

Supporting Information

Squid Suckerin Biomimetic peptides form Amyloid-like Crystals with Robust Mechanical Properties

Shu Hui Hiew¹, Antoni Sánchez-Ferrer², Shahrouz Amini¹, Feng Zhou¹, Jozef Adamcik², Paul Guerette¹, Haibin Su¹, Raffaele Mezzenga^{2} and Ali Miserez^{1,3*}*

¹ School of Materials Science and Engineering, Nanyang Technological University, Singapore 639798

² Department of Health Sciences & Technology, ETH Zurich, Switzerland CH-8092, CH-8093

³ School of Biological Sciences, Nanyang Technological University, Singapore 637551

* Authors for correspondence: ali.miserez@ntu.edu.sg; raffaele.mezzenga@hest.ethz.ch

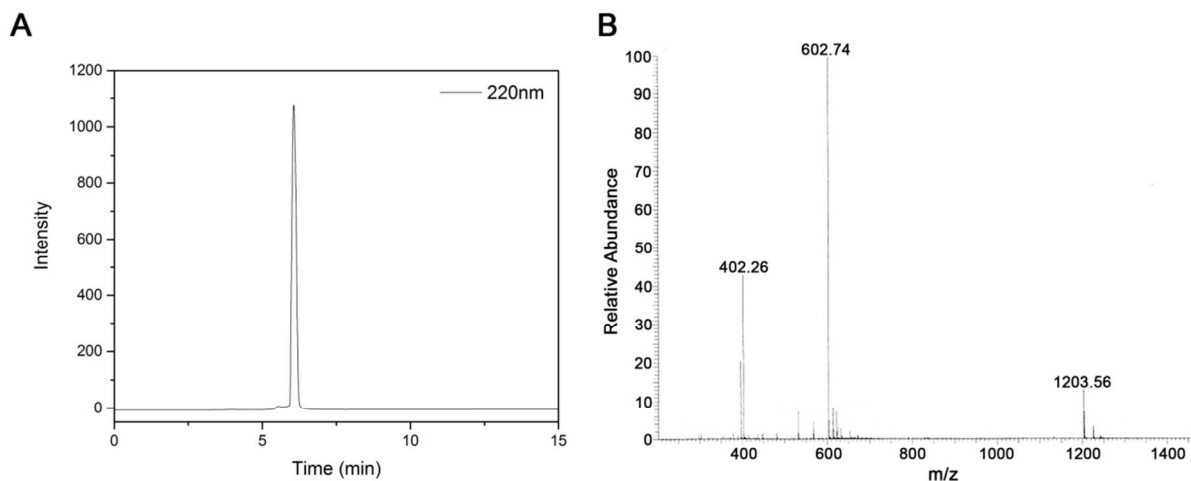


Figure S1. Quality analysis of synthesized peptide A1H1. (A) Trace analysis of purified peptide showing a sharp peak with >95% purity on a HPLC chromatogram. (B) LC/MS analysis of purified peptide showing the mass-charge adducts of the peptide, which has a calculated molecular mass of 1203.27 g/mol.

