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Analytical Data of T-n-T (n = 11 and 12), A-n-A (n = 11 and 12), and T-n-A (n = 11 and 12)

***N, N'*-bis[3-(2,4-dihydroxy-5-methylpyrimidine-1-yl)propionyl]1, 11-diaminoundecane (T-11-T).** This bolaamphiphile was synthesized in the same way as **T-10-T**. mp 200.0–204.0; TLC (silica gel, 1:1 $CHCl_3$/MeOH) R_f = 0.9; ^{1}HNMR (600 MHz, DMSO-d_6, 25°C): δ 1.21–1.34 (m, 18H, $-CH_2-$), 1.71 (s, 6H, CH_3-5), 2.41 (t, J = 6.6 Hz, 4H, $-CH_2CH_2CONH-$), 2.99 (dt, J = 6.6, 6.6 and 7.2 Hz, 4H, $-CONHCH_2-$), 3.80 (t, J = 6.6 Hz, 4H, $-CH_2CH_2CONH-$), 7.37 (s, 2H, H-6), 7.94 (t, J = 5.4 Hz, 2H, –CONH–), 11.2 (s, 2H, NH-3). Anal. Calcd for $C_{27}H_{42}N_6O_6\cdot H_2O$: C, 57.43; H, 7.85; N, 14.88. Found: C, 57.03; H, 7.57; N, 14.73.

***N, N'*-bis[3-(6-aminopurine-9-yl)propionyl]1, 11-diaminoundecane (A-11-A).** This bolaamphiphile was synthesized in the same way as **A-10-A**. mp 224.0–230.0; TLC (silica gel, 2:1 $CHCl_3$/MeOH) R_f = 0.6; ^{1}HNMR (600 MHz, DMSO-d_6, 25°C) δ 1.12–1.31 (m, 18H, $-CH_2-$), 2.65 (t, J = 6.6 Hz, 4H, $-CH_2CH_2CONH-$), 2.97 (dt, J = 6.0, 6.0 and 7.2 Hz, 4H, $-CONHCH_2-$), 4.33 (t, J = 6.6 Hz, 4H, $-CH_2CH_2CONH-$), 7.20 (s, 4H, NH_2-6), 7.88 (t, J = 5.4 Hz, 2H, –CONH–), 7.98 (s, 2H, H-8), 8.13 (s, 2H, H-2). Anal. Calcd for $C_{27}H_{40}N_{12}O_2\cdot H_2O$: C, 55.65; H, 7.26; N, 28.84. Found: C, 55.40; H, 7.33; N, 28.59.

***N, N'*-bis[3-(2,4-dihydroxy-5-methylpyrimidine-1-yl)propionyl]1, 12-diaminododecane (T-12-T).** This bolaamphiphile was also synthesized in the same way as **T-10-T**. mp 211.8–214.4; TLC (silica gel, 1:1 $CHCl_3$/MeOH) R_f = 0.9; ^{1}HNMR (600 MHz, DMSO-d_6, 25°C): δ 1.22–1.34 (m, 18H, $-CH_2-$), 1.71 (s, 6H, CH_3-5), 2.41 (t, J = 6.6 Hz, 4H, $-CH_2CH_2CONH-$), 3.00 (dt, J = 6.6, 6.6 and 7.2 Hz, 4H, $-CONHCH_2-$), 3.81 (t, J = 6.6 Hz, 4H, $-CH_2CH_2CONH-$), 7.37 (s, 2H, H-6), 7.94 (t, J = 5.4 Hz, 2H, –CONH–), 11.2 (s, 2H, NH-3). Anal. Calcd for $C_{28}H_{44}N_6O_6$: C, 59.98; H, 7.91; N, 14.99. Found: C, 59.98; H, 7.93; N, 14.78.

***N, N'*-bis[3-(6-aminopurine-9-yl)propionyl]1, 12-diaminododecane (A-12-A).** This bolaamphiphile was also synthesized in the same way as **A-10-A**. mp 228.9–233.4; TLC (silica gel, 2:1 $CHCl_3$/methanol) R_f = 0.4; ^{1}HNMR (600 MHz, DMSO-d_6, °C) δ 1.20–1.30 (m, 18H, $-CH_2-$), 2.65 (t, J = 6.6 Hz, 4H, $-CH_2CH_2CONH-$), 2.97 (dt, J = 6.0, 6.0 and 7.2 Hz, 4H, $-CONHCH_2-$), 4.33 (t, J = 6.6 Hz, 4H, $-CH_2CH_2CONH-$), 7.19 (s, 4H, NH_2-6), 7.88 (t, J = 5.4 Hz, 2H, –CONH–), 7.98 (s, 2H, H-8), 8.13 (s, 2H, H-2). Anal. Calcd for $C_{28}H_{42}N_{12}O_2 \cdot 1/2H_2O$: C, 57.22; H, 7.37; N, 28.60. Found: C, 57.42; H, 7.32; N, 28.64.

***N*-[3-(2,4-dihydroxy-5-methylpyrimidine-1-yl)propionyl], *N'*-[3-(6-aminopurine-9-yl)propionyl]1,11-diaminoalkane (T-11-A).** This bolaamphiphile was synthesized in the same way as **T-10-A**. mp 204.0–207.0; TLC (silica gel, 1:1 $CHCl_3$/MeOH) R_f = 0.7; ^{1}H-NMR (600 MHz, DMSO-d_6, 25°C): δ 1.20–1.35 (m, 18H, $-CH_2-$), 1.71 (s, 3H, Thy-CH_3-5), 2.41 (t, J = 6.6 Hz, 2H, Thy-$(CH_2)_2CONHCH_2CH_2-$), 2.65 [t, J = 6.6 Hz, 2H, Ade-$(CH_2)_2CONHCH_2CH_2-$], 3.00 (dt, J = 6.2, 3.7, 6.2, and 6.6 Hz, 4H, $-CONHCH_2-$), 3.81 (t, J = 6.6 Hz, 2H, Thy-CH_2CH_2CONH-), 4.33 (t, J = 6.6 Hz, 2H, Ade-CH_2CH_2CONH-), 7.16 (s, 2H, Ade-NH_2-6), 7.36 (s, 1H, Thy-H-6), 7.86 [t, J = 5.5 Hz, 1H, Ade-$(CH_2)_2CONH-$], 7.92 [t, J = 5.4 Hz, 1H, Thy-$(CH_2)_2CONH-$], 7.98 (s. 1H, Ade-H-8), 8.13 (s. 1H, Ade-H-2), 11.2 (s, 1H, Thy-NH-3). Anal. Calcd for $C_{27}H_{41}N_9O_4$: C, 58.36; H, 7.44; N, 22.69. Found: C, 58.55; H, 7.56; N, 22.35.

