Electronic Supplementary information for

Multiple and Configurable Optical Logic Systems Based on Layered Double Hydroxides and Chromophore Assemblies

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1. Experimental section

1.1 Materials

Sodium salt of dsDNA of salmon spermary (biochemistry grade), sulfonated poly(p-phenylene) (APPP), fluorescein (FLU) and ethidium bromide (EB) were purchased from Sigma-Aldrich Company. Analytical grade chemicals containing $Mg(NO_3)_2{\cdot}6H_2O$, $Al(NO_3)_3{\cdot}9H_2O$, formamide, H_2O_2 and H_2SO_4 were used without further purification. The deionized and decarbonated water was used in all the experimental processes.

1.2 Fabrication of fluorescein modified LDH nanosheets (LDH-FLU nanosheets)

The Mg_2Al-NO_3 LDH precursor was synthesized by a hydrothermal method. Subsequently, the FLU intercalated LDH colloid composite was prepared following the

ion-exchange method. FLU (10^{-3} mol) was dissolved in 150 mL of water. Freshly prepared Mg_2Al-NO_3 LDH colloid (40 mL, 0.025 g mL^{-1}) was dispersed in the FLU solution thoroughly. The suspension was agitated at room temperature under N_2 atmosphere for 48 h. The FLU-LDH nanoparticles were obtained by washing extensively with water and then drying at 70 °C for 24 h. Subsequently, a 0.1 g of FLU-LDH was shaken in 100 mL of formamide solution for 24 h to obtain a colloidal suspension of exfoliated FLU-LDH nanosheets.

1.3 Fabrication of the (APPP/LDH-FLU)$_n$ UTFs

The quartz glass substrate was cleaned in concentrated NH_3/30% H_2O_2 (7:3) and concentrated H_2SO_4 for 30 min each. After each procedure, the quartz substrate was rinsed and washed thoroughly with deionized water. The substrate was dipped in a colloidal suspension (0.1 g mL^{-1}) of LDH-FLU nanosheets for 10 min followed by washing thoroughly, and then the substrate was treated with a 100 mL of APPP aqueous solution (0.05 wt%) for another 10 min followed by washing. Multilayer films of the (APPP/LDH-FLU)$_n$ were fabricated by alternate deposition of LDH-FLU nanosheets suspension and APPP solution for *n* cycles. The resulting films were dried with a nitrogen gas flow for 2 min. The (LDH-FLU/DNA)$_n$ UTFs, the (LDH/DNA)$_n$ UTFs, the (LDH-FLU/DNA)$_n$ UTFs, the (APPP/LDH-FLU/DNA)$_n$ UTFs and the (APPP/LDH/DNA)$_n$ UTFs were fabricated following the same procedure.

1.4 Fabrication of the (LDH/DNA-EB)$_n$ UTFs

The (LDH/DNA)$_n$ UTFs were immersed into 1 mM aqueous solution of EB at 50 °C for 1 min, followed by washing sufficiently with deionized water. The (LDH-FLU/DNA-EB)$_n$ UTFs,

the $(APPP/LDH/DNA\text{-}EB)_n$ UTFs and the $(APPP/LDH\text{-}FLU/DNA\text{-}EB)_n$ UTFs were fabricated by the same procedure.

1.5 Characterization techniques

The UV-vis absorption spectra were collected in the range from 200 to 700 nm on a Shimadzu T-9201 spectrophotometer, with the slit width of 2.0 nm. The fluorescence spectra were performed on a RF-5301PC fluorospectrophotometer with the excitation wavelength of 500 nm. The fluorescence emission spectra range in 400–750 nm, and both the excitation and emission slits were set to 5 nm. X-ray diffraction patterns (XRD) of the $(DNA/LDH)_n$ UTFs were recorded using a Rigaku 2500 VB2+PC diffractometer under the conditions: 40 kV, 50 mA, Cu Kα radiation (λ =0.154 nm) step-scanned with a scanning rate of 0.5 °/min, and a 2θ angle ranging from 2 to 70°. The morphology of UTFs was investigated by using a scanning electron microscope (SEM ZEISS), and the accelerating voltage applied was 20 kV. The surface roughness and thickness data were obtained by using the atomic force microscopy (AFM) software (Digital Instruments, version 6.12).