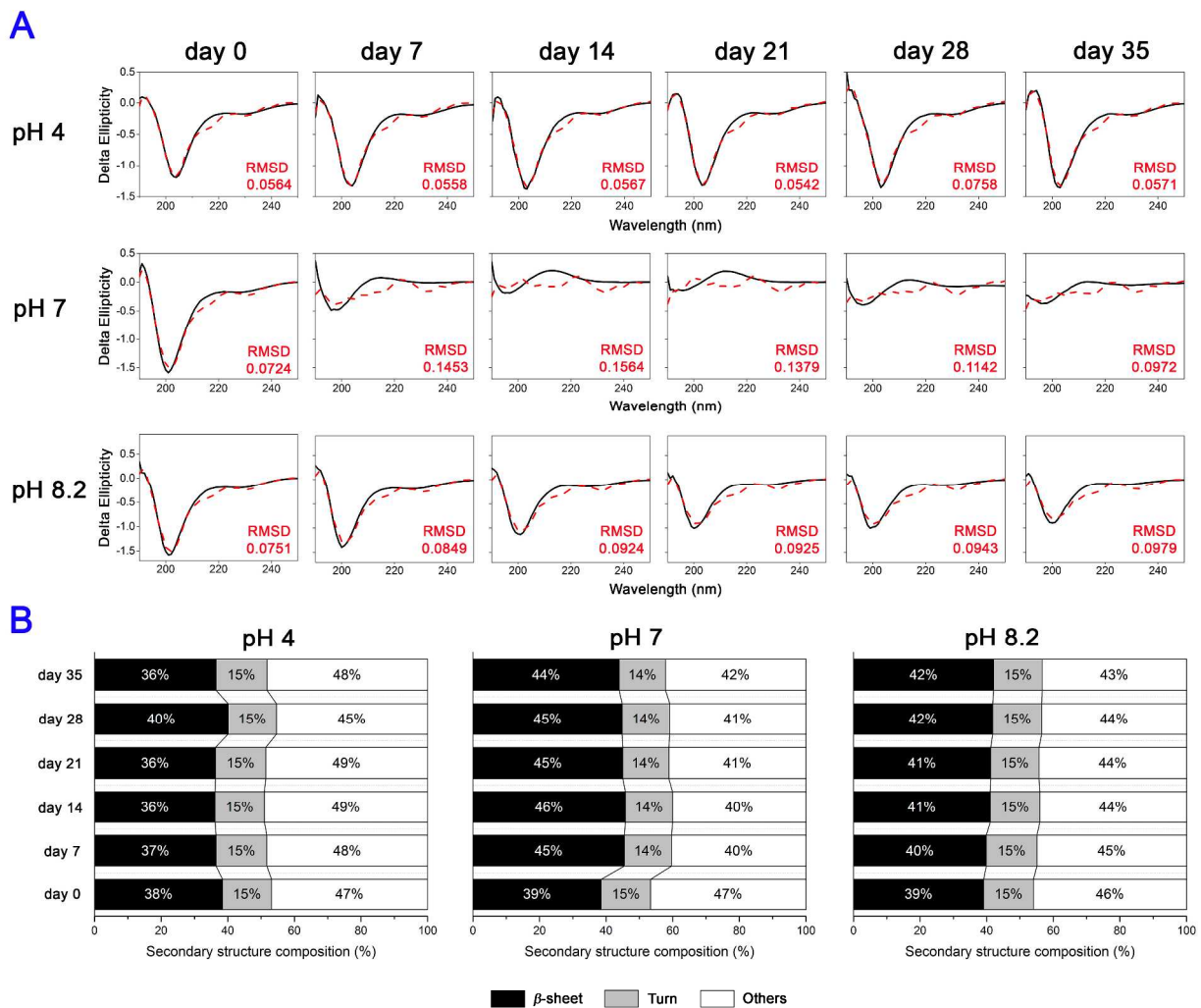


Figure S2. BeStSel fitting and deconvolution of 35-day time series CD spectra of peptide A1H1 in aqueous buffers. (A) Fitted and original CD spectra of A1H1 peptide solution incubated at pH 4, pH 7 and pH 8.2 respectively, with RMSD values indicated. (B) Secondary structure composition obtained from BeStSel deconvolution.