***N*-[3-(2,4-dihydroxy-5-methylpyrimidine-1-yl)propionyl], *N'*-[3-(6-aminopurine-9-yl)propionyl]1,12-diaminoalkane (T-12-A).** This bolaamphiphile was also synthesized in the same way as **T-10-A**. mp 228.0–228.6; TLC (silica gel, 1:1 $CHCl_3$/MeOH) R_f = 0.7; ^{1}H-NMR (600 MHz, DMSO-d_6, 25°C): δ 1.20–1.32 (m, 20H, $-CH_2-$), 1.71 (s, 3H, Thy-CH_3-5), 2.41 (t, J = 6.6 Hz, 2H, Thy-$(CH_2)_2CONHCH_2CH_2-$), 2.65 [t, J = 6.6 Hz, 2H, Ade-

$(CH_2)_2CONHCH_2CH_2-$], 2.98 (dt, J = 6.2, 3.7, 6.2, and 6.6 Hz, 4H, $-CONHCH_2-$), 3.81 (t, J = 6.6 Hz, 2H, Thy-CH_2CH_2CONH-), 4.33 (t, J = 6.6 Hz, 2H, Ade-CH_2CH_2CONH-), 7.16 (s, 2H, Ade-NH_2-6), 7.36 (s, 1H, Thy-H-6), 7.86 [t, J = 5.5 Hz, 1H, Ade-$(CH_2)_2CONH-$], 7.92 [t, J = 5.4 Hz, 1H, Thy-$(CH_2)_2CONH-$], 7.98 (s. 1H, Ade-H-8), 8.13 (s. 1H, Ade-H-2), 11.2 (s, 1H, Thy-NH-3). Anal. Calcd for $C_{28}H_{43}N_9O_4$: C, 59.03; H, 7.61; N, 22.13. Found: C, 59.22; H, 7.56; N, 22.30.

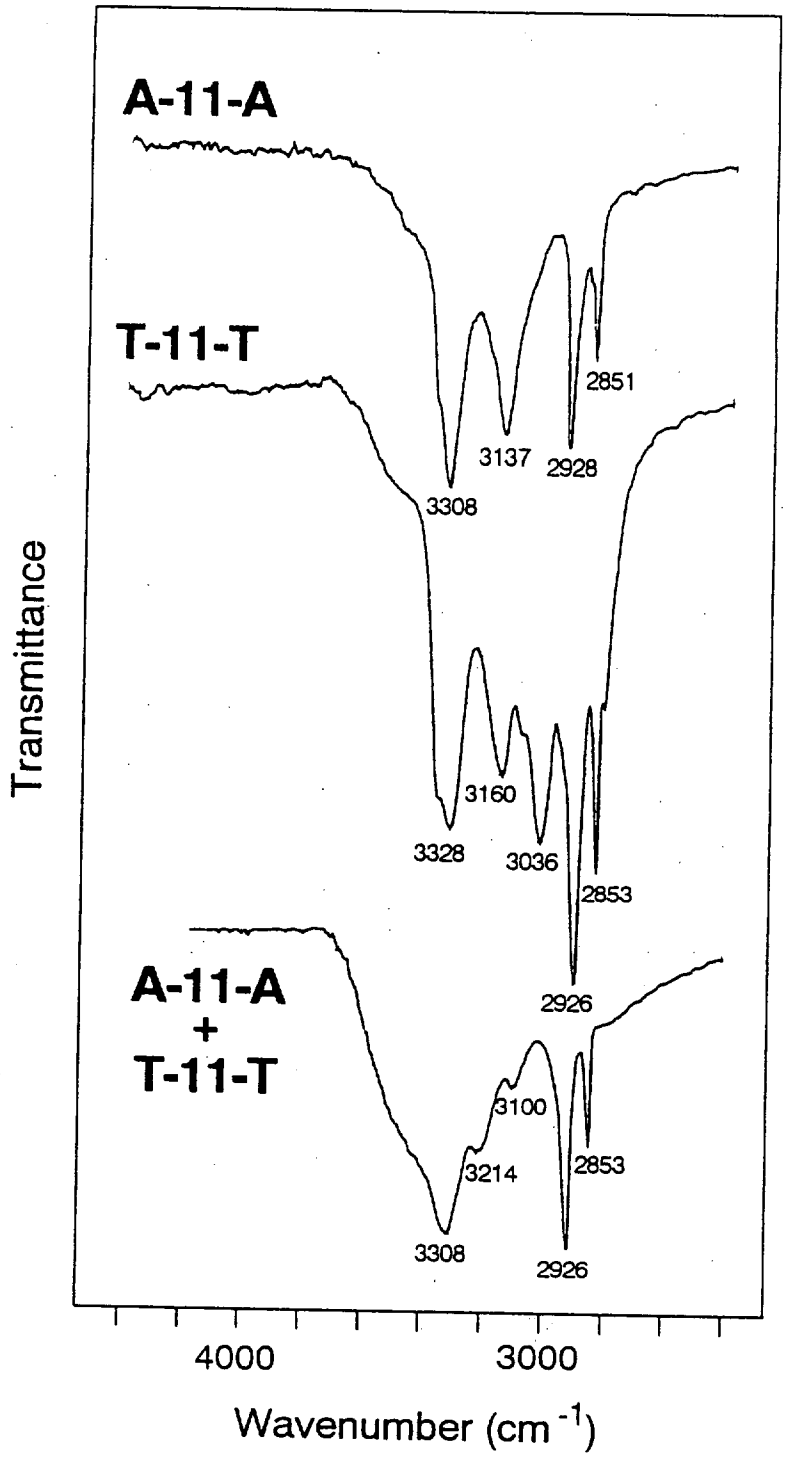

Fig. S-1. FT-IR Spectra (2700 –4400 cm^{-1}) of **A-11-A**, **T-11-T**, and their 1:1 Self-(hetero)-Assembly

