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Supporting Information

Chitin-Amyloid Synergism and Their Use as Sustainable Structural Adhesives

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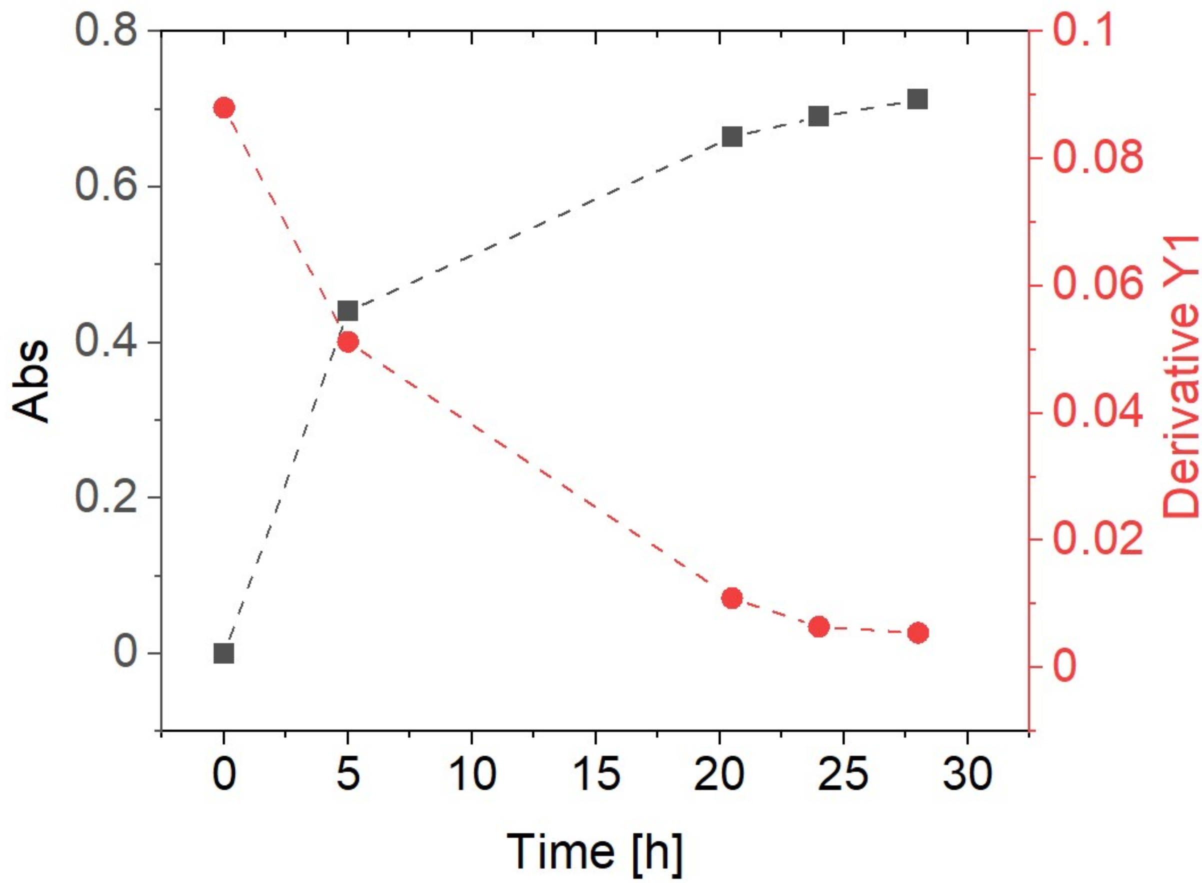

Fig.S1 - UV-Vis absorbance at 280 nm from the supernatant collected during the deproteinization step. The secondary Y axis correspond to the first derivative of the absorbance, showing negligible change between 24 and 28 h. The dashed line is used as a guide to the eye.

