

Supporting Information

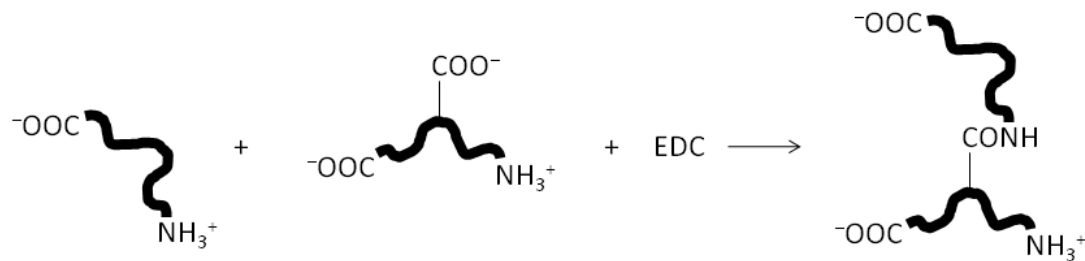
**A Synthetic Polypeptide Electrospun Biomaterial**

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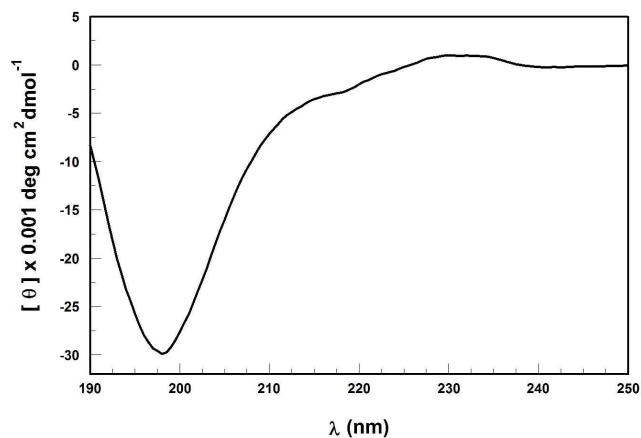
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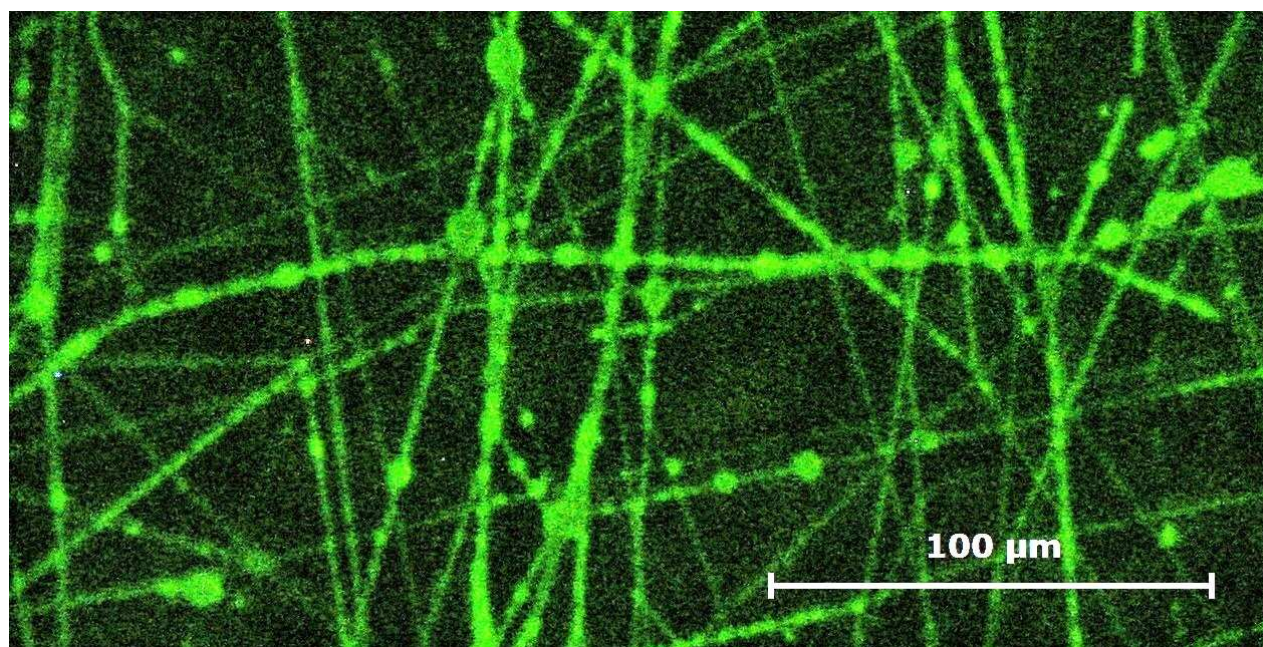
†CIFM.



