

Supplementary materials

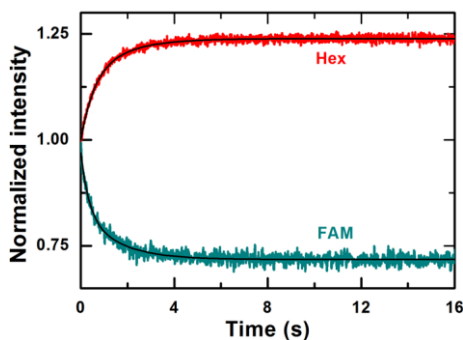


Figure S1 The FRET effect caused by G-quadruplex folding

Fluorescence intensities of FAM (donor) at 525 nm and Hex (acceptor) at 595 nm verse time during the G-quadruplex folding process in 100 mM NaCl buffer at room temperature (23 °C). The two measurements were performed separately (i.e., not simultaneously), because we had to change the optical filters. The solid lines are double-exponential fits of the data, yielding $A_{\text{fast}} = 0.205 \pm 0.003$, $A_{\text{slow}} = 0.059 \pm 0.002$, $k_{\text{fast}} = 1.63 \pm 0.02 \text{ s}^{-1}$, and $k_{\text{slow}} = 0.26 \pm 0.01 \text{ s}^{-1}$ for FAM; and $A_{\text{fast}} = -0.193 \pm 0.003$, $A_{\text{slow}} = -0.060 \pm 0.001$, $k_{\text{fast}} = 1.52 \pm 0.02 \text{ s}^{-1}$, and $k_{\text{slow}} = 0.22 \pm 0.01 \text{ s}^{-1}$ for Hex.

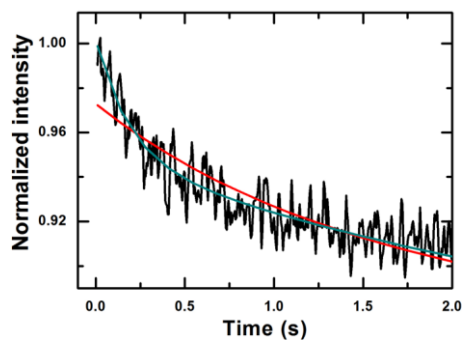


Figure S2 Double- or triple-exponential fittings of the transition kinetics of G-quadruplex structures from Na⁺ form to K⁺ form

The black line is the time-course (only the initial 2 s was shown) of the fluorescence intensity of FAM after the basket-type G-quadruplex preformed in 100 mM NaCl was rapidly mixed with 100 mM KCl at room temperature (23 °C). The red line represents a double-exponential fitting, yielding $A_{\text{fast}} = 0.066 \pm 0.001$, $A_{\text{slow}} = 0.128 \pm 0.001$, $k_{\text{fast}} = 1.90 \pm 0.04 \text{ s}^{-1}$, and $k_{\text{slow}} = 0.12 \pm 0.01 \text{ s}^{-1}$. The blue line represents a triple-exponential fitting, with $A_{\text{fast}} = 0.061 \pm 0.004$, $A_{\text{slow1}} = 0.042 \pm 0.002$, $A_{\text{slow2}} = 0.117 \pm 0.002$, $k_{\text{fast}} = 6.97 \pm 0.08 \text{ s}^{-1}$, $k_{\text{slow1}} = 0.62 \pm 0.04 \text{ s}^{-1}$, and $k_{\text{slow2}} = 0.11 \pm 0.01 \text{ s}^{-1}$.

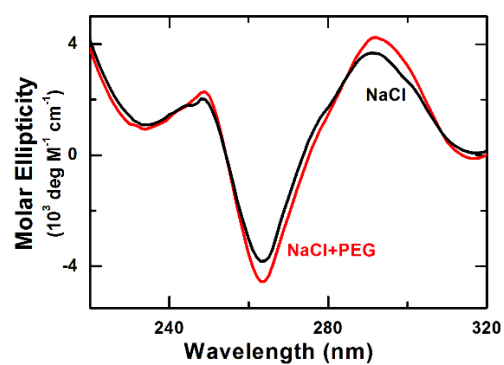


Figure S3 Effect of PEG200 on Na⁺-induced G-quadruplex folding

CD spectra of the G-quadruplex sequence in 100 mM NaCl buffer and 100 mM NaCl buffer with 40% PEG200 at room temperature (23 °C).