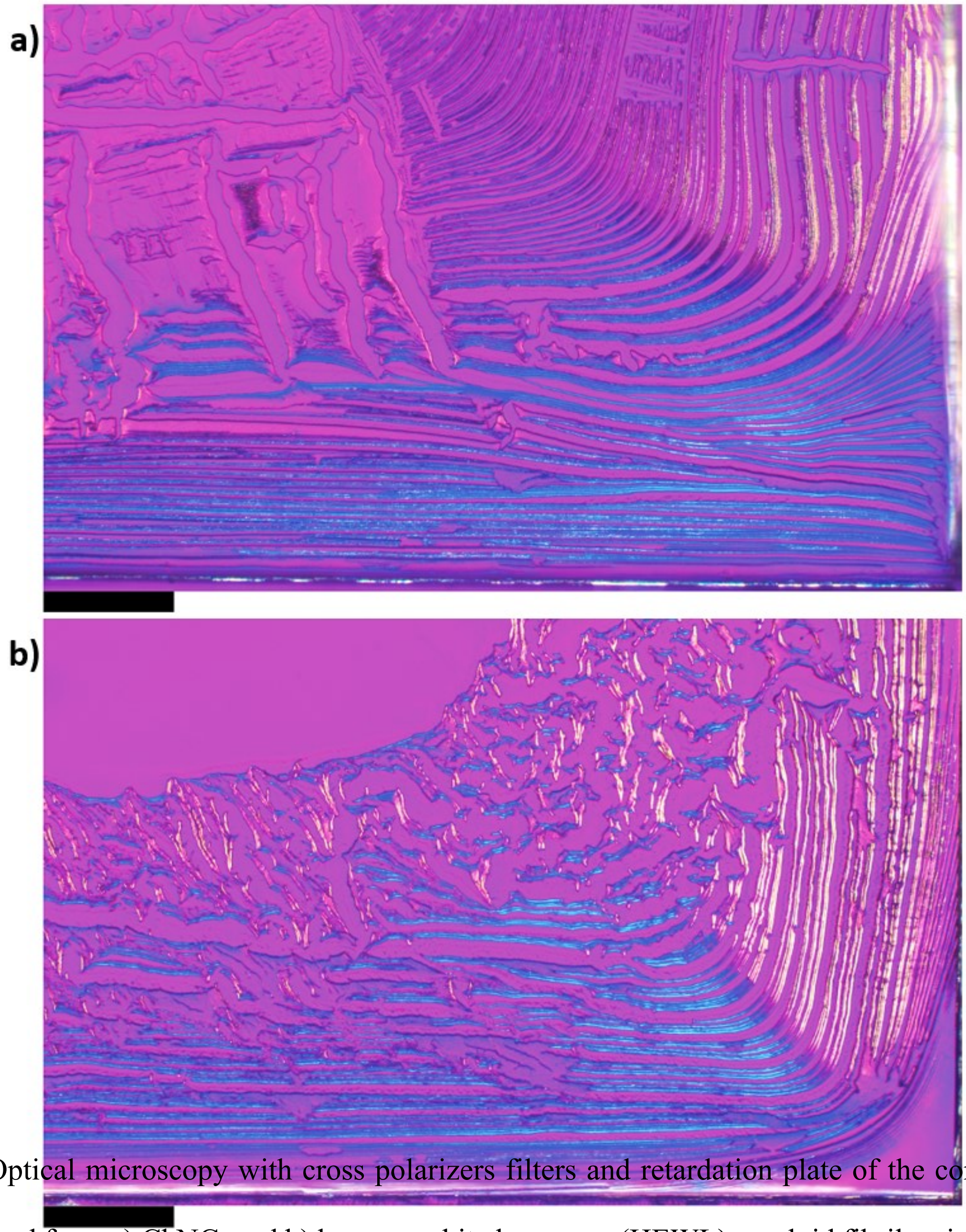

Fig.S2 – Optical microscopy with cross polarizers filters and retardation plate of the corners of bonds formed from a) ChNC, and b) hen egg white lysozyme (HEWL) amyloid fibrils mixed with ChNC at 1:10 ratio. Scale bar represents 400 μm.

