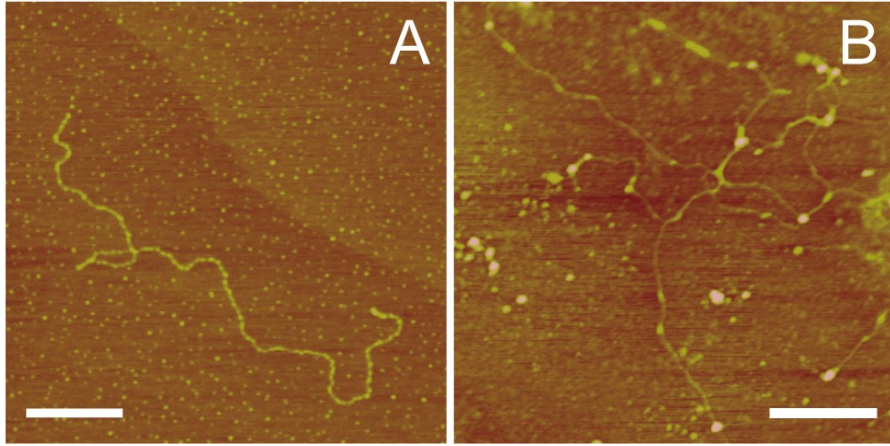


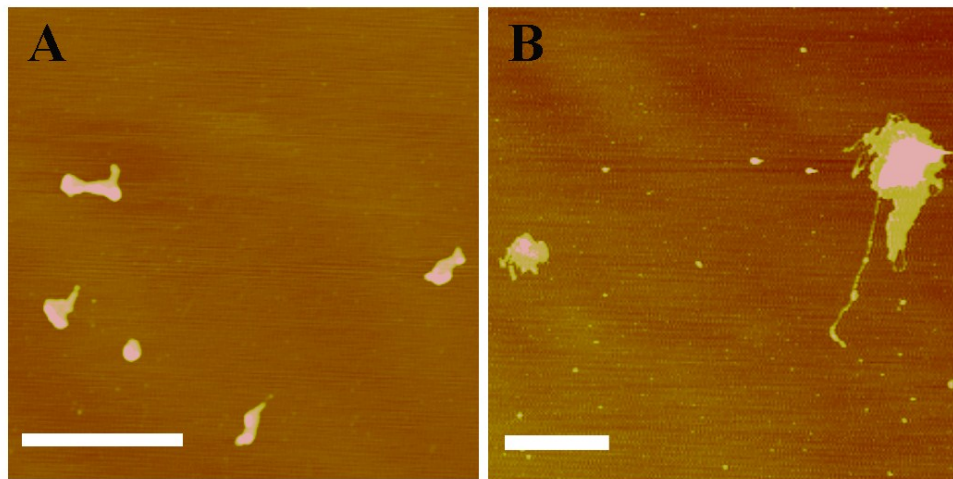
**Peptide Nucleic Acid-Ionic Self-Complementary Peptide  
Conjugates: Highly Efficient DNA Condensers with Specific  
Condensing Mechanism**

