Electronic Supplementary Information

Induction of a higher-ordered architecture in glatiramer acetate improves its biological efficiency in an animal model of multiple sclerosis

Ziyuan Song[#a,b], Yee Ming Khaw[#a,c], Lazaro A. Pacheco[b], Kuan-Ying Tseng[b], Zhengzhong Tan[b], Kaimin Cai[b], Ettigounder Ponnusamy[d], Jianjun Cheng*[b], Makoto Inoue*[a,c]

[a]Department of Comparative Biosciences, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, USA. *Email*: makotoi@illinois.edu

[b]Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, USA. *Email*: jianjunc@illinois.edu

[c]University of Illinois at Urbana-Champaign, Neuroscience Program, 405 North Matthews Avenue, Urbana, Illinois 61801, USA

[d]MilliporeSigma, 545 South Ewing, St. Louis, Missouri 63103, USA

*Corresponding author. Department of Comparative Biosciences, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801, United States.

[#]These authors contributed equally to this work.

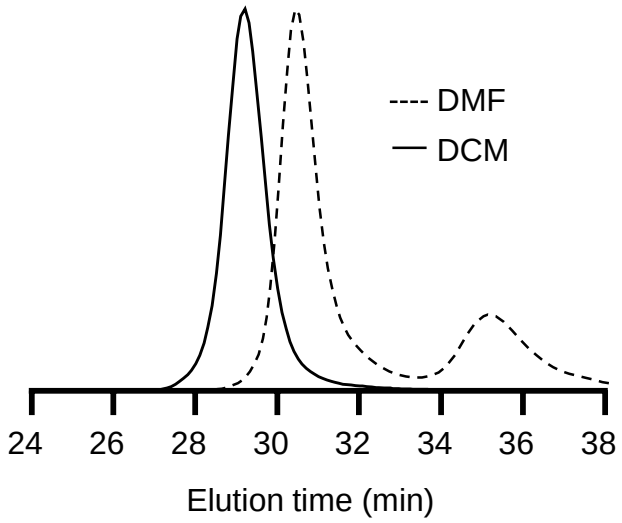

Fig. S1. Normalized GPC-dRI traces of the resulting polypeptides from PAMAM-initiated polymerization of BLG-NCA in DMF (M_n = 65.0 kDa, $Đ$ = 3.22) and DCM (M_n = 345 kDa, $Đ$ = 1.10). $[M]_0/[I]_0$ = 50. $[M]_0$ = 400 and 100 mM for the polymerization in DMF and DCM, respectively.

