Electronic Supplementary Information

Label-free and enzyme-free platform for the construction of advanced DNA logic devices based on the assembly of graphene oxide and DNA-templated AgNCs

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Table S1. Sequences of DNA strands used in this work.(poly-C regions to form AgNCs were colored in red; G-quadruplex sequences were underlined; hybridization region of P strand and complementary region were italicized; toehold region was colored in blue; hybridization region of G-quadruplex was colored in purple; poly-C region to inhibit G-quadruplex was colored in yellow, respectively.)

Strand Name	Sequence (5' to 3')
P	CCCACCCACCCTCCCA *TGCA AAGTAAGTTAAAGTCGTATATAA*
2D0	ACGTTTCATTCAATTTCAGCATATATT
2D1	*TTATATACGACTTTAACTTACTTTGCA*
4G3	TATGCTTGAAACTATTCTTAT GGGTGGGTGGGT
4D0	ACGTTTCATTCAATTTCAGCATATATT
4D1	*TTATATA CGACTTTAACTTAC TTT GCA*
4D2	TGGG ATAAGAATAGTTTCAAGCATA GTTGTAA
4D3	TGGG ATAAGAATAGTTTCAAGCATA TGG *ATATACGACTTTAACTTACTTTGCA*
1G8	TATGCTTGAAACTATTCTTAT GGGTGGGTGG
1G4	GTGGG ATAAGAATAGTTTCAAGCATA GTTGTAA
1D0	TTACAAC TATGCTTGAAACTATTCTTAT CCCAC *TATAT TTATATACGACTTTAACTTACTTTGCA*
C	CCCACCCACCCTCCCA ATTCGTCACTTTCGTATCACGTTTGTGTGCAAT
Co-1	CCCAAAACCCAAAACCC CGATGAACCAAACGTTAGACGAAAGTATATAAT
Co-2	ATTATATACAAACGTCTAACGAAAGGTTCATCG GGGTTTTGGGTTTTGGGTTTTGGG

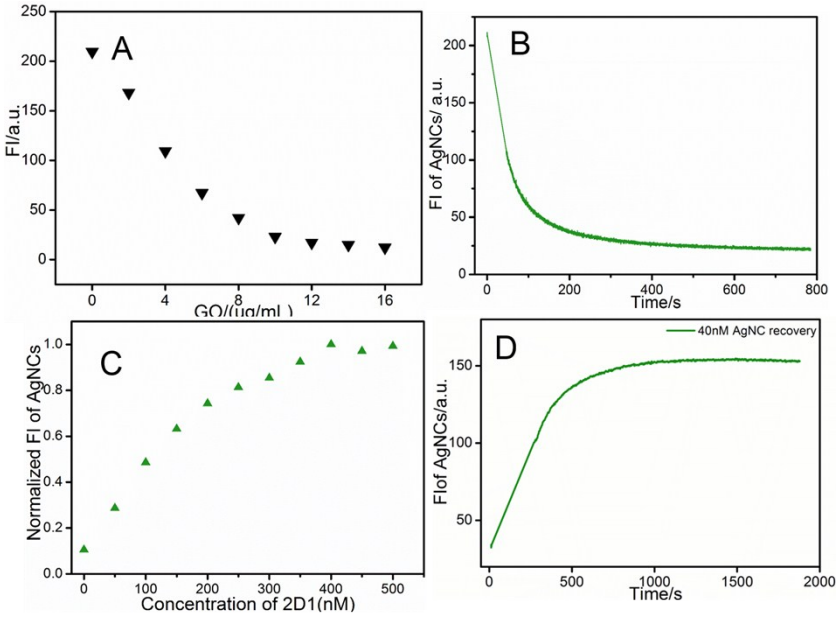

Figure S1. (A) Fluorescence values at 790 nm of 40 nM P-strand stabilized AgNCs in the presence of different concentrations of GO (0、2、4、6、8、10、12、14、16 ug/mL); **(B)** The kinetic quenching curve of 40nM P-strand stabilized AgNCs in the presence of 9 ug/mL GO. **(C)** Normalized fluorescence recovery responses of the mixture of 40 nM AgNCs, 9 ug/mL GO in the presence of different concentrations of complementary strand 2D1 (The highest fluorescence value at 400 nM was taken as 1.0). **(D)** The kinetic recovery curve of 40nM P-strand stabilized AgNCs after quenching by 9 ug/mL GO in the presence of 400 nM complementary strand.