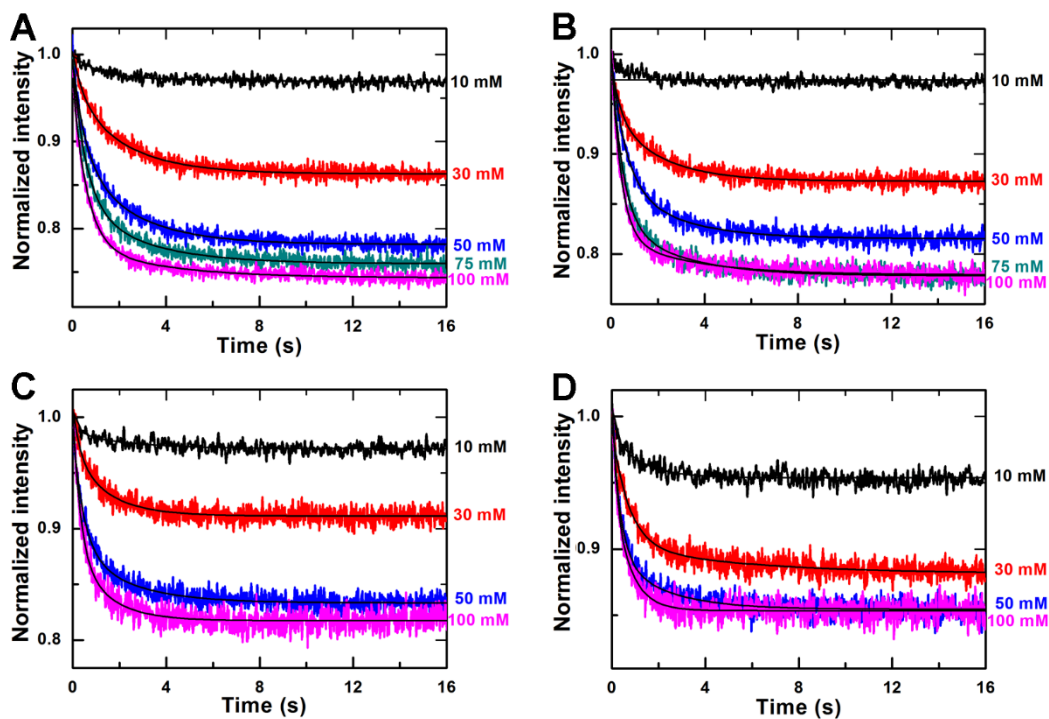


Figure S4 Effect of Li^+ on Na^+ -induced G-quadruplex folding

Time-courses of the fluorescence intensity of FAM after the unfolded G-quadruplex DNA was rapidly mixed with different concentrations of NaCl and (A) 30 mM, (B) 50 mM, (C) 75 mM and (D) 100 mM LiCl at room temperature (23 °C). The solid lines are double-exponential fits of the data, with some fitting parameters given in Figures 4B and 4C.

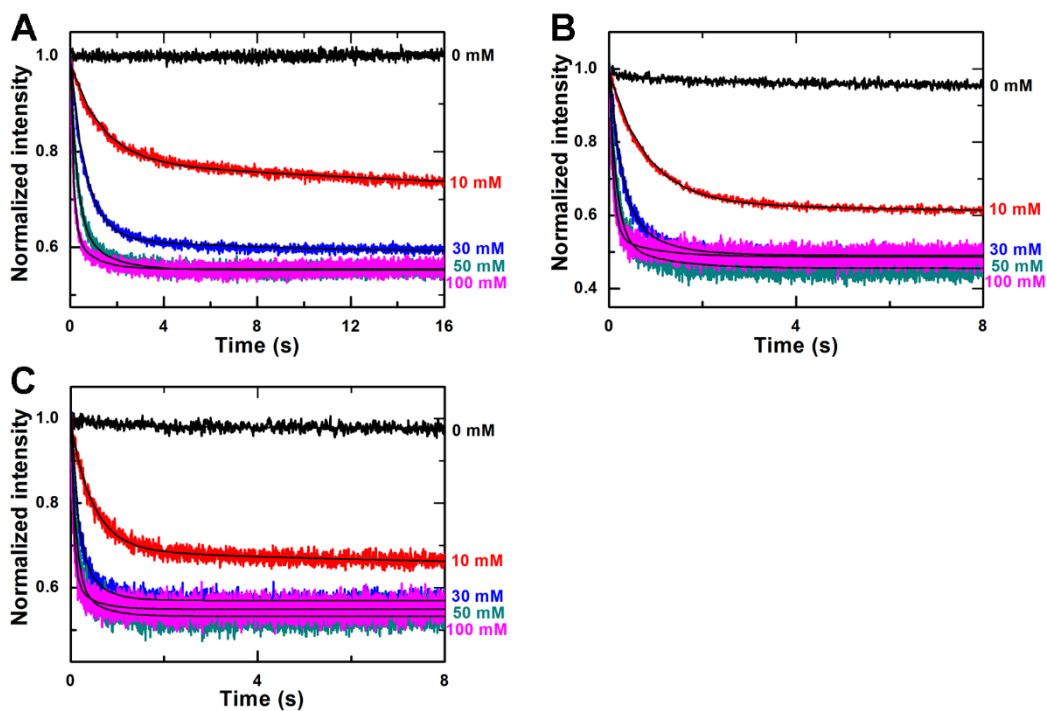


Figure S5 Effect of Li^+ on K^+ -induced G-quadruplex folding

Time-courses of the fluorescence intensity of FAM after the unfolded G-quadruplex DNA was rapidly mixed with different concentrations of KCl and (A) 30 mM, (B) 50 mM, and (C) 100 mM LiCl at room temperature (23 °C). The solid lines are double-exponential fits of the data, with some fitting parameters given in Figures 5C and 5D.

