

Supplementary Information

Supramolecular Multiplexes from Collagen Mimetic Peptide-PNA(GGG)₃ Conjugates and C-rich DNA: pH-induced reversible switching from triplex-duplex to triplex-i-motif

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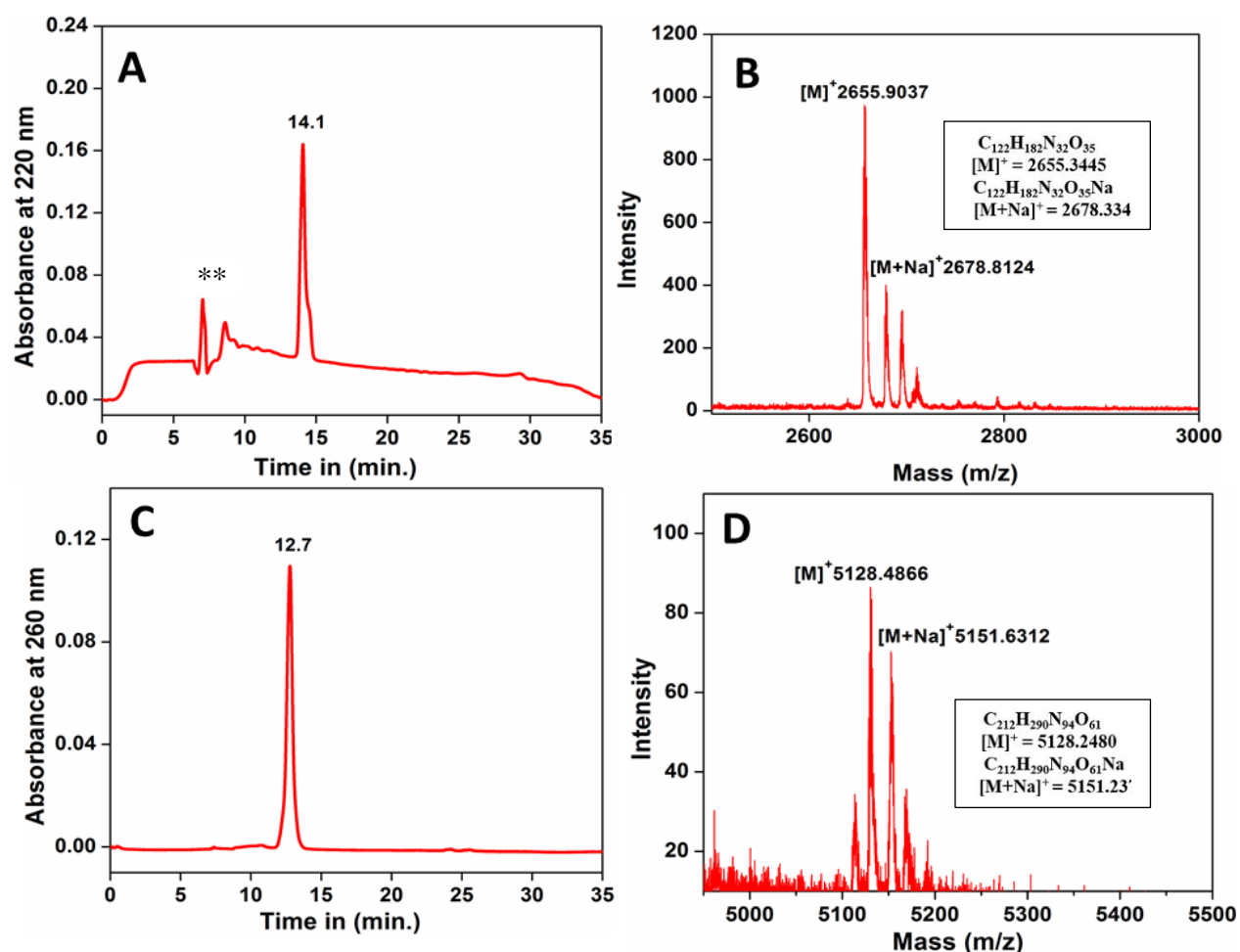
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Table of Contents

