

## Supporting Information

### Boosting the Photodynamic Degradation of Islet Amyloid

### Polypeptide Aggregates via a “Bait-Hook-Devastate” Strategy

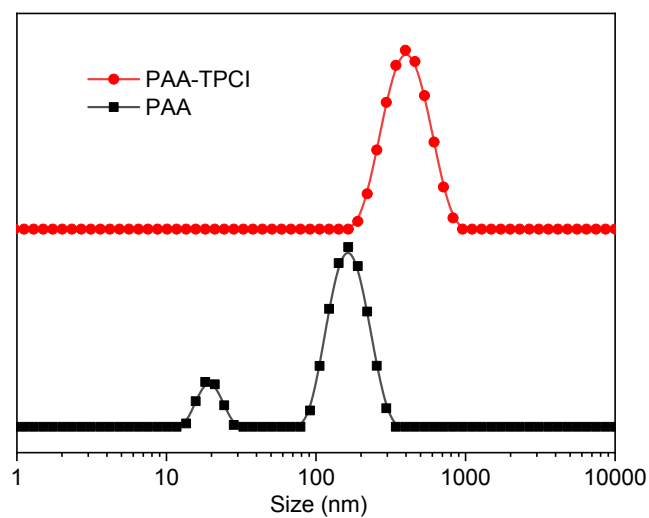
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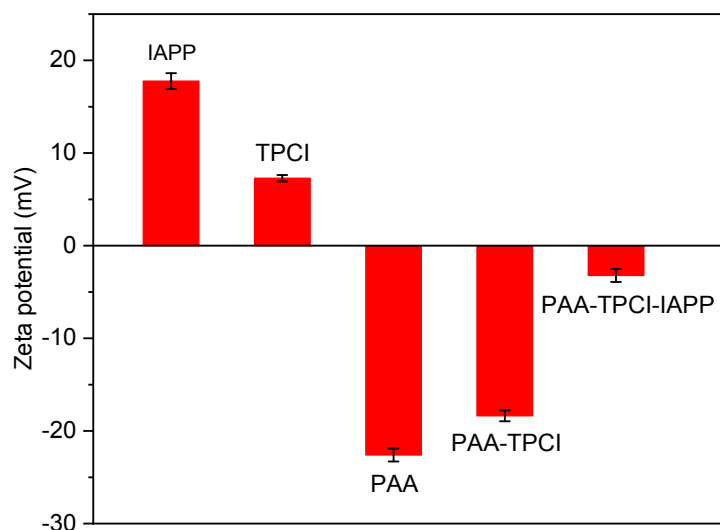
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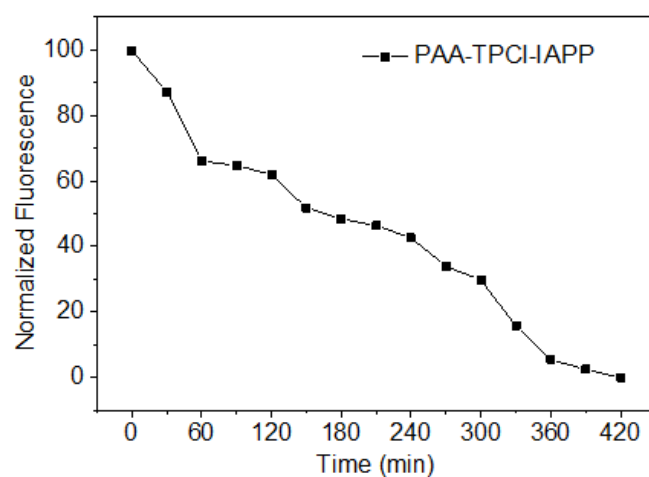
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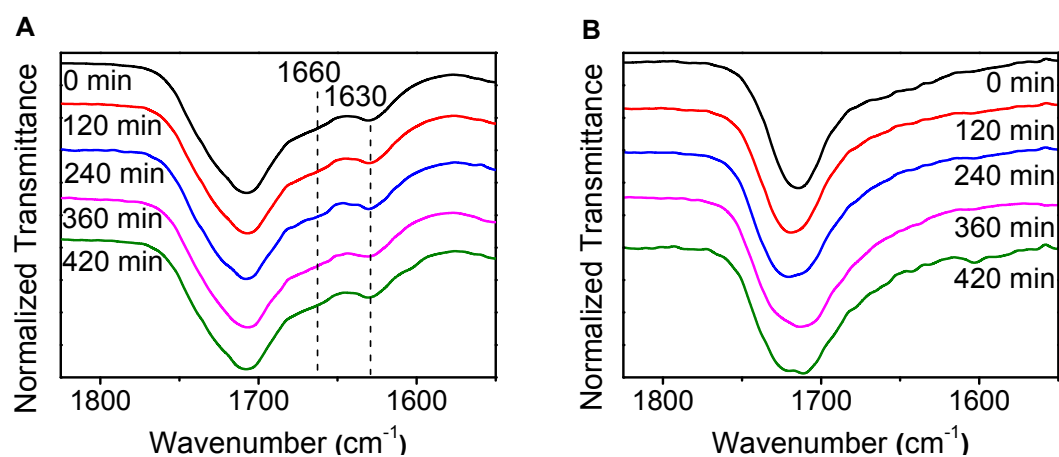
**Figure S1.** Particle size distributions of PAA and PAA-TPCI measured by DLS. The concentrations of PAA and TPCI were  $1.28 \mu\text{M}$  and  $8 \mu\text{M}$  respectively.