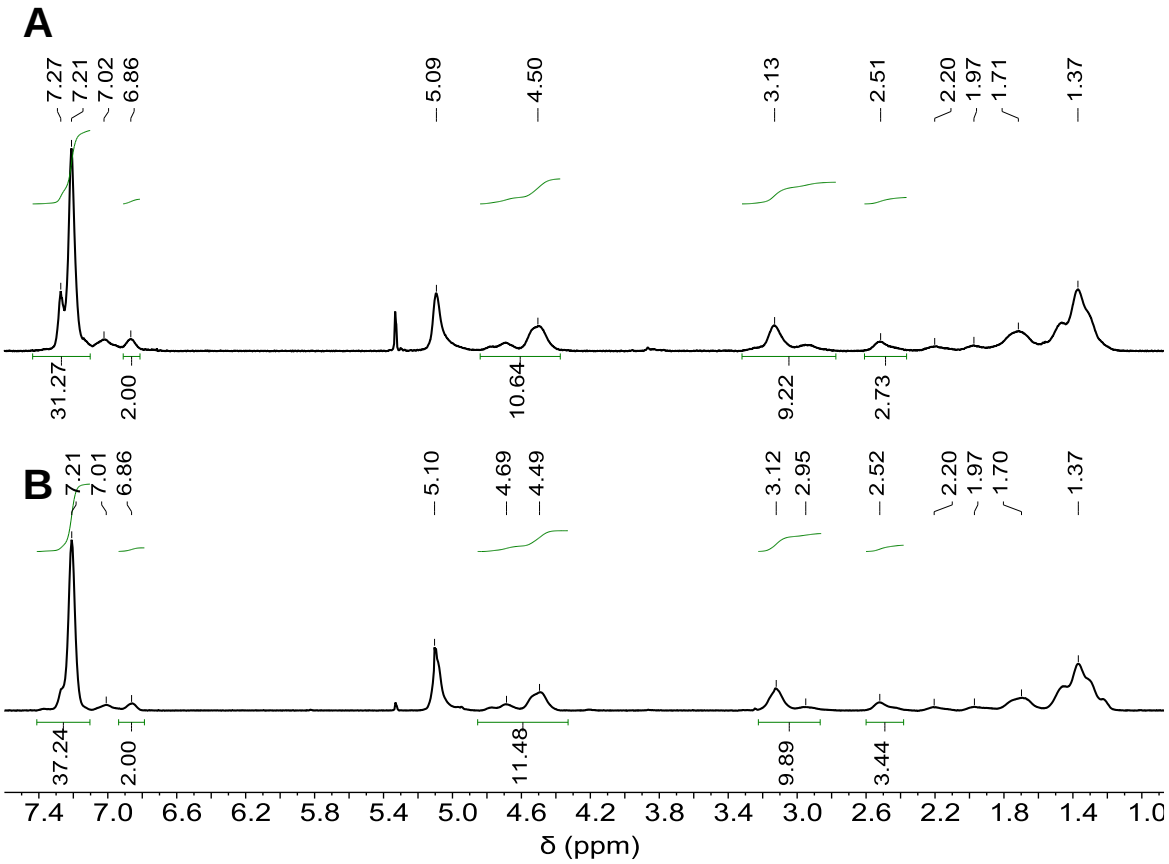

Fig. S2. ^{1}H NMR spectra (500 MHz) of the polypeptide precursors for sGA (A) and GA (B) in TFA-*d*. The fractions of amino acid residues were calculated based on the integration of peaks at 6.86 (Ar-H of BLT residues), 4.85–4.30 (α-H of all residues), 3.25–2.80 (ε-H of ZLL residues and β-H of BLT residues), and 2.51 (γ-H of BLG residues) ppm.