S1.	HPLC and MALDI-TOF spectra of Collagen-(PNA-GGG) ₃ peptide CMP-3 & CMP-4	2
S2.	CD spectra and melting of collagen-(PNA-GGG) ₃ peptide (CMP-3) at different salt concentrations	3
S3.	CD spectra of Collagen-(PNA-GGG) ₃ (CMP-3) with DNA 1 in 0.33 and 0.66 equivalents	3
	S3a: CD spectra of Collagen CMP-3 single strand (5 μM) + DNA 1	4
S4.	CD spectra of Collagen-(PNA-GGG) ₃ (CMP-3) with mismatch DNA - T ₉	4
S5.	CD Melting of triplex in CMP-3:DNA 1 complex monitored at A ₂₂₅ nm (pH 7.2)	5
	S5a: Raw CD Data for S5A	5
S6.	CD melting of <i>i</i> -motif of DNA 1 at A ₂₈₉ nm at pH 4.5	6
	S6a: Raw CD Data for S6	6
S7.	CD Melting of triplex in Collagen-(PNA-GGG) ₃ :DNA 1 complex monitored at A ₂₂₅ nm (pH 4.5) at different ratios	7
S8.	CD Melting of <i>i</i> -motif in Collagen-(PNA-GGG) ₃ :DNA 1 complex monitored at A ₂₈₉ nm (pH 4.5) at different ratios	7
	S8a: Raw CD Graphs for Figures S7 and S8	8
S9	Collagen-(PNA-GGG) ₃ CMP-3:DNA 1 gel electrophoresis at pH 4.5	8
S10	TEM images of CMP-3 (triplex) +DNA1 at pH 7.2	9

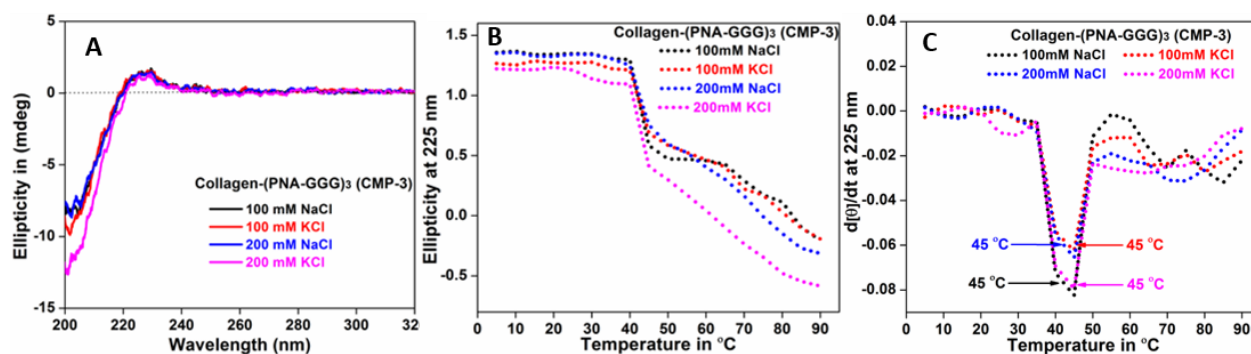
	S10a Collagen CMP-3 (Single strand) +DNA 1 complex	9
	S10b PNA-GGG (15 μM) +DNA 1 complex	10
	S10c PNA-GGG (30 μM) +DNA 1 complex	10
S11	HPLC and MALDI-TOF spectra of PNA-(G) ₃	10
S12	ThT Emission Spectra for PNA-(G) ₃ , and PNA-(G) ₃ +DNA1	11
S13	Reversible switching of ThT Emission Spectra for the Collagen-(PNA-GGG) ₃ Peptide-3:DNA1 between pH 7.2 & pH 4.5	11

S1. HPLC and MALDI-TOF spectra of Collagen-(PNA-GGG)₃ peptide CMP-3 & CMP-4



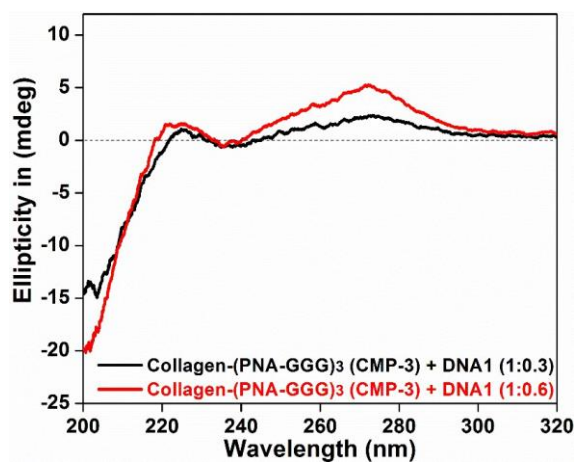
HPLC profiles (A, C) and MALDI-TOF spectra (B, D) for Collagen-(PNA-GGG)₃ peptide (CMP-3) and CMP-4 respectively. Inset in (B) and (D) are molecular formulae and calculated mass;
 * Impurity from solvent.

