Supporting Information

Reconfigurable and resettable arithmetic logic units based on magnetic bead and DNA

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Detailed Experimental Procedures

Preparation of molecular beacons modified Fe₃O₄@SiO₂@Au magnetic nanoparticles

The Fe₃O₄ magnetic beads with the size of 200 nm were synthesized using solvothermal method. FeCl₃•6H₂O (0.54 g), Na acrylate (1.5 g), NaOAc (1.5 g), were dissolved in ethylene glycol (20 mL) under magnetic stirring. The mixture was transferred into a Teflon stainless-steel autoclave and heated at 200 °C for 10 h. The products were washed four times with ethanol and water. Then the magnetic Fe₃O₄ nanoparticles were dispersed in a mixture of ethanol (80 mL) and water (20 mL) under ultrasonication for 5 min. To prepare Fe₃O₄@SiO₂ core-shell nanostructures, ammonia water (1 mL) and TEOS (200 μL) was added into the mixture solution under...
mechanical stirring for 12 h. After being separated, the product was mixed with
APTES (100μL) and ammonia (2 mL) under mechanical stirring for another 12 h.
Then 50 mL of Au NPs solution (13 nm, synthesized with citrate reduction) was
added into the dispersed solution. The diameter of the Fe$_3$O$_4$@SiO$_2$@Au was about
200 nm evaluated by TEM measurements.

The molecular beacons functionalized Fe$_3$O$_4$@SiO$_2$@Au was prepared by
mixing Fe$_3$O$_4$@SiO$_2$@Au suspension with thiol-molecular beacons (final
concentration of oligonucleotides 10 μM) in PBS buffer (0.3 M NaCl, 0.2 PBS, pH=7)
under continuous shaking for 16 hours. To remove excess thiol-molecular beacons,
these molecular beacons-functionalized Fe$_3$O$_4$@SiO$_2$@Au nanoparticles were
magnetic separated and washed three times using Tris-HCl buffer (20 mM Tris-HCl,
200 mM KCl, 10 mM MgCl$_2$, pH 8.0). The functionalized Fe$_3$O$_4$@SiO$_2$@Au
nanoparticles were stored in the buffer solution and used as the initial platform of
the half adder and half subtractor.
**Fig. S1.** Transmission electron micrograph (TEM) of (A) Fe₃O₄ microspheres, (B) magnetic Fe₃O₄@SiO₂ core/shell composites, (C) and (D) magnetic Fe₃O₄@SiO₂@Au composites.

As exhibited by transmission electron microscopy (TEM), Fig. S1A shows the magnetic nanoparticles have an average diameter of about 200 nm. Through the hydrolysis and condensation of TEOS in the ethanol–ammonia mixture, the silica layer was gradually coated onto the surface of Fe₃O₄ nanosphere. The core–shell structure of Fe₃O₄@SiO₂ nanosphere was clearly exhibited in Fig. S1B. Then the magnetic Fe₃O₄@SiO₂@Au nanoparticles were prepared based on Au–N binding between the amine modified on the surface of Fe₃O₄@SiO₂ and gold nanoparticles (Fig. S1C and Fig. S1D). The prepared Fe₃O₄@SiO₂@Au nanoparticles had long-time stability and would not be separated under ultrasonic condition.
**Fig. S2.** The UV-Vis absorption spectra of (a) Fe$_3$O$_4$@SiO$_2$@Au (black curve) and (b) Fe$_3$O$_4$@SiO$_2$@Au@DNA (red curve).

To endow reconfigurable function, molecular beacons are modified on the Fe$_3$O$_4$@SiO$_2$@Au surface. After removing the excess molecular beacon, the functionalized magnetic bead is characterized by measuring the UV absorbance spectra (Fig. S2). Except that the feature peaks of gold nanoparticle are monitored at 530 nm from Fig. S2a and S2b, a new peak at 260 nm is found for functionalized magnetic bead in Fig. S2b, indicating the successful anchoring of molecular beacon on the surface of magnetic bead.
Fig. S3. CD spectra of products for characterizing the DNA structural conversion of the half adder (A) and half subtractor (B), demonstrating G-quadruplex formation. The concentration of each strand (MB, HA-1, HA-2, HS-1 and HS-2) was 1 μM.

In the HA logic gate, G-quadruplex is formed owing to the hybridization of HA-1 and HA-2. As shown in Fig. S3A, the CD spectra of the random oligonucleotides, molecular beacon, HA-1 and HA-2 is of relatively low amplitude, indicating that the DNA strands possess no obvious G-quadruplex structure. Once the two inputs oligonucleotides hybridize together, distinct peaks appear at 242 nm for a negative peak and 264 nm for a positive peak, respectively, indicating the formation of a parallel G-quadruplex with the aid of K⁺ in the solution. In the HS logic gate, HS-1 is a random single strand DNA and presents low amplitude. While HS-2 is a G-riched oligonucleotide and can form a parallel G-quadruplex structure according to obvious negative peak at 242 nm and positive peak at 267 nm in CD spectra. Once adding HS-1 into HS-2 (curve c), the obvious peaks disappear, indicating that the hybridization between HS-1 and HS-2 forms a more stable double-stranded structure and influences the G-quadruplex configuration of HS-2.
Fig. S4 The FAM fluorescence response of magnetic bead/MB at 518 nm with increasing the concentration of (A) HA-1, HA-2 and (B) HS-1, HS-2.

The fluorescent signal of FAM is gradually recovered and reaches a platform with the increasing concentration of input DNA. Here, 250 nM was chosen as the appropriate concentration for all the input DNA.