Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.

Complex Coacervates of Oppositely Charged Co-Polypeptides Inspired by the Sandcastle Worm Glue

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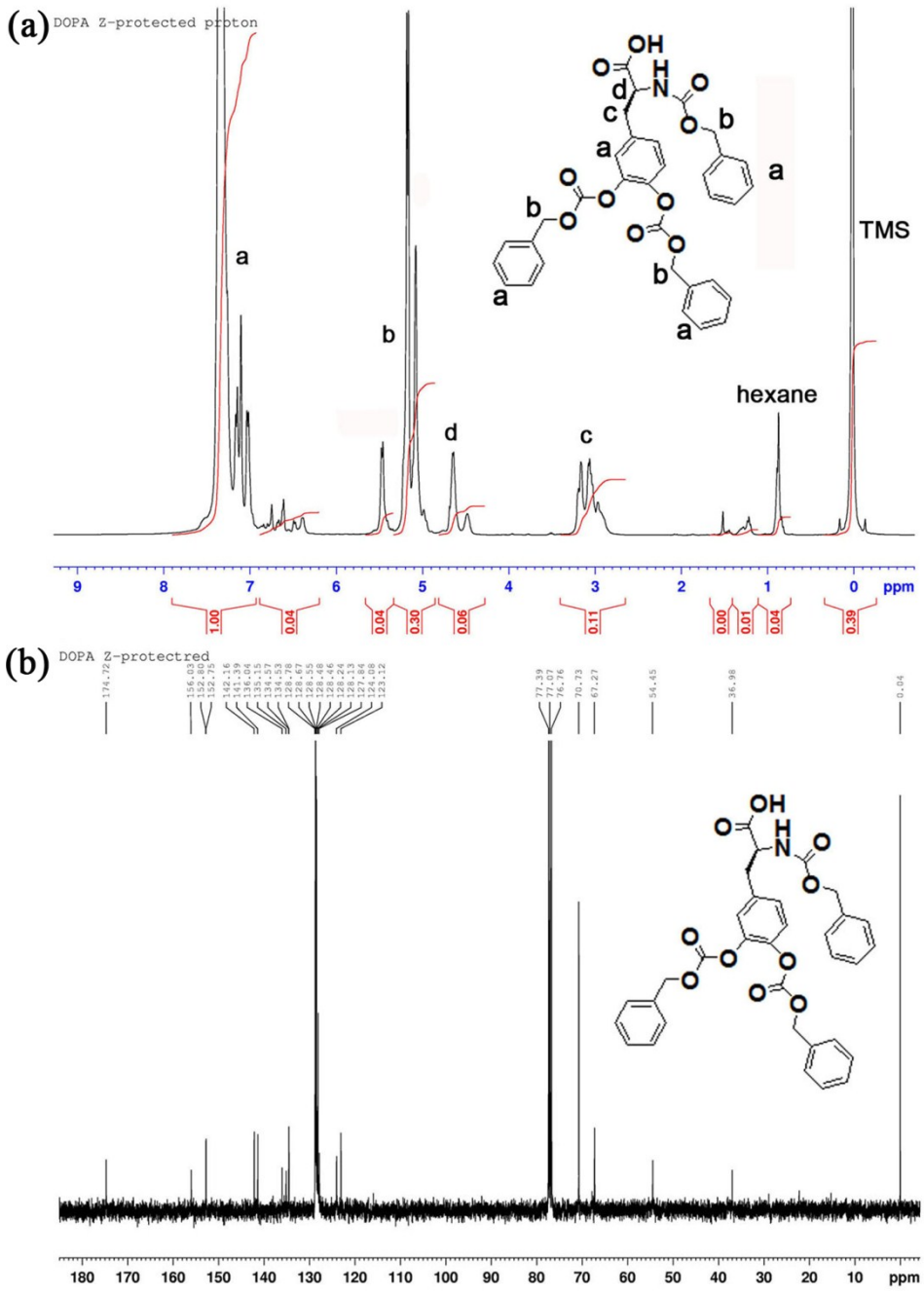
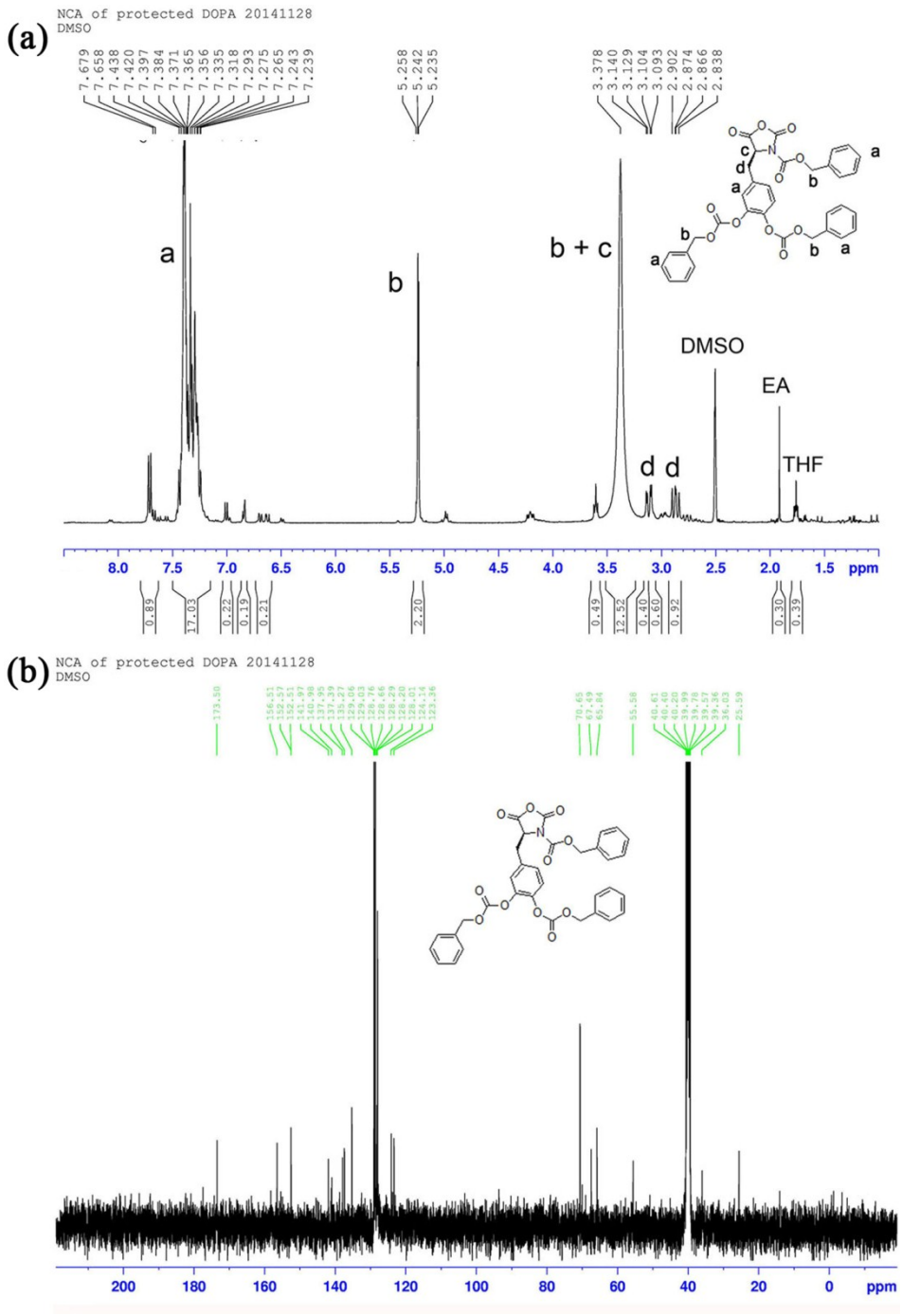
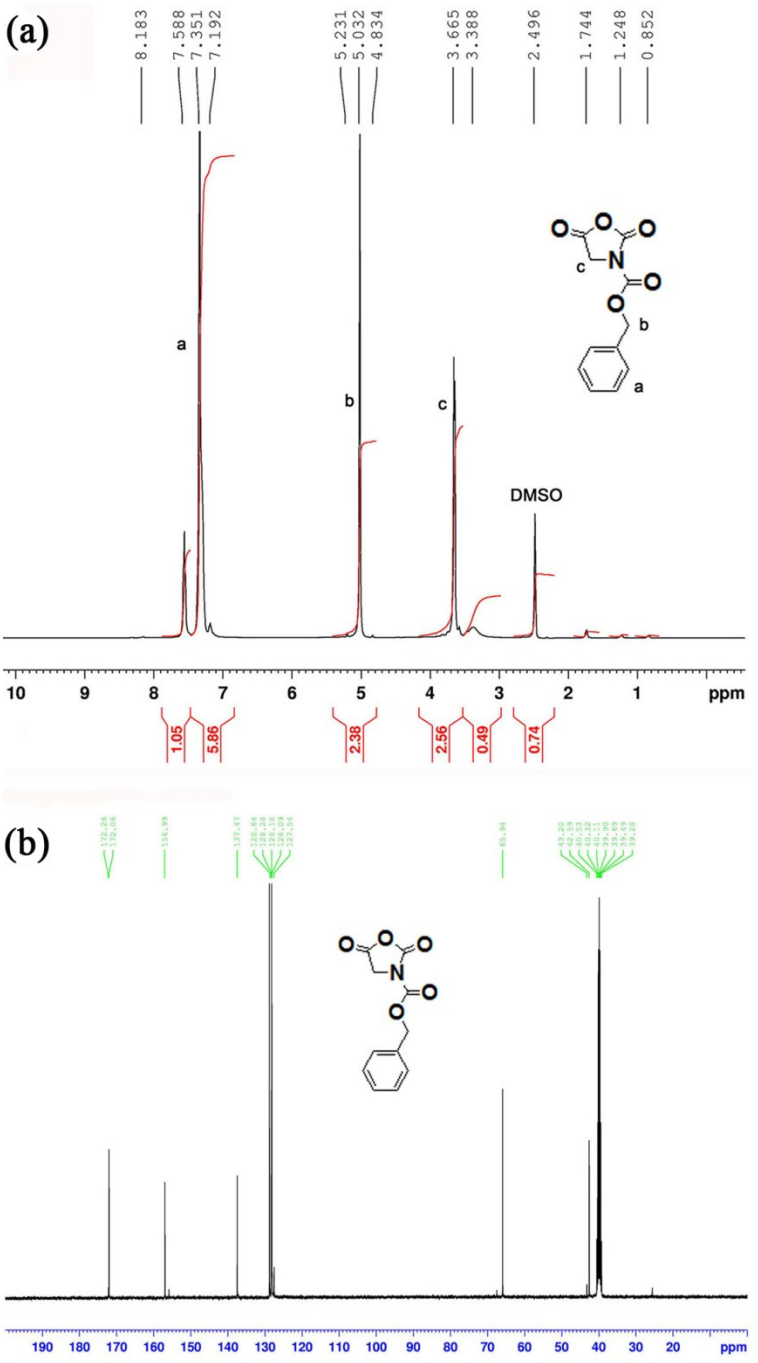
Supplementary information



