Supporting Information

Crystallization and Self-assembly of Shape-Complementary Sequence-defined Peptoids

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Table S1. Characterization data of diblock copolypeptoids **1-6**.

	Peptoid Nomenclature	Theoretical M	Observed[a]		Purity(%)[b]
			$[M+2H]^{2+}$	$[M+3H]^{3+}$	
1	Ac-DDNNOOHH-Nde_8	2741.0	1371.5	914.7	>90
2	Ac-HHOONNDD-Nde_8	2741.0	1371.5	914.7	>90
3	Ac-DHDHDHDH-Nde_8	2741.0	1371.5	914.7	>90
4	Ac-HDHDHDHD-Nde_8	2741.0	1371.5	914.7	>90
5	Ac-DDHHDDHH-Nde_8	2741.0	1371.5	914.7	>90
6	Ac-HHDDHHDD-Nde_8	2741.0	1371.5	914.7	>90

[a]Mass was obtained from LC-MS. [b]Purity was determined from LC-MS. *N*-decyl glycine is denoted as D and *N*-heptyl glycine is denoted as H.

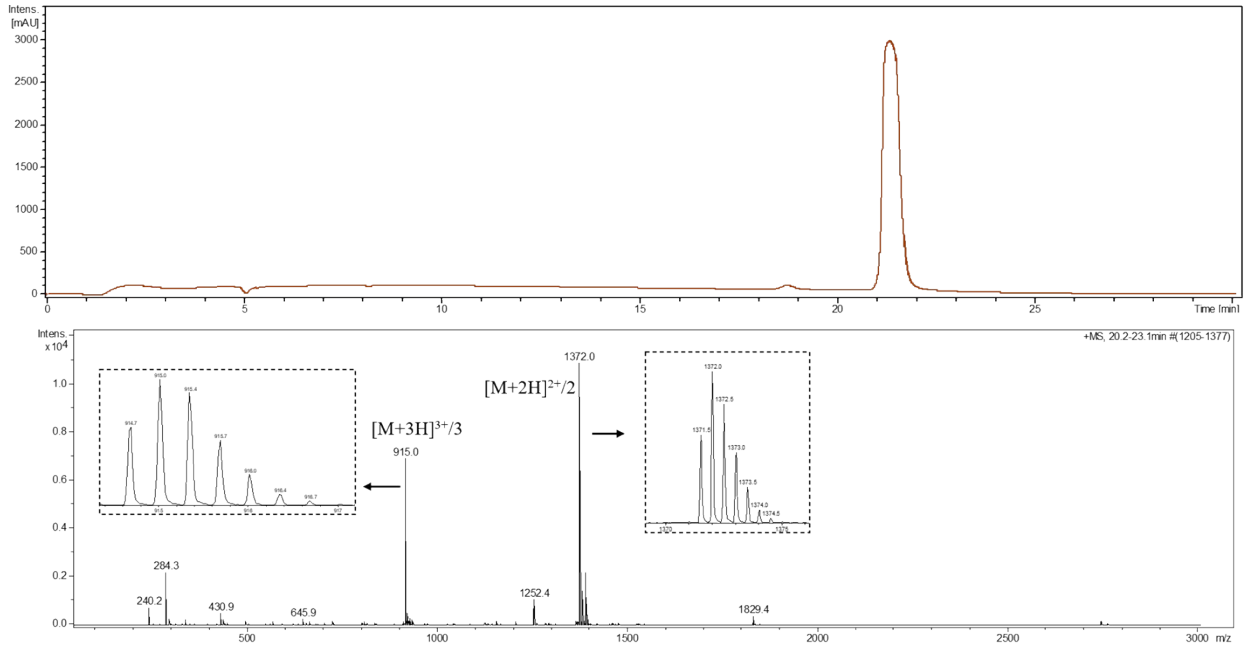

Figure S1. LC-MS trace of purified DDNNOOHH-Nde$_8$ (**1**) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of **1** (top image) and mass spectrum of **1** (bottom image).

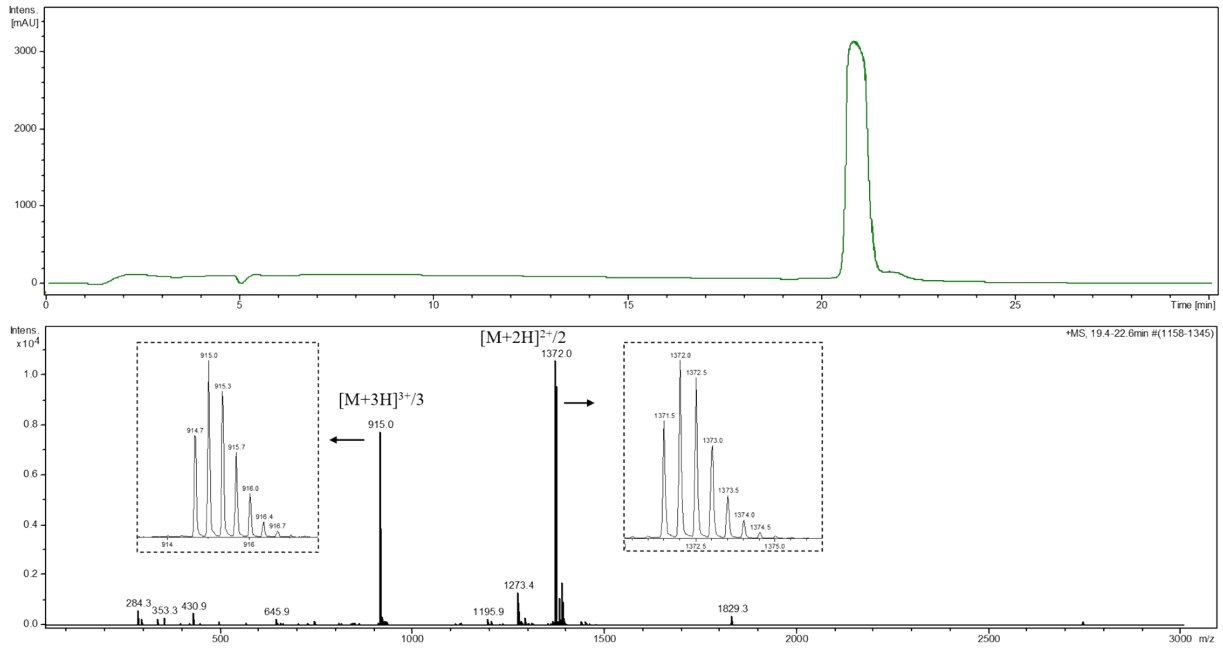

Figure S2. LC-MS trace of purified HHOONNDD-Nde$_8$ (**2**) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of **2** (top image) and mass spectrum of **2** (bottom image).