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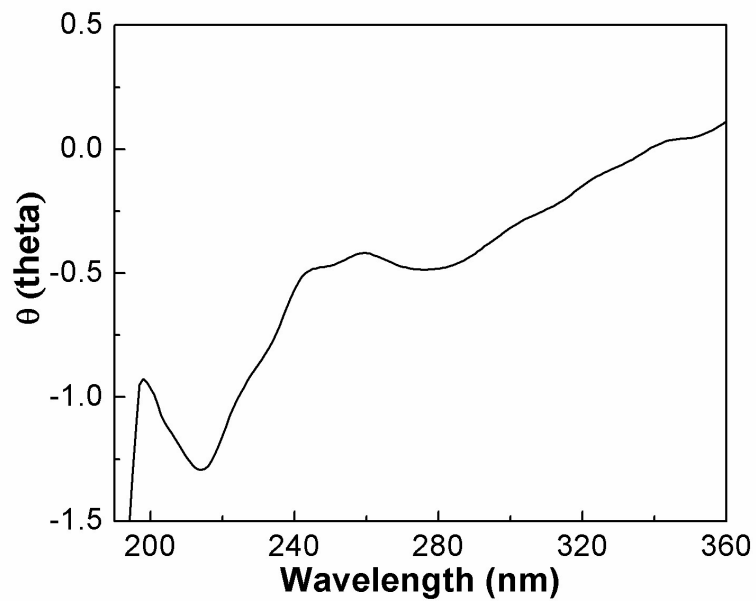
**Figure S1.** AFM height images showing the structures of  $\lambda$ -DNA ( $1.6 \times 10^{-5} \mu\text{M}$ ) in the presence of (A) PNA molecule of T'T'T' ( $1.0 \mu\text{M}$ ) and (B) ISCP molecule of (AKAE)<sub>2</sub> ( $2.0 \mu\text{M}$ ) after mixing for 4 h. Scale bar represents 500 nm for both images. The results showed that the elongated thread-like structure of  $\lambda$ -DNA still remained, suggesting that T'T'T' and ISCP molecules did not induce DNA condensation individually.



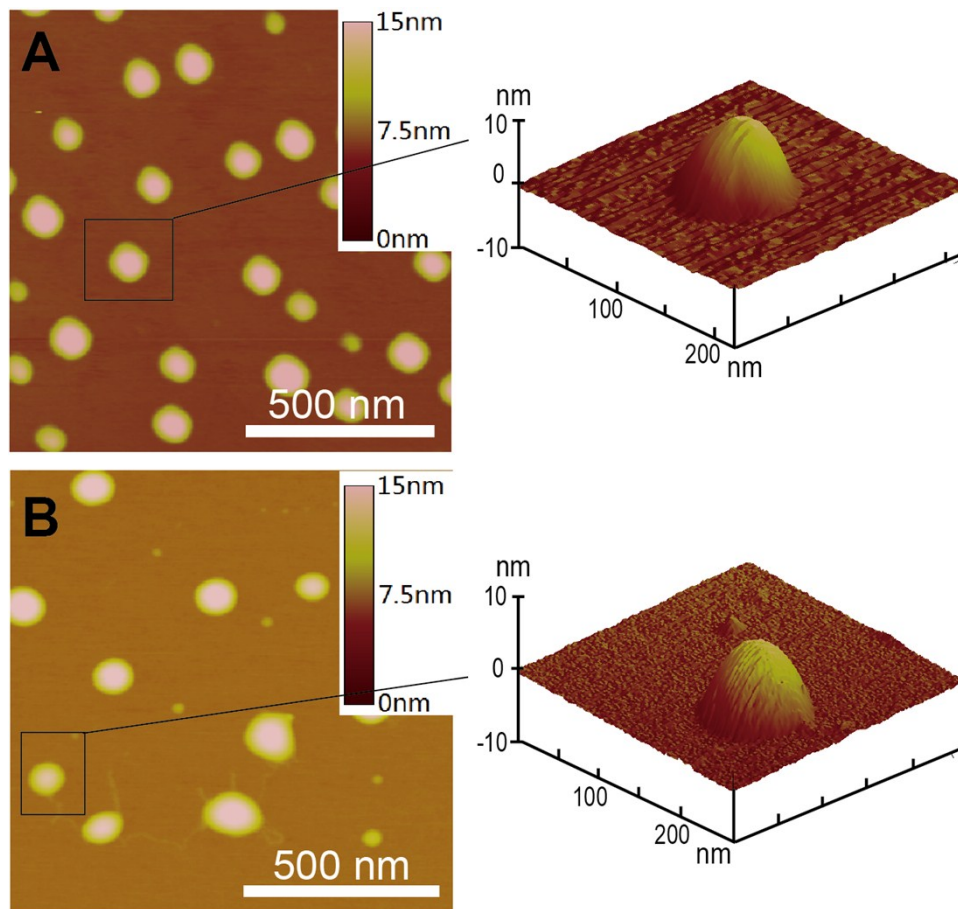
**Figure S2.** AFM height images showing the structures of  $\lambda$ -DNA ( $1.6 \times 10^{-5} \mu\text{M}$ ) in the presence of (A) CTAB and (B) PEI (+/- charge ratio 2:1) after mixing for 4 h. The scale bar represents 1000 nm for all the images.