2. Results and discussion

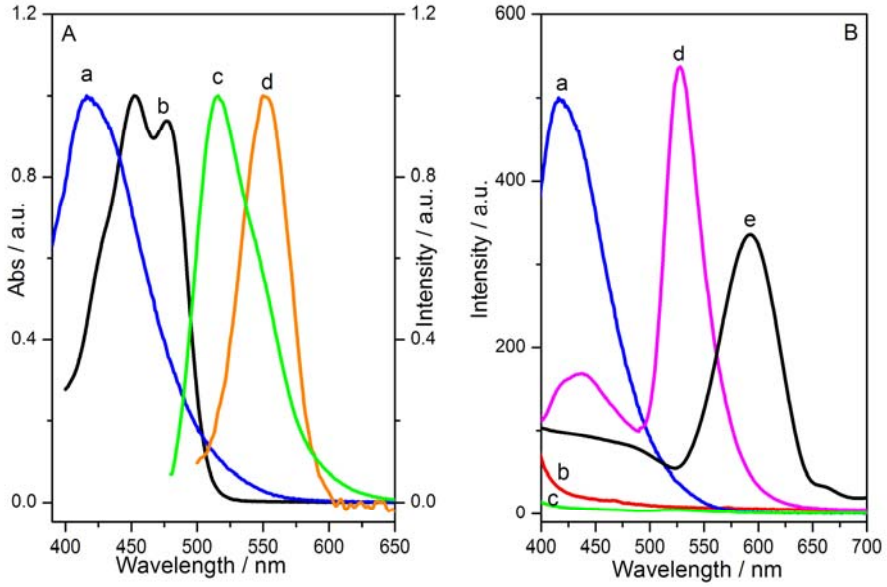

Fig. S1 (A) Normalized spectra of (a) fluorescence emission of APPP aqueous solution (λ_{ex} = 350 nm), (b) UV-vis absorption and (c) fluorescence emission of FLU aqueous solution (λ_{ex} = 490 nm), (d) UV-vis absorption of EB aqueous solution. (B) Fluorescence emission spectra of (a) APPP aqueous solution, (b) FLU aqueous solution, (c) EB aqueous solution, (d) APPP and FLU mixture solution, (e) APPP, FLU and EB mixture solution (λ_{ex} = 350 nm).

Fig. S1A shows the spectrum superimposition between fluorescence emission of APPP solution (curve a) and UV-vis absorption of FLU solution (curve b), indicating the fluorescence resonance energy transfer (FRET) may occur between APPP and FLU. Similar spectrum overlap between fluorescence emission of FLU solution (curve c) and UV-vis absorption of EB solution (curve d) is also observed. The phenomena imply the possibility of a dual FRET: energy transfer from APPP to FLU, followed by a further energy transfer from FLU to EB. This was confirmed from the fluorescence emission of several mixture solutions (Fig. S1B). The absence of emission peak of FLU (curve b) and EB (curve c) solution shows that both FLU and EB can not be excited at λ_{ex} = 350 nm. As a result, the presence of emission at 525 nm for APPP+FLU mixture solution (curve d) as well as the one at 600 nm

for APPP+FLU+EB mixture solution (curve e) testifies the occurrence of dual FRET process (λ_{ex} = 350 nm).

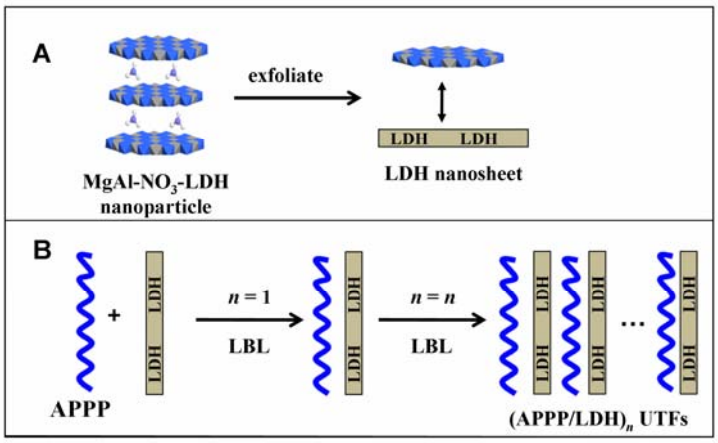

Fig. S2 A schematic representation of (A) an LDH nanoparticle and an LDH nanosheet, (B) the $(APPP/LDH)_n$ UTFs.