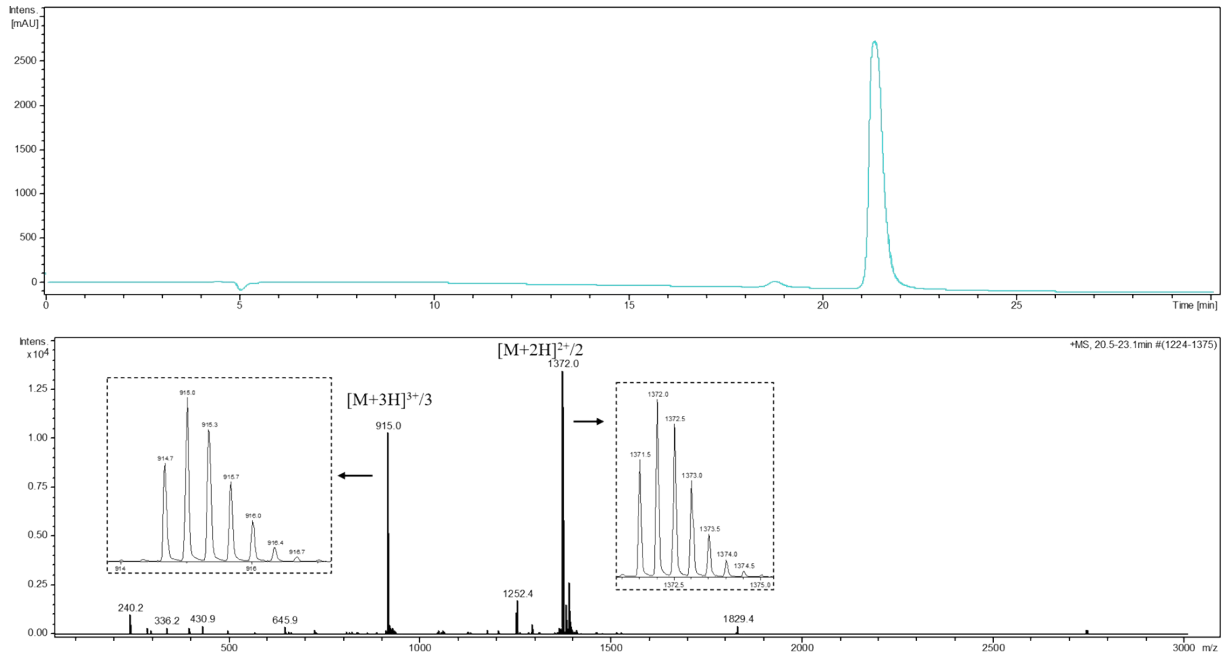

Figure S3. LC-MS trace of purified DHDHDHDH-Nde$_8$ (**3**) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of **3** (top image) and mass spectrum of **3** (bottom image).

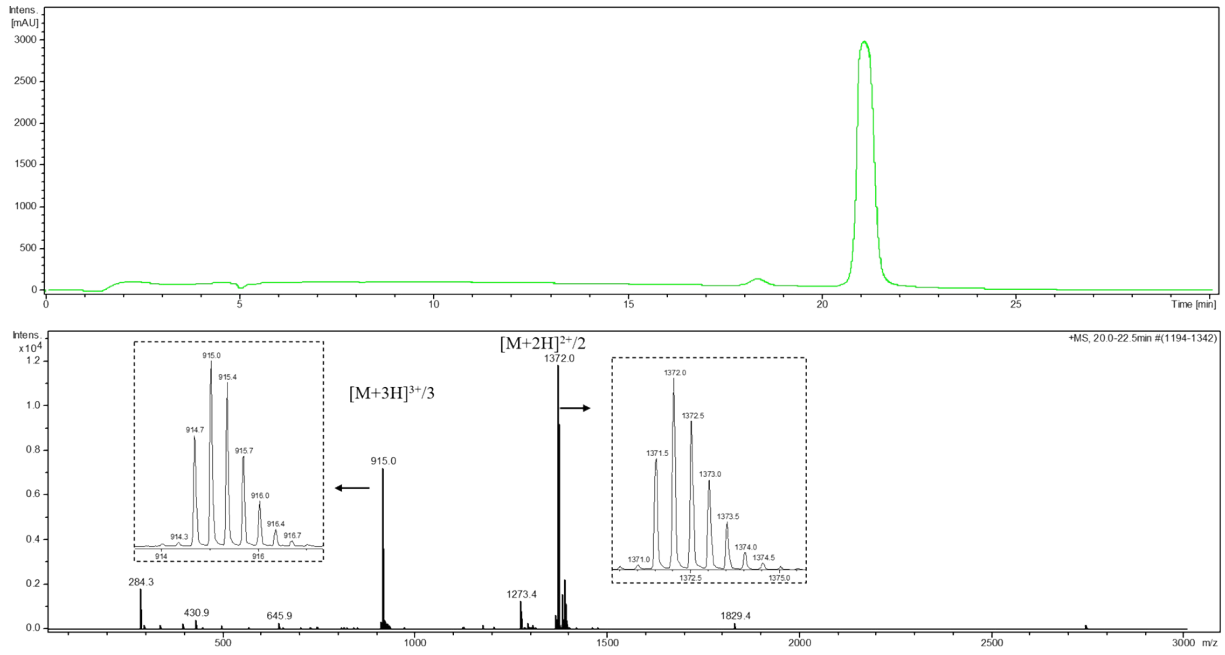

Figure S4. LC-MS trace of purified HDHDHDHD-Nde$_8$ (**4**) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of **4** (top image) and mass spectrum of **4** (bottom image).

