

## Supporting Information

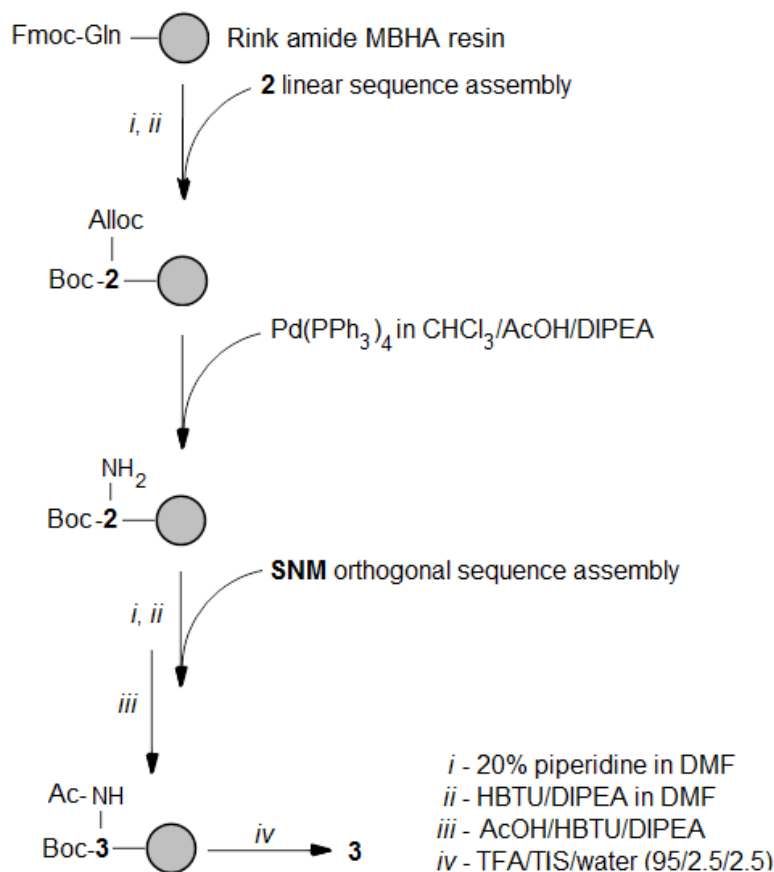
### Linear and orthogonal peptide templating of silicified protein fibres

Bella et al.

