Supporting Materials and Methods

Analysis of time profiles based on the multistep hopping mechanism was performed with numerical analysis by using MATLAB software. A kinetic model of the multistep hole-transfer process is shown in Scheme 1.

Charge recombination process (k_{cr}) can be ignored because the charge-separated state persisted over several hundred microseconds when naphthalimide (NI) and the nearest G are separated by six A bases. According to Scheme 1, simultaneous differential equations are shown as follows:

$$\begin{aligned}
\frac{d[\mathrm{G}_1]}{dt} &= -k[\mathrm{G}_1] + k[\mathrm{G}_2] \\
\frac{d[\mathrm{G}_2]}{dt} &= k[\mathrm{G}_1] - 2k[\mathrm{G}_2] + k[\mathrm{G}_3] \\
&\vdots \\
\frac{d[\mathrm{G}_\mathrm{n}]}{dt} &= k[\mathrm{G}_{\mathrm{n}-1}] - (k + k_1)[\mathrm{G}_\mathrm{n}] \\
\frac{d[\mathrm{PTZ}]}{dt} &= k[\mathrm{G}_\mathrm{n}]
\end{aligned} \qquad [2]$$

where [G_i (i = 1…n)] corresponds to the hole population at each G-site, k is the hole-transfer rate constant between Gs, and k_1 is hole transfer from $G_n^{\bullet+}$ to PTZ. Fitting results for 5'-$(GA)_n$ and $(GT)_n$ (n = 12) according to Eq. **2** are presented in Fig. 8, providing the rate constants of $k = 4 \times 10^7\ s^{-1}$ and $k_1 = 6 \times 10^7\ s^{-1}$ for $(GA)_{12}$ and $k = 6.5 \times 10^5\ s^{-1}$ and $k_1 = 5 \times 10^6\ s^{-1}$ for $(GT)_{12}$, respectively. The obtained values of k for $(GA)_{12}$ and $(GT)_{12}$ were similar with the values obtained for $(GT)_2$ and $(GA)_2$ with single-exponential analysis according to Eq. **1** in the main text, showing the validity of approximate analysis performed with single-exponential fitting. Difference in the k_1 values between $(GA)_{12}$ and $(GT)_{12}$ is attributed to the difference in the nucleobase that is stacked to PTZ.