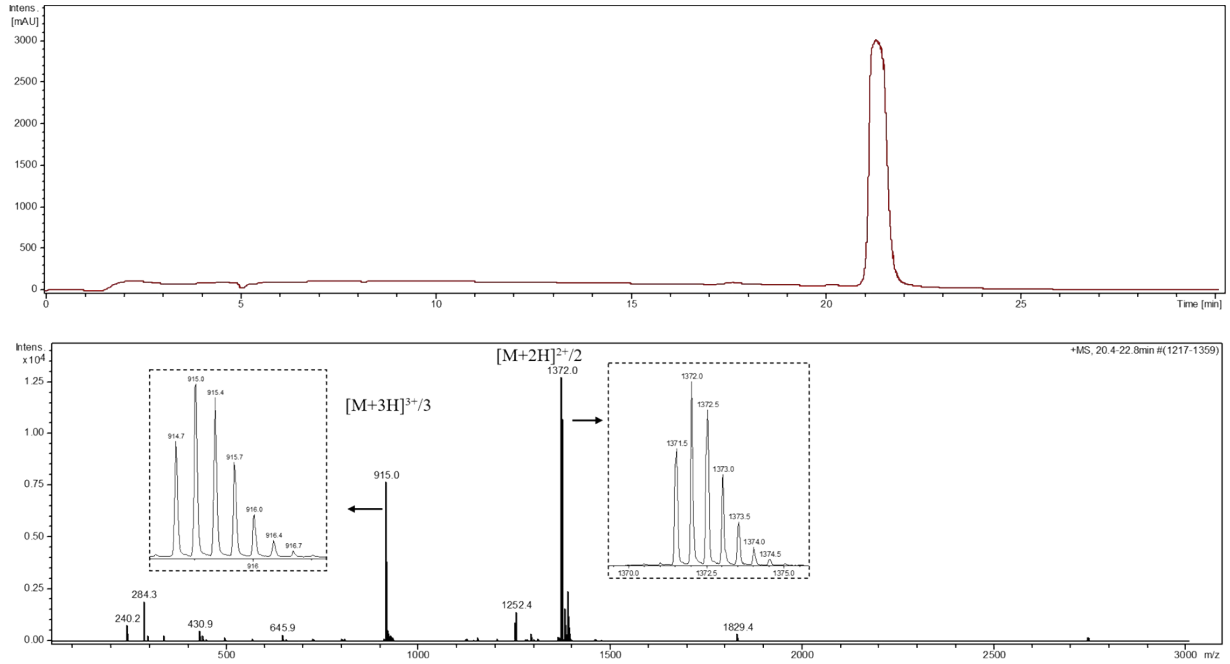

Figure S5. LC-MS trace of purified DDHHDDHH-Nde$_8$ (**5**) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of **5** (top image) and mass spectrum of **5** (bottom image).

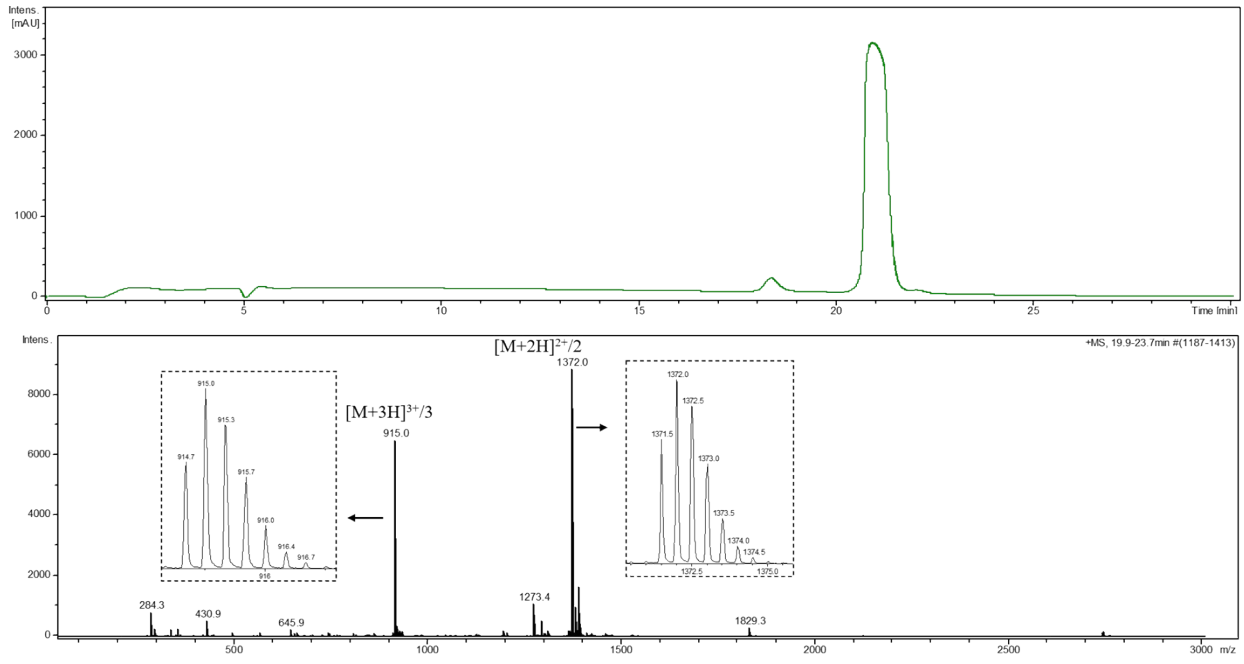

Figure S6. LC-MS trace of purified HHDDHHDD-Nde$_8$ (**6**) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of **6** (top image) and mass spectrum of **6** (bottom image).