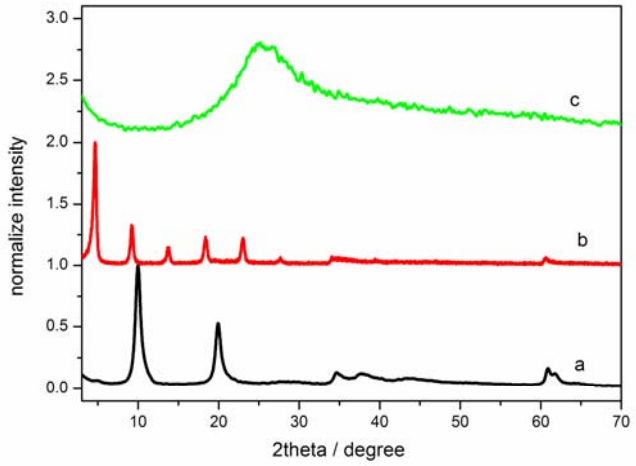

Fig. S3 The XRD patterns of (a) Mg_2Al-NO_3 LDH, (b) Mg_2Al-FLU LDH, (c) Mg_2Al-FLU LDH nanosheets.

The XRD patterns of Mg_2Al-NO_3 LDH, Mg_2Al-FLU LDH and Mg_2Al-FLU LDH nanosheets are shown in Fig. S3. Both of the two powder samples (curve a and b) can be indexed to a hexagonal lattice. The interlayer spacing can be calculated from averaging the positions of the

three harmonics: c = 1/3 (d_{003}+ 2d_{006}+ 3d_{009}). The (003) reflection of Mg_2Al-NO_3 LDH sample at 2θ 9.9° (curve a) shows an interlayer distance of 0.88 nm; the basal spacing of Mg_2Al-FLU LDH (curve b) increases to ~1.9 nm compared with Mg_2Al-NO_3 LDH precursor, indicating that FLU was intercalated into the galleries of LDH. The absence of LDH characteristic reflections in curve c indicates a successful exfoliation of Mg_2Al-FLU LDH nanoparticles to the resulting Mg_2Al-FLU LDH nanosheets.

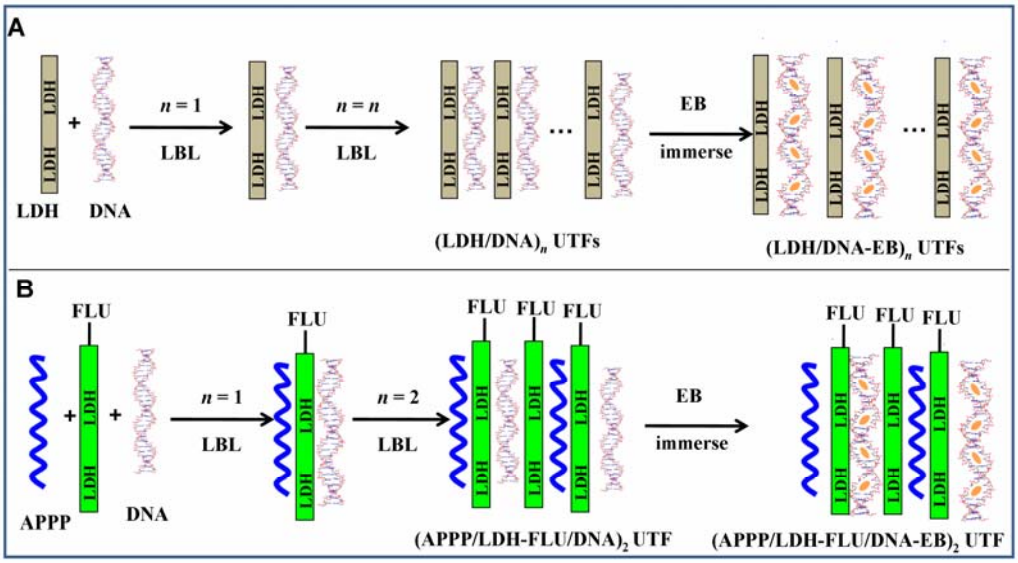

Fig. S4 A schematic representation of (A) the (LDH/DNA-EB)$_n$ UTFs and (B) the (APPP/LDH-FLU/DNA-EB)$_2$ UTF.