S2. CD spectra and melting of collagen-(PNA-GGG)₃ peptide (CMP-3) at different salt concentrations



(A) CD spectra of collagen-(PNA-GGG)₃ (CMP-3) at 100 mM and 200 mM salt concentrations; (B) CD Melting (ellipticity vs temperature) monitored at λ_{225} nm in different salt concentrations (100, 200 mM; NaCl & KCl) (C) First derivative plots for data in (B). Temperature range is 5 °C to 90 °C; Buffer conditions: 10 mM Tris with 100 or 200 mM NaCl or KCl, pH 7.2.

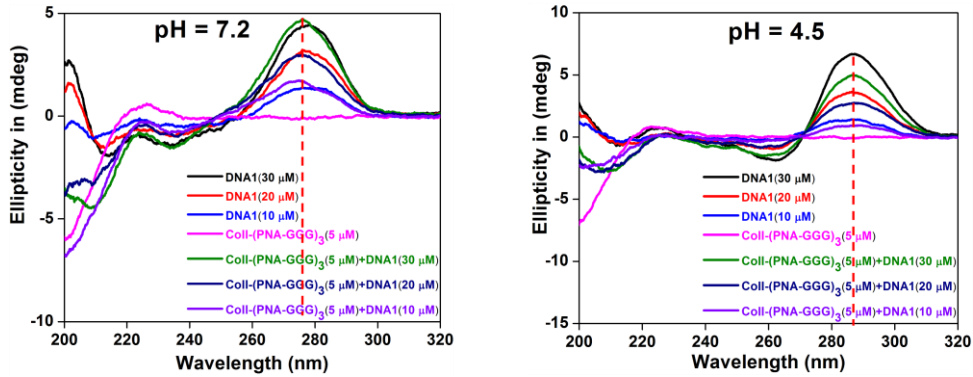
S3. CD spectra of Collagen-(PNA-GGG)₃ (CMP-3) with DNA 1 in 0.33 and 0.66 equivalents



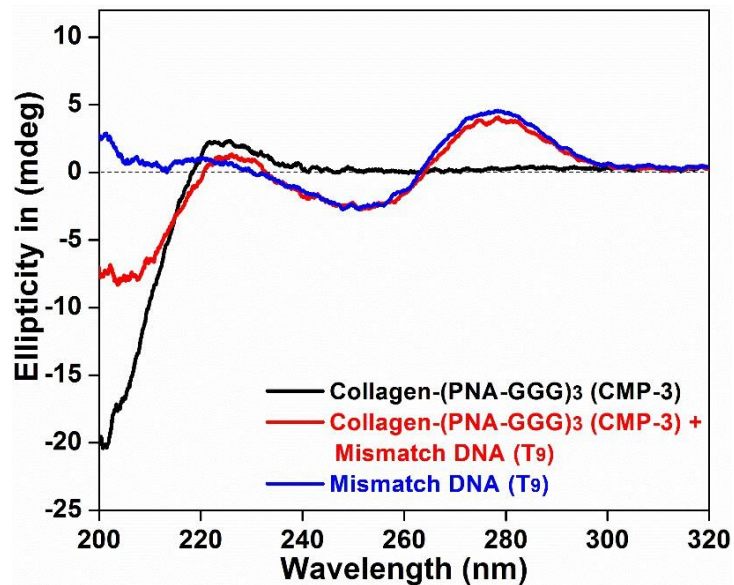
CD spectra of collagen-(PNA-GGG)₃ (CMP-3) with DNA 1 in 0.33 (black) and 0.66 (red) equivalents. Buffer 10 mM Tris, 10 mM NaCl, pH 7.2.