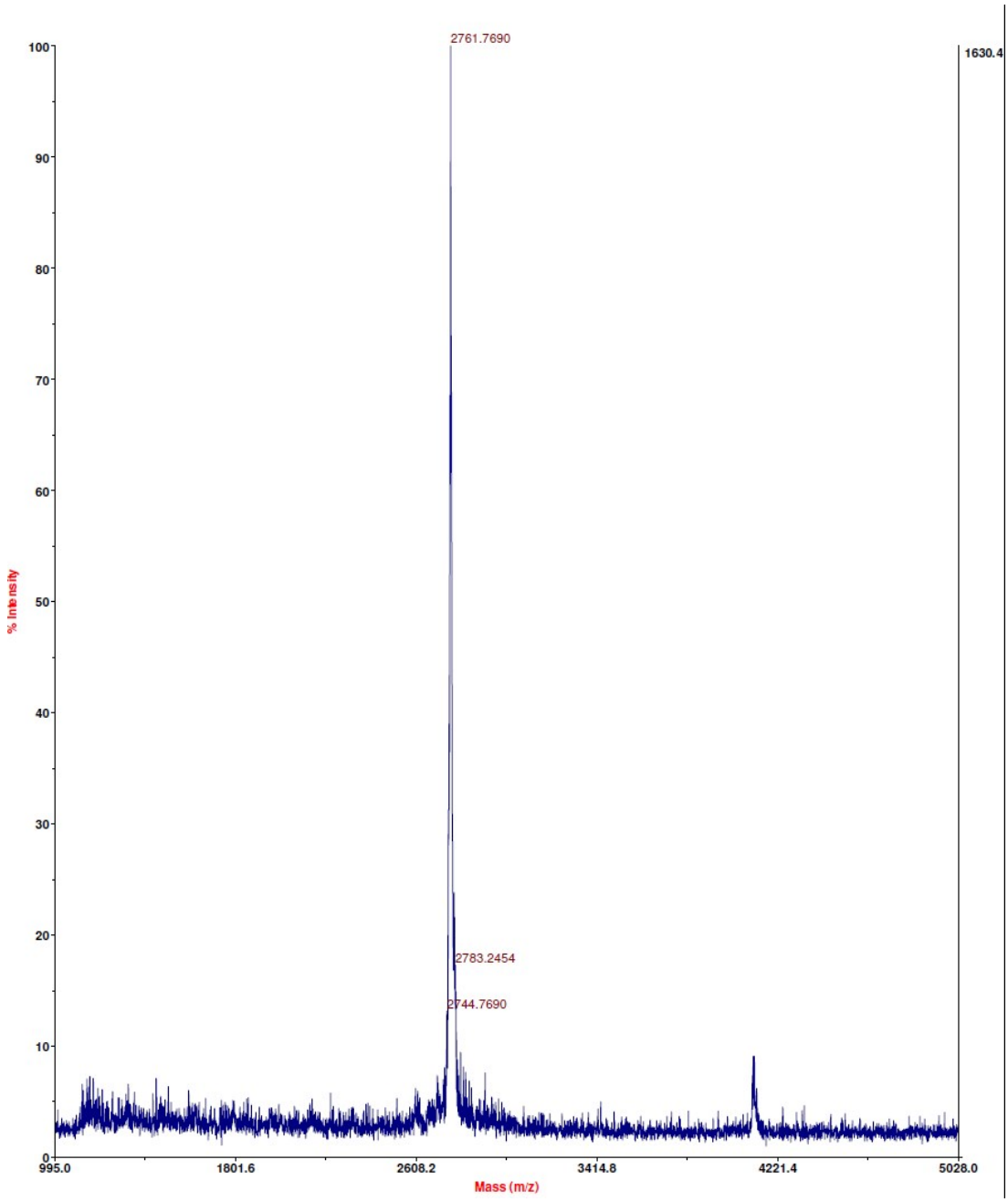

Figure S7. MALDI-TOF spectrum of Ac-DDNNOOHH-Nde_8 (**1**).

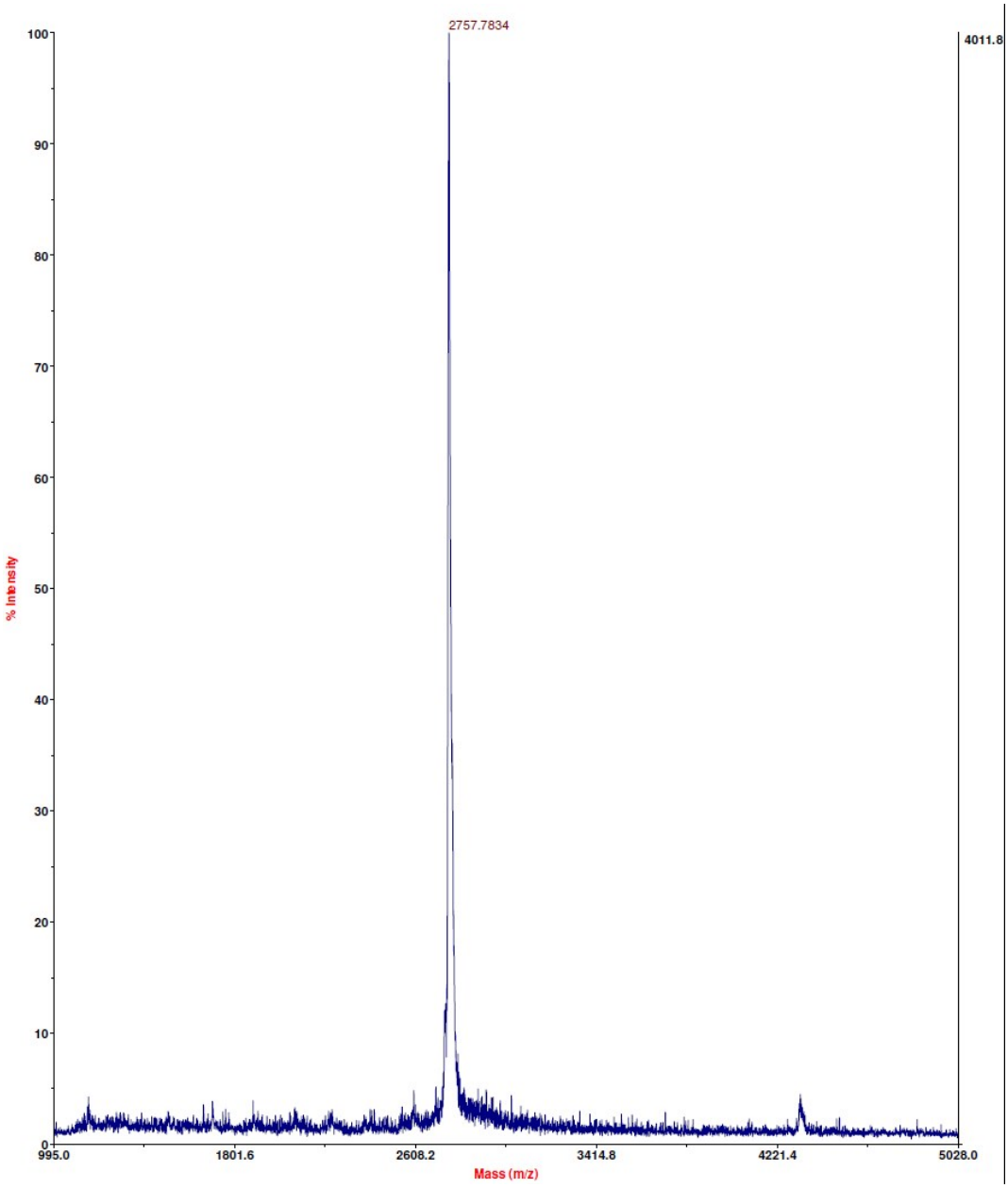

Figure S8. MALDI-TOF spectrum of Ac-HHOONNDD-Nde_8 (**2**).

