Supporting Information

Manifold of Self-Assembly of a De Novo Designed Peptide: Amyloid Fibrils, Peptide Bundles, and Fractals

Yu-Jo Chao, Kan Wu, Hsun-Hui Chang, Ming-Jou Chien, and Jerry Chun Chung Chan

Department of Chemistry, National Taiwan University, No. 1, Section 4, Roosevelt Road, Taipei, 10617, Taiwan.

FIGURES AND TABLE

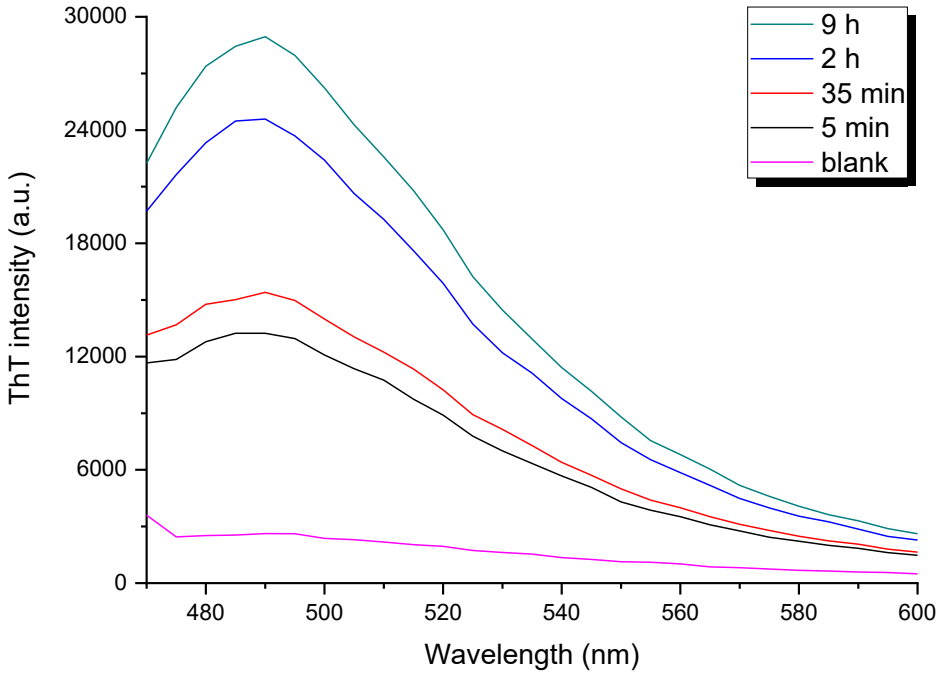

Figure S1: ThT results of the EASZ peptides at different incubation periods under the condition of pH 2.0. The rapid increase in ThT fluorescence is consistent with the fast kinetics of the aggregation of short amyloidogenic peptides.