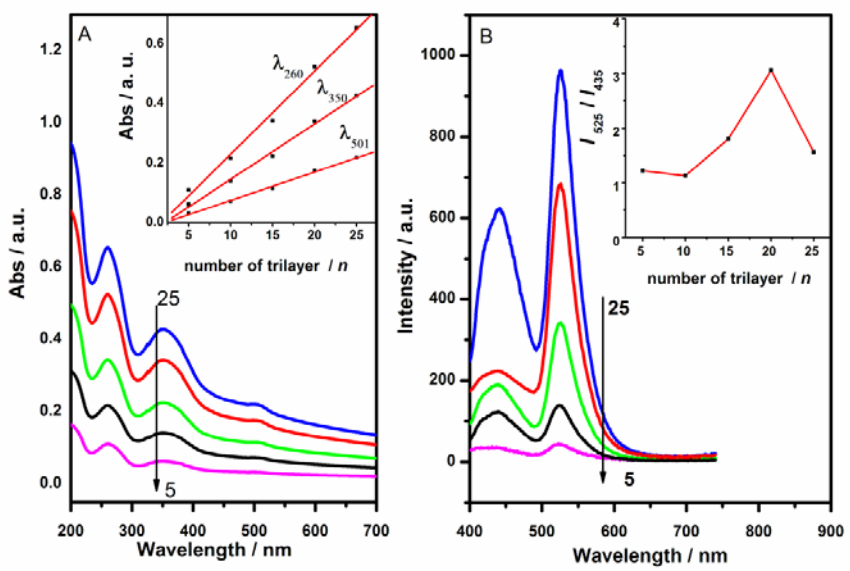

Fig. S5 (A) UV-vis absorption spectra of the (APPP/LDH-FLU/DNA)$_n$ UTFs (n=5–25) (inset: plots of the absorbance at 260, 350 and 501 nm *vs*. *n*, respectively). (B) Fluorescence emission spectra of the (APPP/LDH-FLU/DNA)$_n$ UTFs (n=5–25) (inset: fluorescence emission ratio I_{525}/I_{435} *vs*. *n*).

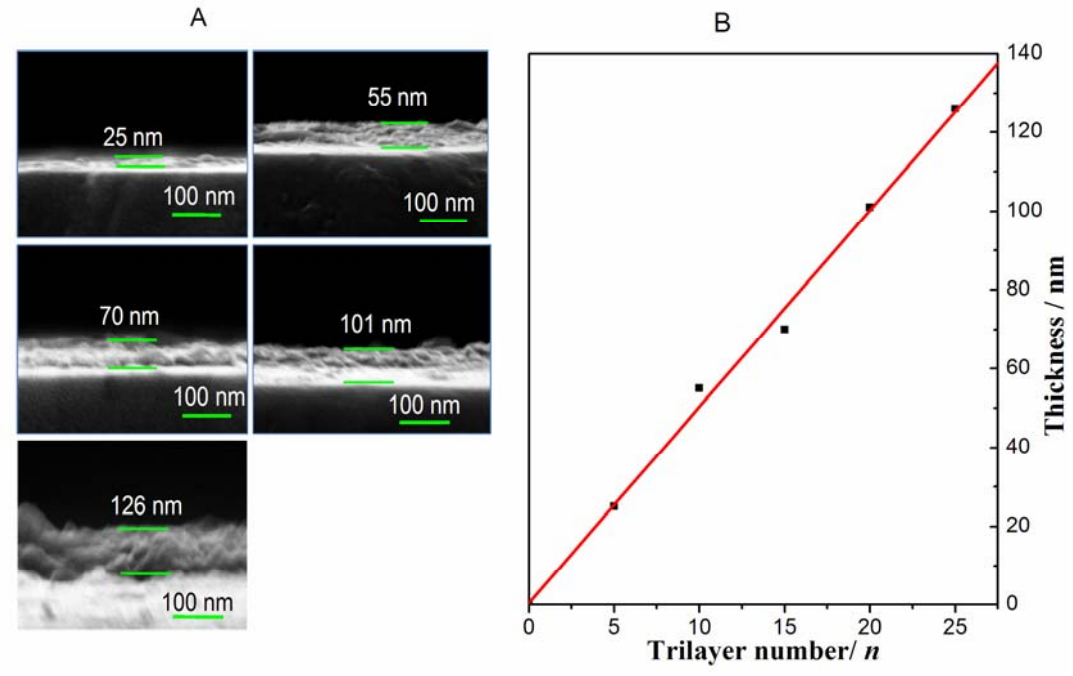

Fig. S6 (A) Side-view of SEM images for the (APPP/LDH-FLU/DNA)$_n$ UTFs with n=5, 10, 15, 20 and 25 respectively. (B) Film thickness of these UTFs as a function of *n*.

