Supporting information for manuscript entitled Dimension Control on self-assembly of crystalline core-forming polypeptoid block copolymer: 1D nanofiber versus 2D nanosheet Yuhan Wei^{*}, Fujun Liu, Min Li, Zhibo Li,^{*} Jing Sun^{*} Key Laboratory of Biobased Polymer Materials, Shandong Provincial Education Department; School of Polymer Science & Engineering, Qingdao University of Science and Technology,

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Scheme S1: Synthetic route of PEG-*b*-PNPE block copolymer.



Figure S1. Representative ¹HNMR spectra of PEG-*b*-PNPE. ¹H NMR (500 MHz, DMSO-d6) δ 7.35 – 7.08 (m, -C₆<u>H</u>₅), 3.89 – 4.30 (s, COCH₂N), 3.50 (s, CH₂CH₂O), 3.23 (s, CH₃O), 2.17 (t, NC**H**₂CH₂), 1.92 – 1.83 (m, NCH₂C**H**₂).



Figure S2. GPC traces of PEG₁₁₃-*b*-PNPE_m block copolymers, (a): PEG₁₁₃-*b*-PNPE₄;
(b): PEG₁₁₃-*b*-PNPE₁₀, (b): PEG₁₁₃-*b*-PNPE₁₇.



Figure S3. TEM images of the self-assembled structure of PEG_{44} -*b*-PNPE₄ in deionized water quenched in liquid N₂ after treatment at 70 °C for 7 h.



Figure S4. TEM images of the self-assembled structure of PEG_{44} -*b*-PNPE₉ in deionized water quenched in liquid N₂ after treatment at 70 °C for 7 h.



Figure S5. DLS curves of the PEG_{44} -*b*-PNPE₉ in deionized water as increasing the solution temperature, (a) at room temperature, (b) at 70°C.



Figure S6. TEM images of the PEG₄₄-*b*-PNPE₉ after dispersed in deionized water with the assist of ultrasonic unit for 20 min.



Figure S7. TEM images of the (a): PEG_{44} -*b*-PNPE₁₆ and (b) PEG_{44} -*b*-PNPE₂₄ after dispersed in deionized water at room temperature with the assist of ultrasonic unit.



Figure S8. (a) First cooling and (b) second heating DSC curves of the four PEG_{113} -*b*-PNPE_m block polymers at 10°C/min.



Figure S9. AFM height image of the spin-coated film from (a, b) PEG_{113} -*b*-PNPE₄, (c) PEG_{113} -*b*-PNPE₁₀, (d) PEG_{113} -*b*-PNPE₁₇. The inserted image in (a) is the respective profile image of the nanosheet indicated by the white line.



Figure S10. Cryo-TEM images of the self-assembled structure of the block copolymer (a) PEG₁₁₃-*b*-PNPE₄ (b) PEG₁₁₃-*b*-PNPE₁₀ (c) PEG₁₁₃-*b*-PNPE₁₇ (d) PEG₁₁₃-*b*-PNPE₂₂ in deionized water.