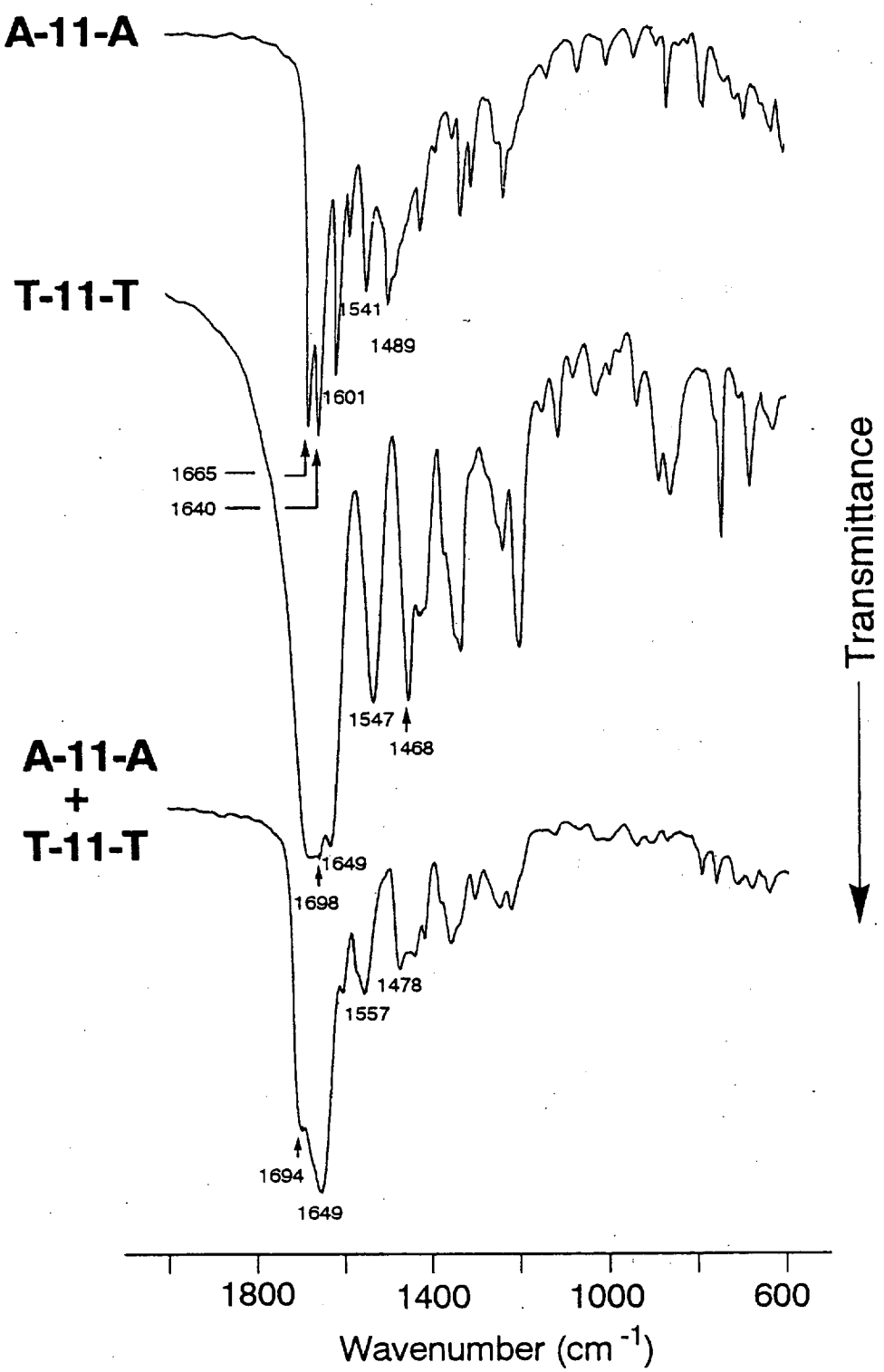

Fig. S-2. FT-IR Spectra (600 –1800 cm $^{-1}$) of **A-11-A**, **T-11-T**, and their 1:1 Self-(hetero)-Assembly