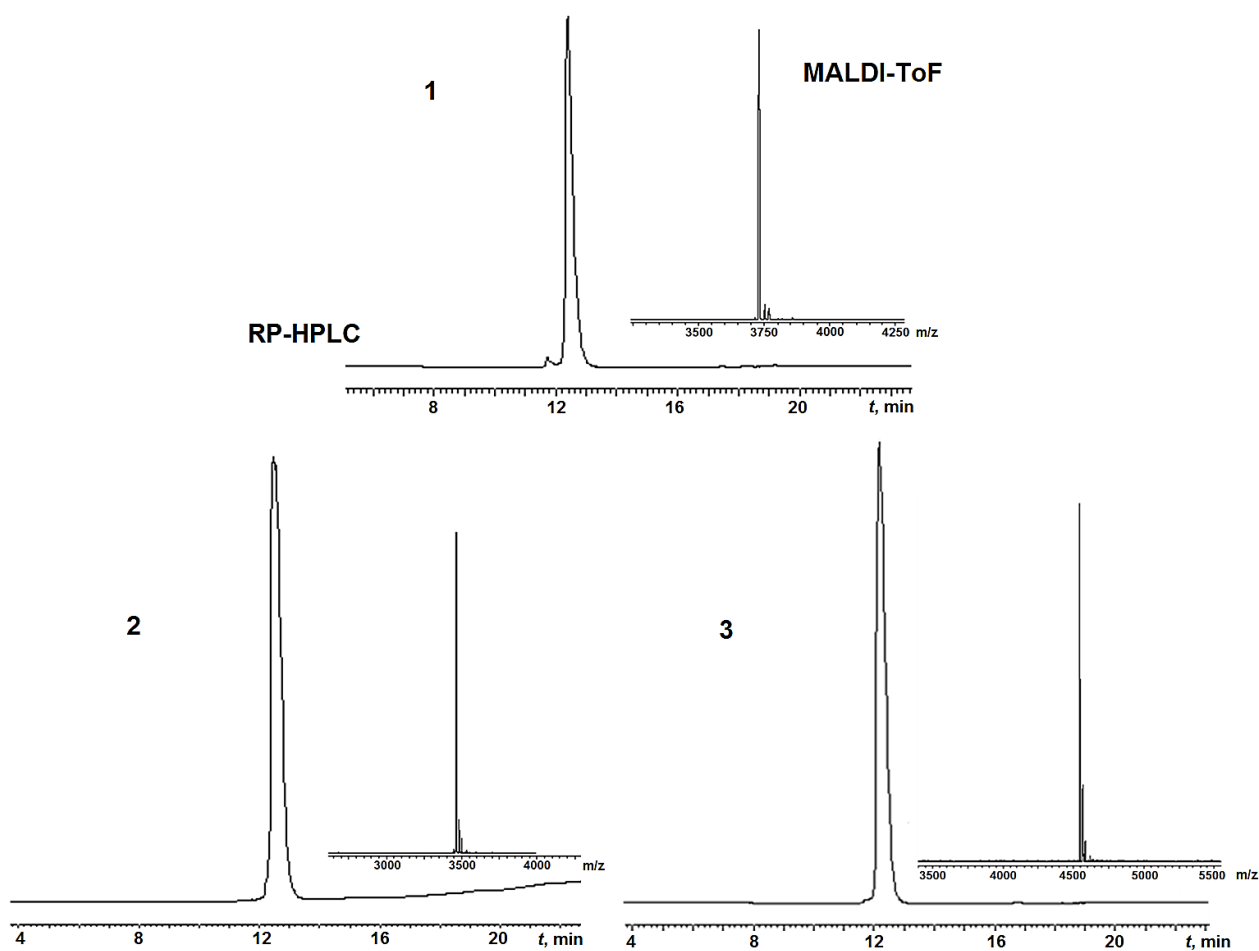
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**Abbreviations** – DIPEA – diisopropylethylamine; Fmoc – 9-fluorenylmethoxycarbonyl; HBTU – *O*-benzotriazole-*N,N,N',N'*-tetramethyl-uronium-hexafluoro-phosphate; tetraethyl orthosilicate – TEOS; RP-HPLC – reversed phase high pressure liquid chromatography; MALDI-ToF – matrix-assisted laser desorption/ionization time of flight; MOPS – 3-(*N*-morpholino) propanesulfonic acid; TIS – triisopropyl silane; TFA – trifluoroacetic acid; XPS – X-ray Photoelectron Spectroscopy.