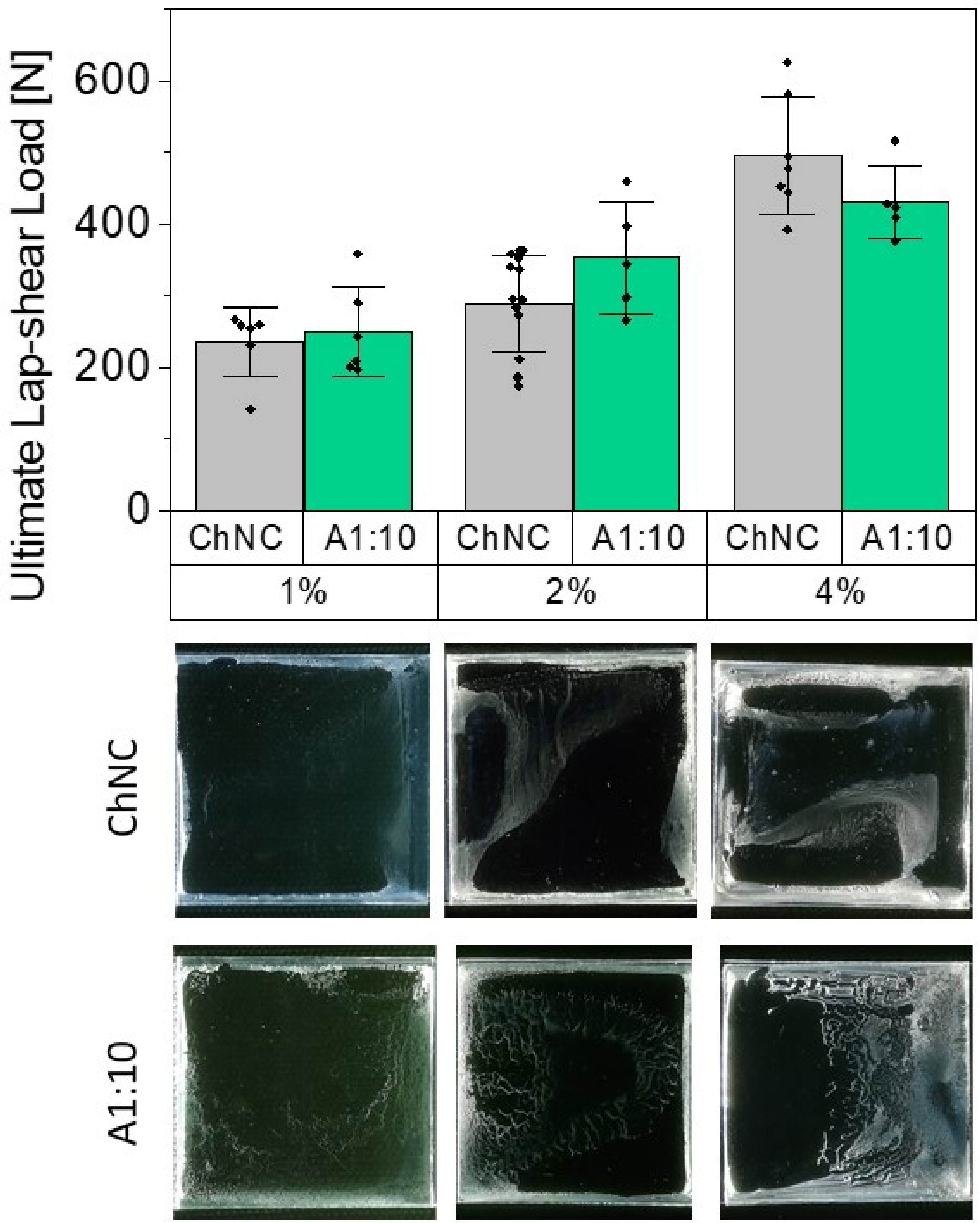

Fig.S3 – Ultimate lap-shear load from bonds assembled from ChNC or HEWL amyloids:ChNC at different concentrations.

