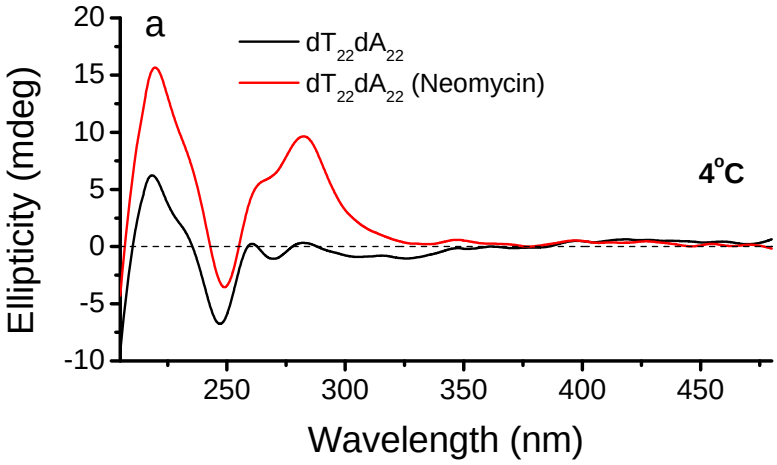
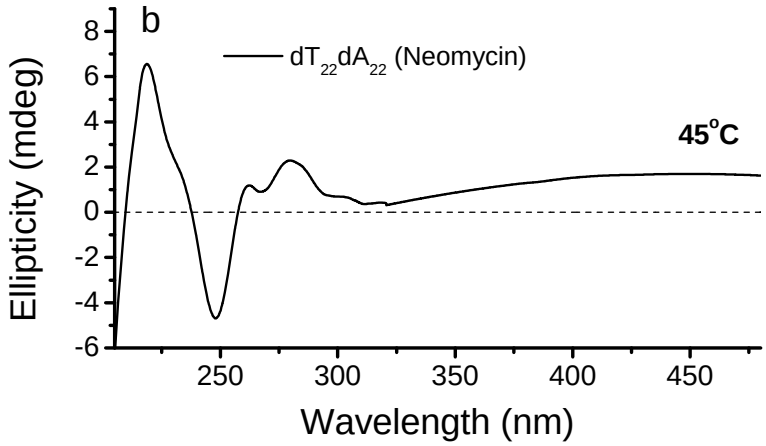
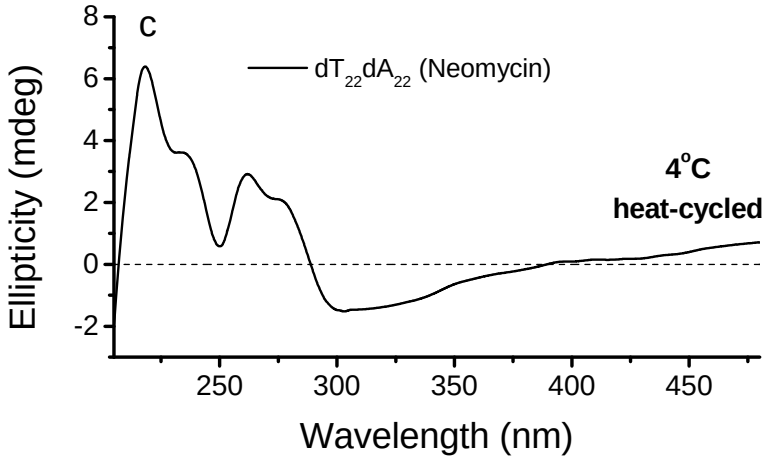
Supporting Figures