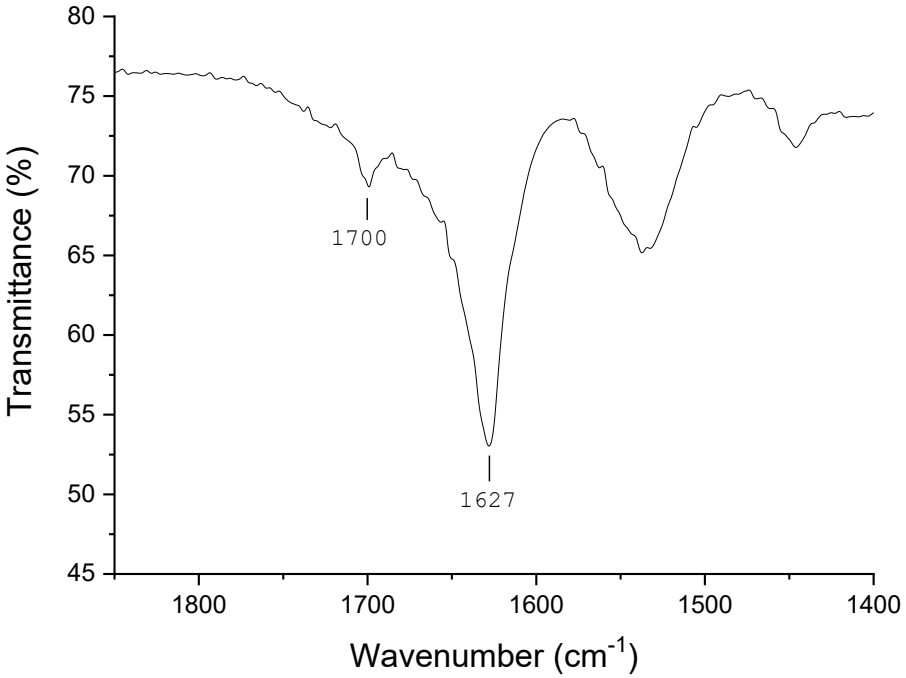

Figure S2: FT-IR spectrum of the EASZ peptides incubated under acidic conditions. The sample was recorded in a KBr pellet at room temperature. The splitting of the amide I band (mainly CO stretching) into a stronger and weaker bands at ca. 1627 and 1700 cm^{-1}, respectively, suggested that the EASZ peptides either adopt an anti-parallel β-sheet structure or a parallel β sheet with the neighboring parallel β sheets stacking in an antiparallel fashion.[1,2]

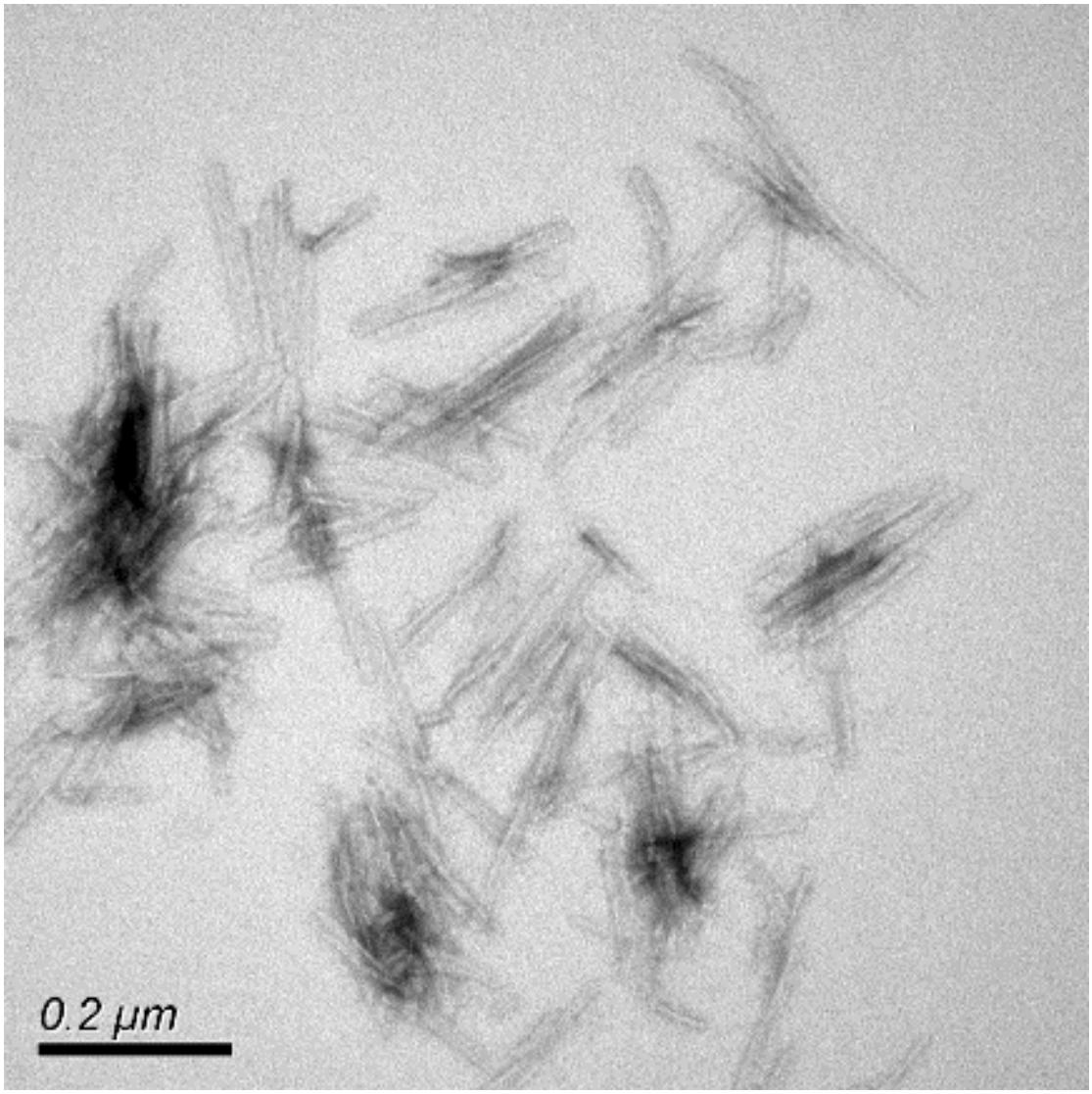

Figure S3: TEM images of the fibrillar aggregates formed by the EASZ peptides under acidic conditions. The sample was stained with uranyl acetate.