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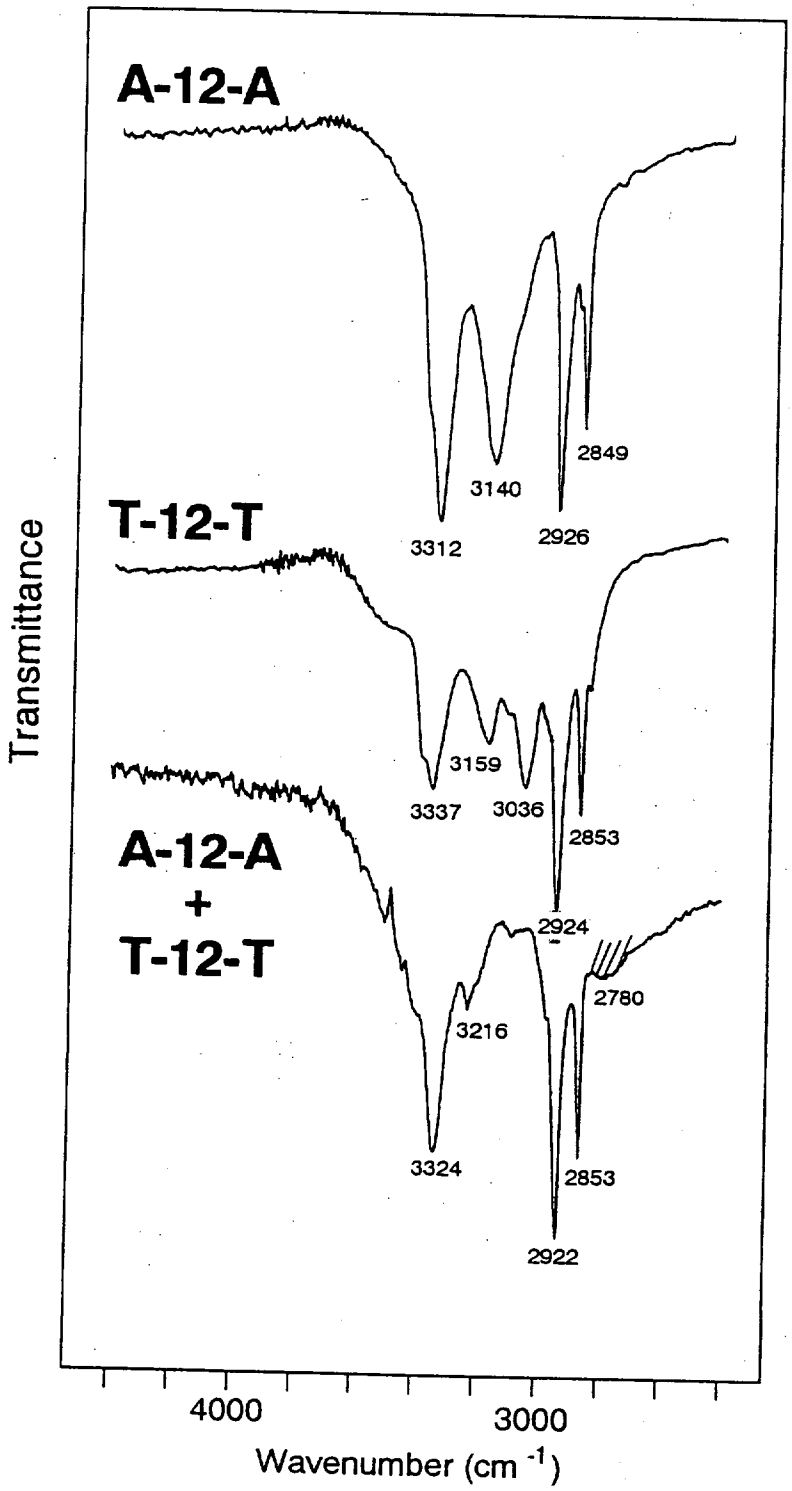

Fig. S-3. FT-IR Spectra (2700 –4400 cm^{-1}) of **A-12-A**, **T-12-T**, and their 1:1 Self-(hetero)-Assembly

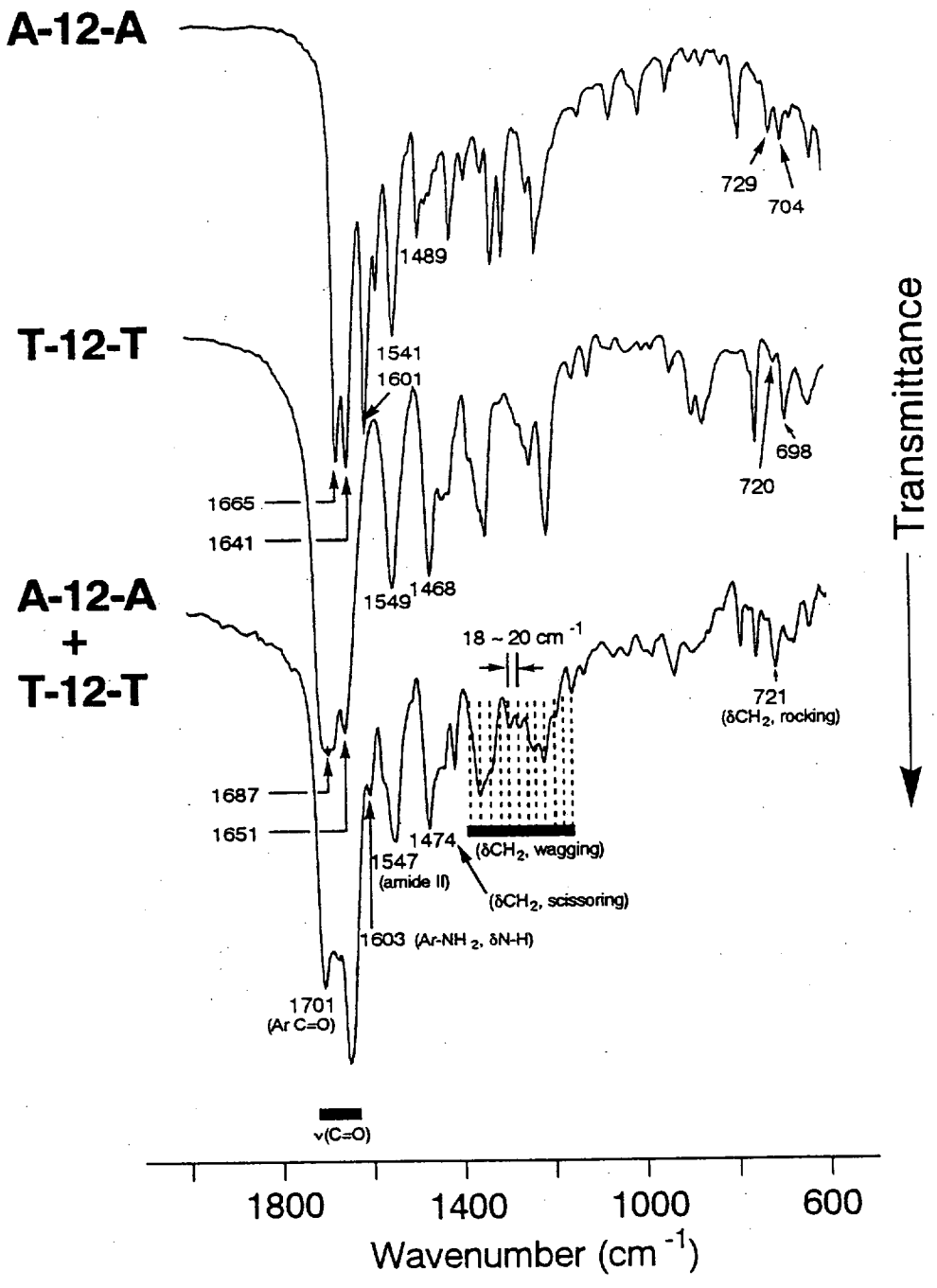

Fig. S-4. FT-IR Spectra (600 –1800 cm^{-1}) of **A-12-A**, **T-12-T**, and their 1:1 Self-(hetero)-Assembly