S3a: CD spectra of Collagen CMP-3 single strand (5 μM) + DNA 1

Collagen-(PNA-GGG)₃ 5 μM + DNA 1

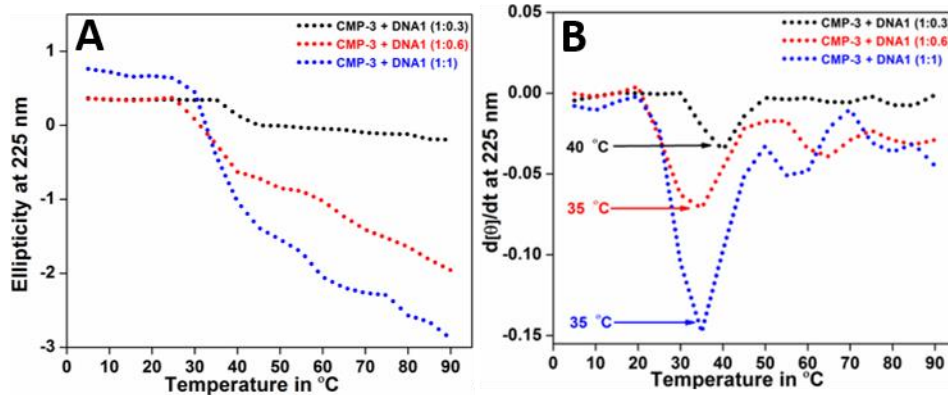


S4. CD spectra of Collagen-(PNA-GGG)₃ (CMP-3) with mismatch DNA - T₉



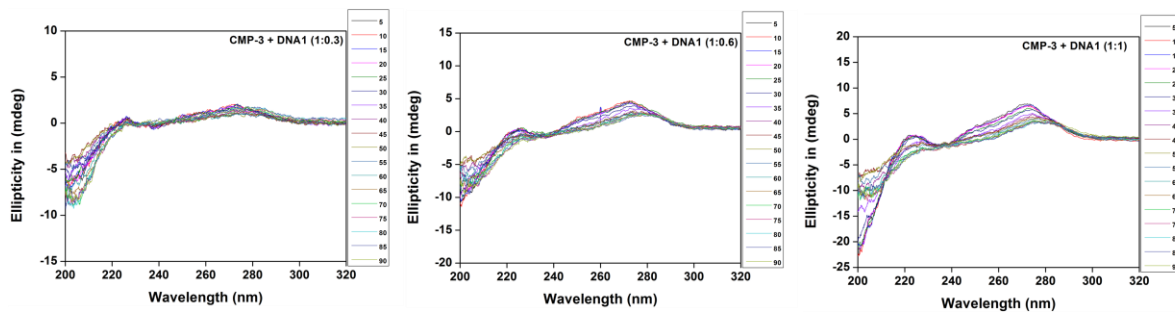
CD spectra of collagen-(PNA-GGG)₃ (CMP-3) with complete mismatch-DNA-T₉. Buffer conditions: 10 mM Tris, 10 mM NaCl, pH 7.2.

S5. CD Melting of triplex in CMP-3:DNA 1 complex monitored at $\lambda = 225$ nm (pH 7.2)



(A) Ellipticity at λ_{225} nm vs temperature plot for collagen-(PNA-GGG)₃ (CMP-3) with 0.33, 0.66 and 1.0 equivalents of DNA 1 (B) First derivative plots of data in (A). Buffer conditions: 10 mM Tris, 10 mM NaCl at pH 7.2.