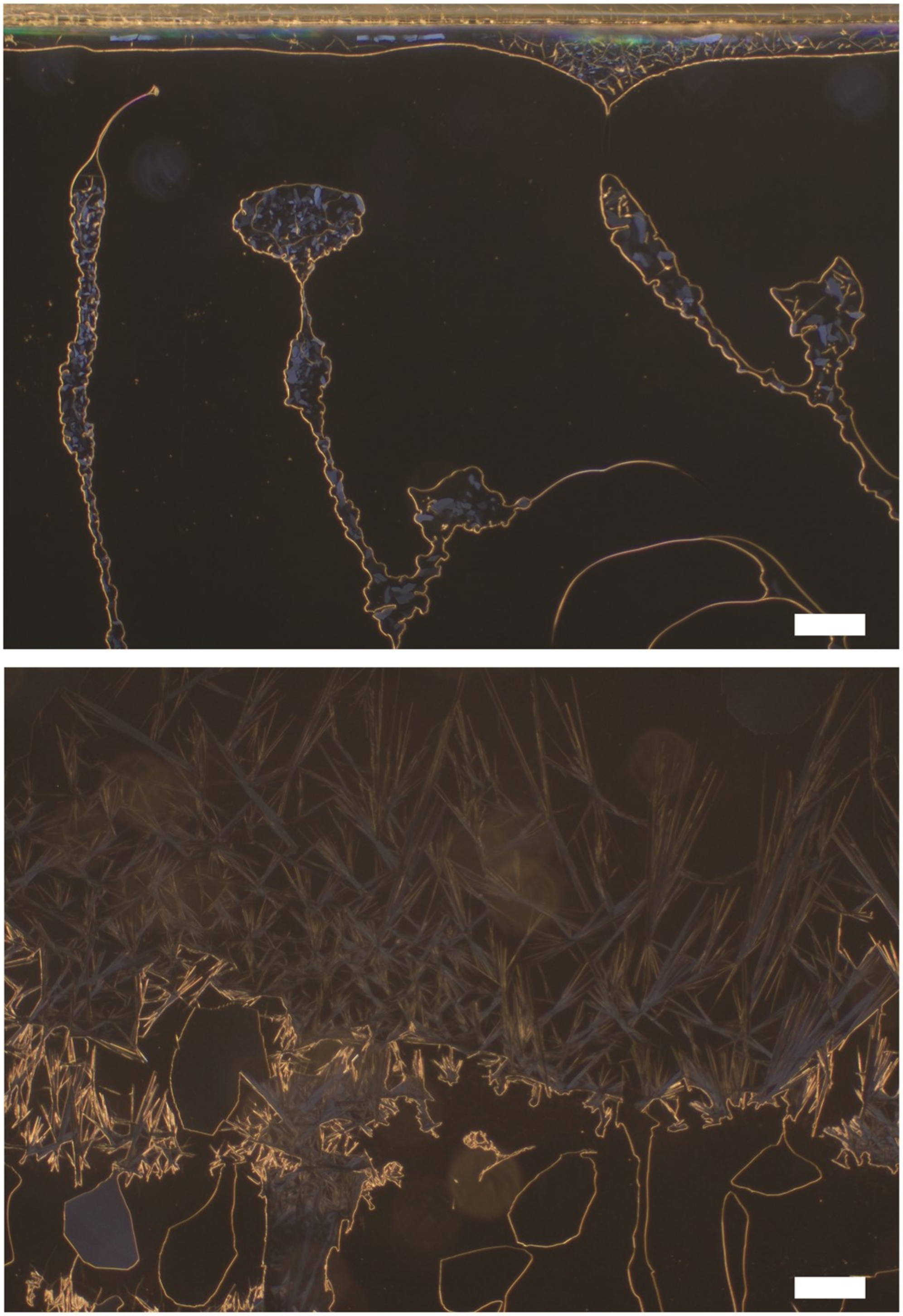

Fig.S4 – Polarized optical microscopies of bonds formed from HEWL monomers. Top, image taken near the edge of the bond. Bottom, image taken in the central part of the bond. Scale bars represent 200 µm.

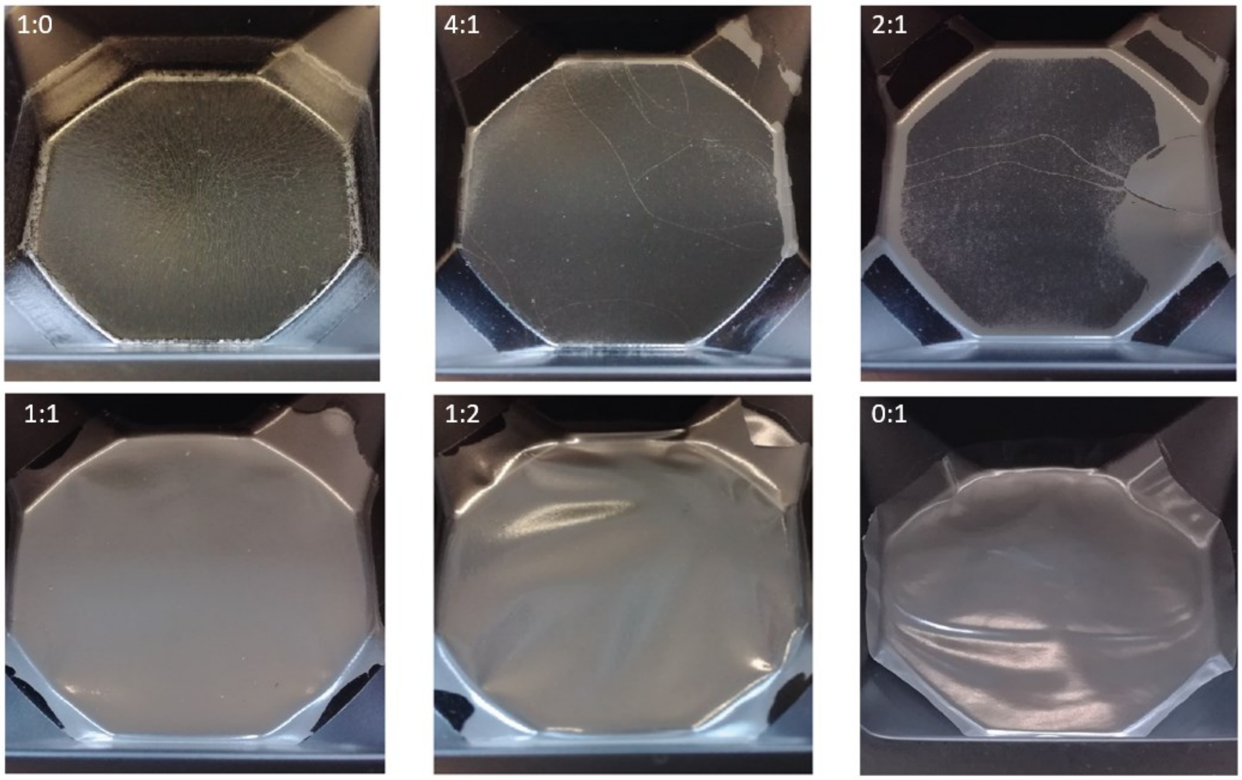

Fig.S5 - Films produced from 2.8 mL of suspension at 1 wt%, and at different mixing ratios of HEWL amyloid fibrils and ChNC. Higher amounts of HEWL amyloids resulted in stronger adhesion to the polypropylene substrate, as well as in higher brittleness of the films.