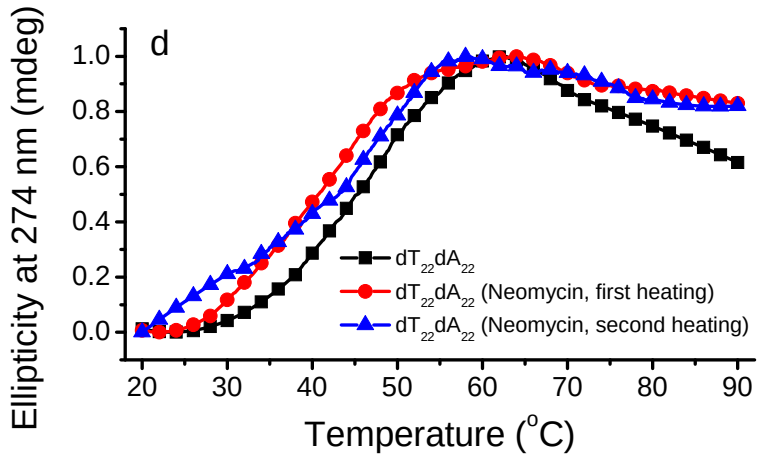
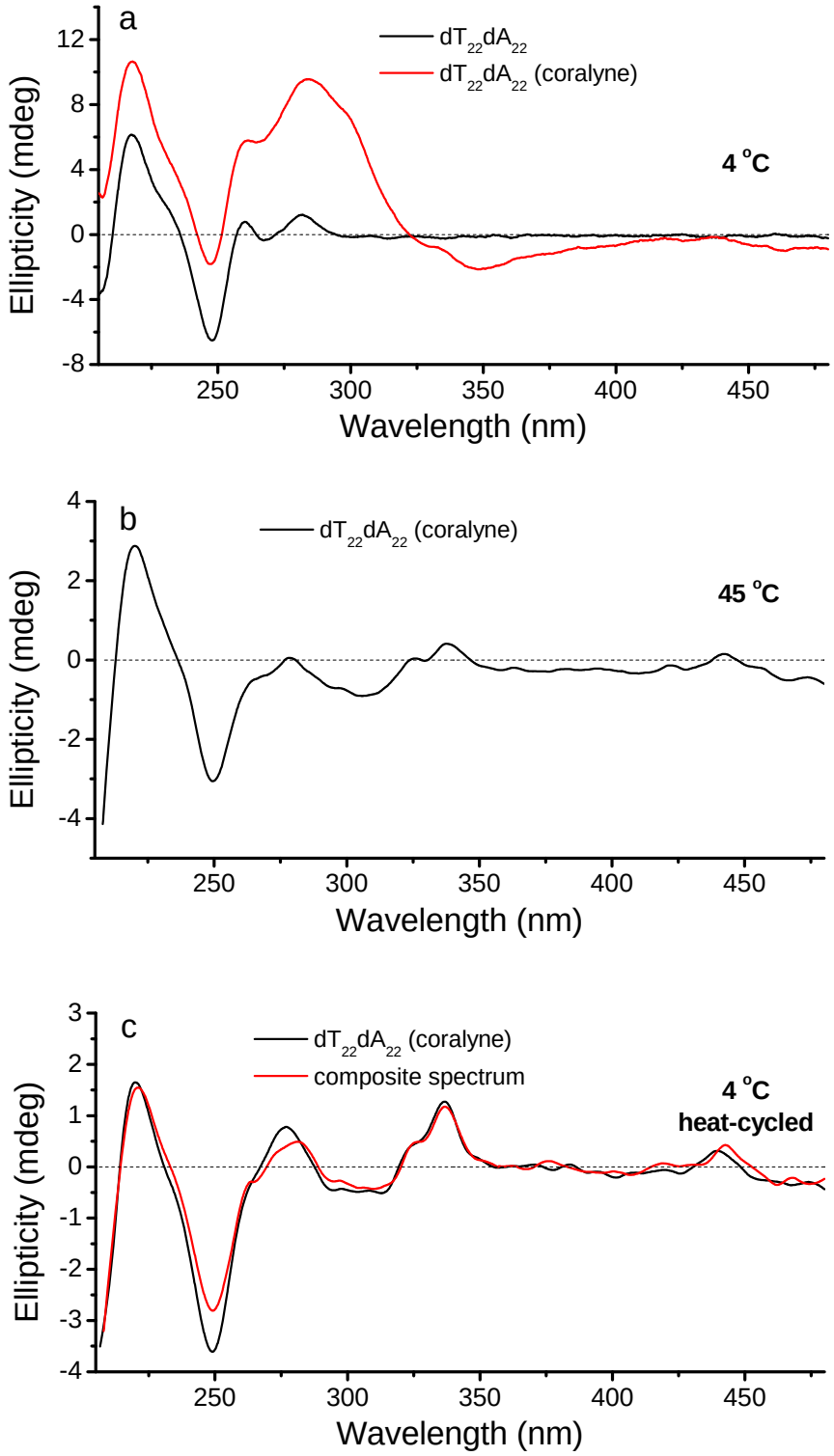