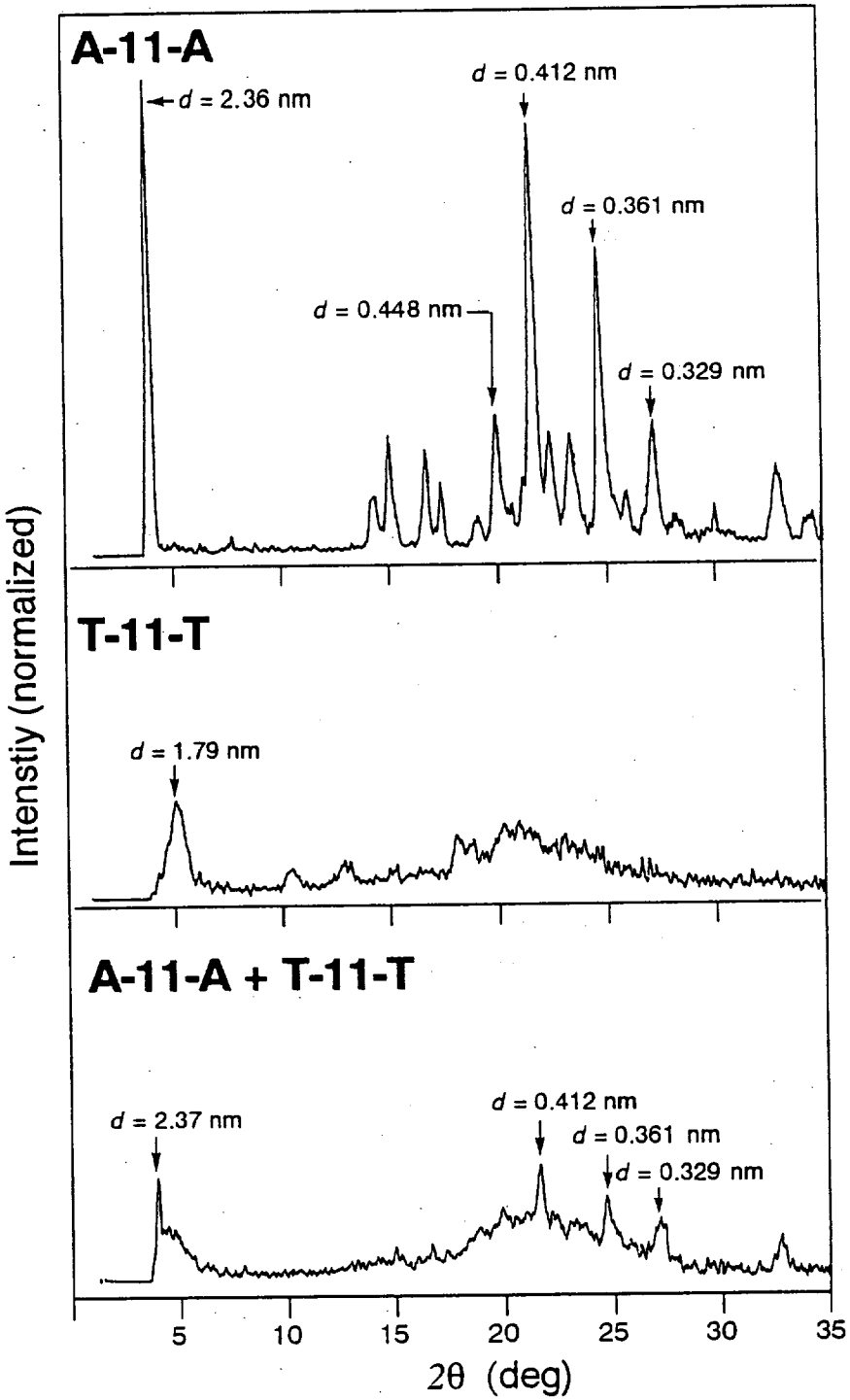

Fig. S-5. XRD diagrams of **A-11-A**, **T-11-T**, and their 1:1 Self-(hetero)-Assembly

