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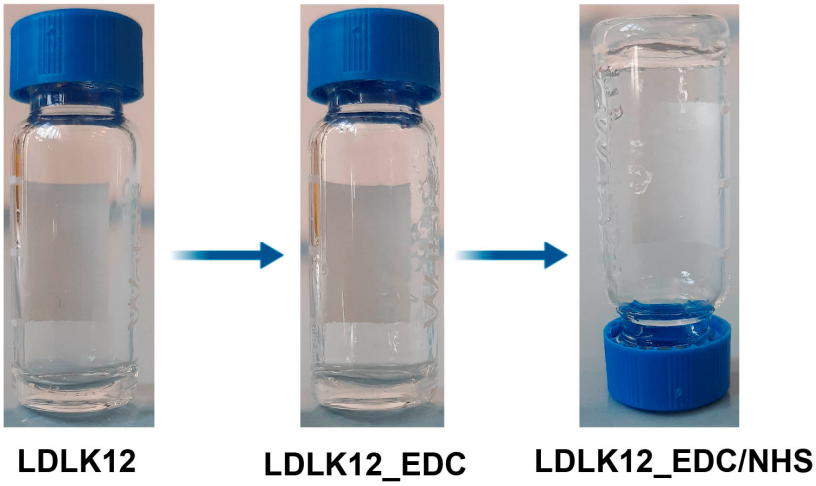
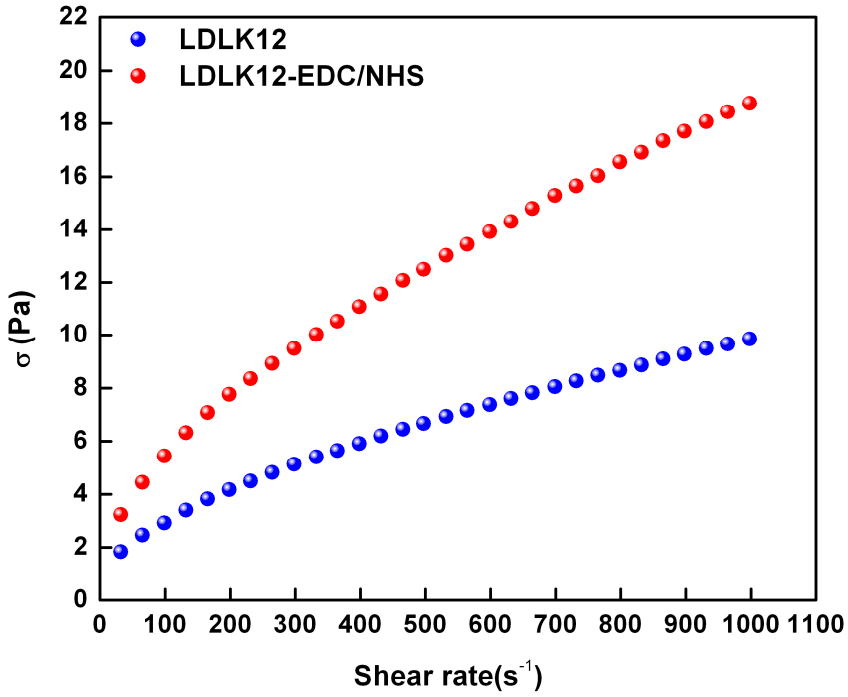
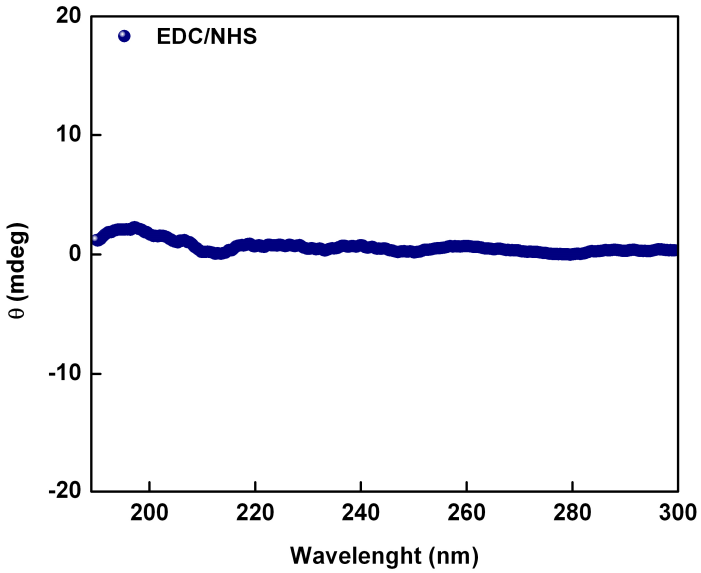
Supplementary Figures