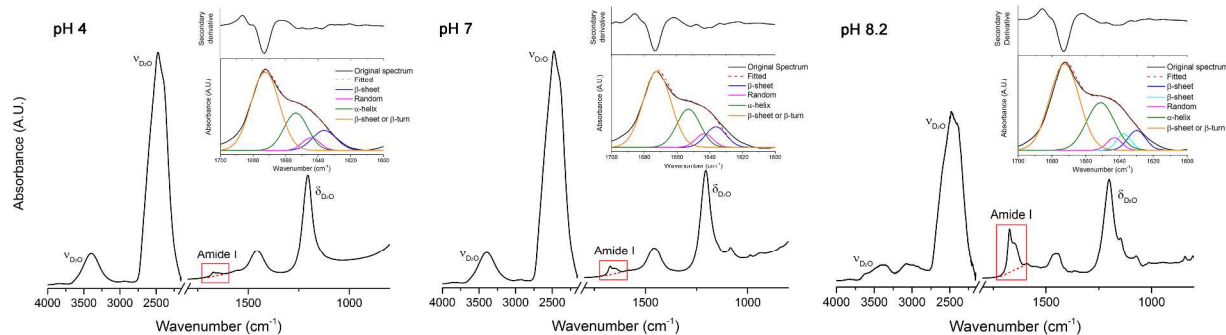


Figure S3. FTIR spectra of peptide A1H1 in D₂O buffers at 3 different pHs. Deconvolution of Amide I peaks was done via obtaining secondary derivative of the spectra after baseline correction.

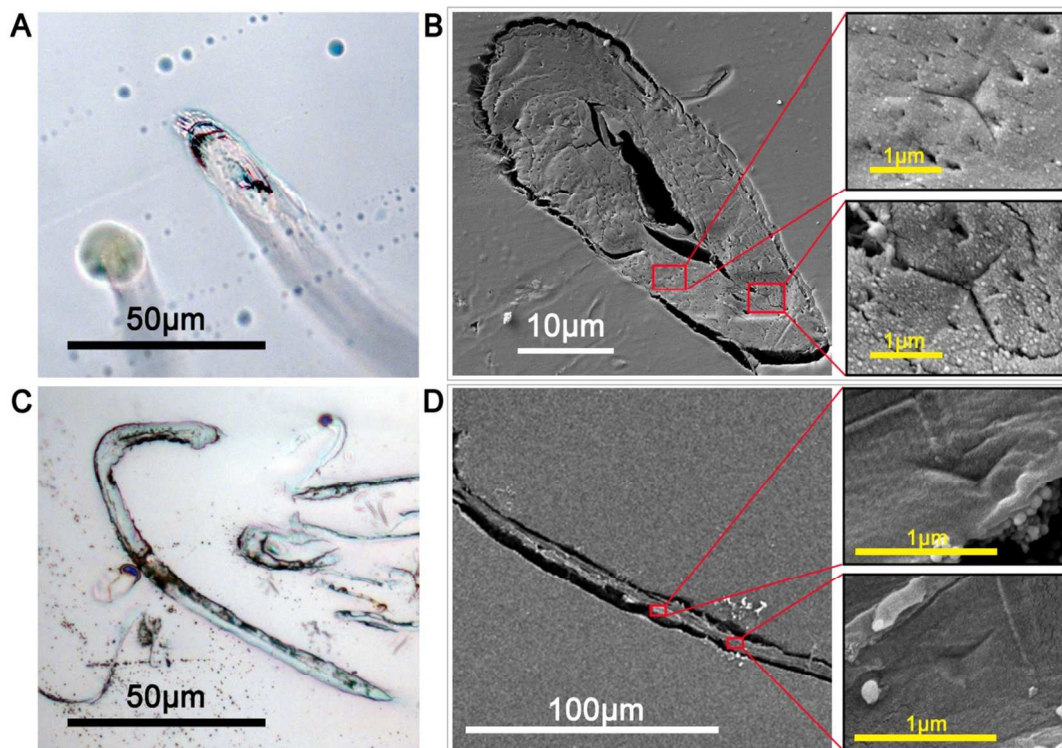


Figure S4. A1H1 peptide fibers embedded in epoxy. (A) Optical microscopic image of fiber cross section (xx direction), (B) SEM image of same fiber cross section, with magnified images indicated by red boxes, showing indentations performed on the fiber under dry state. (C) Optical microscopic image of fiber cross section (yy direction), (D) SEM image of same fiber cross section, with magnified images indicated by red boxes, showing indentations performed on the fiber under hydrated state.