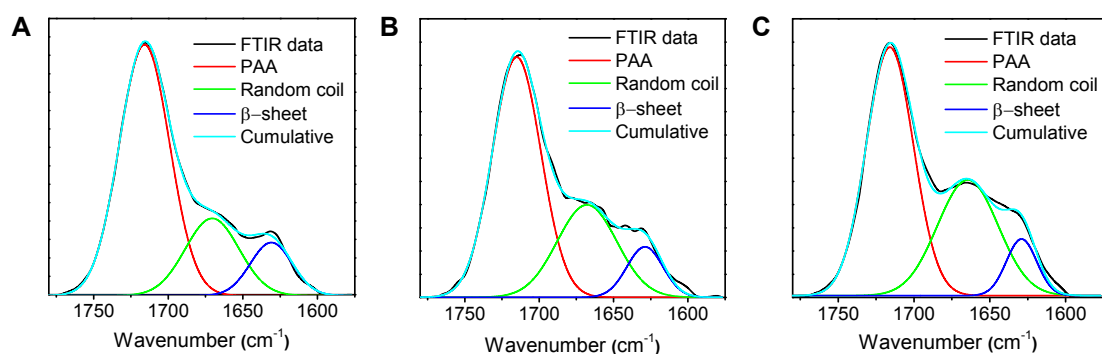
**Figure S2.** Zeta potential of IAPP aggregates (IAPP), TPCI, PAA, and complexes. The concentrations of IAPP aggregates, PAA, TPCI were 32  $\mu\text{M}$ , 1.28  $\mu\text{M}$  and 8  $\mu\text{M}$ , respectively when measured separately. The concentrations of PAA and TPCI in PAA-TPCI were 0.018  $\mu\text{M}$  and 0.11  $\mu\text{M}$  respectively. The concentrations of PAA, TPCI and IAPP aggregates in PAA-TPCI-IAPP were 0.018  $\mu\text{M}$ , 0.11  $\mu\text{M}$ , 32  $\mu\text{M}$  respectively.