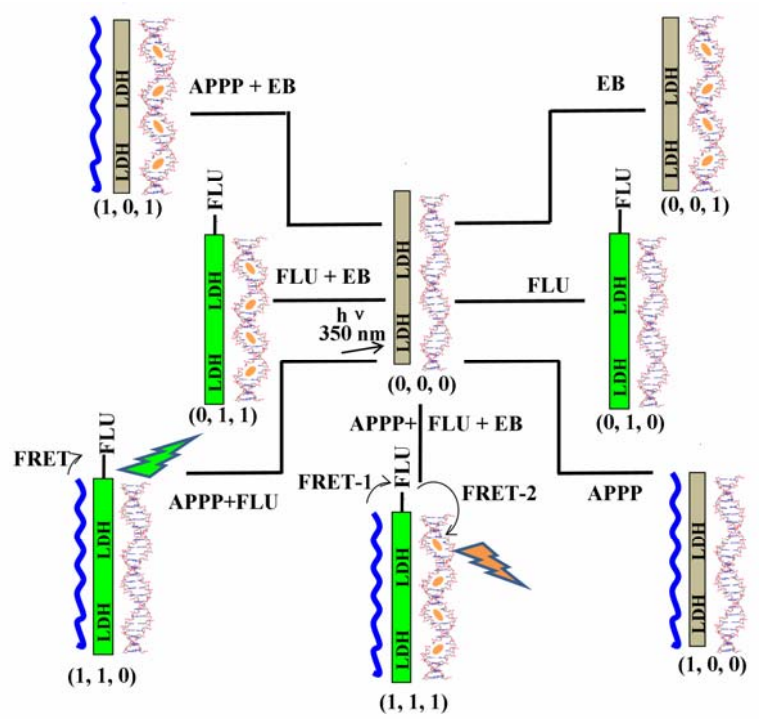

Fig. S7 A schematic representation for the molecular basis of the AND+AND, INH and INHIBIT logic operation.

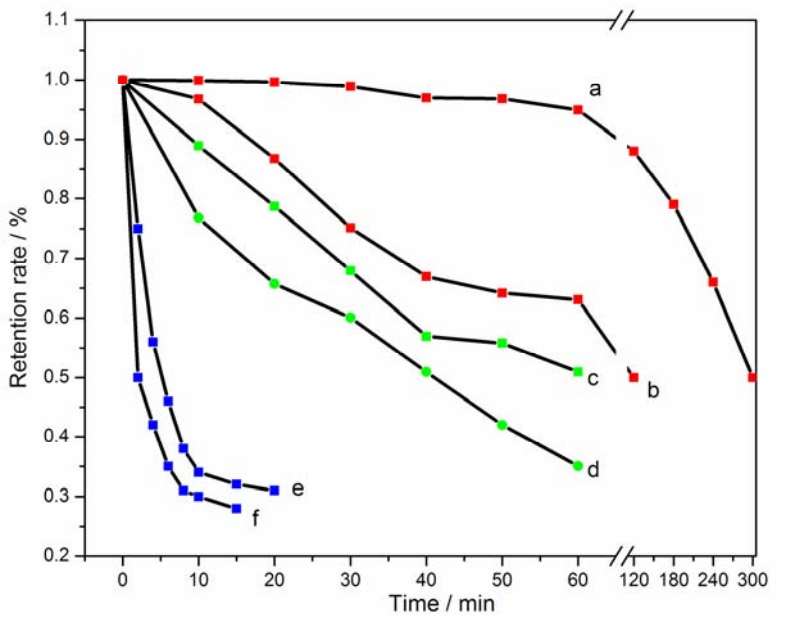

Fig. S8 The retention rate of fluorescence intensity as a function of UV illumination time: (a) the $(LDH\text{-}FLU/DNA)_{20}$ UTF, (b) FLU aqueous solution, (c) the $(LDH/DNA\text{-}EB)_{20}$ UTF, (d) EB aqueous solution, (e) the $(APPP/LDH/DNA)_{20}$ UTF, (f) APPP aqueous solution. Indicated values are the mean of three experiments with the standard error less than 1.5%.