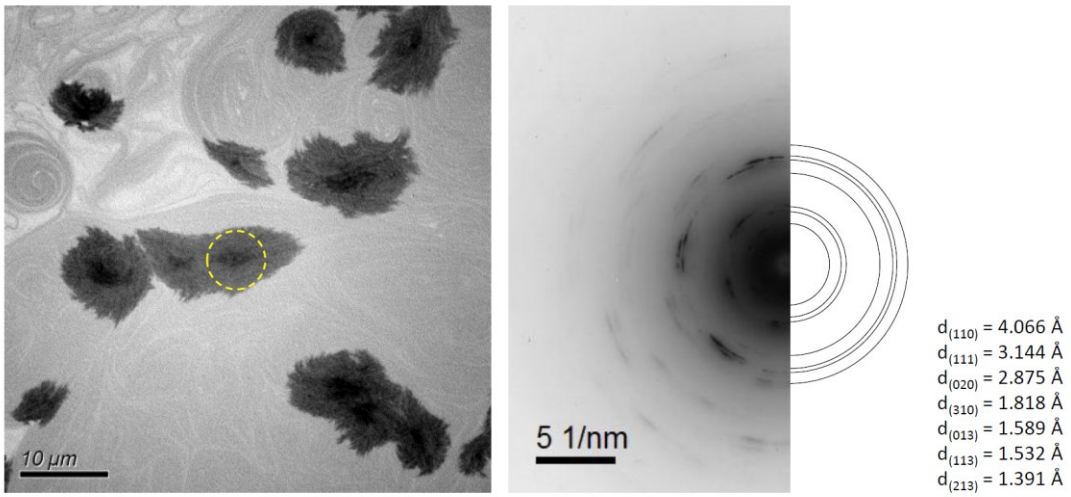

Figure S4: Unstained TEM image (left) and SAED pattern (right) of the EASZ peptides after incubating under the atmosphere of NH_3 only. The SAED pattern, which was taken for the area highlighted by a dashed circle, was assigned with reference to the crystal structure of ammonium nitrate (IV).[3] The pH adjustment was carried out with $HNO_{3(aq)}$.

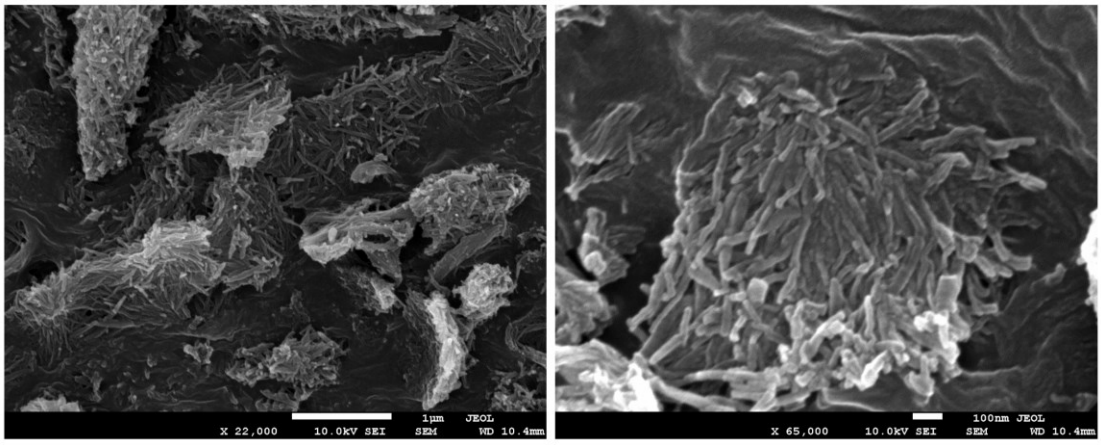

Figure S5: SEM images of the peptides after lyophilizing the solution of the EASZ peptides in the buffer of sodium bicarbonate at different magnifications.