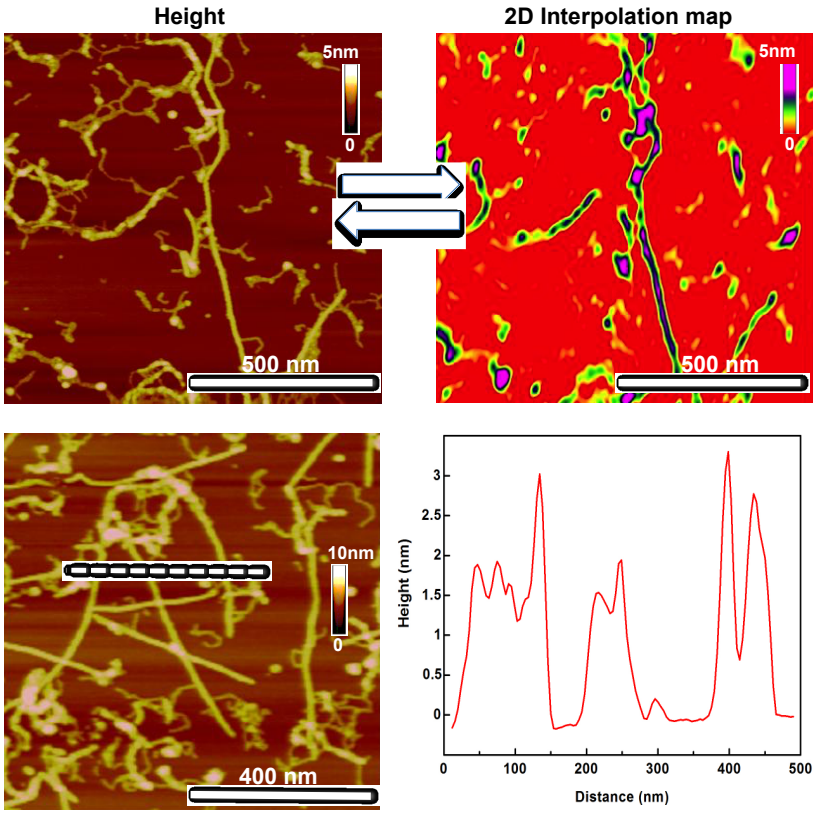

**Supplementary Figure S1: EDC/sulfo-NHS cross-linked LDLK12 peptide.** One-pot *in situ* LDLK12 (1% *w*/*v*) cross-linking via EDC/sulfo-NHS coupling: after cross-linking the self-supporting SAP scaffold can be appreciated.