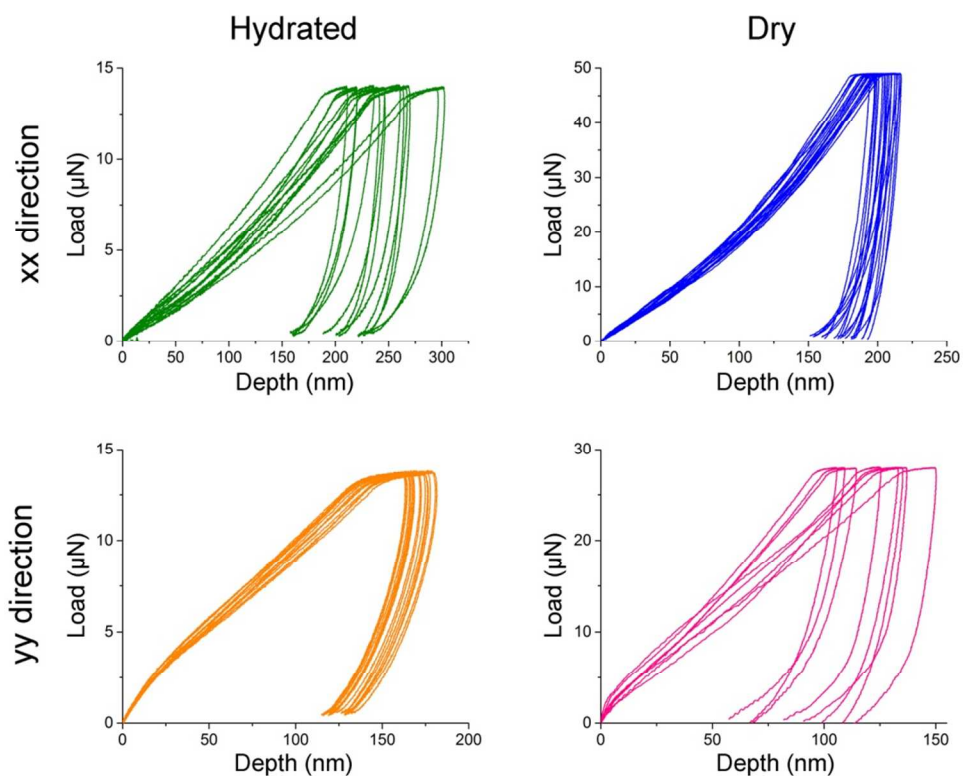


Figure S5. Loading and unloading curves obtained of indentations performed on A1H1 peptides in hydrated and dry states in the *xx* and *yy* directions.

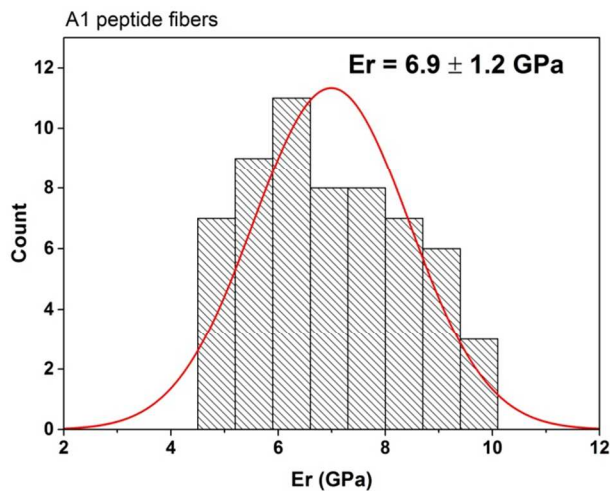


Figure S6. Indentations performed on short A1 peptide fibers under dry conditions. An average reduced elastic (E_r) modulus of 6.9 ± 1.21 GPa was obtained for the short fibers (A1: Ac-AATAVS-NH₂).