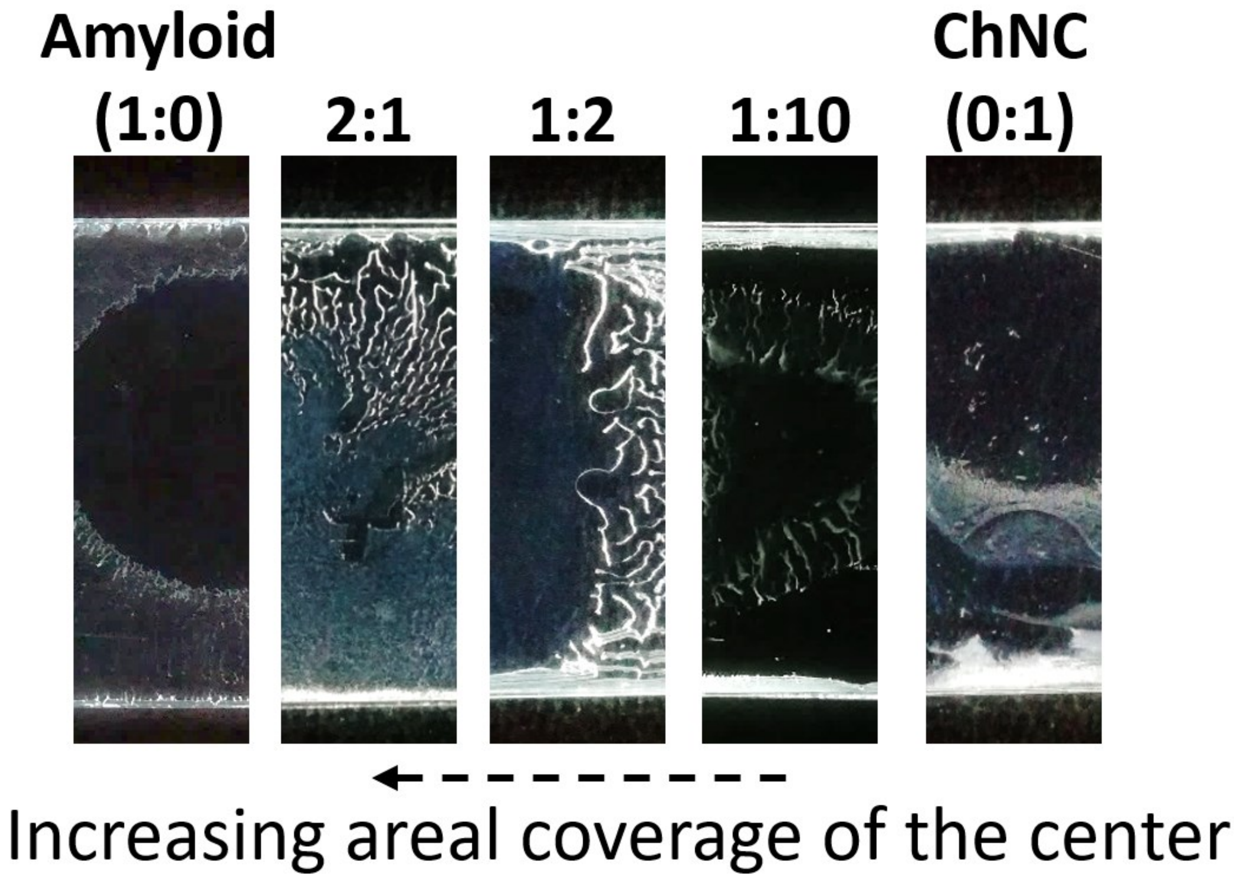

Fig.S6 – Digital photographs of the central part of the bonds formed at different mixing ratios of amyloids and ChNC. In general, the areal coverage at the central part of the bonds increased upon addition of amyloids.