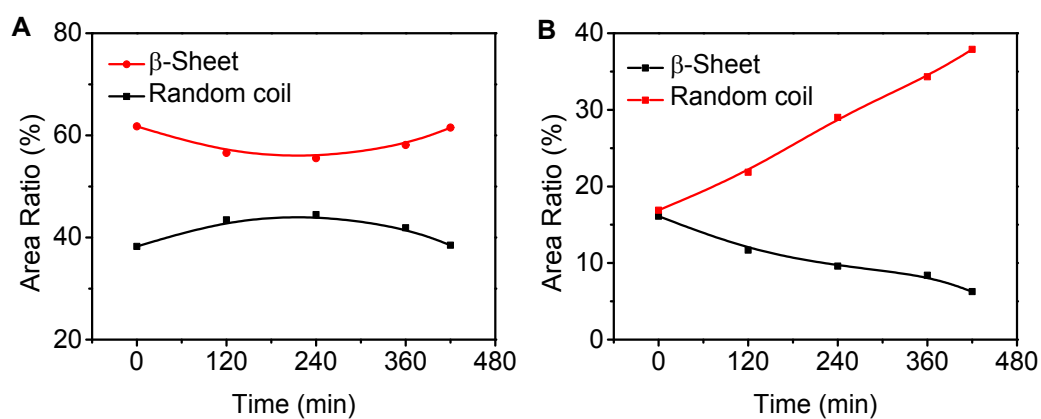
**Figure S3.** ThT fluorescence kinetics of IAPP aggregates in the presence of PAA and TPCI (PAA-TPCI-IAPP) upon LED light illumination in water instead of tris-HCl buffer. The concentrations of IAPP aggregates and TPCI were 32  $\mu$ M and 8  $\mu$ M respectively.



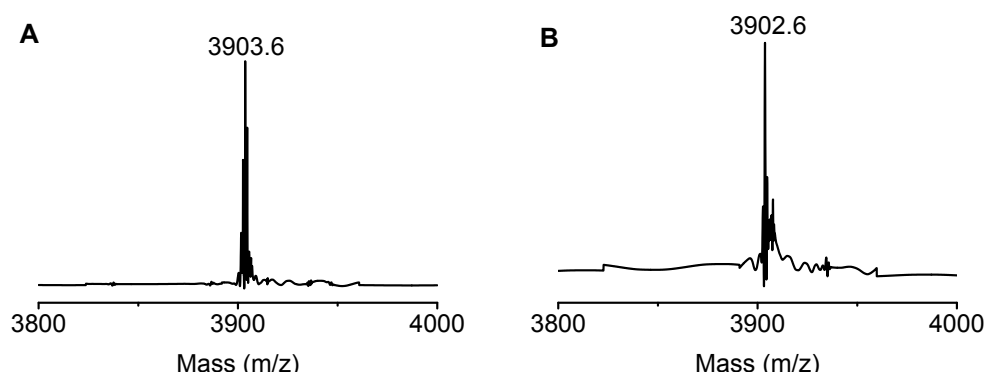
**Figure S4.** FTIR spectra of PAA-IAPP (A), PAA-TPCI (B) under different illumination time points. The concentrations of IAPP aggregates and TPCI were 32  $\mu\text{M}$  and 8  $\mu\text{M}$  respectively.



**Figure S5.** FTIR multiple-peak fit spectra of the PAA-TPCI-IAPP sample after being irradiated for 120 min (A), 240 min (B) and 360 min (C). The concentrations of IAPP aggregates and TPCI were 32  $\mu$ M and 8  $\mu$ M respectively.