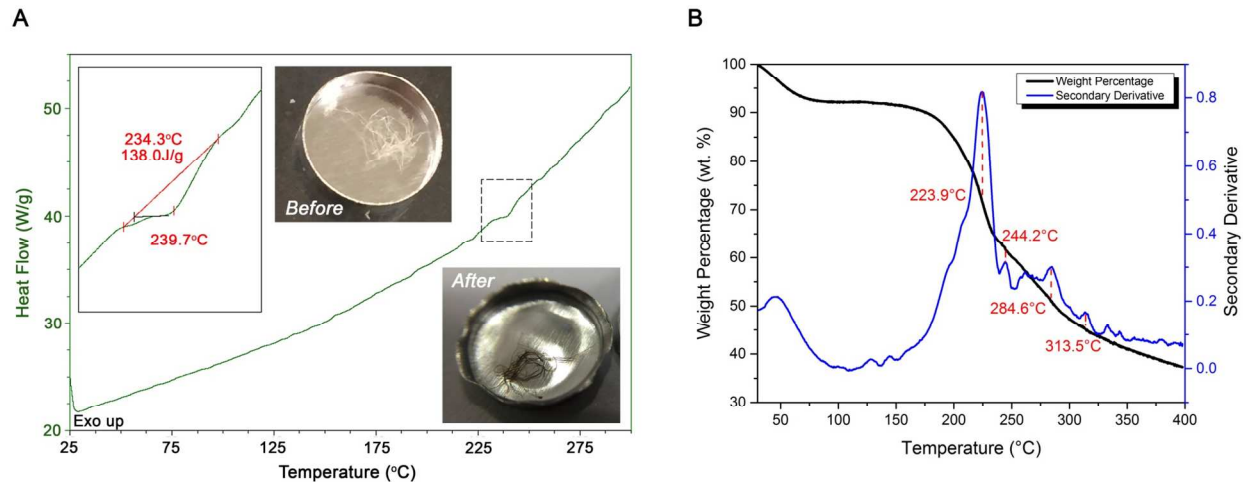


Figure S7. Thermal stability properties of A1H1 peptide fibers and powder. (A) DSC performed on dried A1H1 peptide fibers showing an endothermic peak with a minima at 239.66°C (T_m) and an energy requirement of 138 J/g to melting the fibers. Insets show the zoom-in region of endothermic peak and fibers before and after DSC experiment, in Tzero pans. (B) TGA scans was performed on the precursor A1H1 peptide powder. A degradation temperature of 223.9°C was obtained.

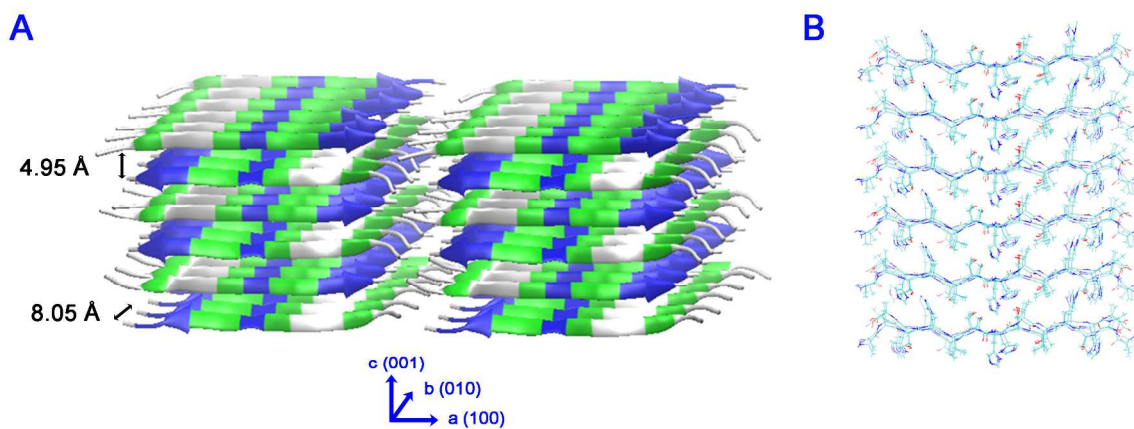


Figure S8. MD simulations of A1H1 peptides in an antiparallel configuration. The assembly, presented as (A) ribbon and (B) stick models, was allowed to relax for 200 ns before obtaining the inter-sheet (8.05 Å) and inter-strand (4.95 Å) distances.