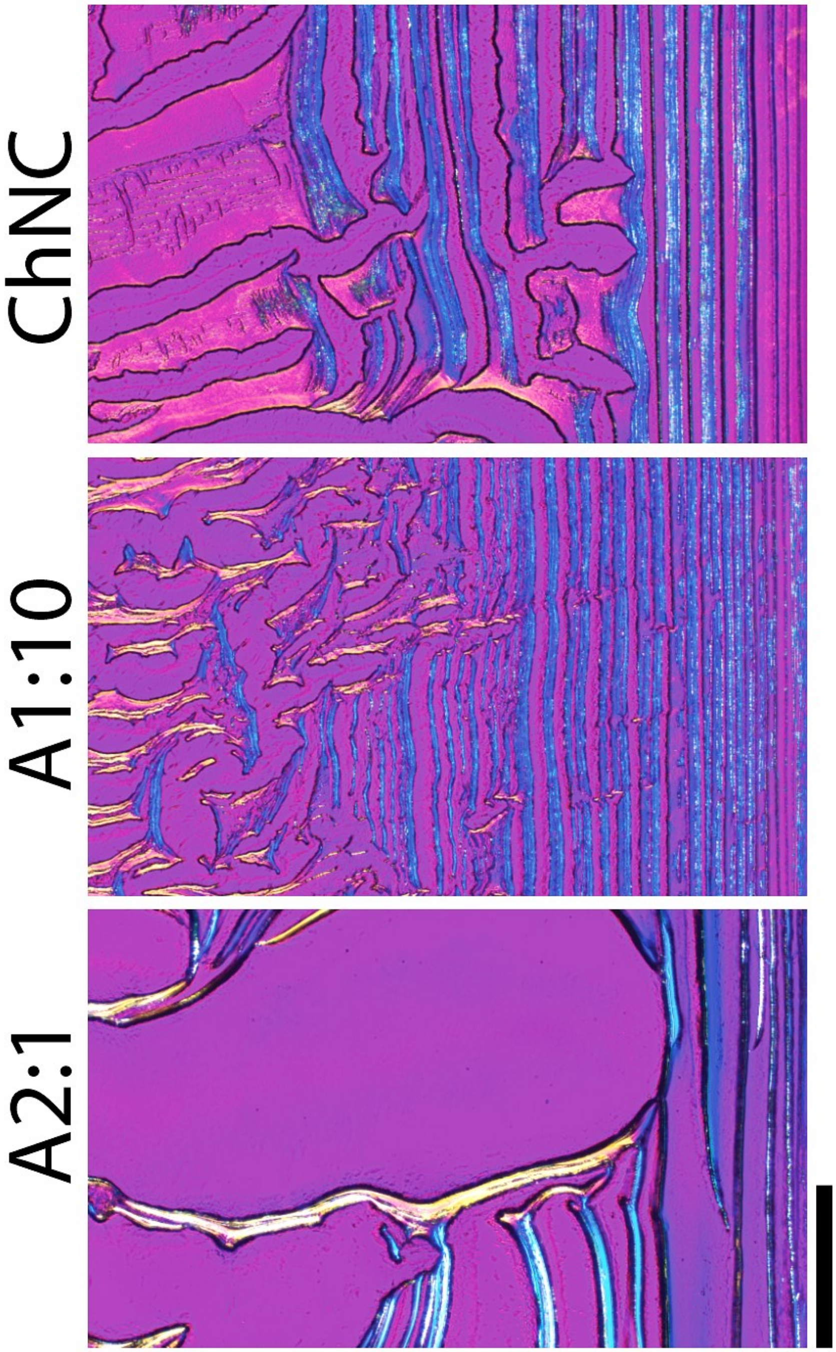

Fig.S7 - Optical microscopy with cross polarizers filters and retardation plate of edges of bonds formed from ChNC, and their mixtures with HEWL amyloid fibrils at 2:1 and 1:10 ratios (amyloid-to-ChNC ratios). Scale bar represents 200 μm.