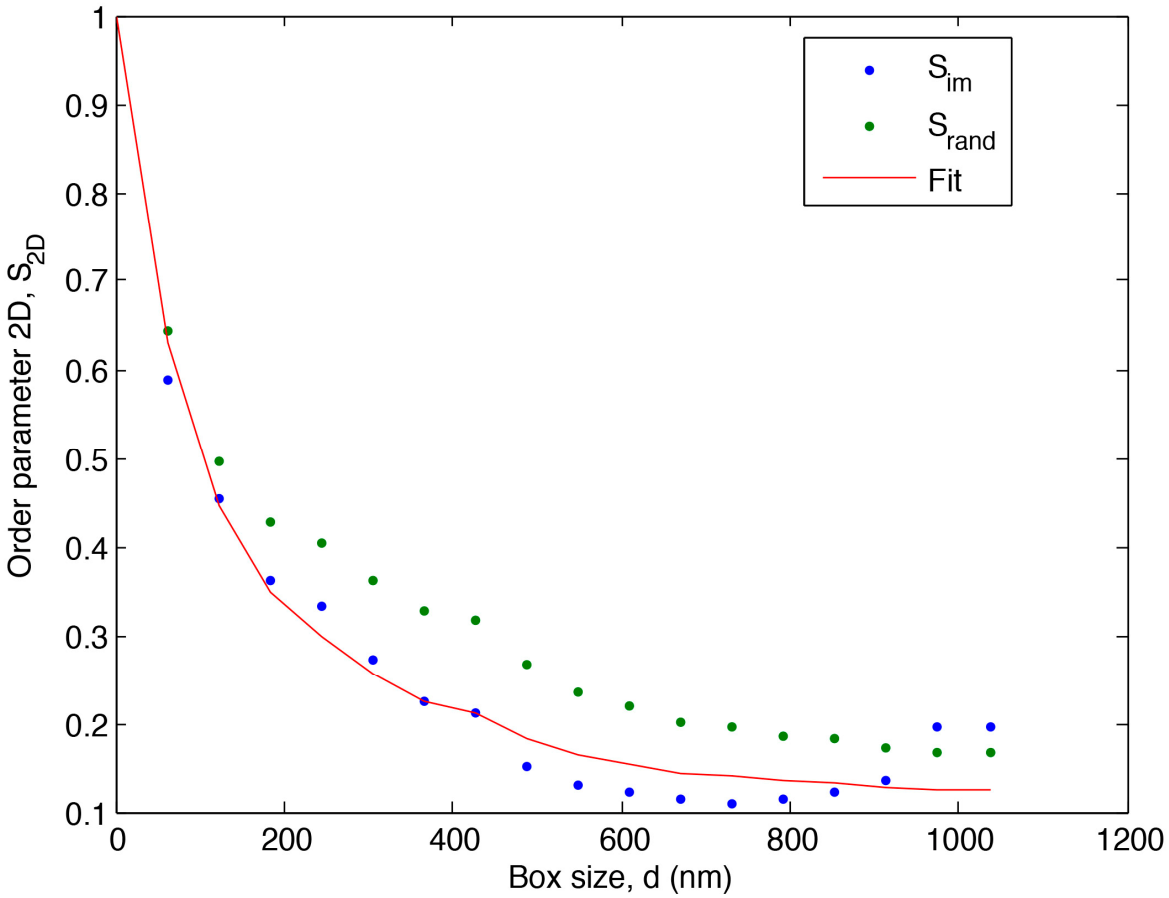

**Supplementary Figure S2: Shear stress (σ) measurements.** Shear stress (σ) measurements at increasing shear rate of the wildtype and cross-linked LDLK12 peptides. Both SAPs exhibited non-Newtonian shear-thinning behavior.



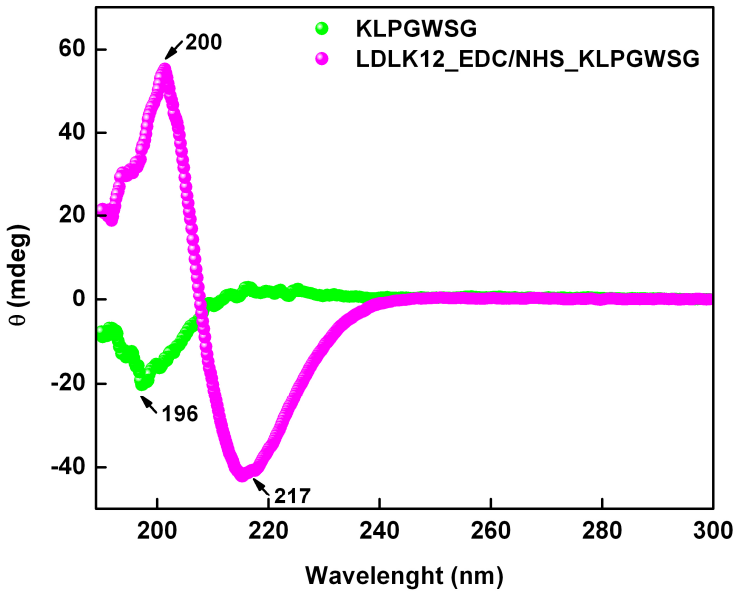

**Supplementary Figure S3: CD spectrum of EDC/sulfo-NHS.** No CD signal was observed in EDC/sulfo-NHS alone, suggesting no assembly propensity for the cross-linker alone.