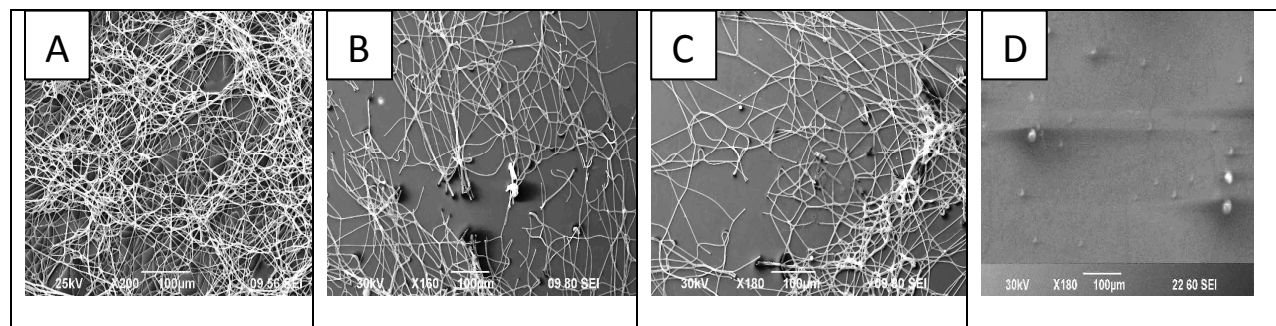
*Scheme S1. The main crosslinking reaction of the present study. A polypeptide has distinctive end groups, amino and carboxyl. These groups will be ionized with high probability in the vicinity of neutral pH. EDC reacts with a carboxyl group in a glutamic acid side chain in the second peptide on the left, forming an amine-reactive O-acylisourea intermediate (not shown). If the intermediate reacts with the amino-terminal amino group on the first peptide on the left, the two peptides become joined by an amide bond, as shown on the right. EDC could join a side chain to the amino-terminus of the same peptide.*



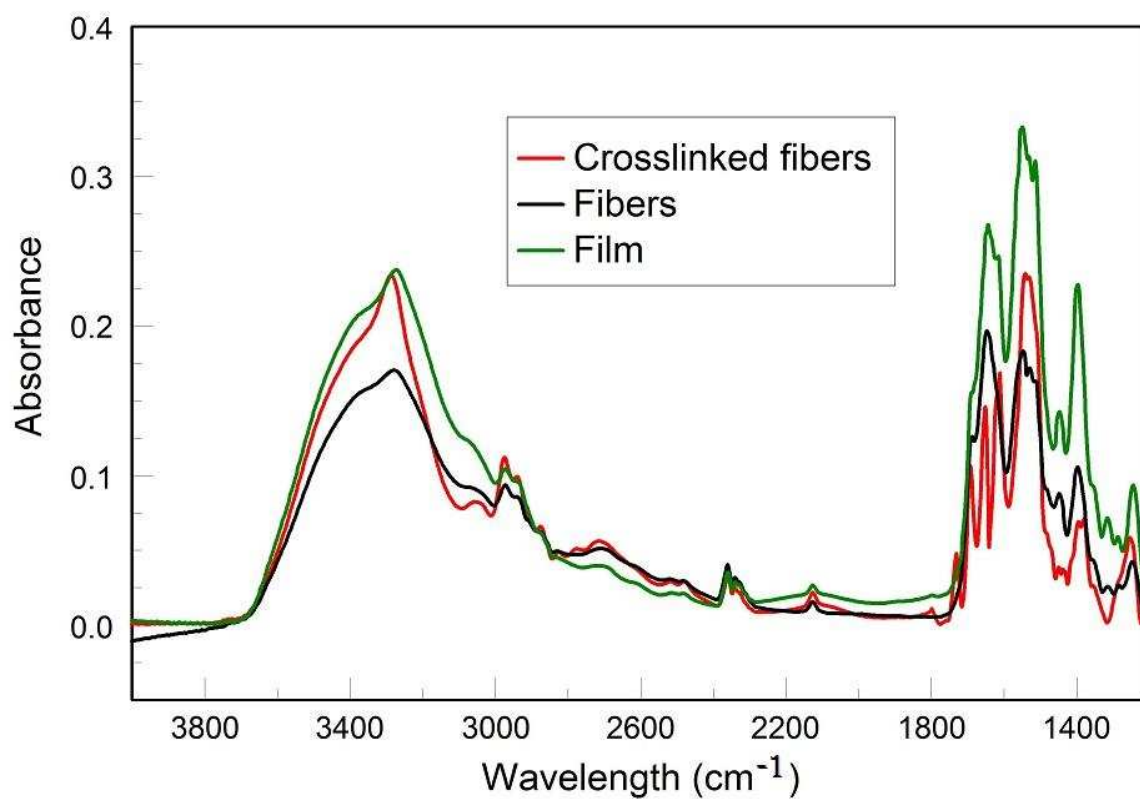
*Figure S1. Far-UV CD spectrum of PLEY in aqueous solution. The spectrum is essentially that of a model random coil. Note especially the sharp minimum just below 200 nm. See Greenfield, N.; Fasman G. D. Biochemistry **1969**, 8, 4108-4116.*



*Figure S2. Fluorescence micrograph of crosslinked electrospun PLEY fibers labeled with FITC-PLL. Details of sample preparation are provided in the paper. The figure shows an example of “unattractive” fibers. Beads are present. The fibers are not smooth and continuous. The image was obtained with a 20× objective on a Leica DM IL HC inverted microscope with a EL6000 external light source.*



*Figure S3. SEM micrographs of crosslinked electrospun 55% PLEY fibers following proteolytic digestion for 5 h at 37 °C. E) 0%, F) 0.02%, G) 0.2%, H) 2% (w/v) protease XIV in pH 7.4 PBS buffer. The accelerating potential was 25 kV or 30 kV. The scale bar is 100 μm in each case.*



*Figure S4. In-situ FTIR spectra of an as-cast film and electrospun fibers of PLEY before and after crosslinking. The spectra were obtained in ATR mode. The full range of data acquisition is shown, 1200-4000 cm<sup>-1</sup>.*