Figure S1. CD spectra of duplex $dA_{22}{\cdot}dT_{22}$ samples. (a) Duplex $dA_{22}{\cdot}dT_{22}$ sample with and without added neomycin at 4 °C prior to heating. (b) Spectra of duplex $dA_{22}{\cdot}dT_{22}$ sample with neomycin at 45 °C. (c) Heat-cycled spectrum of $dA_{22}{\cdot}dT_{22}$ sample in the presence of neomycin at 4 °C. (d) CD melting profiles for 22mer duplex $dA_{22}{\cdot}dT_{22}$ sample at wavelengths selected and the first and second heating of duplex $dA_{22}{\cdot}dT_{22}$ after the addition of neomycin. DNA concentrations were 22 μM in base pair. Neomycin concentration was 0.5 molar equivalents per base pair (11 μM).

The CD spectrum of duplex $dA_{22}{\cdot}dT_{22}$ at 4 °C changes dramatically upon the addition of 0.5 molar equivalents of neomycin per DNA base pair (Figure S1a). These changes include the appearance of a substantial positive CD band (or bands) near 280 nm and 225 nm. However, typical CD bands of triplex DNA with intercalated compound at ~340 and ~440 nm are non-existed. This indicates that neomycin does not cause the formation of triplex DNA at 4 °C. Upon heating of the $dA_{22}{\cdot}dT_{22}$ sample with added neomycine to 45 °C dramatically reduces the magnitude of the duplex-specific neomycin CD bands at 280 and 225 nm (Figure S1b); small positive bands of triplex at ~340 and ~440 nm (Figure S1b) are also not observed. Additionally, the melting profile of the $dA_{22}{\cdot}dT_{22}$ sample in the presence of neomycin exhibits a transition at 46 °C (Figure S1d), which is similar with the duplex $dA_{22}{\cdot}dT_{22}$ itself. Thus, the 46 °C transition in the duplex sample with neomycin can be assigned

to the melting of duplex $dA_{22}{\cdot}dT_{22}$. So, these results clearly indicate that neomycin can not cause the strands of $dA_{22}{\cdot}dT_{22}$ to repartition into an equimolar amount of triplex $dT_{22}{\cdot}dA_{22}{\cdot}dT_{22}$ and dA_{22}.