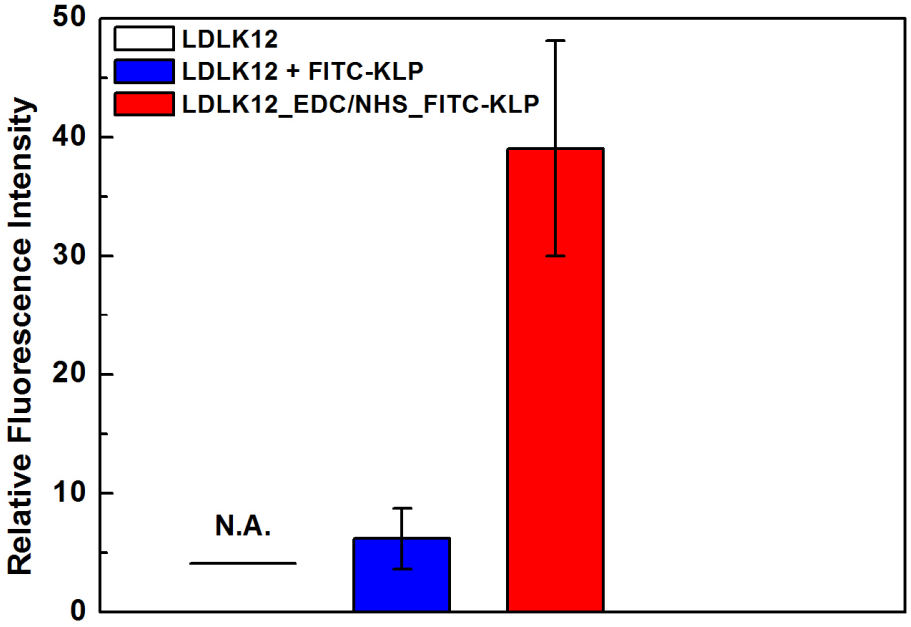

**Supplementary Figure S4: 2D heights interpolation map.** 2D heights interpolation map and height measurements of EDC/sulfo-NHS cross-linked LDLK12 peptide.




**Supplementary Figure S5: Nematic order parameter.** Nematic order parameter of cross-linked LDLK12 nanostructured scaffold, obtained by FiberApp software [60,61]. To calculate the 2D order parameter (i.e. $S_{2D}$) we divided the whole AFM image into square blocks of a certain size ($d$). Calculating and averaging $S_{2D}$ values for all blocks results in one mean number, which is parametric with ($d$), yielding to the length scale-dependent $S_{2D}(d)$[62]. $S_{2D}(d)$ function is further expressed as sum of the weighted components $S_{2D}^{align}$ and $S_{2D}^{rand}$, corresponding to the alignment of the nematic and isotropic components, respectively: $S_{2D}(d) = a S_{2D}^{align}(d) + (1 - a)\, S_{2D}^{rand}(d)$; where $a$ is the relative surface fraction of the aligned (nematic) domains. By applying this analysis to the tracked nanofibrils, it is possible to quantify isotropic–nematic transitions rigorously.[61, 62].




**Supplementary Figure S6: CD spectra of KLPGWSG and LDLK12_EDC/NHS_KLPGWSG.** LDLK12 peptide after the EDC/sulfo-NHS reaction with KLPGWSG showed the presence of β-sheet secondary structures, while the KLPGWSG alone showed an unstructured conformation.




**Supplementary Figure S7: FITC-KLPGWSG fluorescence intensity.** Quantification of FITC-KLPGWSG fluorescence intensity on LDLK12 peptide nanostructures. EDC/sulfo-NHS-mediated conjugation of FITC-KLPGWSG peptide to LDLK12 nanofibers showed higher fluorescence intensity compared to the non-specific adsorption of FITC-KLPGWSG to nanofibers of standard LDLK12. As expected, LDLK12 alone did not show any detectable signal.