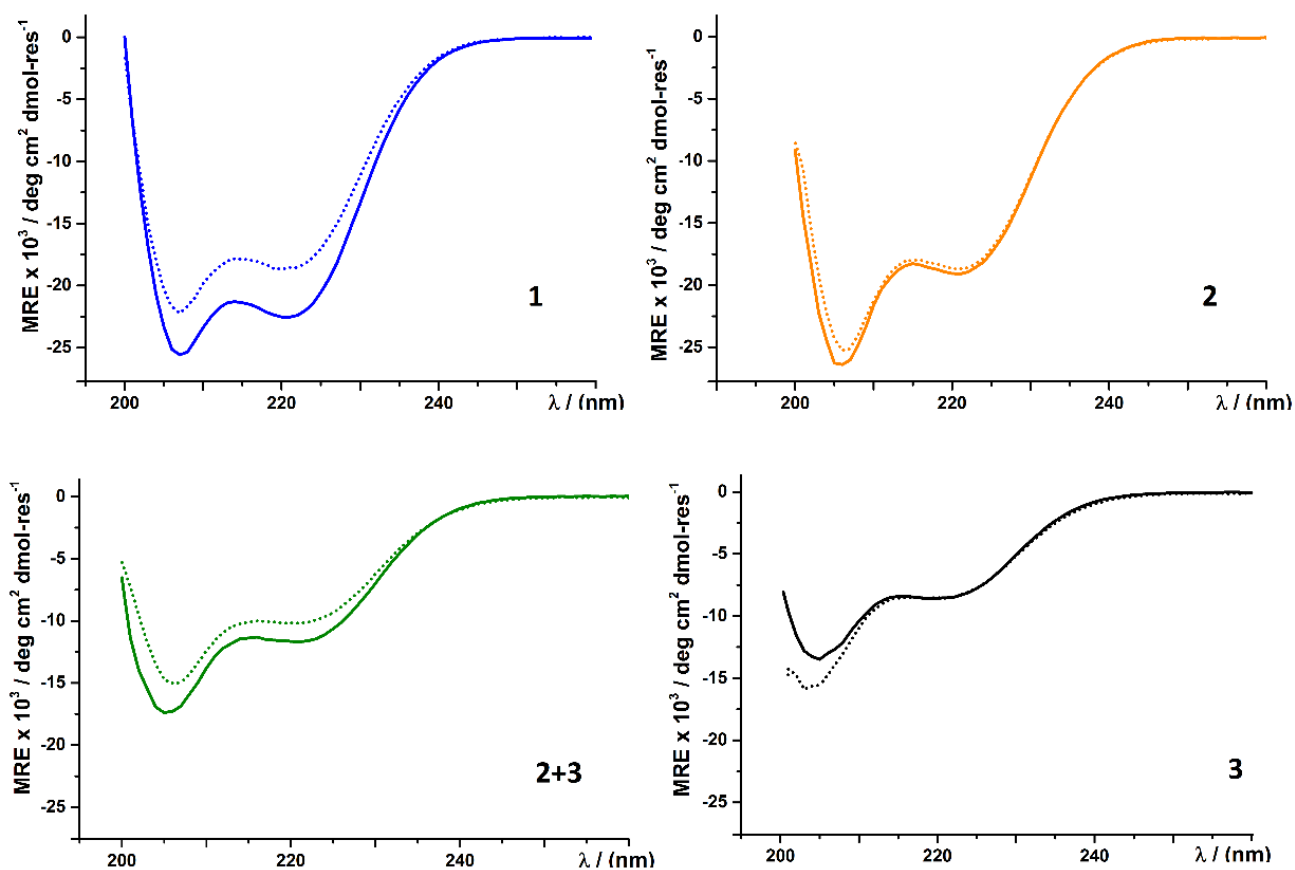
### Figures



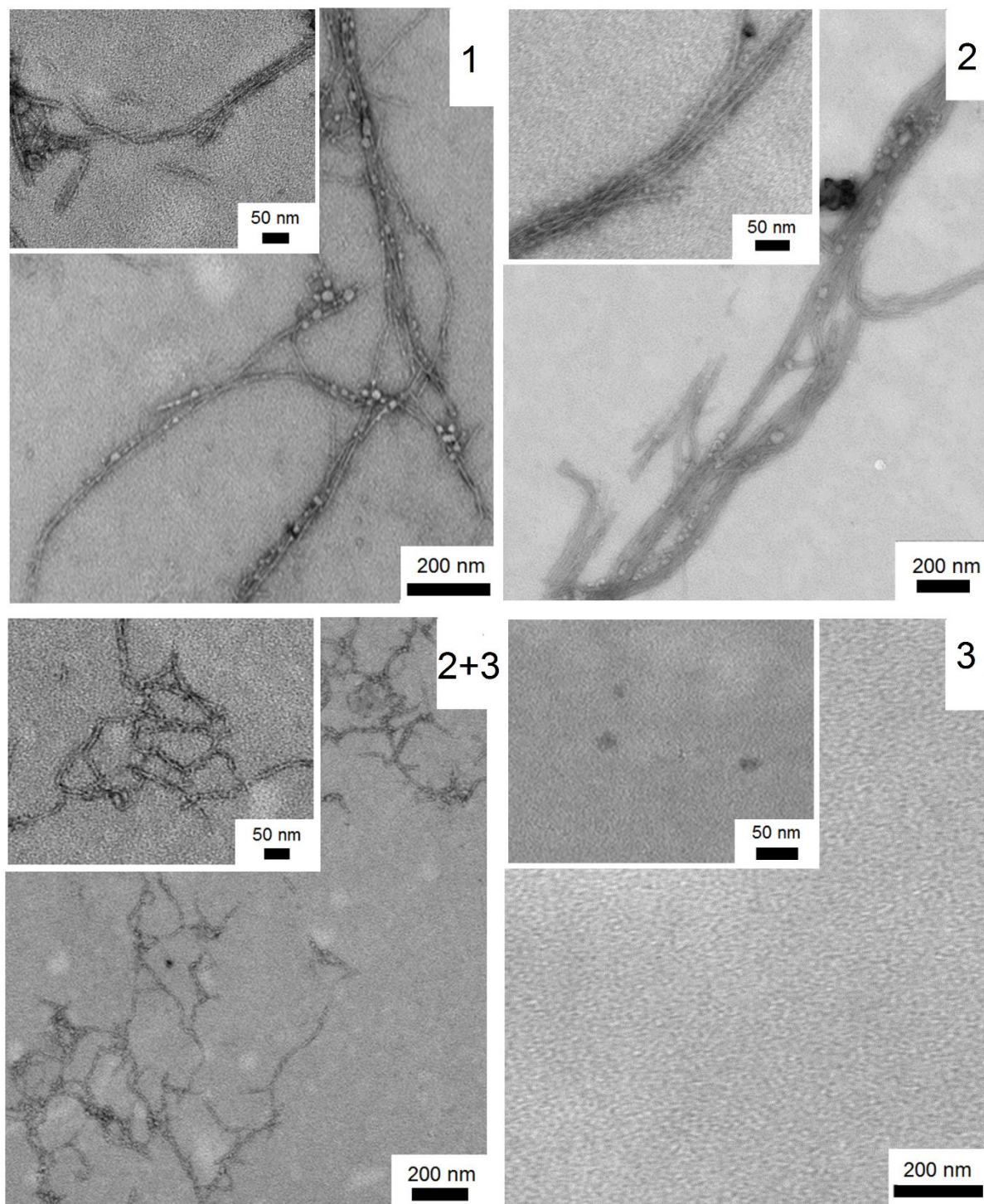
**Figure S1.** Schematic representation of the solid-phase synthesis of **3**. Note: the last residue is Boc-protected, -Lys(Alloc)- was used for the orthogonal assembly of the SNM sequence preceded by a three- $\beta$ alanyl linker.