S5a: Raw CD Data for S5A



S6. CD melting of *i*-motif of DNA1 at λ_{289} nm at pH 4.5

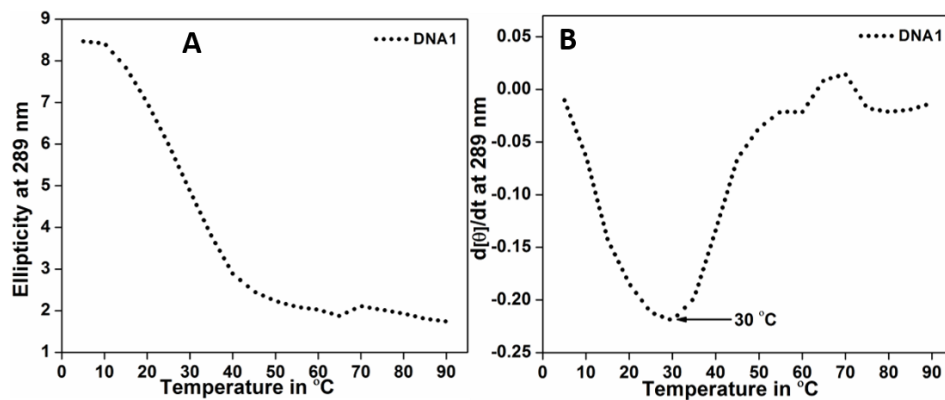
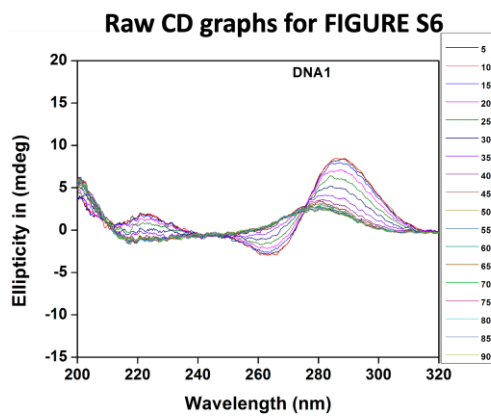
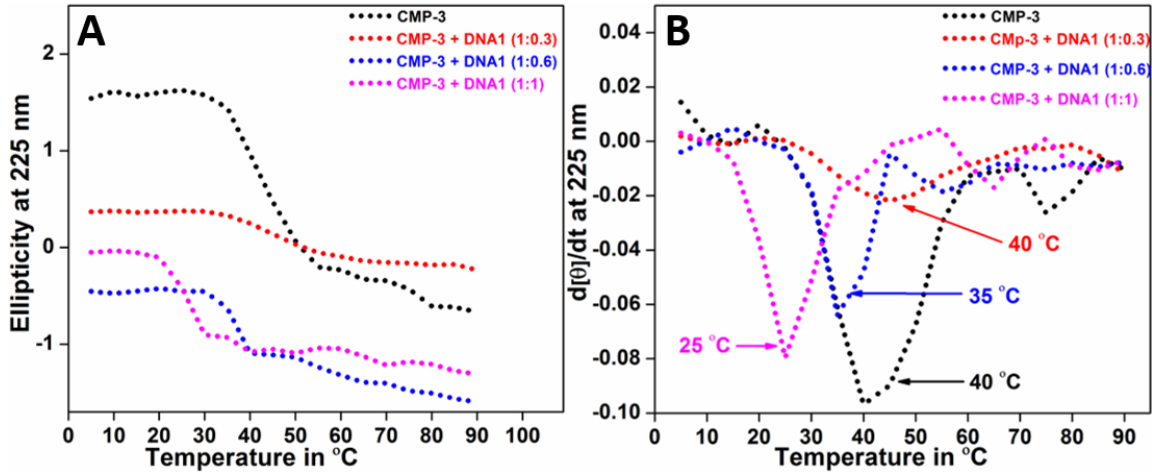


Figure S6: (A) Ellipticity vs temperature plot at λ_{289} nm for DNA 1 (30 μ M); (B) Derivative plots for data in (A). Buffer: 10 mM Tris, 10 mM NaCl at pH 4.5.

S6a: Raw CD Data for S6A

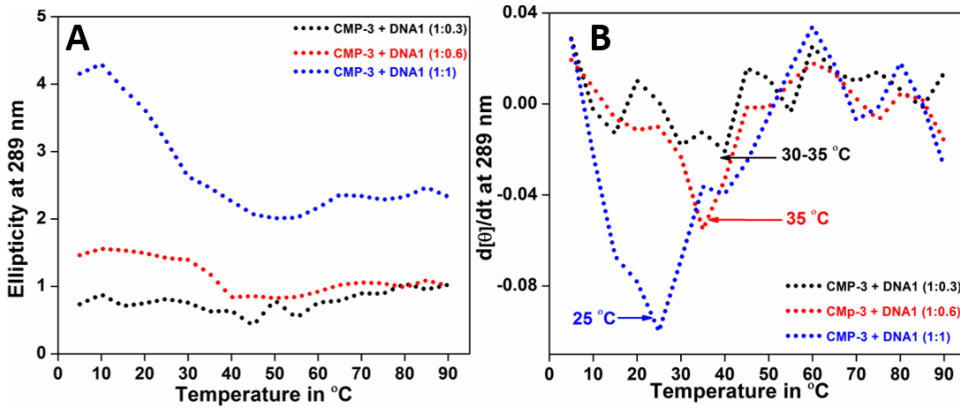


S7. CD Melting of triplex in Collagen-(PNA-GGG)₃:DNA 1 complex monitored at A₂₂₅ nm (pH 4.5) at different ratios



(A) Ellipticity vs temperature plot θ_{225} nm for collagen-(PNA-GGG)₃ (CMP-3) with DNA 1 in 0.33, 0.66 and 1 equivalents (B) Derivative plots for collagen-(PNA-GGG)₃ (CMP-3)+DNA 1 complexes. Buffer conditions: 10 mM Tris, 10 mM NaCl at pH 4.5.