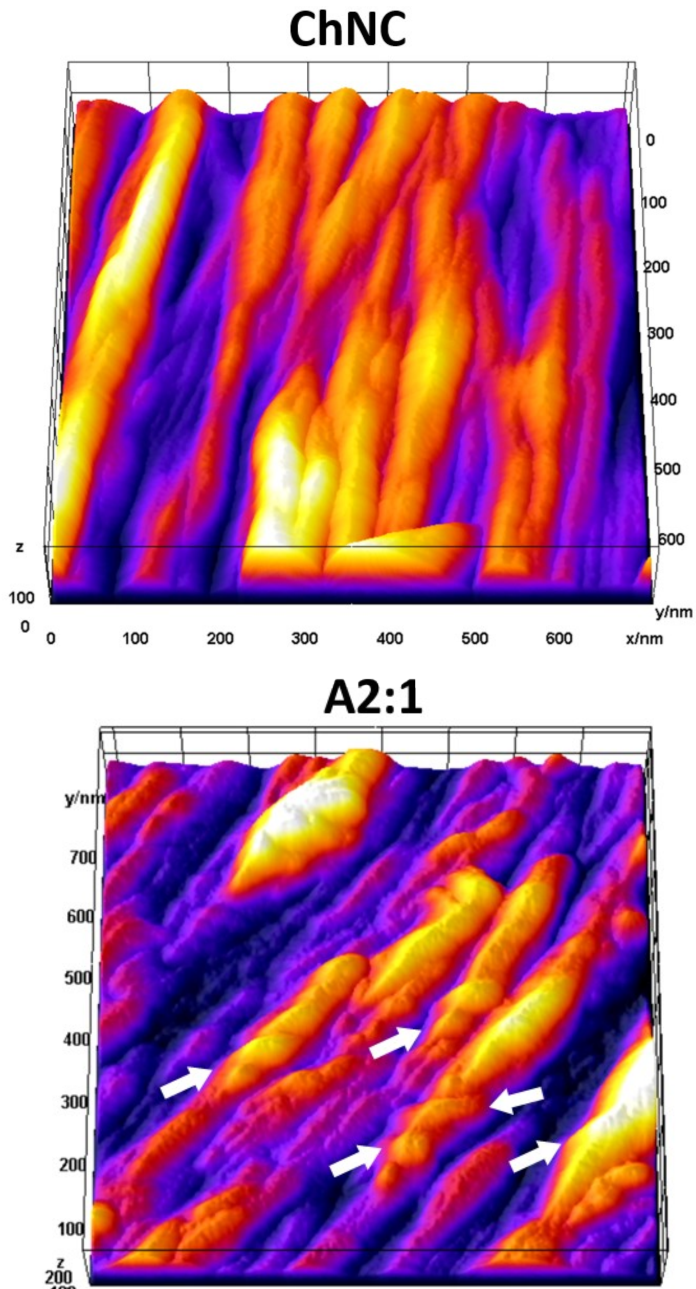

Fig.S8 - Three-dimensional maps generated from AFM height images of the ChNC (top) and amyloid:ChNC lamellae at 2:1 ratio (bottom). Arrows indicate the areas where amyloids crossing ChNC can be distinguished.

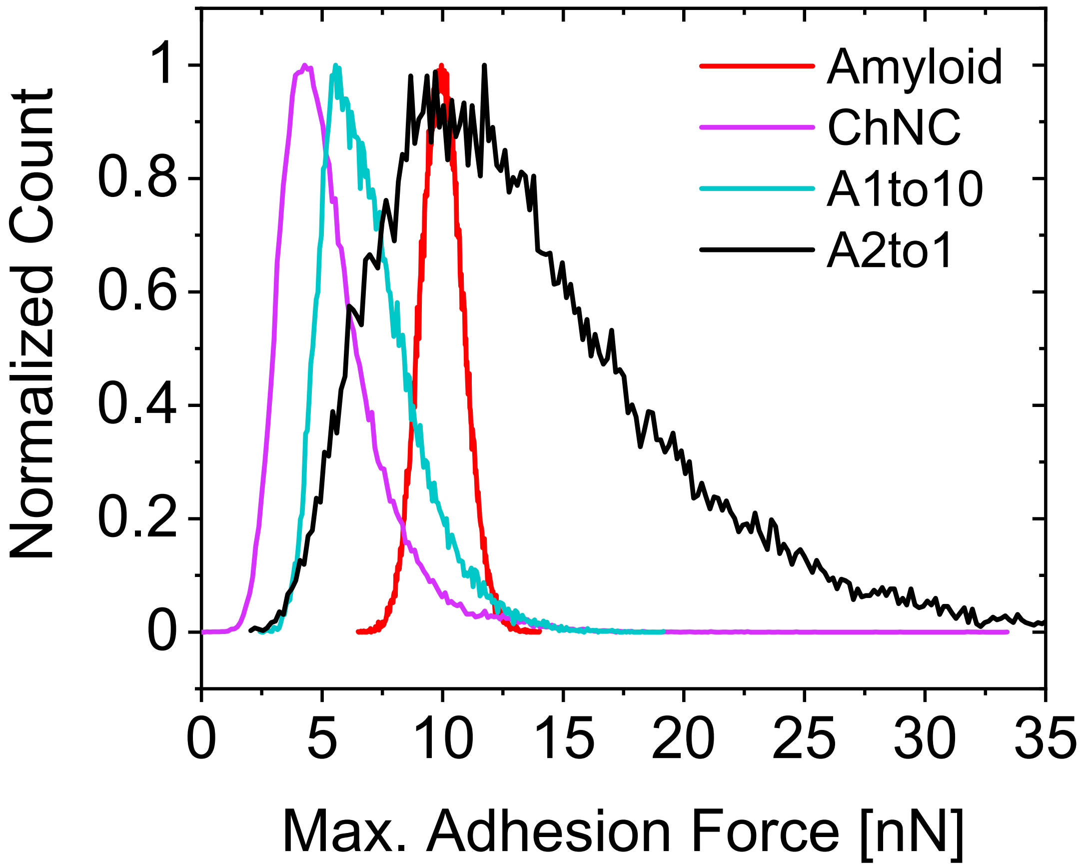

Fig.S9 – Histogram representing the distribution of the maximum adhesion force from the force mapping AFM images represented in **Fig.8**.

