Alignment of Twisted Nanoribbons formed by C₁₇H₃₅CO-Val-

Ala Sodium Salts

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A typical procedure for the preparation of $C_{17}H_{35}CO-L-Val-L-Ala-OH$ ((L, L)-14) was shown as following. Before the reaction, the 2-chlorotrityl chloride resin (2.0 g, 2.0 mmol) of was swelled in 10.0 mL of anhydrous DCM for 30 min. Then, the resin was washed with 10.0 mL of anhydrous DMF four times. Fmoc-L-Ala-OH (1.72 g, 5.49 mmol) was dissolved in 20.0 mL of DMF in the presence of 2.1 mL (12.5 mmol) of DIEA. The mixture was added into the reactor and left for 1.0 hour to ensure the amino acid was fully loaded onto the resin. The resin was washed with 10.0 mL of DMF four times and the possible unreacted sites of the resin were deactivated using a block solution (DCM:MeOH:DIEA = 80:15:5) two times (30 and 15 minutes each). After washing with 10.0 mL of DMF five times, the resin was treated with 20% piperidine in DMF (25.0 mL) two times (30 and 15 minutes each) to deprotect the Fmoc group and then washed with 10.0 mL of DMF five times. Fmoc-L-Val-OH (1.87 g, 5.49 mmol) was dissolved in 20.0 mL of DMF in the presence of 2.1 mL (12.5 mmol) of DIEA and the coupling reagent HBTU (1.88 g, 4.95 mmol). The coupling and deprotection processes were performed as described above. The fluorinated tail was conjugated using a mixed solution of C₁₇H₃₅COOH (2.70 g, 5.00 mmol), DIEA (2.10 mL, 12.5 mmol), and HBTU (1.88 g, 4.95 mmol). After all the elongation steps were carried out, the resin was successively washed in DMF, DCM, methanol, and *n*-hexane. Trifluoroacetic acid (10.0 mL) was applied to cleave the peptide derivatives from the resin for three times. The obtained crude products were precipitated in anhydrous diethyl ether and filtered to give 913 mg of the peptides (yield: 86.6%). The corresponding sodium salt was obtained by adding NaOH aqueous solution.



Fig. S1 The picture of the (L, L)- and (D, D)-14 hydrogels at a concentration of 20 g L^{-1} .

Solvents	(L, L)- 14	(D, D)- 14
H ₂ O	Translucent gel, 20	Translucent gel, 20
Toluene	Transparent gel, 20	Transparent gel, 20
DMF	Translucent gel, 15	Translucent gel, 15
THF	Translucent gel, 20	Translucent gel, 20
DMSO	Translucent gel, 20	Translucent gel, 20

Table S1. MGCs of the lipodipeptide sodium salts in selected solvents.^a

^aThe data in the above table refer to the MGCs (g L⁻¹) of the lipodipeptide sodium salts in the listed solvents at 25 °C.



Fig. S2 The dimer of CH_3CO -D-Val-D-Ala-COOH.



Fig. S3 The HOMOs and LUMOs of a CH_3CO -D-Val-D-Ala-COOH dimmer.



Fig. S4 FT-IR spectra of the aqueous solutions and hydrogels of (L, L)-14 and (D, D)-14 in D_2O at a concentration of 20 g L⁻¹.



Fig. S5 ¹H NMR spectra of (D, D)-**14** and (L, L)-**14** hydrogels (20 g L⁻¹) at 70 °C and solutions (20 g L⁻¹) at 80 °C in H_2O/D_2O (v/v = 85/15).



Fig. S6 FE-SEM images of the aligned self-assemblies of (D, D)-**14** at different concentrations. (a) 0.3 g L⁻¹; (b) 6.0 g L⁻¹.



Fig. S7 (a) SAXRD and (b) WAXRD patterns of the xerogel of (D, D)-14 after being aligned.



Fig. S8 Picture of the (D, D)-**14** hydrogel which was pressed out through a 5[#] needle.



Fig. S9 Picture of the (D, D)-**14** hydrogel colored with methylene blue and being injected through the needle of a syringe in a HCl (1 mM) aqueous solution.



Fig. S10 Rheology measurement of the (D, D)-14 hydrogel at a regular frequency (f = 1 Hz).



Fig. S11 Pictures of the (D, D)-**14** hydrogel. (a) at a concentration of 20 g L^{-1} , (b) being injected through the needle of a syringe, (c) after being injected and (d) after standing for 1.0 min.