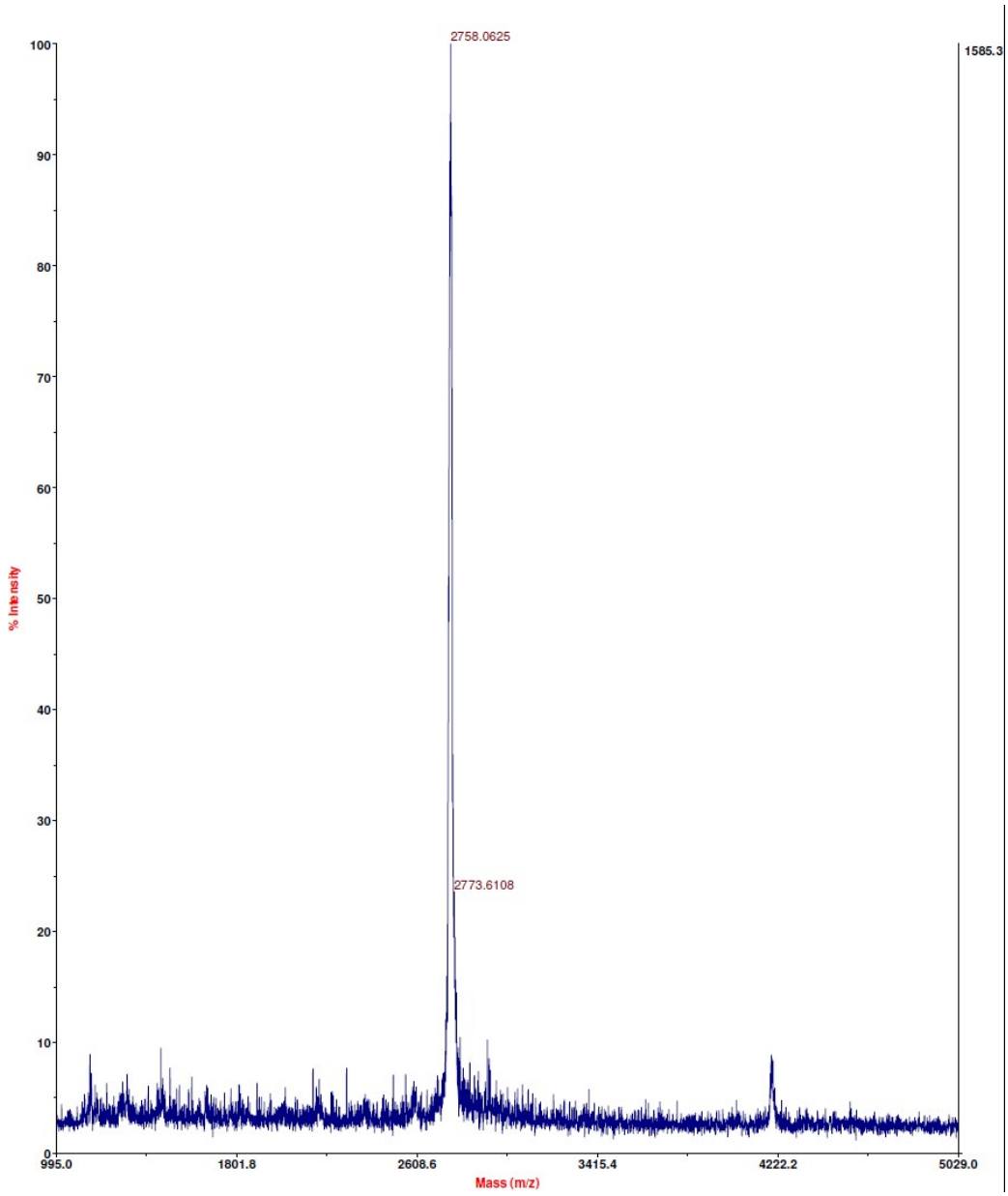

Figure S9. MALDI-TOF spectrum of Ac-DHDHDHDH-Nde_8 (**3**).

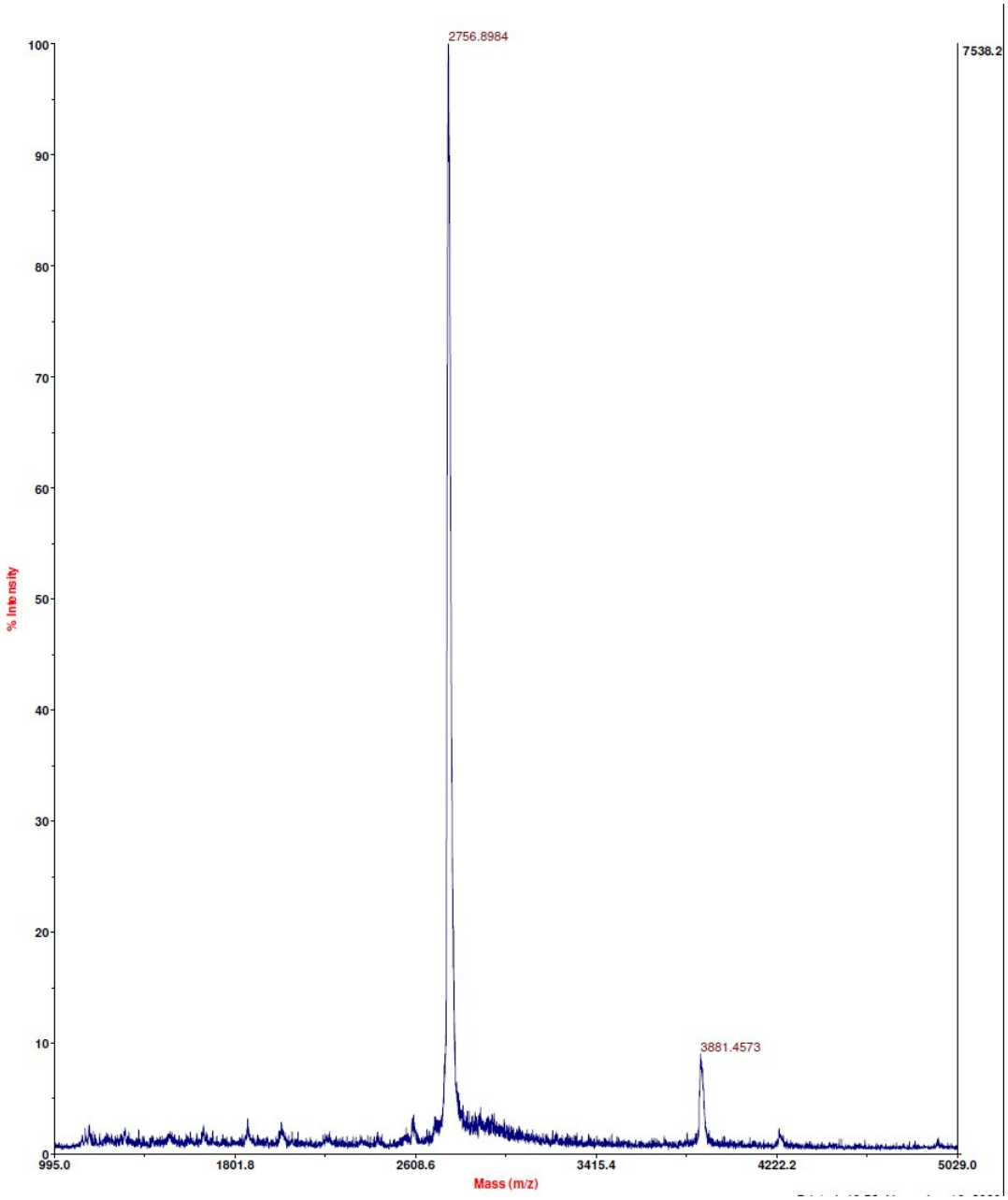

Figure S10. MALDI-TOF spectrum of Ac-HDHDHDHD-Nde_8 (**4**).