As shown in **Figure S1 (A)**, the fluorescence value decreased gradually with the addition of GO, and reached the lowest value in the presence of 10 ug/mL GO. To facilitate the recovery of AgNCs' fluorescence, we selected 9 ug/mL GO in our work. **Figure S1 (B)** shows the quenching kinetic curve of 40 nM AgNCs in the presence of 9 ug/mL GO, the fluorescence exhibited no changes after 10 min, so we selected 10 min to quench the AgNCs in our work. As can be seen in **Figure S1(C)** the fluorescence began to recover with the addition of different concentrations of 2D1 and reached a plateau in the presence of 400 nM complementary strand. **Figure S1 (D)** shows the kinetic recovery curve of 40 nM AgNCs after quenching by 9 ug/mL GO in the presence of 400 nM complementary strand, the fluorescence reached a plateau after 2000s, so we selected 35 min in the recovery.

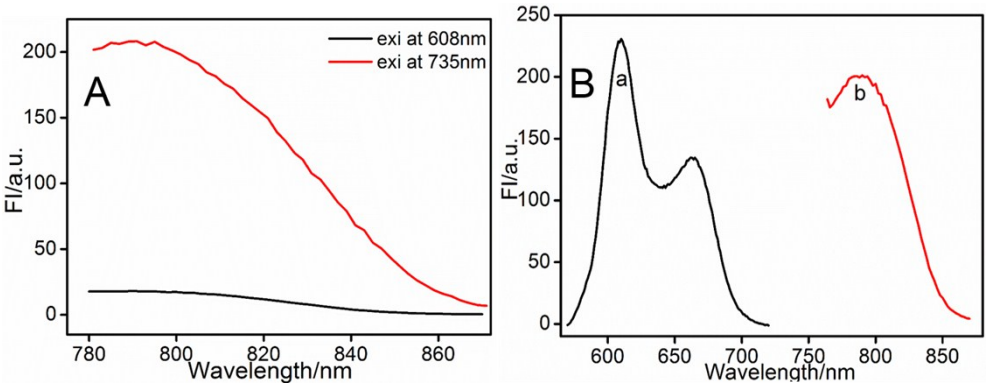

Figure S2. (A) Fluorescence emission spectra of P stabilized AgNCs excited at NMM's emission wavelength 608 nm (black) and at the its excitation wavelength, 735 nm (red);**(B)** Fluorescence emission spectra of NMM enhanced by 150 nM G-quadruplex (a, black) and 40 nM P stabilized AgNCs (b, red) in the same solution.

As can be seen in **Figure S2 (A)**, the fluorescence shows only negligible value excited at NMM's emission wavelength 608 nm, which indicated that there is not FRET phenomenon between AgNCs and NMM. **Figure S2 (B)** shows NMM's emission spectrum (608 nm) enhanced by 150 nM G-quadruplex and AgNCs' emission spectrum (790 nm), indicating that the two signals could coexist in the same solution without interferences between each other.