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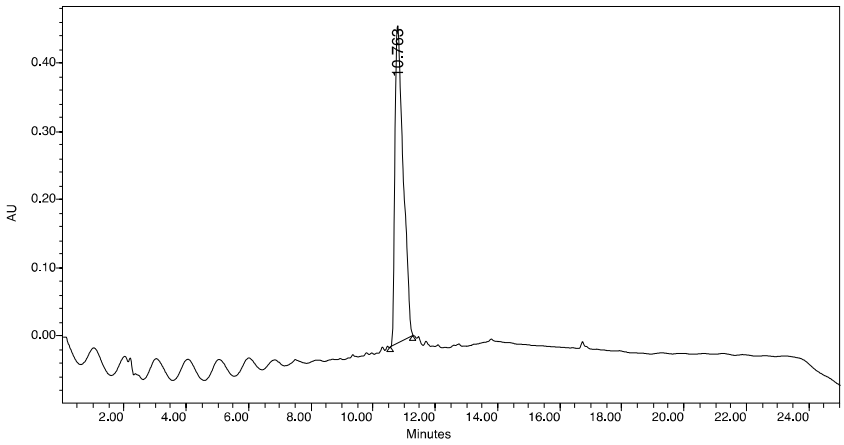
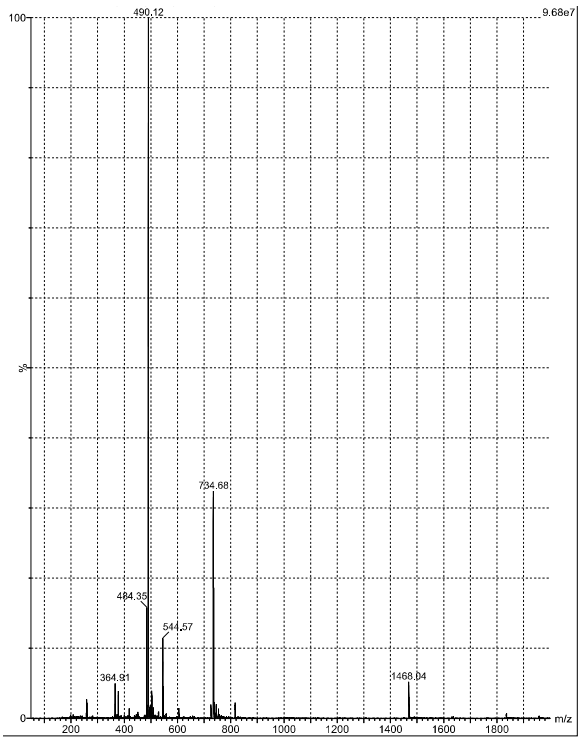
MDPI