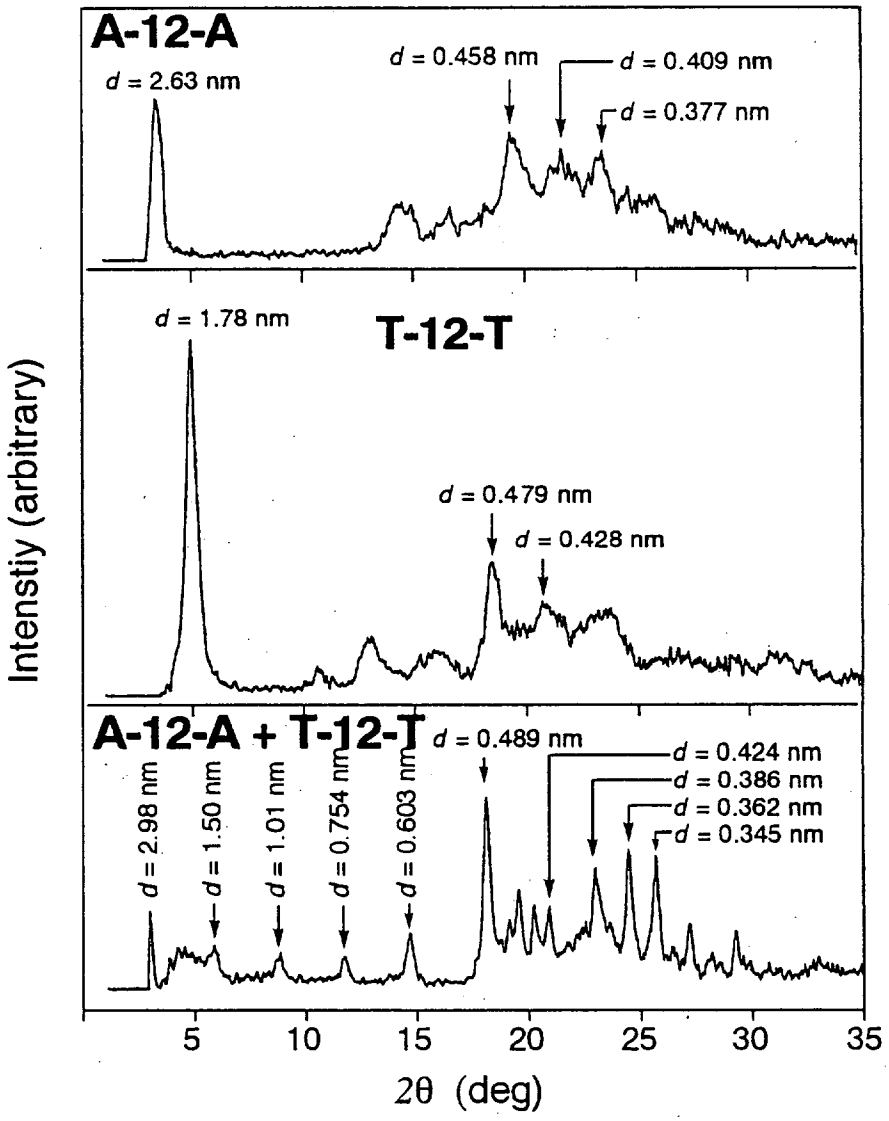

Fig. S-6. XRD diagrams of **A-12-A**, **T-12-T**, and their 1:1 Self-(hetero)-Assembly

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Table S-7. Self-Assembling Morphologies[a] of **T-n-T** (n = 11 and 12), **A-n-A** (n = 11 and 12), and the Equimolar Mixtures of **T-n-T/A-n-A** in Ethanol Solutions

	Homo-Assembly		1:1 Hetero-Assembly
n	**T-n-T**	**A-n-A**	**T-n-T/A-n-A**
11	amorphous gel (200.0–204.0)[b]	crystalline powder (224.0–230.0)[b]	amorphous gel (207.0–209.0)[b]
12	amorphous gel (211.8–214.4)[b]	crystalline powder (228.9–233.4)[b]	crystalline solid (228.2–235.1)[b]
			T-11-T/A-12-A
			crystalline solid
			T-12-T/A-11-A
			amorphous gel

[a]Observed using light microscopy.

[b]Melting point of the dried sample (°C).

Table S-8. FT-IR Absorption Bands (400- 1800 cm^{-1} region) and Their Plausible Assignment for the Homo- and 1:1 Hetero-Assemblies Formed from **T-11-T**, **T-12-T**, **A-11-A**, and **A-12-A**[a]

Assignment	Homo-Assembly				1:1 Hetero-Assembly	
	T-n-T		**A-n-A**		**T-n-T/A-n-A**	
	n = 11	n = 12	n = 11	n = 12	n = 11	n = 12
Aromatic C=O str.	1698	1687	–	–	1694	1701
Amide C=O str. (amide I)	1649	1651	1665	1665	1649	1647
			1640[b]	1641[b]		
NH_2 scissoring plus ring C–H str.	–	–	1603	1601	1603	1603
Amide N–H def. (amide II)	1547	1549	1541	1541	1557	1547
CH_2 scissoring def.	1468	1468	1489[c]	1489[c]	1478[c]	1474
CH_2 rocking def.	721	720	723	729	718	721
			704	704		

[a]Self-assembling was carried out in ethanol solutions.

[b]Hydrogen-bonded NH_2 scissoring plus ring C–H stretching.

[c]Broad band.

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Table S-9. X-ray Diffraction Data for the Homo- and 1:1 Hetero-Assemblies Formed from **T-11-T**, **T-12-T**, **A-11-A**, and **A-12-A**[a]

	Homo-Assembly				1:1 Hetero-Assembly	
	T-n-T		**A-n-A**		**T-n-T/A-n-A**	
	n = 11	n = 12	n = 11	n = 12	n = 11	n = 12
Long-range ordering (nm) and the order of diffraction (parentheses)	1.79 (2) 0.84 (4)	1.78 (2) 0.84 (4)	2.36 (1)	2.63 (1)	2.37 (1)	2.98 (1) 1.50 (2) 1.01 (3) 0.754 (4) 0.603 (5)
Long period (nm)[b]	3.58	3.56	2.36	2.63	2.37	3.01
Short-range ordering (nm)	c	0.479 0.428 0.396 0.360	0.448 0.412 0.361 0.329	0.458 0.409 0.381	c	0.489 0.424 0.386 0.362 0.345
Possible Packing Mode[d]	Dh	T//	O'⊥	O⊥	Dh	T//

[a]Self-assembling was carried out in ethanol solutions. [b]Obtained by reflection method. [c]No well-defined peaks were obtained because of the broadening. [d]T//: triclinic subcell, Dh: distorted hexagonal packing, O⊥ and O'⊥: orthorhombic subcell

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Table S-10. Molar Absorptivity (ε) and Absorption Maxima (λ_{max}) of **T-11-T**, **T-12-T**, **A-11-A**, **A-12-A**, and the Their 1:1 Mixtures in Ethanol Solutions and Their Hypochromicity.

Nucleobase Derivative	ε (λ_{max}, nm)	ε_R (λ_{max}, nm)	H(%)
T-11-T	10600 (269)	7300[a] (269)	27[b]
T-12-T	13800 (269)	7300[a] (269)	5.0[b]
A-11-A	14400 (261)	12600[c] (261)	43[b]
A-12-A	12500 (261)	12600[c] (261)	51[b]
1:1 **T-11-T/A-11-A**	13850 (264)	25000[d]	45[e]
1:1 **T-12-T/A-12-A**	15000 (264)	26300[f]	43

[a]The molar absorptivity of thymine. [b]Evaluated according to the following equation: $H(\%) = 100 \times (1 - \varepsilon/2\varepsilon_R)$. [c]The molar absorptivity of adenine. [d]Summation of the molar absorptivity of **T-11-T** and **A-11-A**. [e]Evaluated according to the following equation: $H(\%) = 100 \times (1 - \varepsilon/\varepsilon_R)$. [f]Summation of the molar absorptivity of **T-12-T** and **A-12-A**.