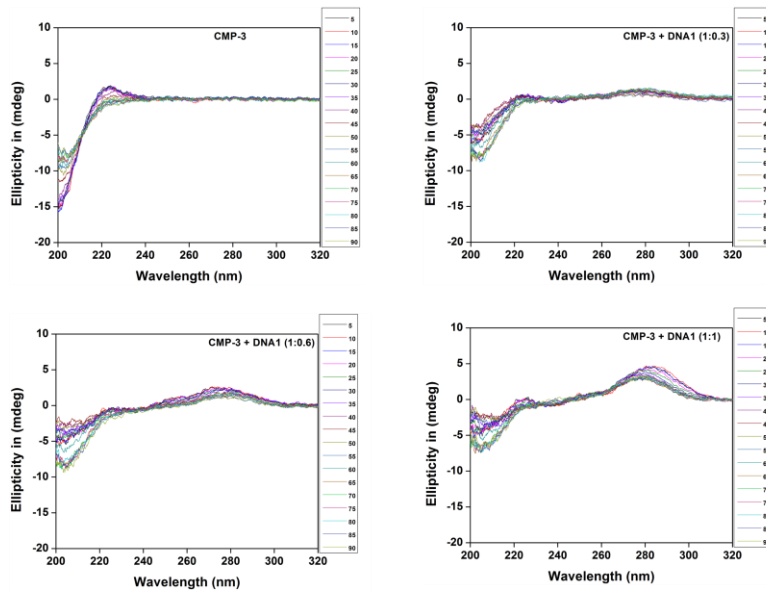
S8. CD Melting of *i*-motif in Collagen-(PNA-GGG)₃:DNA 1 complex monitored at A₂₈₉ nm (pH 4.5) at different ratios



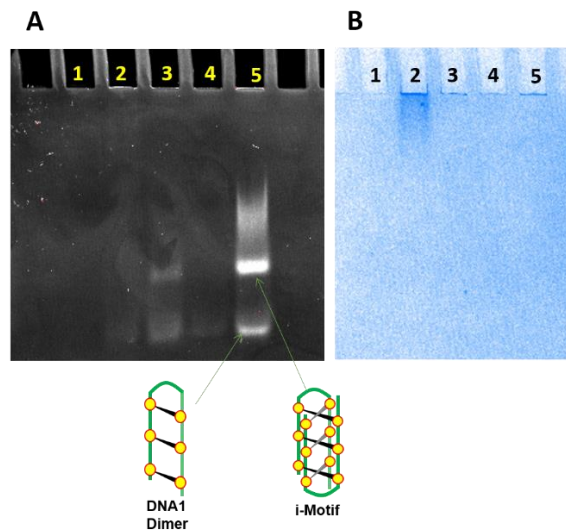
i-motif melting: (A) Ellipticity fall at 289 nm for collagen-(PNA-GGG)₃ (CMP-3) with DNA1 in 0.33, 0.66 and 1 equivalents (B) Derivative plots for collagen-(PNA-GGG)₃ (CMP-3) + DNA1 complexes. Buffer conditions: 10 mM Tris, 10 mM NaCl at pH 4.5.