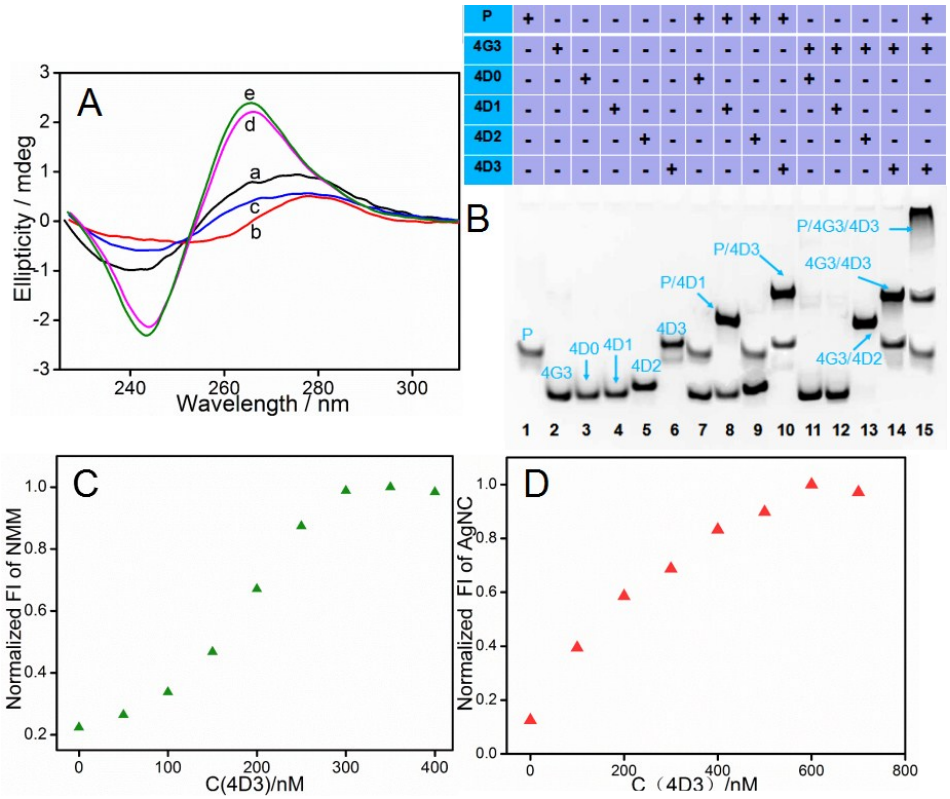

Figure S3. (A) The CD spectrum of different DNA strands: 4G3 (a), 4D2 (b), 4D3 (c), 4D2 and 4G3 (d), 4D3 and 4G3 (e). **(B)** 15% PAGE analysis of the interaction of different DNA strands used in 4 to 2 encoder. (The presence and absence of different DNA strands were represented by "+" and "-", respectively). **(C)** Normalized fluorescence responses at 608 nm of NMM and 150 nM 4G3 in the presence of different concentrations of strand 4D3 (The highest fluorescence value at 300 nM was taken as 1.0). **(D)** Normalized fluorescence responses at 790 nm of 40 nM AgNCs in the presence of different concentrations of strand 4D3 (The highest fluorescence value at 600 nM was taken as 1.0).

As shown in **Figure S3 (A)**, the CD spectrum have no obvious peaks in the presence of 4G3 (a), 4D2 (b) and 4D3(c), while after the addition of either 4D2 or 4D3 into 4G3, a positive peak at 266 nm (264 nm for 4D3) and a negative peak at 244 nm (242 nm for 4D3) appeared, which are the characteristic peaks of a parallel G-quadruplex configuration. Thus, the formation of G-quadruplex was validated.

Figure S3 (B) shows the polyacrylamide gel analysis of the interaction between different DNA strands used in 4 to 2 encoder. Lane 1 to Lane 6 shows the DNA bands of ss-DNA P, 4G3, 4D0, 4D1, 4D2, 4D3, respectively. The two separate bands shown in Lane 7 and Lane 9 suggested that

P will not hybridize with 4D0 and 4D2; while after the addition of 4D1 or 4D3, new bands appeared in Lane 8 and Lane 10, indicated the hybridization between P and 4D1, P and 4D3. Similarly, no new bands produced in Lane 11, Lane 12 suggested the non-hybridization between 4G3 and 4D0; 4G3 and 4D1. And new bands appeared in Lane 13, Lane 14, Lane 15 suggested the formation of duplex 4G3/4D2 and 4G3/4D3. And in the presence of P, 4G3, 4D3, another new band appeared in Lane 15, indicated the formation of triplex configuration of P/4G3/4D3.

As can be seen in **Figure S3 (C),** normalized fluorescence values at 608 nm of NMM with 150 nM 4G3 increased gradually with the addition of different concentrations of 4D3, and reached a plateau at 400 nM. **Figure S3 (D)** shows the normalized fluorescence values at 790 nm of 40 nM AgNCs in the presence of different concentrations of 4D3, the signals reached the highest value at 600 nM. To perform the logic device at optimized conditions, 600 nM 4D3 was used in our work.