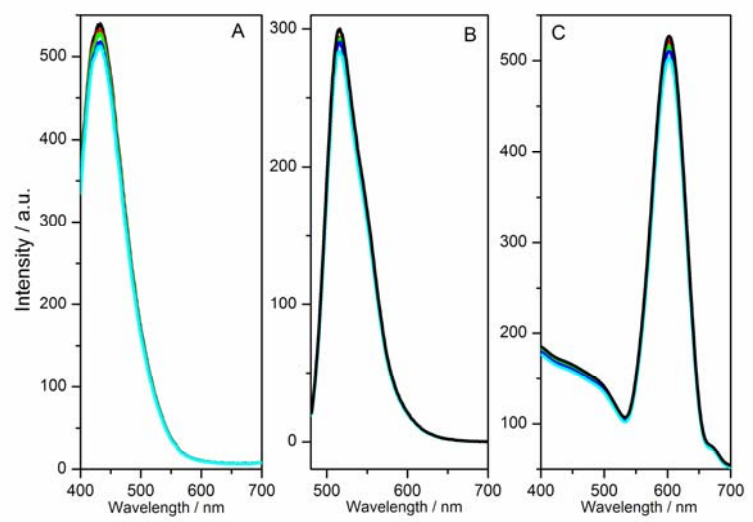

Fig. S9 Fluorescence emission spectra of (A) the $(APPP/LDH/DNA)_{20}$ UTF, (B) the $(LDH\text{-}FLU/DNA)_{20}$ UTF and (C) the $(LDH/DNA\text{-}EB)_{20}$ UTF (stored at 4 °C) recorded weekly in 5 weeks.

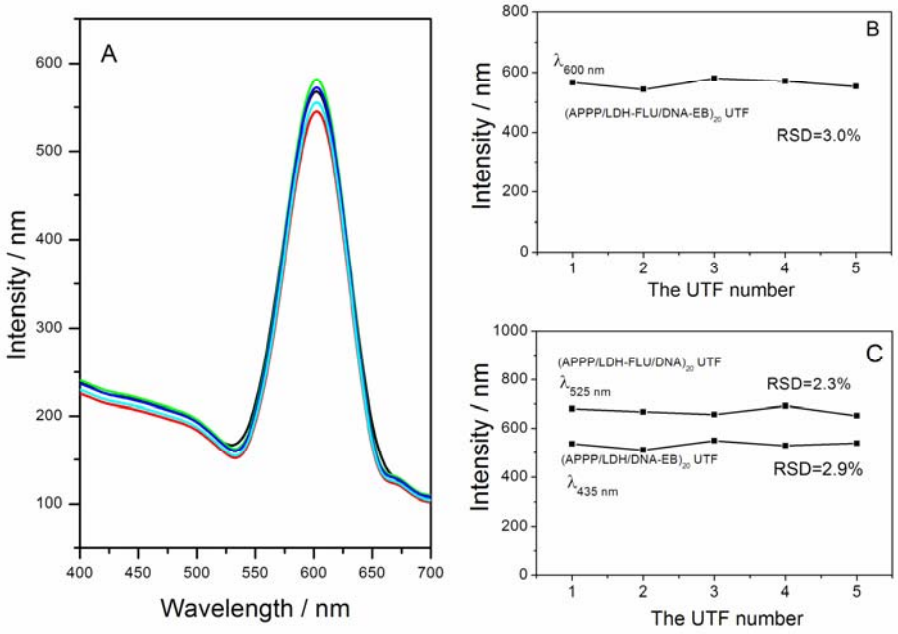

Fig. S10 (A) Fluorescence emission spectra of 5 independent $(APPP/LDH\text{-}FLU/DNA\text{-}EB)_{20}$ UTFs. The reproducibility of (B) the $(APPP/LDH\text{-}FLU/DNA\text{-}EB)_{20}$ UTF, (C) the $(APPP/LDH/DNA\text{-}EB)_{20}$ UTF and $(APPP/LDH\text{-}FLU/DNA)_{20}$ UTF. The fluorescence intensity was measured based on 5 parallel samples.