**Ac-LDLKLDLKLDLK-CONH$_2$**



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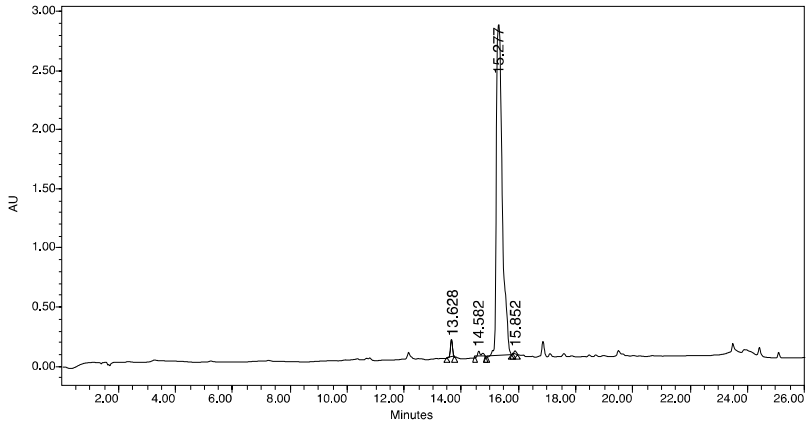
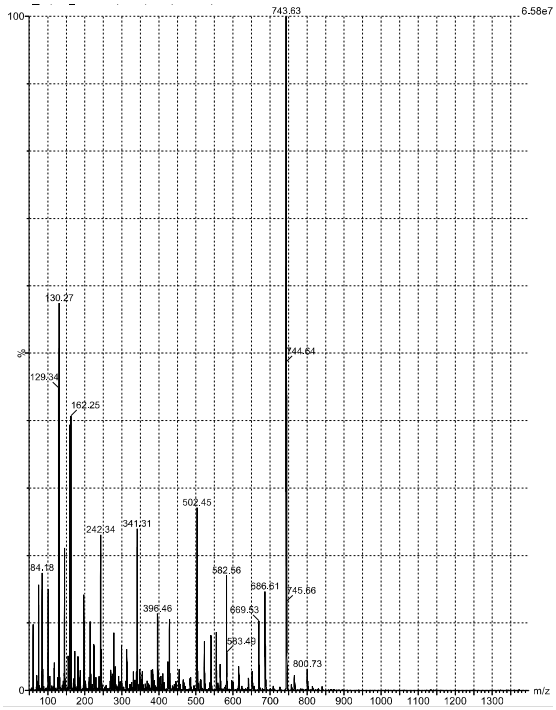
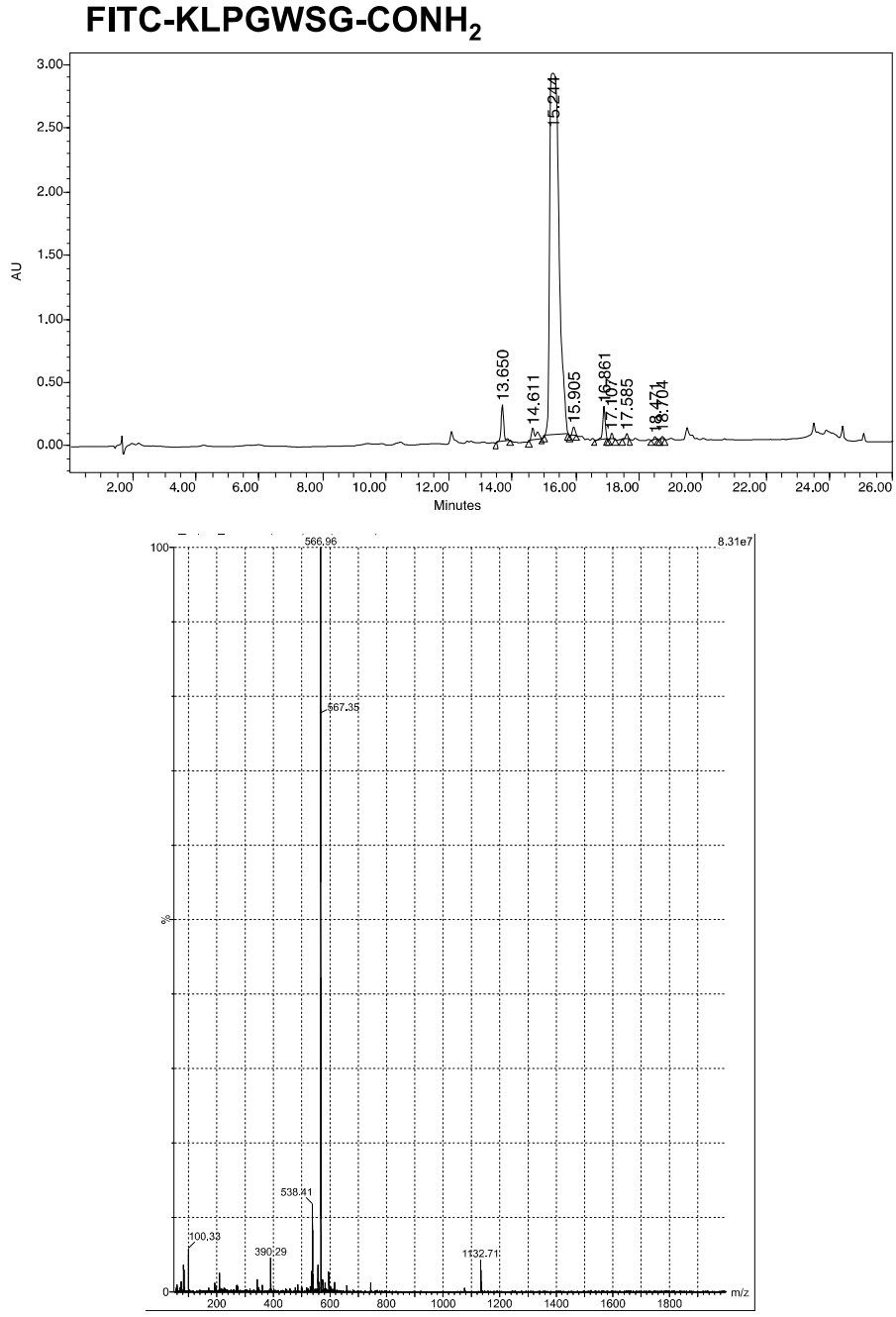
MDPI