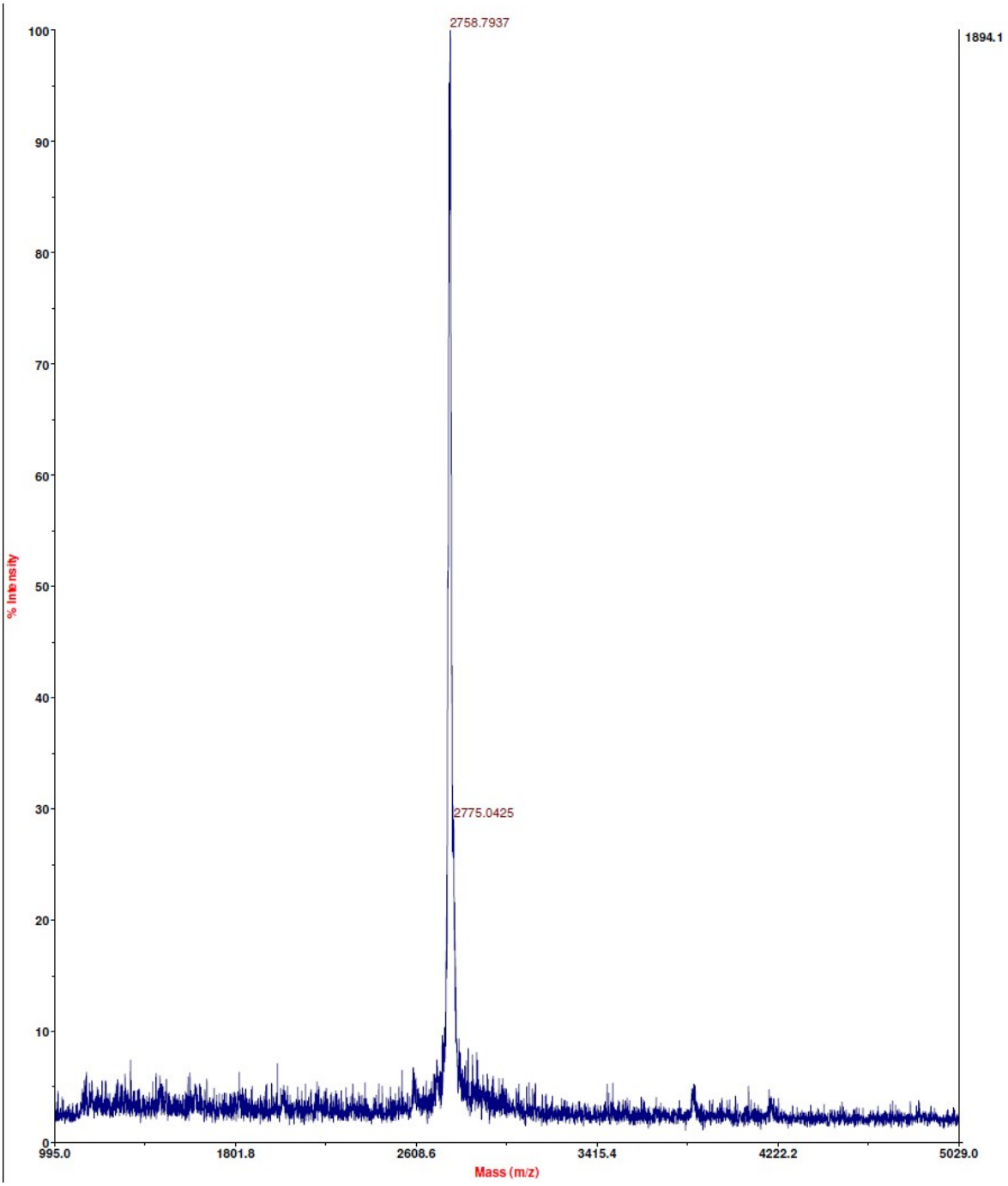

Figure S11. MALDI-TOF spectrum of Ac-DDHHDDHH-Nde_8 (**5**).