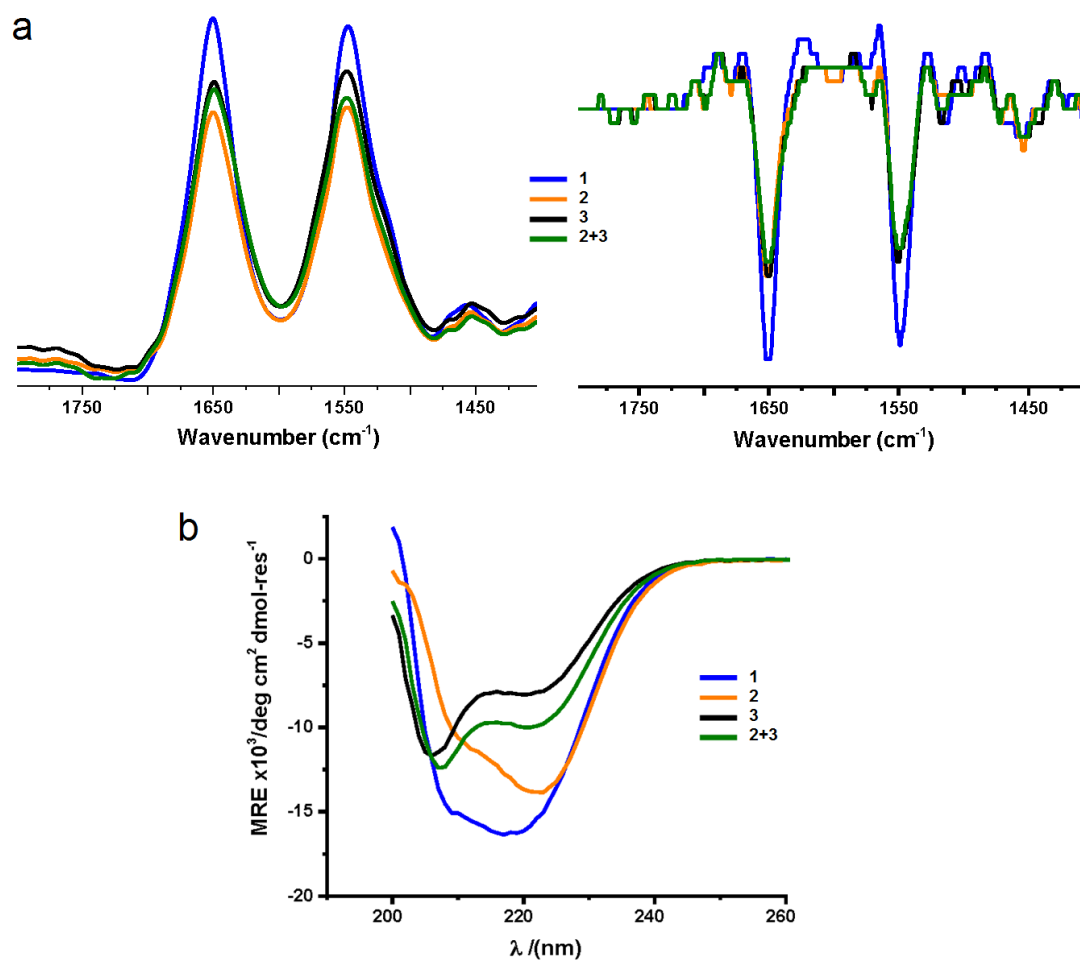
**Figure S2.** RP-HPLC traces and MALDI-ToF spectra for purified peptides used in the study.