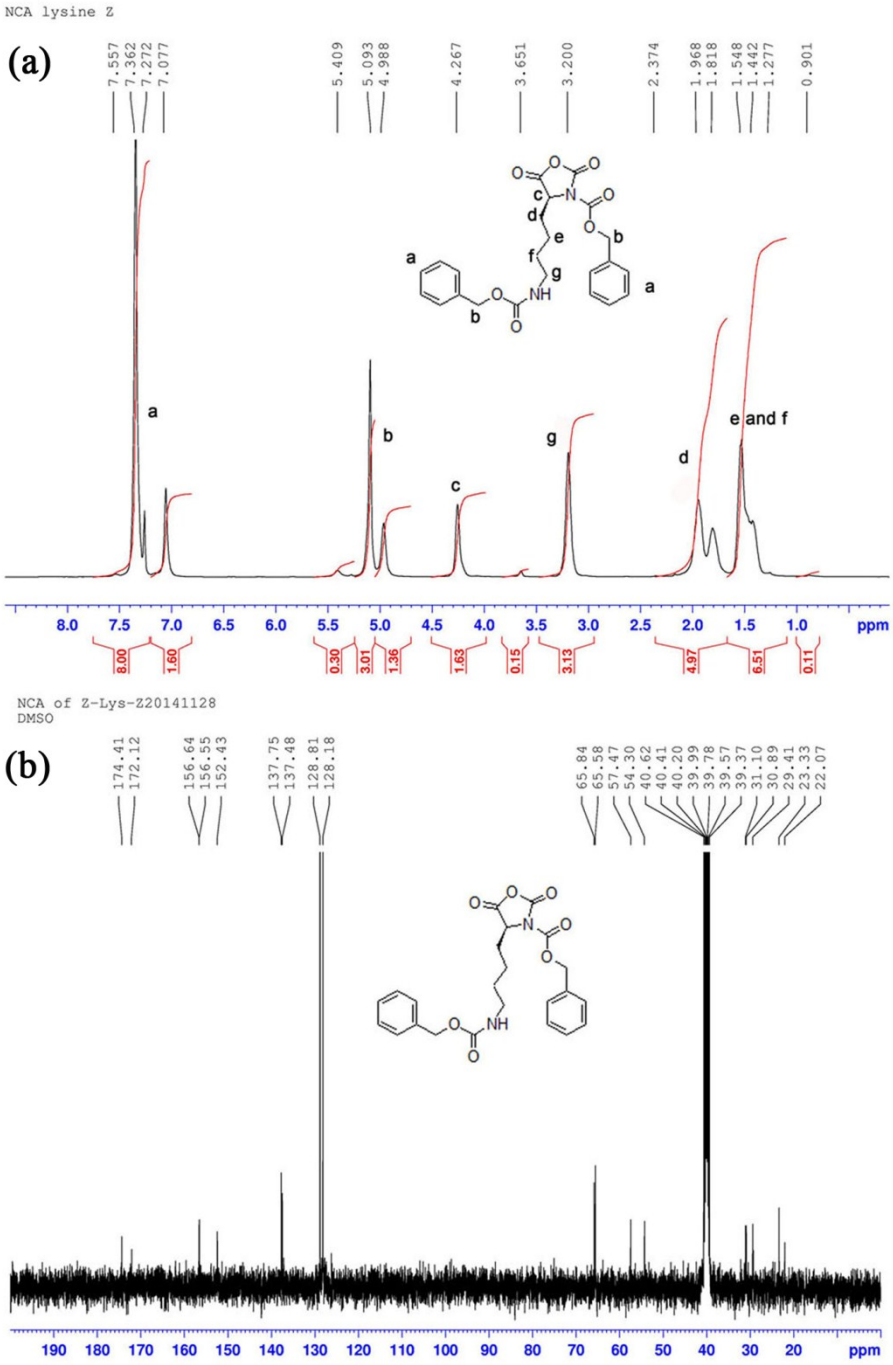
Supplementary Figure 1. **(a)** ^{1}H NMR spectrum of Z-Dopa$(Z)_2$. **(b)** ^{13}C NMR spectrum of Z-Dopa$(Z)_2$.