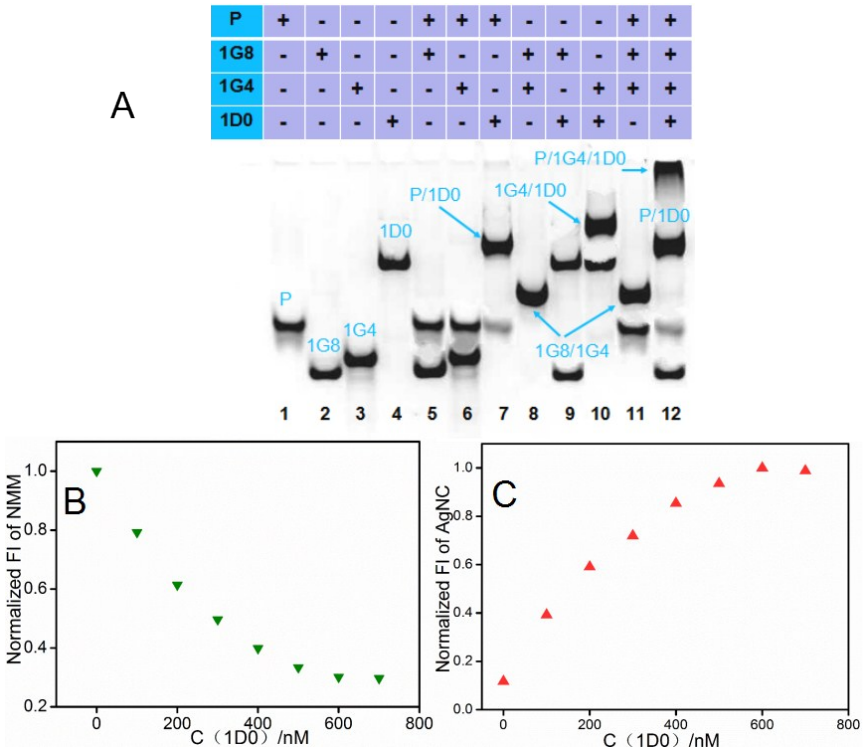

Figure S4. (A) 15% PAGE analysis of the interaction of different DNA strands used in 1 to 2 decoder. (The presence and absence of different DNA strands were represented by "+" and "-", respectively).**(B)** Normalized fluorescence responses at 608 nm of NMM and 150 nM 1G8 and 1G4 in the presence of different concentrations of strand 1D0 (The highest fluorescence value at 0nM was taken as 1.0). **(C)** Normalized fluorescence responses at 790 nm of 40 nM AgNCs in the presence of different concentrations of strand 1D0 (The highest fluorescence value at 600 nM was taken as 1.0).

Figure S4 (A) shows the polyacrylamide gel analysis of the interaction between different DNA strands used in 1 to 2 decoder. Lane 1 to Lane 4 shows the DNA bands of ss-DNA P, 1G8, 1G4, 1D0, respectively. The separate two bands shown in Lane 5, Lane 6 and Lane 9 suggested that P will not hybridize with 1G8 and 1G4; and 1D0 will not hybridize with 1G8. While after the addition of excess 1D0 into P, 1G8 into 1G4, 1D0 into 1G4, new bands appeared in Lane 7, Lane 8 and Lane 11, indicated the hybridization between 1D0 and P, 1G8 and 1G4, 1D0 and 1G4, respectively. And in the presence of P, 1G8, 1G4 and 1D0, another new band appeared in Lane 12, indicated the formation of triplex configuration of P/1G4/1D0.

As can be seen in **Figure S4 (B),** normalized fluorescence signals at 608 nm of NMM with 150 nM G-quadruplex decreased gradually with the addition of different concentrations of 1D0, and reached the lowest value at 600 nM. **FigureS4(C)** shows the normalized fluorescence values at

790 nm of 40 nM AgNCs in the presence of different concentrations of 1D0, the signals reached the highest value at 600 nM. To perform the logic device at optimized conditions, 600 nM 1D0 was used in our work.

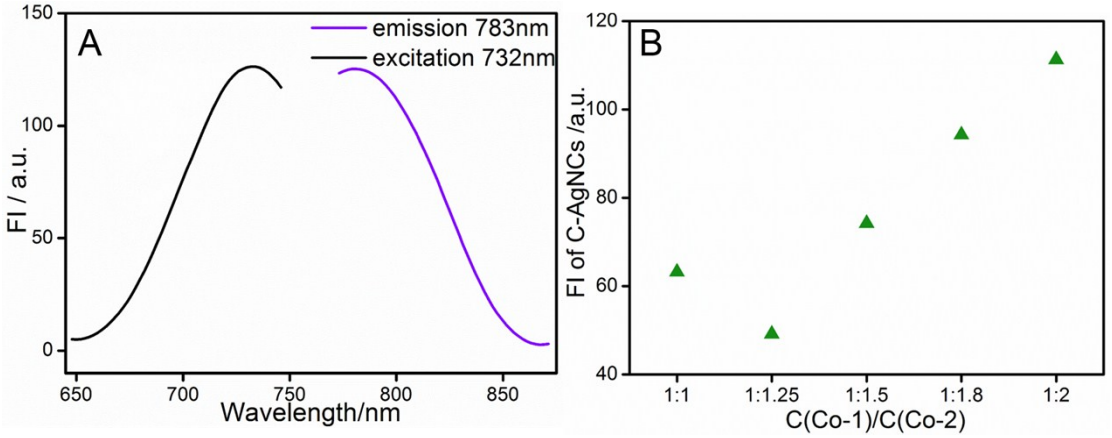

Figure S5. (A) The fluorescence excitation (black, 732 nm) and emission spectrum (purple, 783 nm) of C-strand stabilized AgNCs used in the comparator. **(B)** The fluorescence values of C-strand stabilized AgNCs in the presence of different ratios of Co-1 and Co-2. (400nM Co-1 was used in the comparator to optimize the concentration of Co-2)

As can be seen in **Figure S5 (B)**, different ratios of Co-1 and Co-2 were added into C-strand AgNCs, fluorescence of C-AgNCs showed the lowest value at the ratio of Co-1: Co-2=1: 1.25, so we select this ratio in our experiment.