# Supporting information

## 1. G-quadruplex recognition by the naked eyes



**Figure S1.** G-quadruplex discrimination from duplex, single-stranded and triplex DNAs by the naked eyes. The DNA used in each tube is labelled at the top of the figure. [TMPipEOPP] = 5  $\mu$ M. [DNA] = 10  $\mu$ M (strand concentration).



#### 2. Effects of DNAs on the TMPipEOPP fluorescence spectrum

**Figure S2.** Fluorescence spectra of TMPipEOPP in the absence or presence of different DNAs when the excitation wavelength is held at 421 nm. [TMPipEOPP] = 5  $\mu$ M. [DNA] = 10  $\mu$ M (strand concentration). [BSA] = 10  $\mu$ M. (The excitation and emission silts were set to 5 nm.)



**Figure S3.** Excitation spectra of TMPipEOPP in the absence or presence of different DNAs when the emission wavelength is held at 719 nm. [TMPipEOPP] = 5  $\mu$ M. [DNA] = 10  $\mu$ M (strand concentration). [BSA] = 10  $\mu$ M. (The excitation and emission silts were set to 3 nm.)



**Figure S4.** Fluorescence spectra of TMPipEOPP in the absence or presence of different DNAs when excited at (a) 454 nm or (b) 700 nm. [TMPipEOPP] = 5  $\mu$ M. [DNA] = 10  $\mu$ M (strand concentration). When excited at 454 nm, the excitation and emission silts were set to 5 nm. When excited at 700 nm, the excitation and emission silts were set to 3 nm.



**Figure S5.** Effects of Hum24 or Hum59 on the fluorescence spectrum of TMPipEOPP in the 20  $\mu$ M AT or GC when excited at (a,c) 454 nm or (b,d) 700 nm. [TMPipEOPP] = 5  $\mu$ M. When excited at 454 nm, the excitation and emission silts were set to 5 nm. When excited at 700 nm, the excitation and emission silts were set to 3 nm.

#### 3. Absorption titration of TMPipEOPP with monomeric G-quadruplexes



**Figure S6.** Absorption titration of TMPipEOPP with (a) KRAS and (b) C-MYC. The concentrations of the two G-quadruplexes are increased from 0 to 50  $\mu$ M. Arrows indicate the increasing G-quadruplex concentrations.

#### 4. Absorption titration of TMPipEOPP with multimeric G-quadruplexes



**Figure S7.** Absorption titration of TMPipEOPP with (a) Hum48 and (b) Hum54. The concentrations of the two G-quadruplexes are increased from 0 to 25  $\mu$ M. Arrows indicate the increasing G-quadruplex concentrations.

## 5. Absorption titration of TMPipEOPP with duplex DNAs



**Figure S8.** Absorption titration of TMPipEOPP with GC. The concentrations of GC are labelled in the figure.

#### 6. Absorption titration of TMPipEOPP with single-stranded DNAs



**Figure S9.** Absorption titration of TMPipEOPP with ssDNA2, Hum24-c and C-MYC-c. The concentrations of the three DNAs are labelled in the figure.

## 7. Absorption titration of TMPipEOPP with triplex T\*AT



**Figure S10.** Absorption titration of TMPipEOPP with triplex T\*AT. The concentrations of T\*AT are labelled in the figure.

## 8. Absorption titration of TMPipEOPP with BSA



**Figure S11.** Absorption titration of TMPipEOPP with BSA. The concentrations of **BSA** are labelled in the figure.

# 9. Absorption titration of TMPipEOPP with Hum24 or Hum59 in the presence of 20 $\mu M$ AT or GC



Figure S12. Absorption titration of TMPipEOPP with Hum24 or Hum59 in the presence of 20  $\mu$ M AT or GC. The concentrations of Hum24 or Hum59 are labelled in the figure.



Figure S13. Job plot analysis of the interaction between TMPipEOPP and Hum24 utilizing the absorption signals at (a) 421 nm, (b) 454 nm, (c) 695 nm and (d) the fluorescence signals at 719 nm ( $\lambda_{ex} = 700$  nm), respectively. [TMPipEOPP] + [Hum24] = 5  $\mu$ M.



Figure S14. Job plot analysis of the interaction between TMPipEOPP and KRAS utilizing the absorption signals at (a) 421 nm, (b) 454 nm, (c) 695 nm and (d) the fluorescence signals at 719 nm ( $\lambda_{ex} = 700$  nm), respectively. [TMPipEOPP] + [KRAS] = 5  $\mu$ M.



Figure S15. Job plot analysis of the interaction between TMPipEOPP and C-MYC utilizing the absorption signals at (a) 421 nm, (b) 454 nm, (c) 695 nm and (d) the fluorescence signals at 719 nm ( $\lambda_{ex} = 700$  nm), respectively. [TMPipEOPP] + [C-MYC] = 5  $\mu$ M.



Figure S16. Job plot analysis of the interaction between TMPipEOPP and Hum48 utilizing the absorption signals at (a) 421 nm, (b) 454 nm, (c) 695 nm and (d) the fluorescence signals at 719 nm ( $\lambda_{ex} = 700$  nm), respectively. [TMPipEOPP] + [Hum48] = 5  $\mu$ M.



Figure S17. Job plot analysis of the interaction between TMPipEOPP and Hum54 utilizing the absorption signals at (a) 421 nm, (b) 454 nm, (c) 695 nm and (d) the fluorescence signals at 719 nm ( $\lambda_{ex} = 700$  nm), respectively. [TMPipEOPP] + [Hum54] = 5  $\mu$ M.



Figure S18. Job plot analysis of the interaction between TMPipEOPP and Hum59 utilizing the absorption signals at (a) 421 nm, (b) 454 nm, (c) 695 nm and (d) the fluorescence signals at 719 nm ( $\lambda_{ex} = 700$  nm), respectively. [TMPipEOPP] + [Hum59] = 5  $\mu$ M.

#### 11. Scatchard plots for TMPipEOPP with G-quadruplexes



**Figure S19.** Scatchard plots for TMPipEOPP with Hum24 utilizing the absorption signal at (a) 454 nm and (b) 695 nm. The *n* and  $K_a$  values obtained by linear curve fitting are shown in the figures.



**Figure S20.** Scatchard plots for TMPipEOPP with KRAS utilizing the absorption signal at (a) 454 nm and (b) 695 nm. The *n* and  $K_a$  values obtained by linear curve fitting are shown in the figures.



**Figure S21.** Scatchard plots for TMPipEOPP with C-MYC utilizing the absorption signal at (a) 454 nm and (b) 695 nm. The *n* and  $K_a$  values obtained by linear curve fitting are shown in the figures.



**Figure S22.** Scatchard plots for TMPipEOPP with Hum59 utilizing the absorption signal at (a) 454 nm and (b) 695 nm. The *n* and  $K_a$  values obtained by linear curve fitting are shown in the figures.



#### 11. Stabilization of G-quadruplexes by TMPipEOPP

**Figure 23.** Melting temperature  $(T_{1/2})$  detection of G-quadruplexes in the absence (black) and presence (red) of 5  $\mu$ M TMPipEOPP under molecular crowding conditions. [DNA] = 2.5  $\mu$ M (strand concentration). [K<sup>+</sup>] = 150 mM. Scatter: experimental data, line: fitting curves.