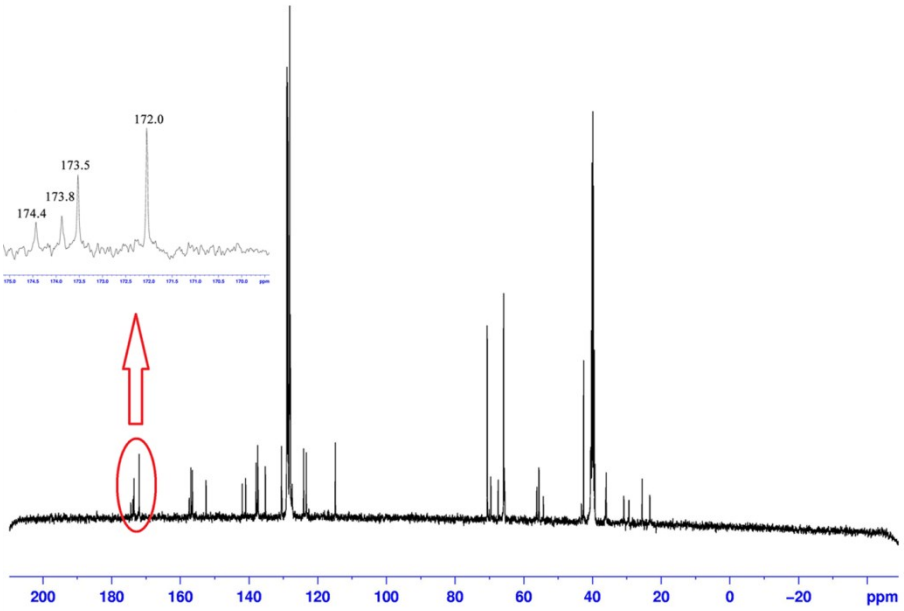
Supplementary Figure 2. **(a)** ^{1}H NMR spectrum of Z-Dopa(Z)$_2$ NCA. **(b)** ^{13}C NMR spectrum of Z-Dopa(Z)$_2$ NCA.