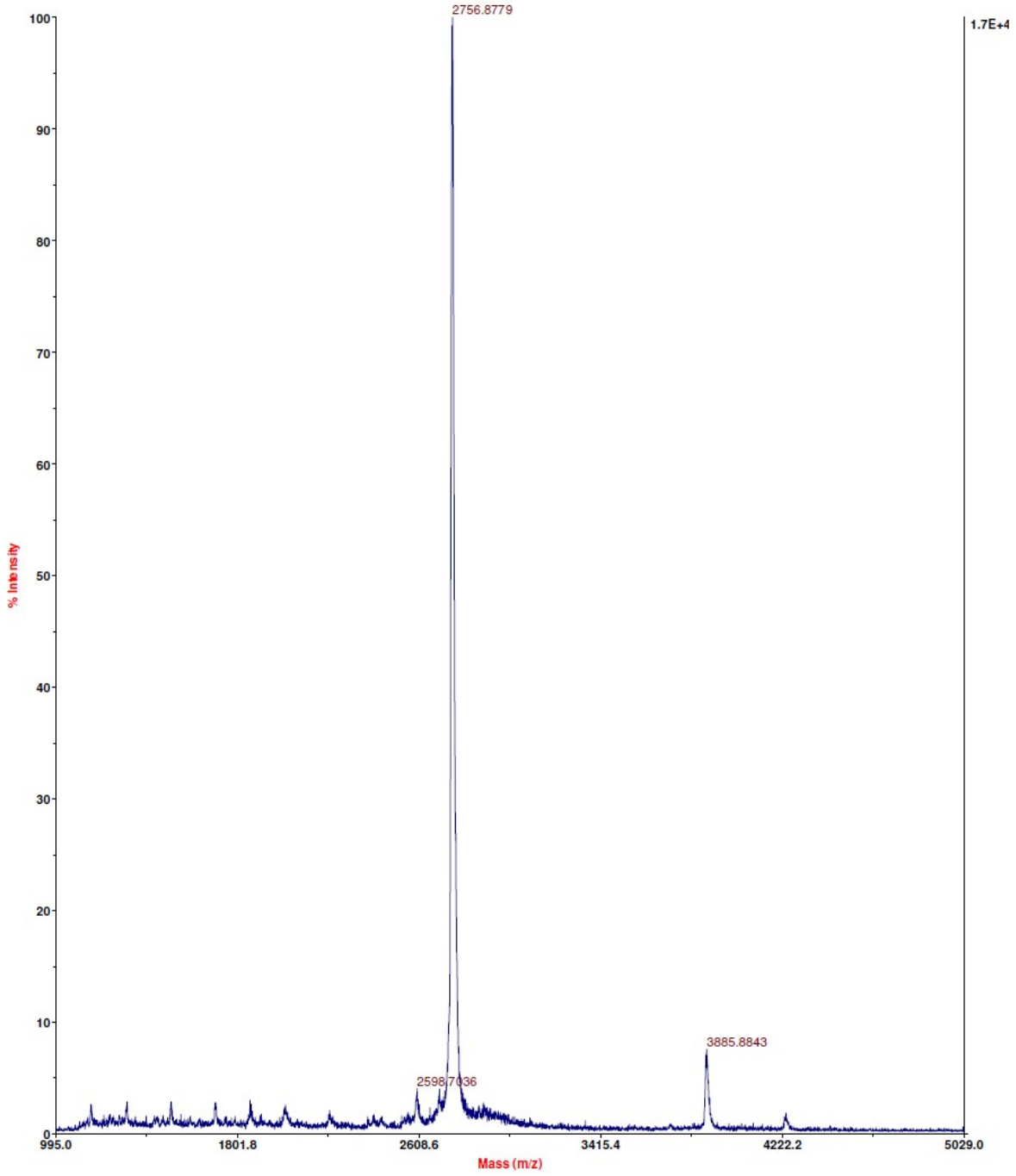

Figure S12. MALDI-TOF spectrum of Ac-HHDDHHDD-Nde_8 (**6**).

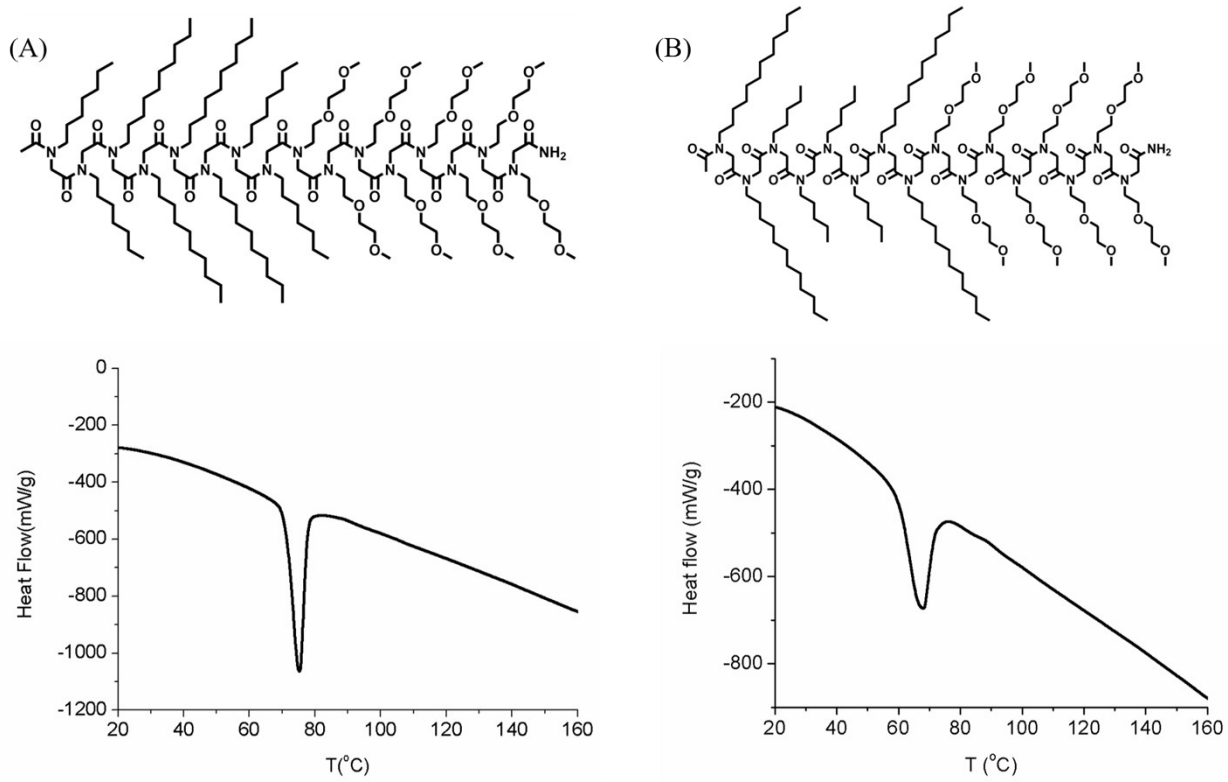

Figure S13. Chemical structures and DSC traces of peptoids with non-complementary molecular shapes: (A) Ac-HHDDDDHH-Nde_8 and (B) Ac-$D_dD_dPPPPD_dD_d$-Nde_8. *N*-decyl glycine is denoted as D, *N*-heptyl glycine is denoted as H, *N*-pentyl glycine is denoted as P, *N*-dodecyl glycine is denoted as D_d.