S8a: Raw CD Graphs for Figures S7 and S8

CD graphs for FIGURE S7 & S8

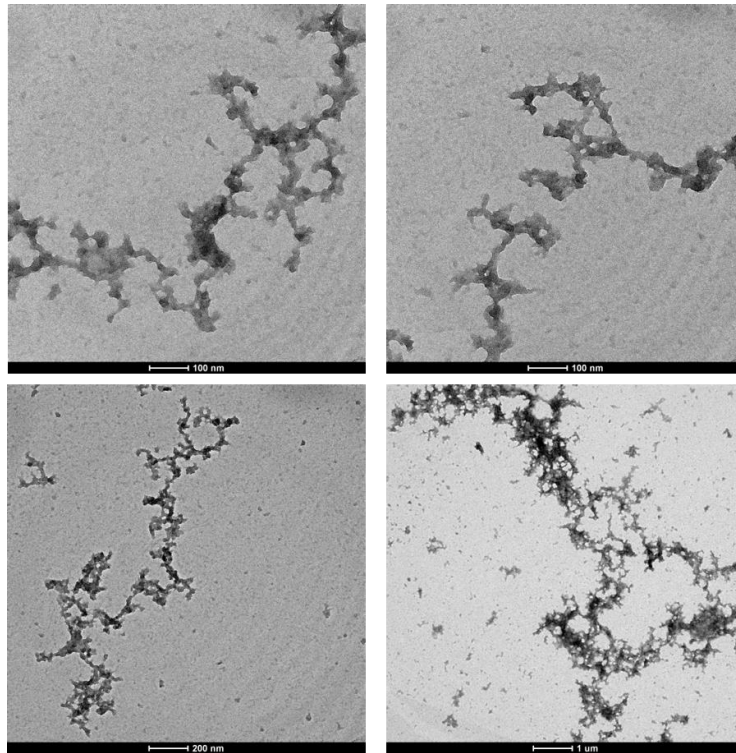


S9. Gel electrophoresis of Collagen-(PNA-GGG)₃ CMP 3:DNA 1 complex at pH 4.5



PAGE: (A) Staining with EB and (B) Staining with Coomassie brilliant blue. Lane 1; Collagen-(PNA-GGG)₃ (CMP-3), lanes 2-4; Collagen-(PNA-GGG)₃ (CMP-3):DNA 1 in 1:0.33, 1:0.6 and 1:1 ratios respectively, lane 5: Single stranded DNA 1 (10 μM). Peptide concentration in lanes 1-4 is 100 μM with equivalent DNA 1 concentration in respective ratios. Lane 5, ssDNA is 10 μM. Buffer: 40 mM Tris-acetate (pH = 4.5) with EDTA (1 mM).

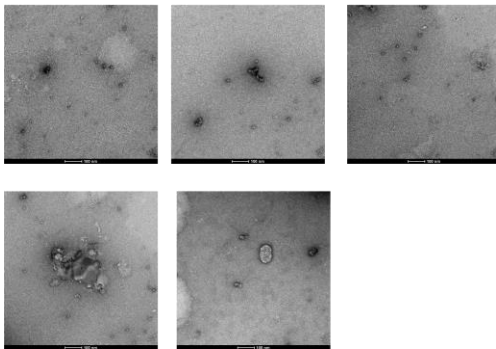
S10. TEM images of CMP-3 20 μ M (triplex) +DNA 1(20 μ M) at pH 7.2



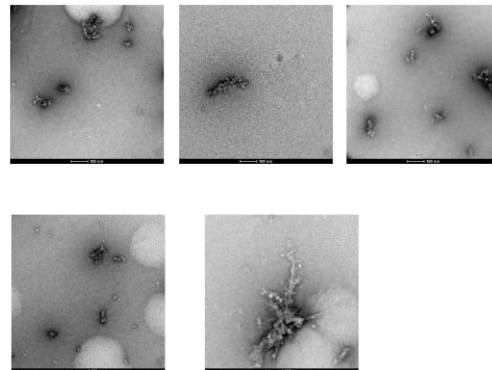
TEM images of CMP-3+DNA1 at pH 7.2.

S10a Collagen CMP-3 (Single strand) +DNA 1 complex

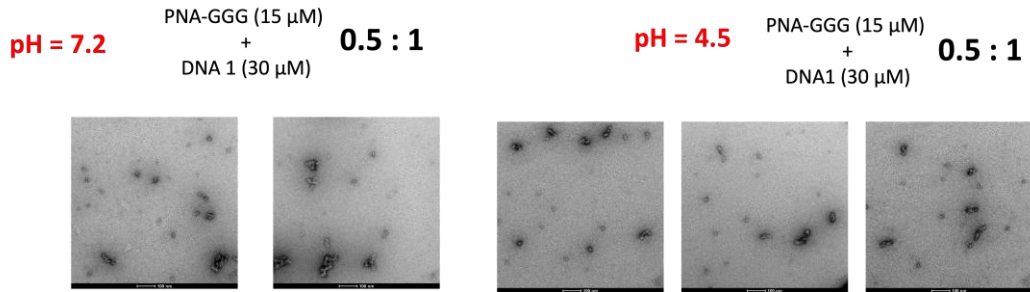
pH = 7.2 Collagen-(PNA-GGG)₃ (2 μ M) Single strand
+
DNA1 (30 μ M)



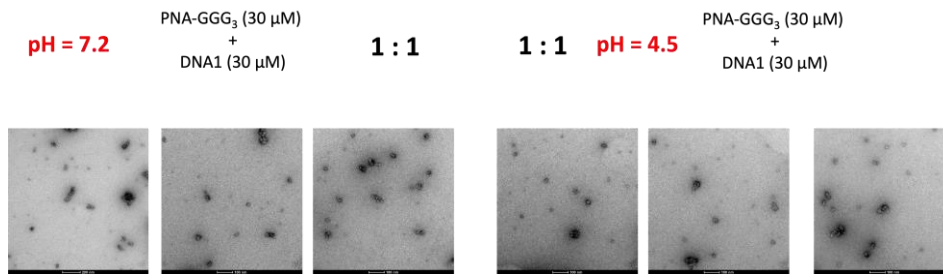
pH = 4.5 Collagen-(PNA-GGG)₃ (2 μ M) Single strand
+
DNA1 (30 μ M)