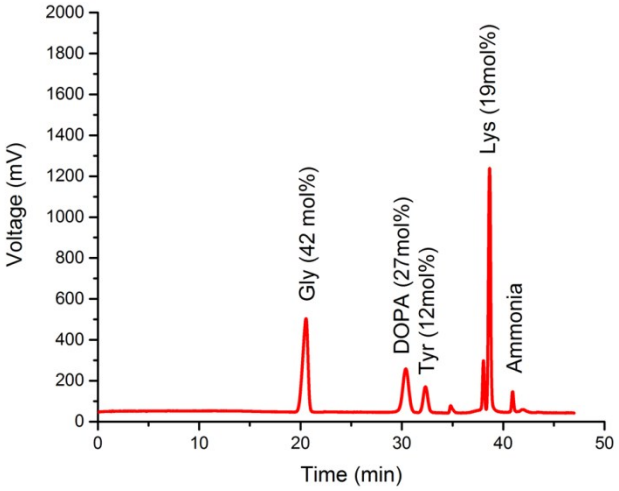
Supplementary Figure 3. **(a)** ^{1}H NMR spectrum of Z-Gly NCA. **(b)** ^{13}C NMR spectrum of Z-Gly NCA.



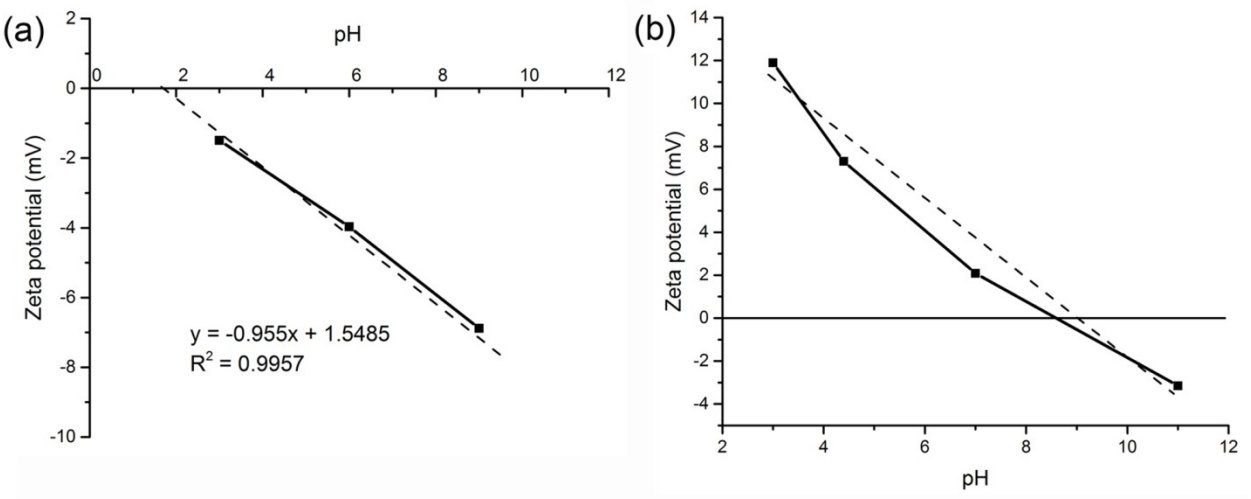
Supplementary Figure 4. **(a)** ^{1}H NMR spectrum of Z-Lys(Z) NCA. **(b)** ^{13}C NMR spectrum of Z-Lys(Z) NCA.