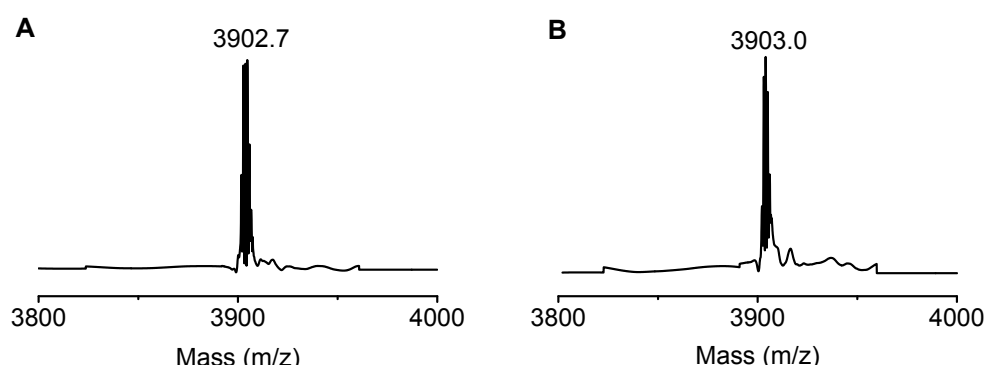


**Figure S6.** The changes of random coil and  $\beta$ -sheet peak area ratio of TPCI-IAPP (A) and PAA-TPCI-IAPP (B) samples in FTIR multi-peak fitting spectrum at different illumination time points. The concentrations of IAPP aggregates and TPCI were  $32\ \mu\text{M}$  and  $8\ \mu\text{M}$  respectively.

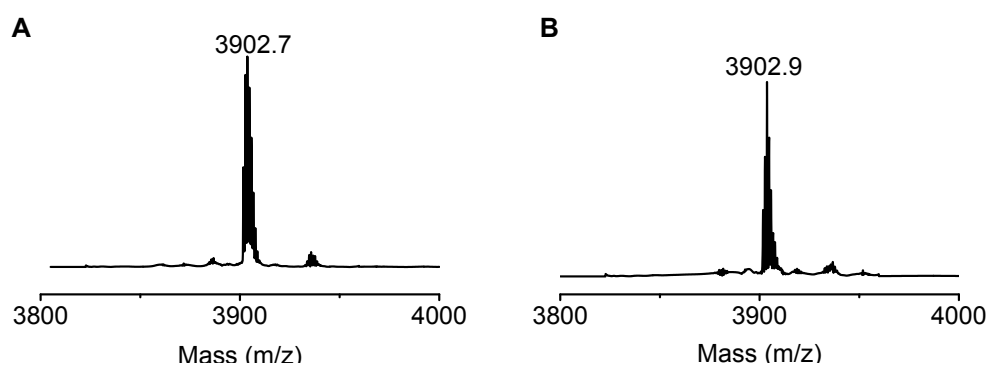


**Figure S7.** MALDI-TOF MS spectra of the PAA-IAPP sample before irradiation (A) and after 90 min of irradiation (B). The concentrations of IAPP aggregates and PAA were 8  $\mu\text{M}$  and 0.32  $\mu\text{M}$  respectively.

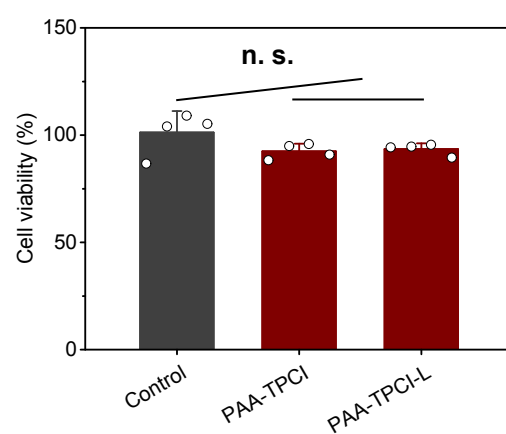




**Figure S8.** MALDI-TOF MS spectra of the TPCI-IAPP sample before irradiation (A) and after 90 min of irradiation (B). The concentrations of IAPP aggregates and TPCI were 8  $\mu$ M and 2  $\mu$ M respectively.



**Figure S9.** MALDI-TOF MS spectra of the IAPP sample before irradiation (A) and after 90 min of irradiation (B). The concentration of IAPP aggregates was 8  $\mu$ M.



**Figure S10.** Cell viability of PAA-TPCI without and with light irradiation (16 mW cm<sup>-2</sup>, 90 min).