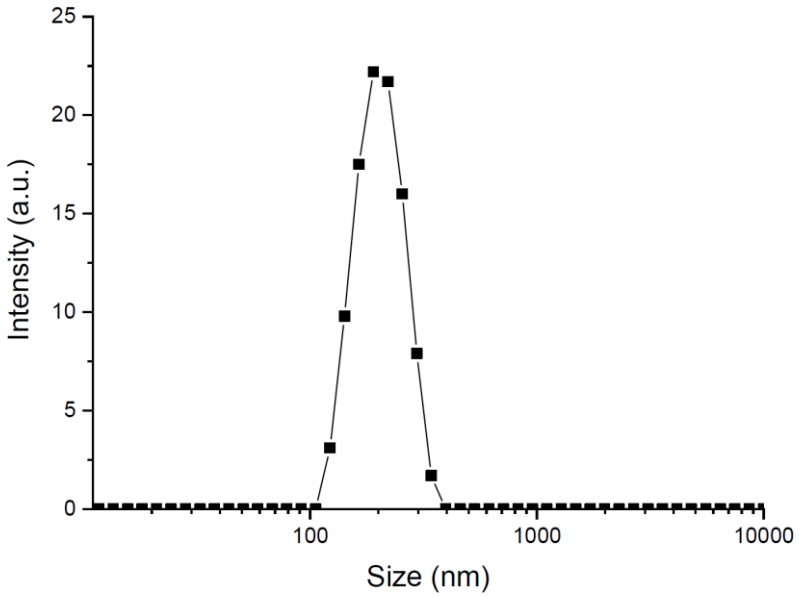

Figure S6: DLS results of the solution of EASZ peptides after dialysis against the buffer of sodium bicarbonate (20 mM, pH 9.0). Because the PdI was as high as 0.42, the data merely provided a qualitative indication for the presence of nanoaggregates.

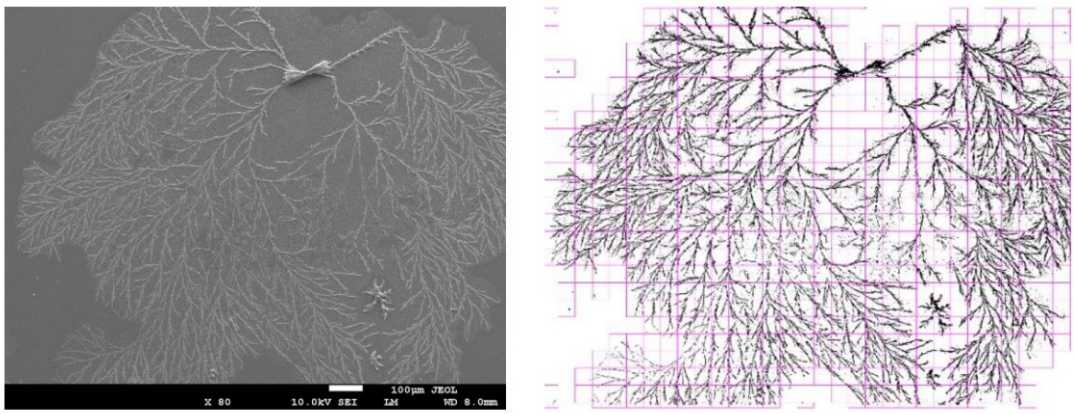

Figure S7: SED image (left) and the processed image for the determination of fractal dimension (right) of the EASZ peptides after dialysis against the buffer of sodium bicarbonate (20 mM, pH 9.0). The peptide solution was dried on a silicon wafer for the SEM measurements.

Table S1: ^{13}C chemical shifts extracted for the EASZ peptides at different aggregation states. The uncertainties in chemical shifts were 0.1−0.3 ppm.

Residues	Amyloid Fibrils (ppm)	Peptide Bundles (ppm)	Fractal Assembly (ppm)
Gly-CO	169.2	169.1	168.7
Gly-C_α	43.1	43.1	42.9
Ala-CO	172.8	172.5	172.4
Ala-C_α	49.5	49.4	49.5
Ala-C_β	21.2	20.9	20.7
Glu-CO	175.8	nd	175.7
Glu-C_α	52.6	53.7	53.3
Glu-C_β	30.0	30.0	31.5
Glu-C_γ	nd	34.5	35.3
Glu-C_δ	nd	181.5	180.4

nd: not determined.

REFERENCES

1 P. Walsh, K. Simonetti and S. Sharpel, Core Structure of Amyloid Fibrils Formed by Residues 106-126 of the Human Prion Protein, *Structure*, 2009, **17**, 417–426.

2 H.-M. Cheng, T. W. T. ; Tsai, W. Y. C. ; Huang, H.-K. Lee, H.-Y. Lian, F.-C. Chou, Y. Mou and J. C. C. Chan, Steric Zipper Formed by Hydrophobic Peptide Fragment of Syrian Hamster Prion Protein, *Biochemistry*, 2011, **50**, 6815–6823.

3 S. B. Hendricks, E. Posnjak and F. C. Kracek, Molecular Rotation in the Solid State. The Variation of the Crystal Structure of Ammonium Nitrate with Temperature, *J. Am. Chem. Soc.*, 1932, **54**, 2766–2786.