 spectrophotometer) both at room temperature and at 90  $^{\circ}\text{C}$ . The difference spectrum (purple curve in Figure 1-S) shows a clear positive peak at 272 nm and a negative peak at 295 nm, which is characteristic of the formation of G-quartets. Such absorption variation cannot be observed for the control DNA.



**Figure 1-S.** UV spectra of  $\text{K}^+$  aptamers. The red and blue curves represent the UV spectra for the random coil and the  $\text{K}^+$ -stabilized G-quartet, respectively. The purple curve is the difference spectrum.



**Figure 2-S.** Selectivity of gold nanoparticles-based colorimetric detection. Four microliters of DNA solutions (8.3  $\mu\text{M}$ ), in the presence of 16.7 mM  $\text{Li}^+$ ,  $\text{Na}^+$ ,  $\text{Rb}^+$ ,  $\text{NH}_4^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  (from left to right), were mixed with 200  $\mu\text{L}$  of gold nanoparticles. The solutions were allowed to react for 4 min at room temperature and then 35  $\mu\text{L}$  of buffered salt (10 mM phosphate, 0.5 M NaCl, pH 7.4) was added to each solution. Note that only  $\text{K}^+$  induced the formation of G-quartet structure and led to the aggregation of gold nanoparticles (purple color).

#### References:

- [1] K. C. Grabar, R. G. Freeman, M. B. Hommer, M. J. Natan, *Anal. Chem.* **1995**, *67*, 735.
- [2] H. Ueyama, M. Takagi, S. Takenaka, *J. Am. Chem. Soc.* **2002**, *124*, 14286.
- [3] J.-L. Mergny, A.-T. Phanb, L. Lacroixa, *FEBS Letters* **1998**, *435*, 74.