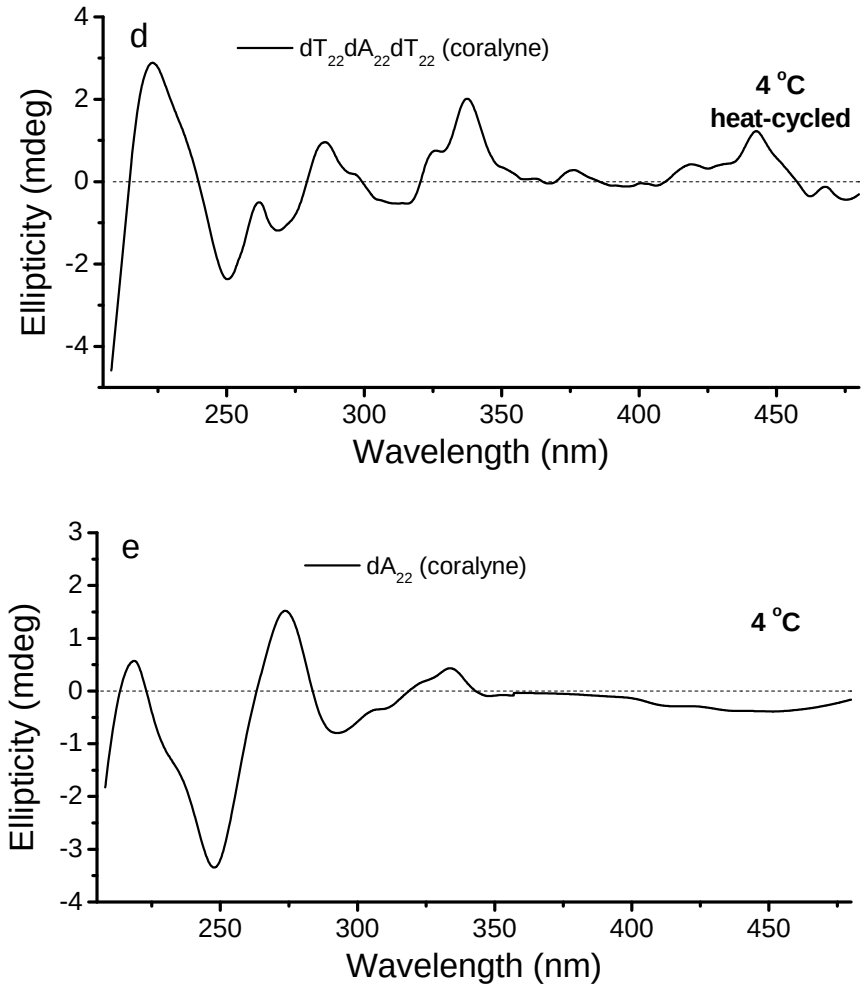


Figure S2. CD spectra of duplex $dA_{22}{\cdot}dT_{22}$ samples that illustrate duplex disproportionation. (a) Duplex $dA_{22}{\cdot}dT_{22}$ sample with and without added coralyne at 4 °C prior to heating. (b) Spectra of duplex $dA_{22}{\cdot}dT_{22}$ sample with coralyne at 45 °C. (c) Heat-cycled spectrum of disproportioned $dA_{22}{\cdot}dT_{22}$ sample in the presence of coralyne at 4 °C and composite spectrum {sum of spectra acquired at 4 °C: [0.5 × triplex $dT_{22}{\cdot}dA_{22}{\cdot}dT_{22}$ with 0.5 molar equivalents of coralyne] + [0.50 × single stranded dA_{22} with 0.5 molar equivalents of coralyne]}. (d) Spectra of triplex $dT_{22}{\cdot}dA_{22}{\cdot}dT_{22}$ at 4 °C after heat cycling with coralyne. (e) Spectra of dA_{22} sample with coralyne at 4 °C. DNA concentrations were 22 μM in nucleotide, base pair or base triplet, respectively. Coralyne concentration was 0.5 molar equivalents per nucleotide, base pair or base triplet, respectively.