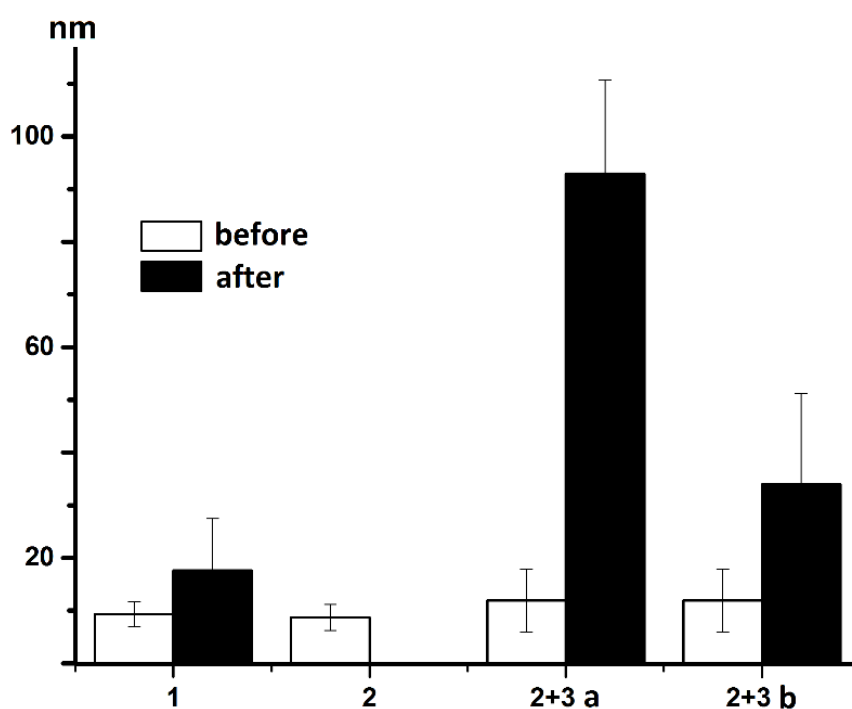
**Figure S3.** CD spectra for the peptide templates before (solid lines) and after (dotted lines) the melt. Folding conditions: 100  $\mu\text{M}$  total peptide in 10 mM MOPS, pH 7.4, rt, overnight.



**Figure S4.** Electron micrographs of peptide templates with (1, 2 and 2+3) and without (3) fibre formation. Assembly conditions: 100  $\mu$ M total peptide in 10 mM MOPS, pH 7.4, rt, overnight.



**Figure S5.** Peptide folding. (a) FT-IR spectra (left) and their second derivatives (right) for **1**, **2**, **3** and **2+3** preparations before silicification. (b) CD spectra for **1**, **2**, **3** and **2+3** preparations after silicification. Assembly conditions: 100  $\mu\text{M}$  total peptide in 10 mM MOPS, pH 7.4, rt, overnight.



**Figure S6.** Diameter distributions for fibres before and after silicification, in each case measured over 50 individual structures. Two dominating size populations for **2+3** templates are shown separately as **a** and **b**.