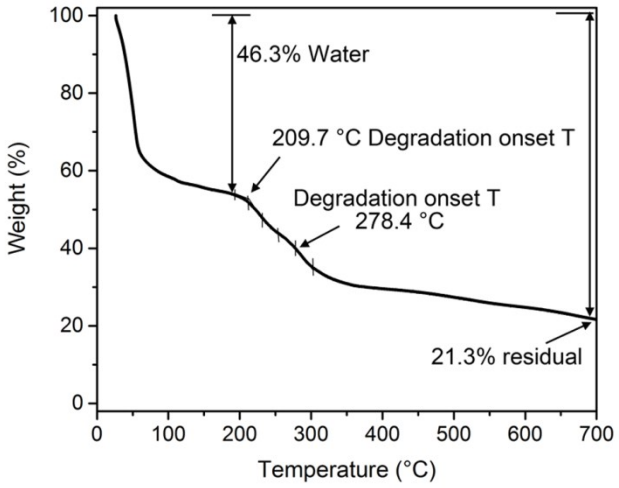
Supplementary Figure 5. ^{13}C NMR spectrum of the co-polypeptide **B** after ring-opening polymerization. The four peaks from left to right correspond to the signal of carbonyl groups of Lys-Cbz, Tyr-O-Bzl, Dopa-$(Cbz)_2$, Gly-H, respectively.



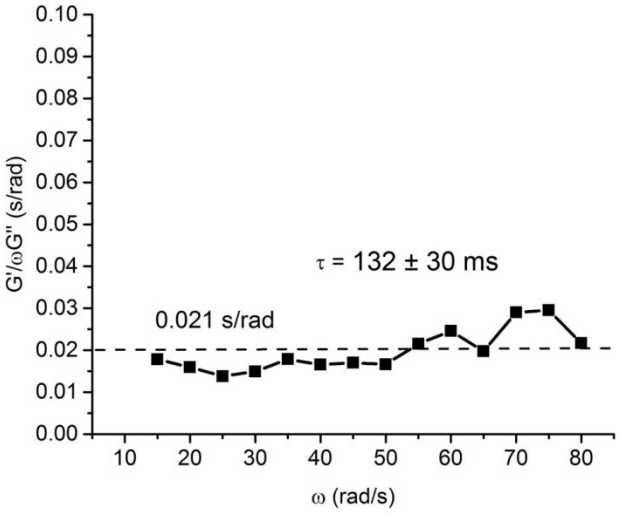
Supplementary Figure 6. Ninhydrin-based amino acid analysis for co-polypeptide **B** after deprotction with 24 hours hydrolysis time.



Supplementary Figure 7. **(a)** Plot of zeta potential ζ *vs.* pH for polypeptide **A2** indicating a IEP of 1.62. **(b)** Plot of ζ *vs.* pH for polypeptide **B** indicating a IEP of ~ 8.9.



Supplementary Figure 8. Thermal gravimetric analysis (TGA) of the coacervate (which **B**:**A2** = 60:40 wt. pct, at pH 6.8) from room temperature to 700 °C.



Supplementary Figure 9. Plot of (G′/ωG″) *vs*. ω for coacervates at 25 °C indicating a relaxation time of 132 ± 30 ms.