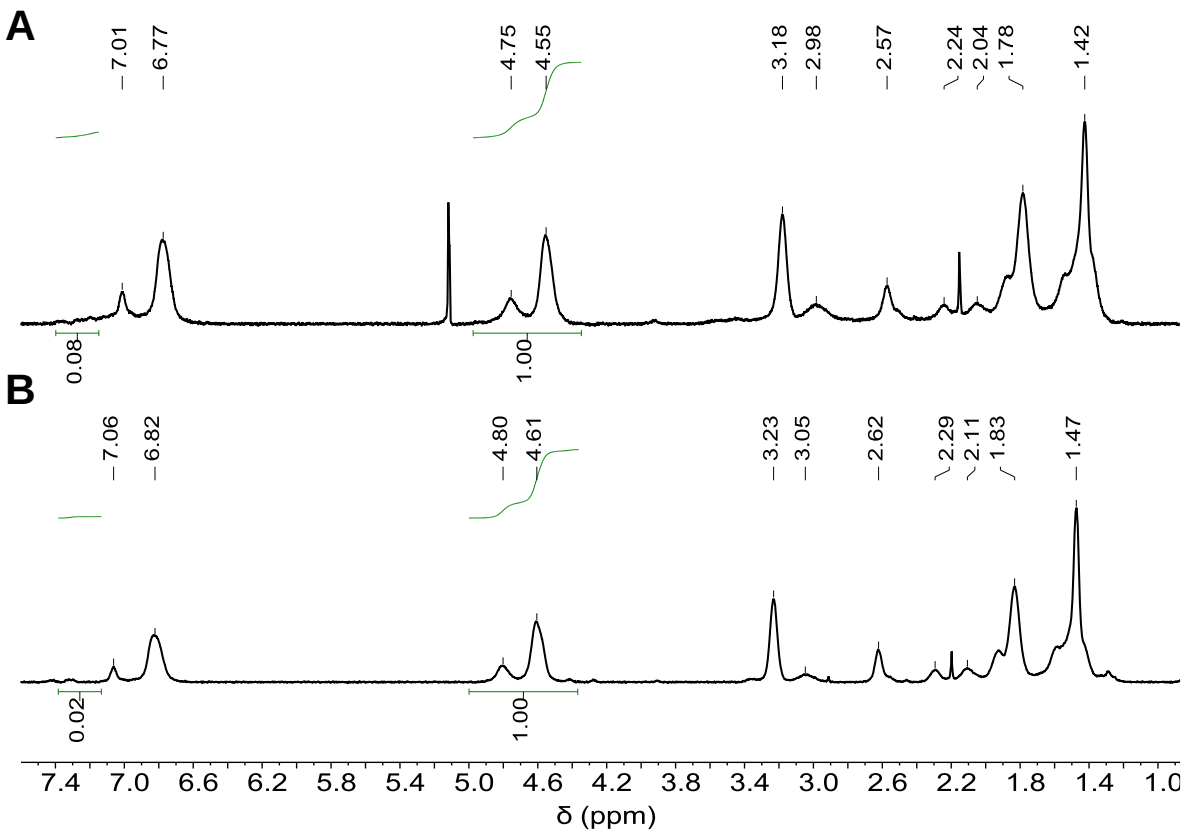

Fig. S3. ^{1}H NMR spectra (500 MHz) of sGA (A) and GA (B) in TFA-*d*. The deprotection efficiency was determined by comparing the integration of peaks at 7.4–7.1 ppm (Ar-H of protecting groups) before and after deprotection.

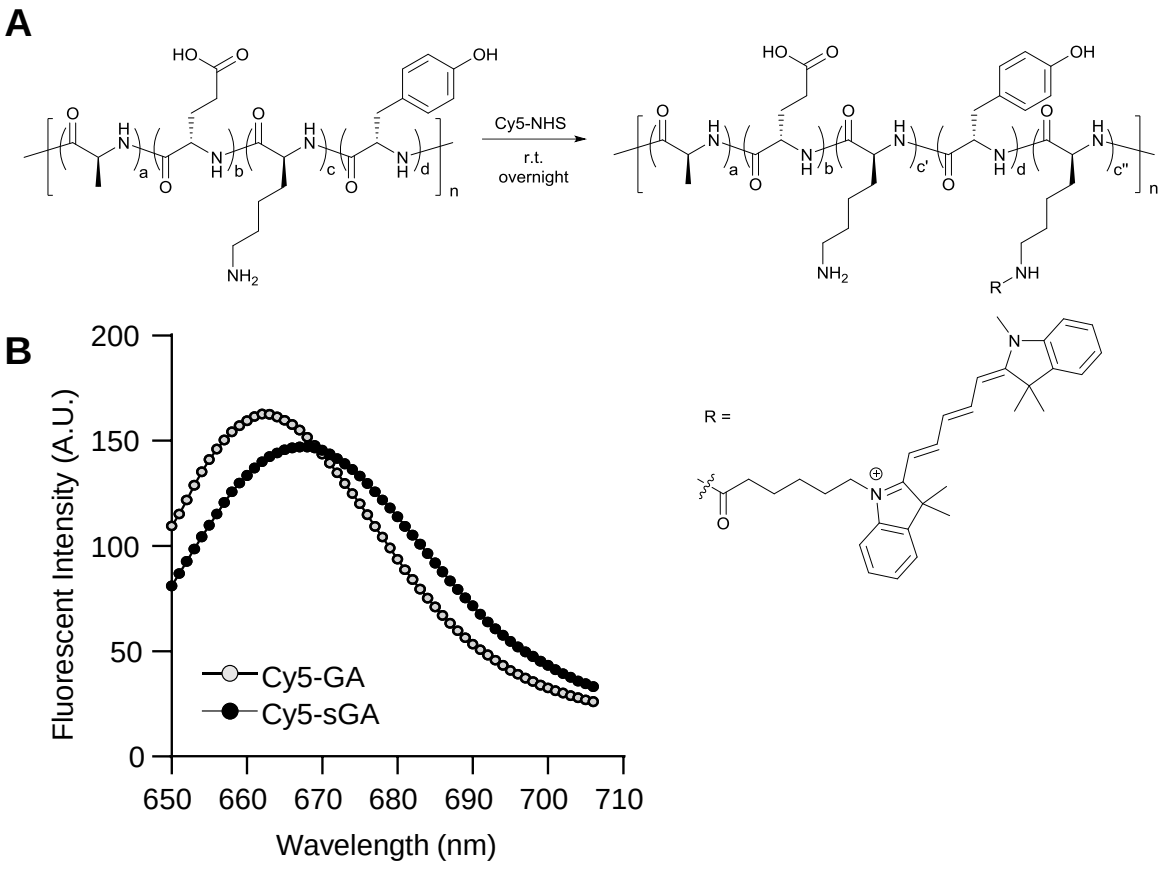

Fig. S4. Fluorescent labelling of sGA and GA with Cy5. (A) Synthetic route to Cy5-labelled sGA and GA. (B) Representative fluorescent spectra of Cy5-labelled sGA and GA at 20 μg/mL in a PBS buffer.