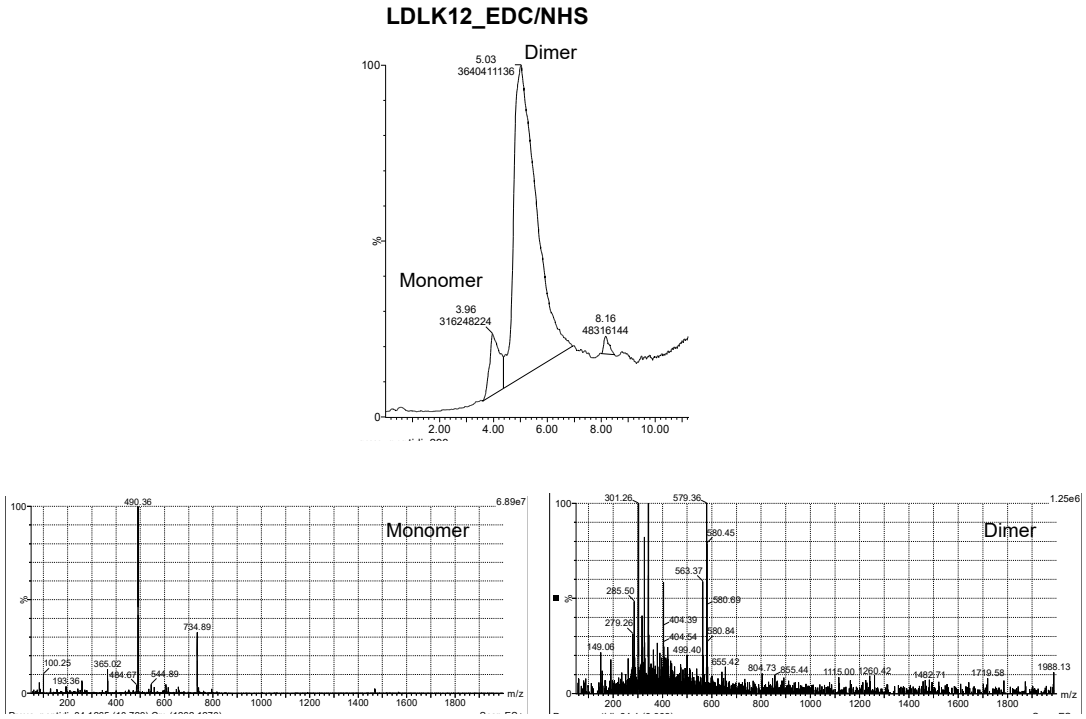
**$H_2N$-KLPGWSG-$CONH_2$**



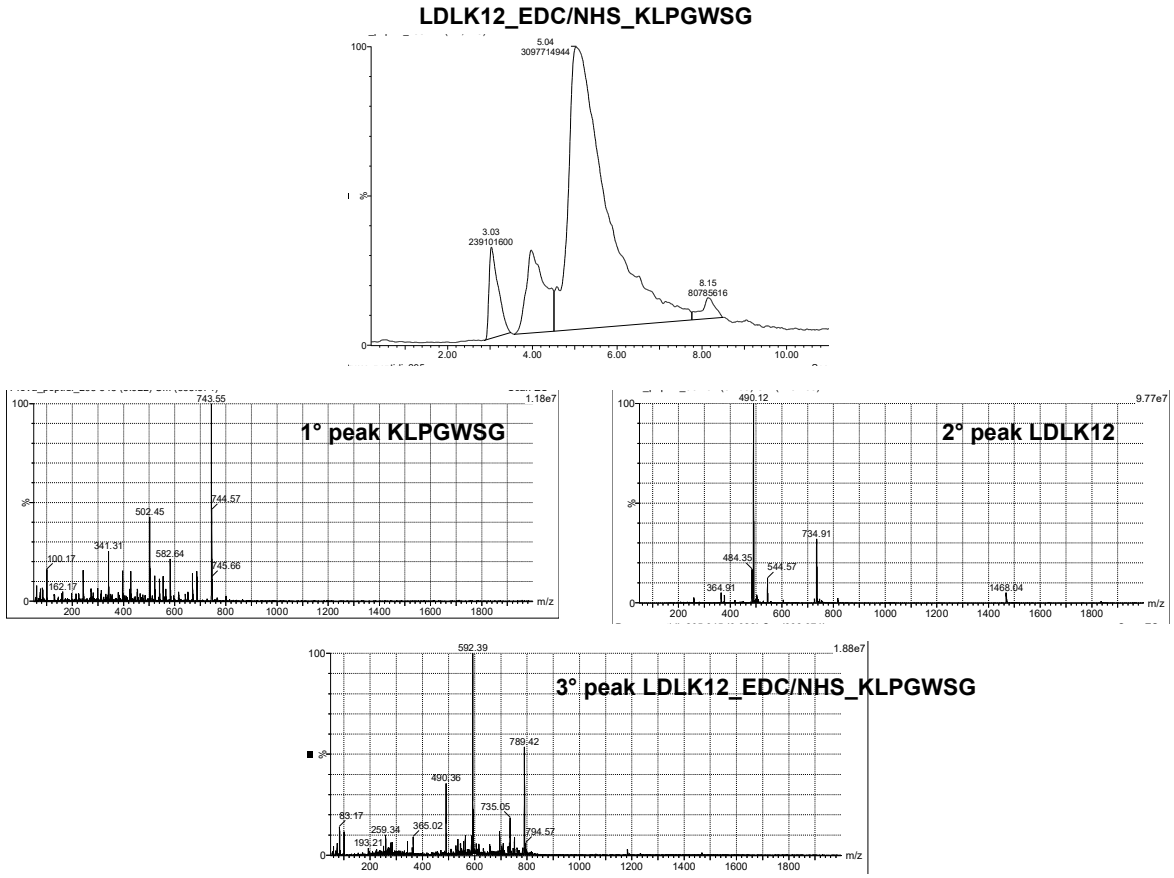






**Supplementary Figure S8: HPLC and LC-MS analyses of LDLK12, KLPGWSG and FITC-KLPGWSG.** LDLK12: LC-MS calc. = 1468.89 g/mol, obs. = 1468.04 g/mol. KLPGWSG: LC-MS calc. = 744.4 g/mol, obs. = 743.63 g/mol. FITC- KLPGWSG: LC-MS calc. = 1133.32 g/mol, obs. = 1132.71 g/mol.




**Supplementary Figure S9: HPLC and LC-MS analyses of cross-linked LDLK12.** Monomer: LC-MS calc. = 734.95 g/mol, obs. = 734.89 g/mol. Dimer: LC-MS calc. = 2076 g/mol [1M+1H]+1, 1438 g/mol [1M+2H]+2 obs. = 1988.13 g/mol, 1482 g/mol. .




**Supplementary Figure S10: HPLC and LC-MS analyses of LDLK12 post-assembly functionalization with KLPGWSG.** 1°peak (KLPGWSG) LC-MS calc. = 744.4 g/mol, obs. = 743.55

g/mol; 2° peak (LDLK12) LC-MS calc. = 1468.89 g/mol, obs. = 1468.04 g/mol.; 3° peak (LDLK12_EDC/sulfoNHS_KLPGWSG) LC-MS calc. = 789.77 [1M + 4H]+ 4 g/mol, obs. = 798.42 g/mol.

## Reference

60. Jordens, S.; Schwenke, K.; Usov, I.; Del Gado, E.; Mezzenga, R. Nematic field transfer in a two-dimensional protein fibril assembly. Soft Matter 2016, 12, 1830–1835.
61. Usov, I.; Mezzenga, R. FiberApp: An Open-Source Software for Tracking and Analyzing Polymers, Filaments, Biomacromolecules, and Fibrous Objects. Macromolecules 2015, 48, 1269–1280.
62. Jordens, S.; Isa, L.; Usov, I.; Mezzenga, R., Non-equilibrium nature of two-dimensional isotropic and nematic coexistence in amyloid fibrils at liquid interfaces. *Nature Commun.* **2013,** *4*, 1917.