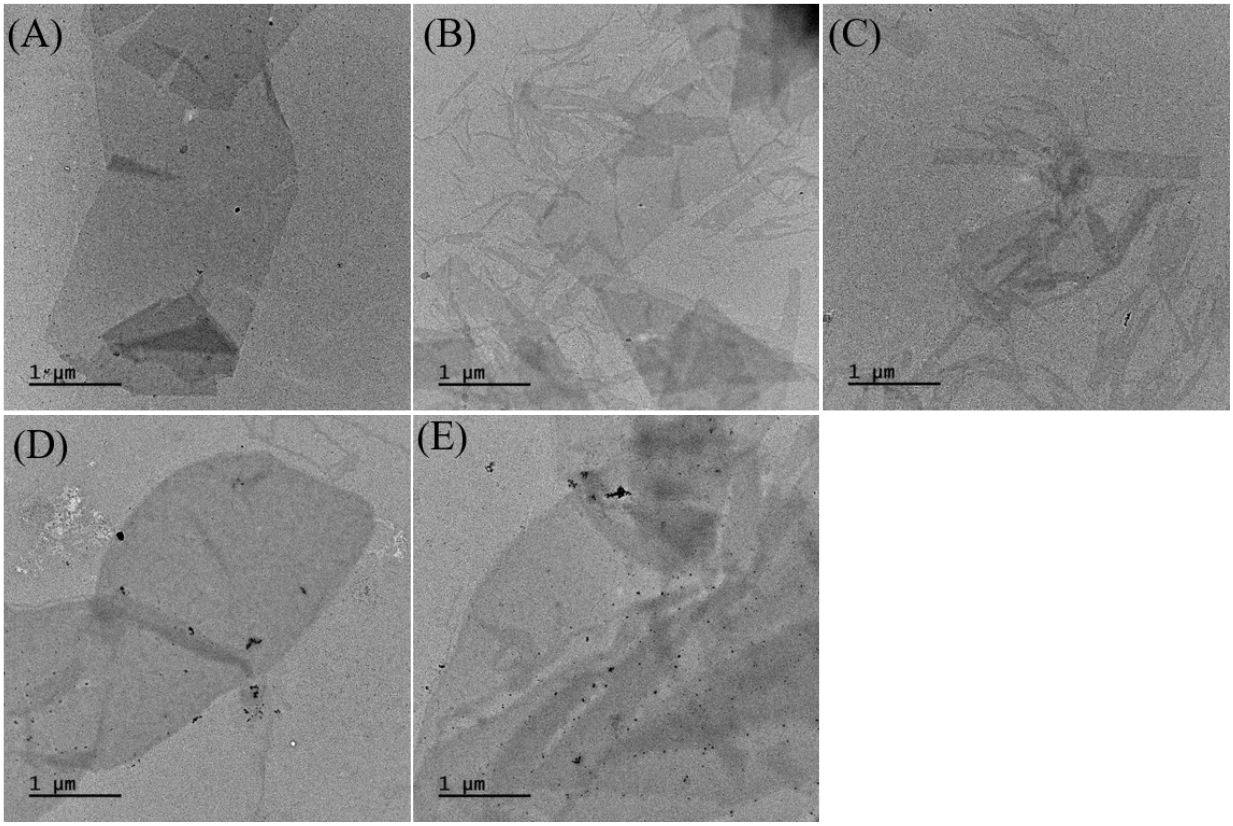

Figure S14. TEM images of sheet **2-6** (A-E).

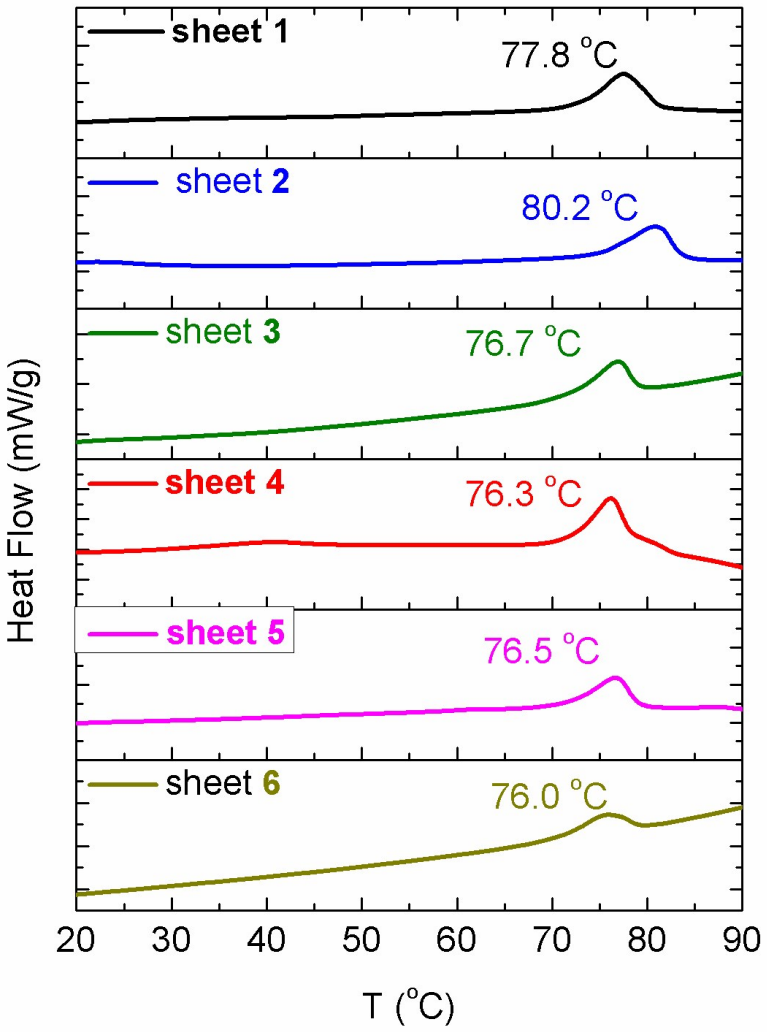

Figure S15. NanoDSC traces of diluted nanosheets in milliQ water at 2 mg/mL.

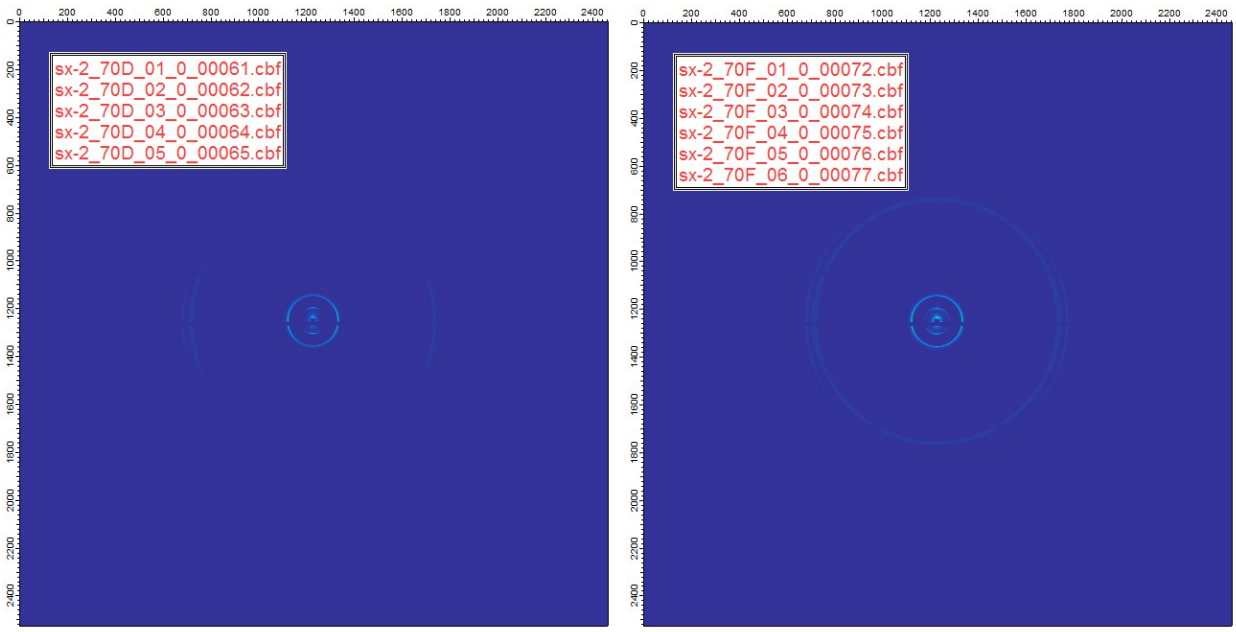

Figure S16. Diffraction patterns of sheet **3** and **4**.