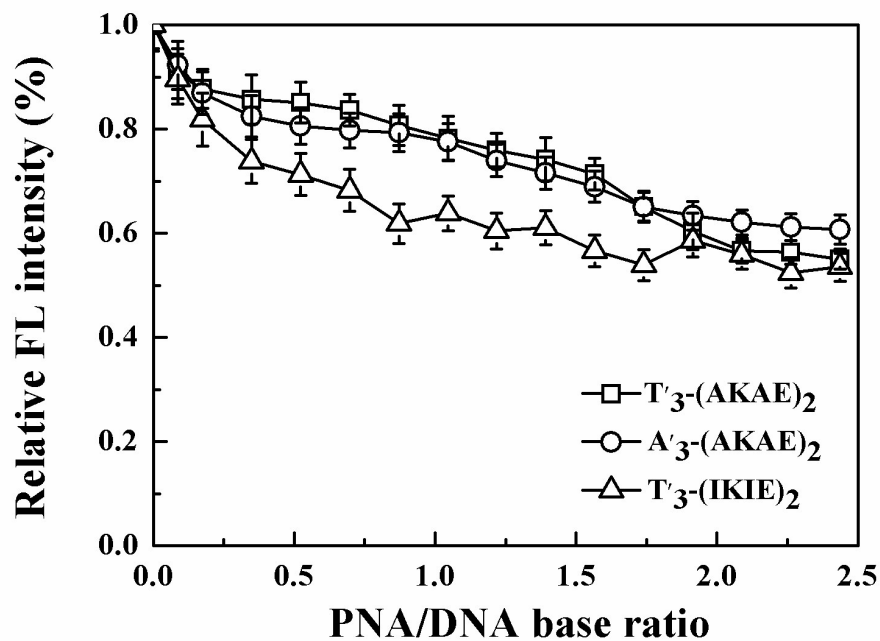
**Figure S3.** Polyacrylamide gel electrophoresis of  $d(A)_{36}$  (the left lane, 200 nM  $d(A)_{36}$ ) and the  $T'_3$ -(AKAE) $_2$ / $d(A)_{36}$  complexes (the right lane, 200 nM  $d(A)_{36}$  + 1.6  $\mu$ M  $T'_3$ -(AKAE) $_2$ ).