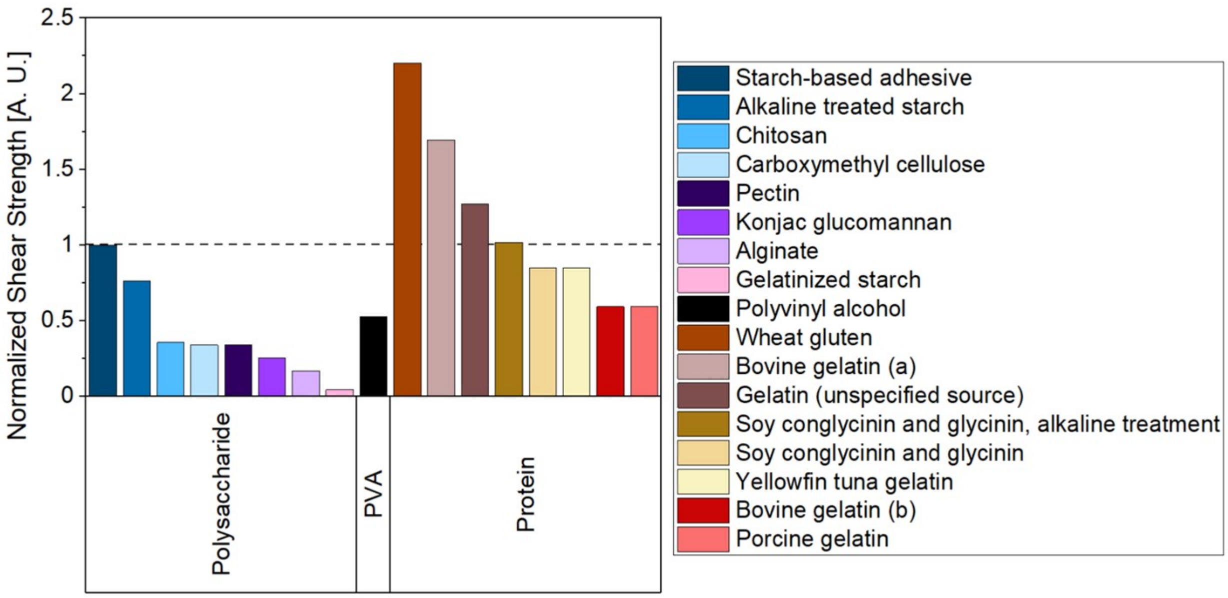

Fig.S10 – Qualitative comparison of the lap shear strength of non-crosslinked (supramolecular) biopolymeric adhesives. The strength values were normalized to the strongest polysaccharide-based adhesive (starch-based) to allow a relative comparison between the different adhesive classes as the absolute value may bear significant experimental bias. A detailed comparative table for the values herein showcased is available in **Table S2.**

Adhesive type	Load [N]	Length [cm]	Areal Density [mg cm^{-2}]	Mass [mg]	Load per adhesive mass [N mg^{-1}]
S1:20	363 ± 36	2.5	0.13	0.8	454 ± 45
A1:10	353 ± 78	2.5	0.13	0.8	441 ± 98
N1:20	333 ± 147	2.5	0.13	0.8	416 ± 184
CNC (0.18)[1]	169 ± 87	1	0.18	0.45	375 ±193
ChNC	289 ± 67	2.5	0.13	0.8	361 ± 84
Lysozyme (N)	266 ± 108	2.5	0.13	0.8	333 ± 135
CNC (0.41)[1]	269 ± 99	1	0.41	1.1	245 ± 90
Short Lys. Amyloid (S)	116 ± 28	2.5	0.13	0.8	145 ±35
Lys. Amyloid (A)	98 ± 28	2.5	0.13	0.8	123 ± 35
Lignin Nanoparticles[2]	81 ± 27	2.6	0.14793	1.0005	81 ± 27

Table S1 – Comparative values for the parameters used for the assembly of bio-colloidal adhesives between glass slides, and the resulting strength values for the assemblies.

Class	Description	Shear Strength MPa	Reference
Polysaccharide	Starch-based adhesive	6	[3]
Polysaccharide	Alkaline treated starch	4.6	[4]
Polysaccharide	Chitosan	2.1	[5]
Polysaccharide	Carboxymethyl cellulose	2	[6]
Polysaccharide	Pectin	2	[6]
Polysaccharide	Konjac glucomannan	1.5	[5]
Polysaccharide	Alginate	1	[6]
Polysaccharide	Gelatinized starch	0.3	[7]
PVA	Polyvinyl alcohol	3.2	[8]
Protein	Wheat gluten	13.2	[9]
Protein	Bovine gelatin (a)	10.2	[10]
Protein	Gelatin (unspecified source)	7.6	[11]
Protein	Soy conglycinin and glycinin, alkaline treatment	6.1	[12]
Protein	Soy conglycinin and glycinin	5.1	[12]
Protein	Yellowfin tuna gelatin	5.1	[13]
Protein	Bovine gelatin (b)	3.6	[13]
Protein	Porcine gelatin	3.6	[13]

Table S2 - Comparison of the lap shear strength of non-crosslinked biopolymeric adhesives represented normalized in **Fig.10**.

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