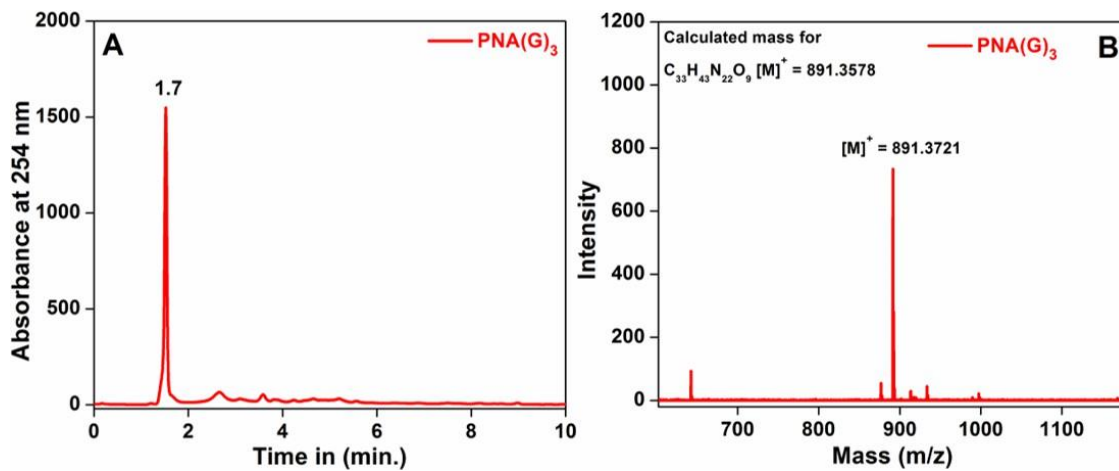
S10b PNA-GGG (15 μ M) +DNA 1 complex



S10c PNA-GGG (30 μ M) +DNA 1 complex

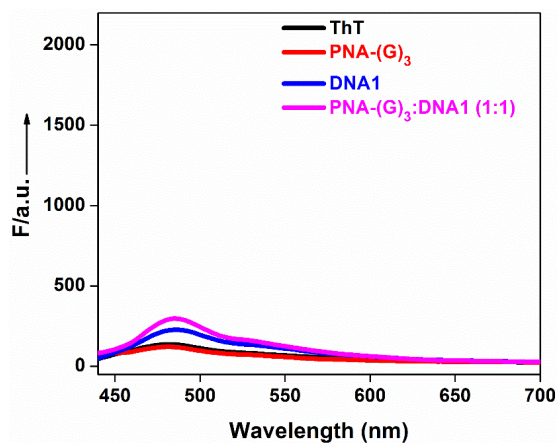


S11. HPLC and MALDI-TOF spectra of PNA-(G)₃



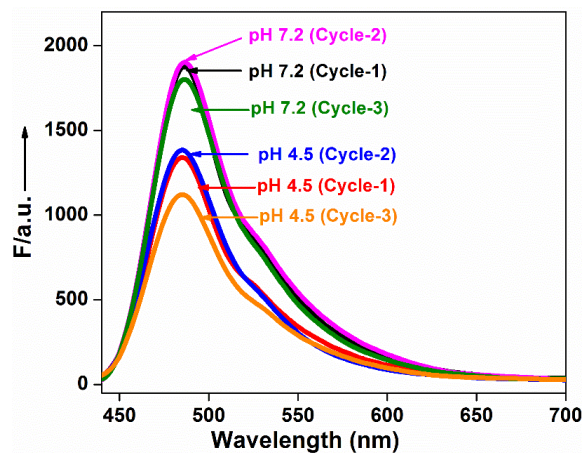
HPLC profiles (A) and MALDI-TOF spectra (B) for PNA-(G)₃.

S12. ThT Emission Spectra for PNA-(G)₃, and PNA-(G)₃+DNA1



Thioflavin ThT emission spectra for PNA-(G)₃, PNA-(G)₃+DNA1 at pH 7.2

S13. Reversible switching of ThT Emission Spectra for the Collagen-(PNA-GGG)₃ Peptide-3+DNA1 system between pH 7.2 & pH 4.5



Reversible Thioflavin ThT emission spectra for collagen-(PNA-GGG)₃ (CMP-3)+DNA1 in between pH 7.2 and pH 4.5. ($\lambda_{ex} = 412$ nm, $\lambda_{em} = 450-700$ nm). Peptide concentration 20 μ M. Buffer conditions: 10 mM Tris, 10 mM NaCl at pH 7.2.