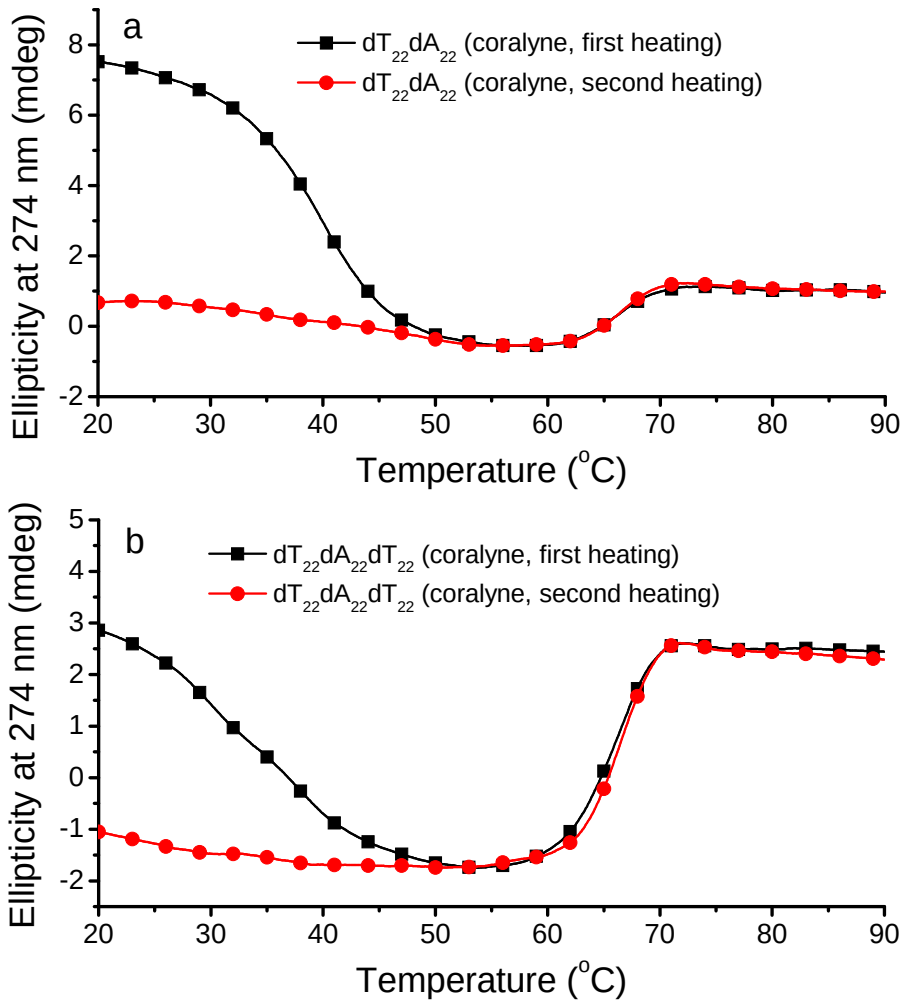


Figure S3. CD melting profiles for 22mer DNA samples at wavelengths selected to show structural transitions. (a) First and second heating of duplex $dA_{22}{\cdot}dT_{22}$ after the addition of coralyne; the duplex disproportionation transition is observed at 39.5 °C during the first heating and triplex melting is observed at 66.1 °C in both the first and second heating of the sample. (b) First and second heating of a triplex $dT_{22}{\cdot}dA_{22}{\cdot}dT_{22}$ sample after the addition of coralyne indicates triplex melting at 66.2 °C. The first heating of this sample after the addition of coralyne also shows a transition centered at 33.6 °C, which is assigned to the reorganization of DNA strands from partial duplex and partial triplex to complete triplex. DNA concentrations were 22 μM in base pair or base triplet, respectively. Coralyne concentration was 0.5 molar equivalents per base pair or base triplet, respectively.

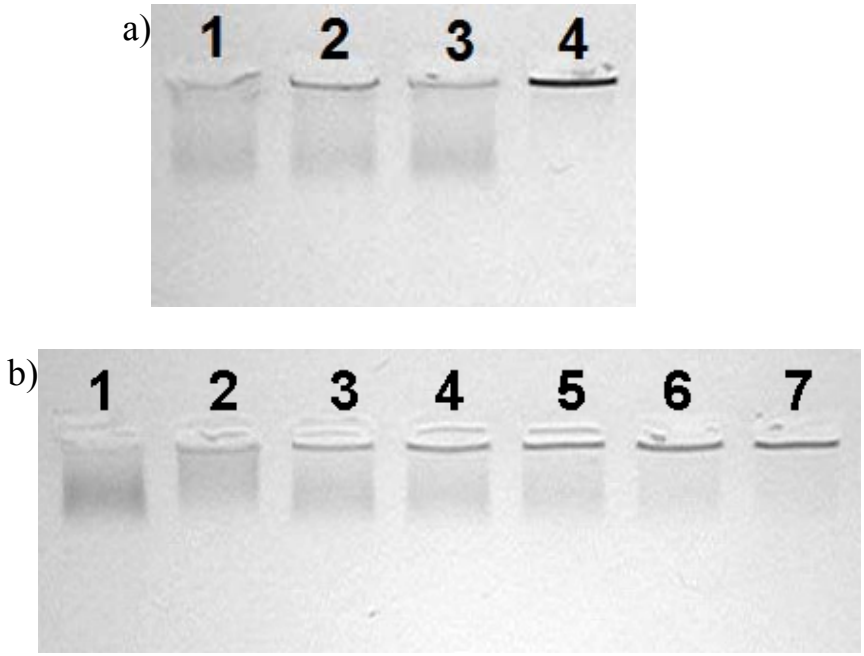


Figure S4. Electrophoretic mobility of DNA-SWNTs conjugates in the absence or presence of coralyne. a) SWNTs-dT_{22} alone (lane 1) and in the presence of 1 μM coralyne (lane 3); SWNTs-dT_{22}·dA_{22} alone (lane 2) and in the presence of 1 μM coralyne (lane 4); b) SWNTs-dT_{22}·dA_{22} alone (lane 1) and in the presence of 0.1 μM (lane 2), 0.2 μM (lane 3), 0.4 μM (lane 4), 0.6 μM (lane 5), 0.8 μM (lane 6), 1 μM (lane 7) coralyne.