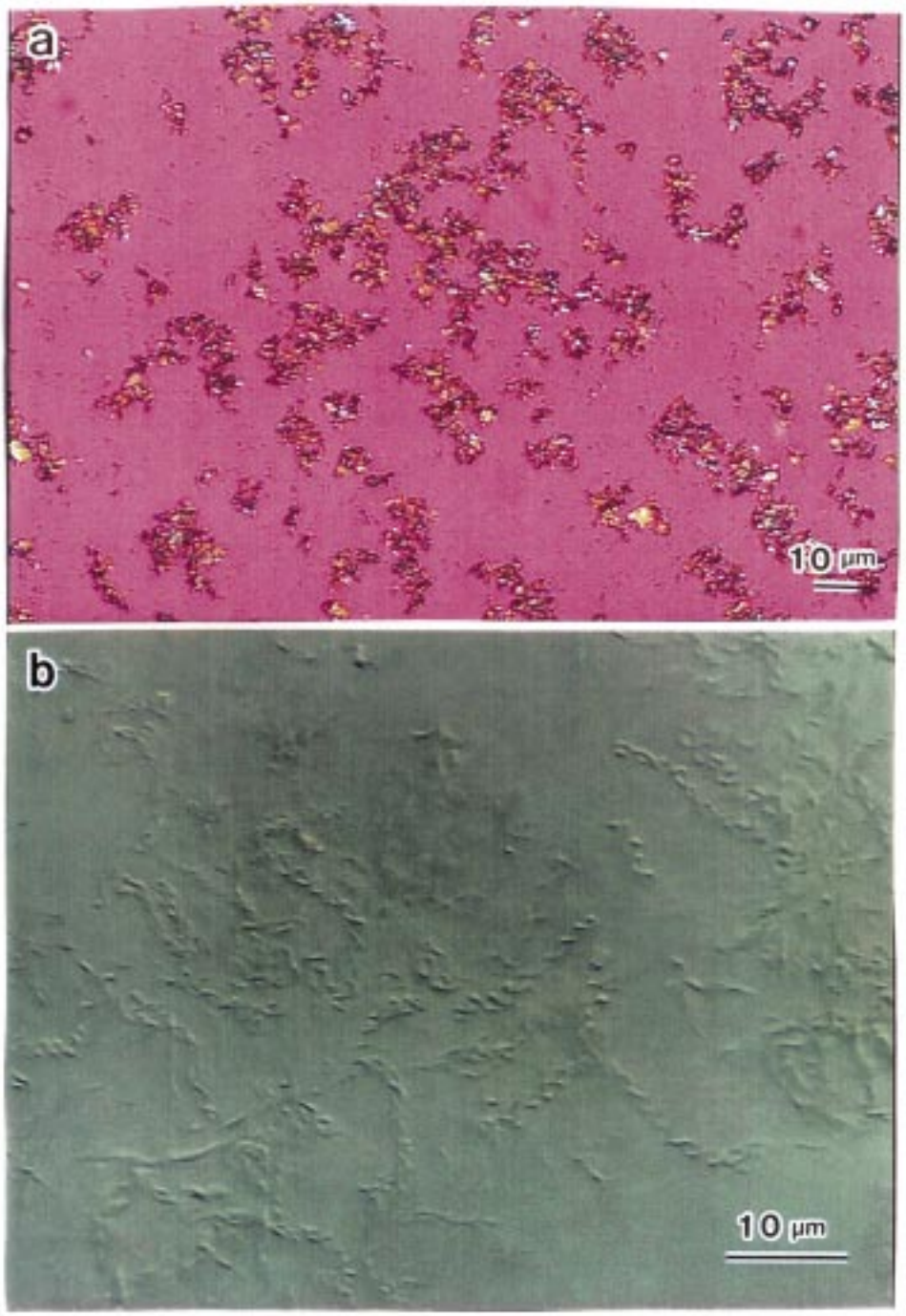

Fig. S-12. (a) Microcrystalline Solids Formed from **A-10-A** and (b) a Swarm of Double-Helical Ropes from **T-10-T** Observed Using (a) Polarized and (b) Dark-Field Light Microscopy (at 25°C in 10%-Ethanolic Aqueous Solutions.