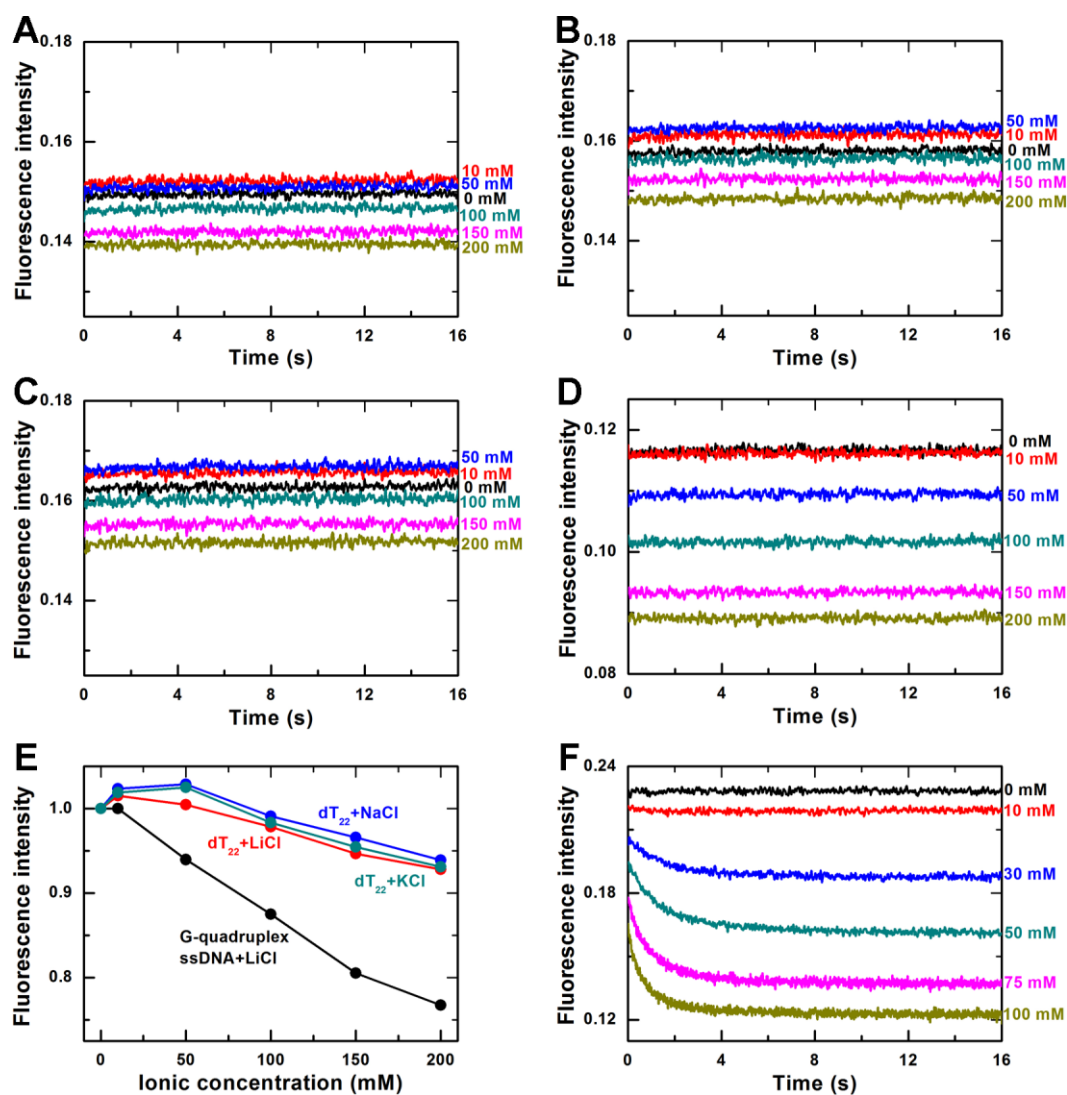


Figure S6 Control experiments with dT₂₂ and G-quadruplex ssDNA in different ionic environments, and one set of the original kinetic data curves

Time-courses of the fluorescence intensity of FAM after the ssDNA rapidly mixed with different cations **at room temperature (23 °C)**. (A) dT₂₂ with LiCl; (B) dT₂₂ with NaCl; (C) dT₂₂ with KCl; (D) G-quadruplex ssDNA with LiCl; (E) Fluorescence intensity versus the ionic concentration, where each data point was obtained by averaging all the data points in each of the kinetic curves shown above, and then normalized; (F) The original kinetic data curves corresponding to Figure 4A.