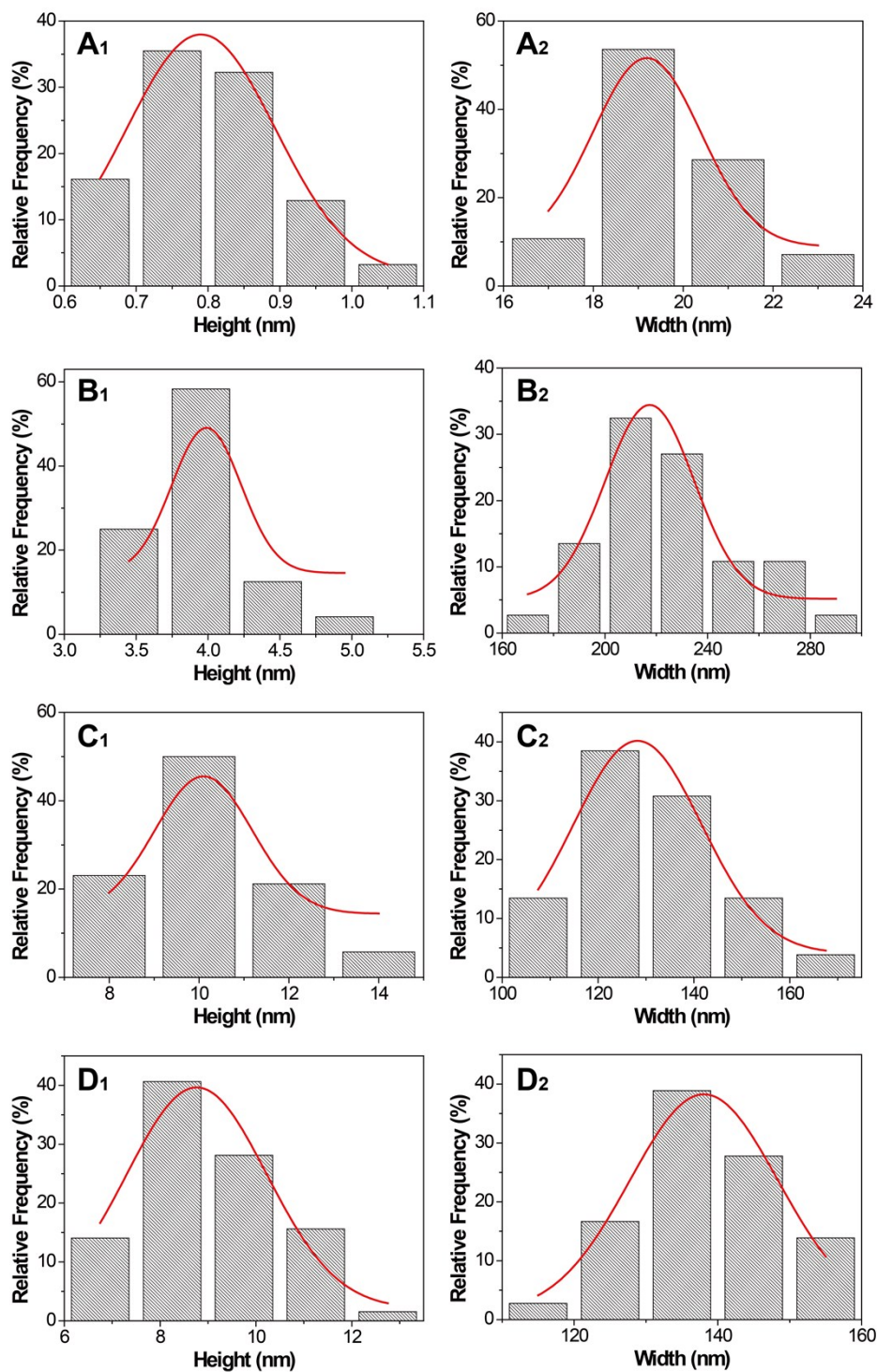
**Figure S4.** CD spectrum of  $T'_3$ -(AKAE) $_2$  (10.0  $\mu$ M).



**Figure S5.** 2D and 3D AFM height images showing the structures of  $\lambda$ -DNA ( $1.6 \times 10^{-5} \mu M$ ) in the presence of (A)  $T'_3-(IKIE)_2$  ( $0.2 \mu M$ ) and (B)  $A'_3-(AKAE)_2$  ( $0.2 \mu M$ ) after mixing for 4 h. The base ratio of PNA-ISC/P/ $\lambda$ -DNA was about 1/2 in these cases.



**Figure S6.** EtBr exclusion assay showing the decrease in fluorescence intensity at 610 nm due to complexation of  $\lambda$ -DNA with T'3-(AKAE)<sub>2</sub> or A'3-(AKAE)<sub>2</sub> or T'3-(IKIE)<sub>2</sub>. The fluorescence of uncomplexed  $\lambda$ -DNA was set as maximum, i.e., 100%.



**Figure S7.** Statistical results of the size parameters obtained from AFM measurements:

(A<sub>1</sub>, A<sub>2</sub>)  $\lambda$ -DNA, (B<sub>1</sub>, B<sub>2</sub>)  $\lambda$ -DNA/ $T'_3$ -(AKAE)<sub>2</sub> globules, (C<sub>1</sub>, C<sub>2</sub>)  $\lambda$ -DNA/ $T'_3$ -(IKIE)<sub>2</sub> globules, and (D<sub>1</sub>, D<sub>2</sub>)  $\lambda$ -DNA/ $A'_3$ -(AKAE)<sub>2</sub> globules.