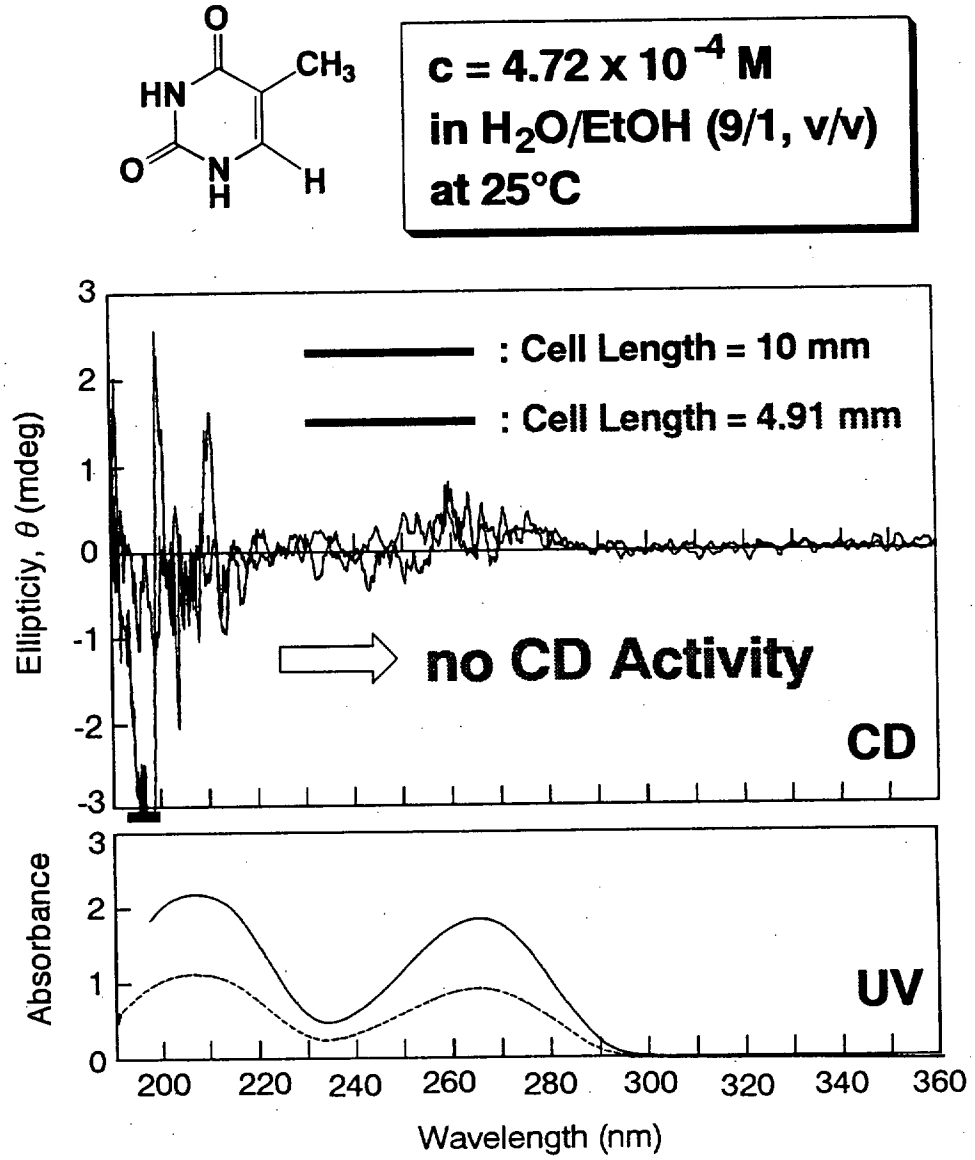

Fig. S-13. Circular Dichroism and Ultraviolet Spectra of Thymine (JASCO J-725)

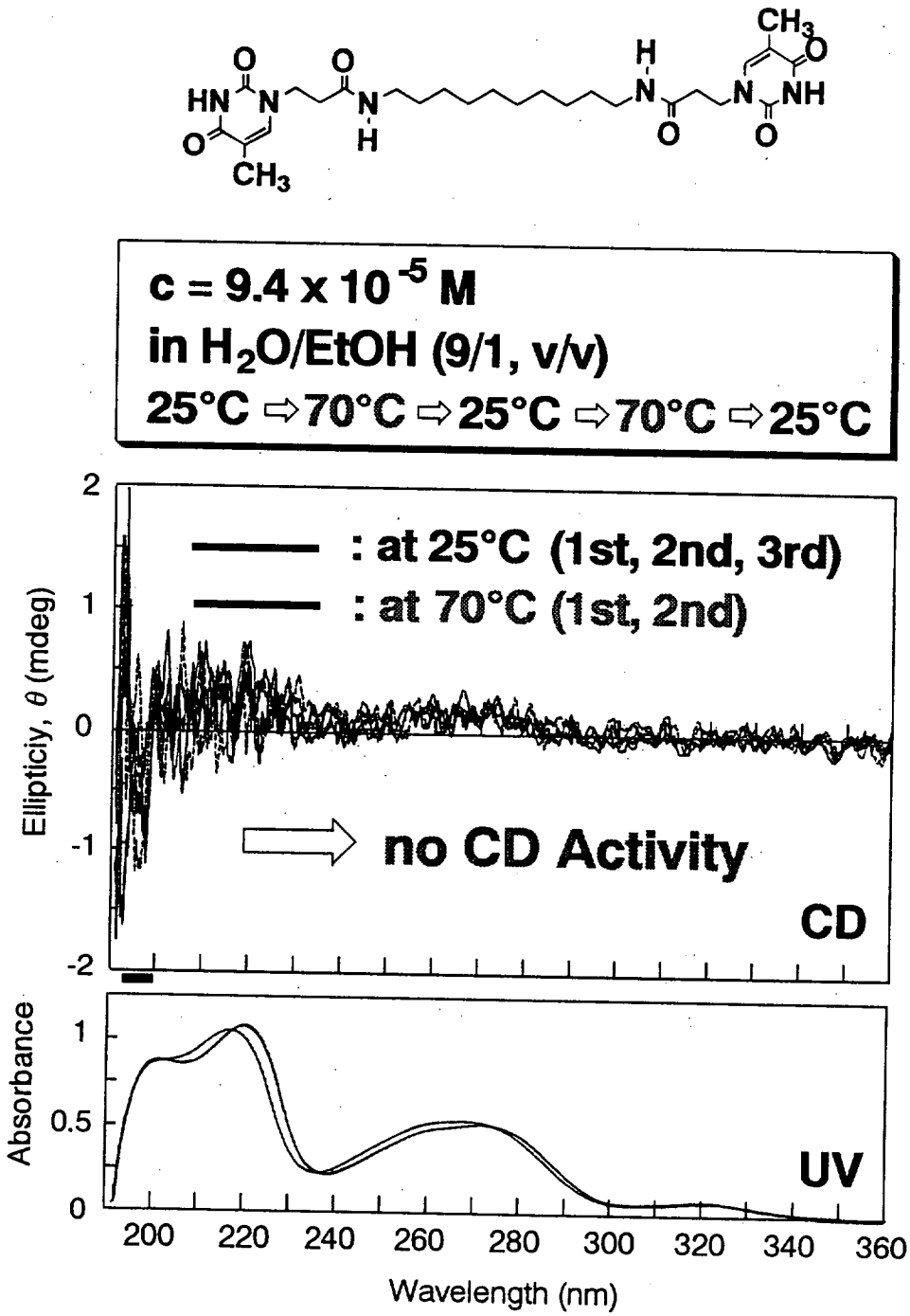

Fig. S-14. Circular Dichroism and Ultraviolet Spectra of Thymine (JASCO J-725)