

## Supporting Information

### Engineering peptide-based biomimetic enzymes for enhanced catalysis

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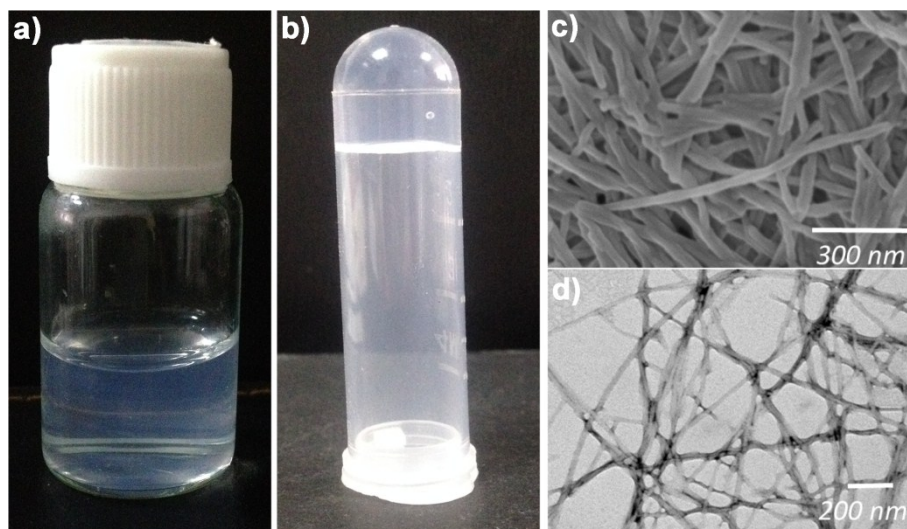
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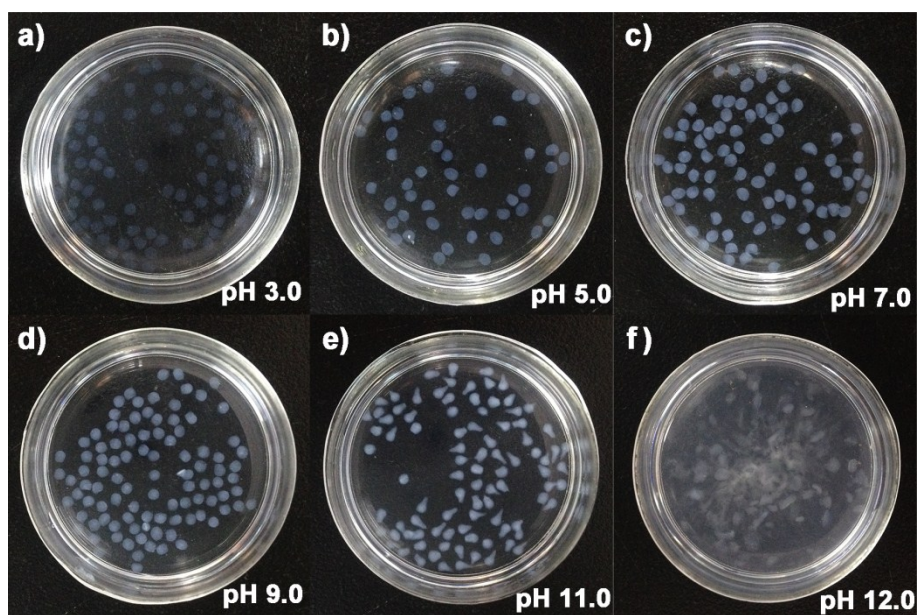
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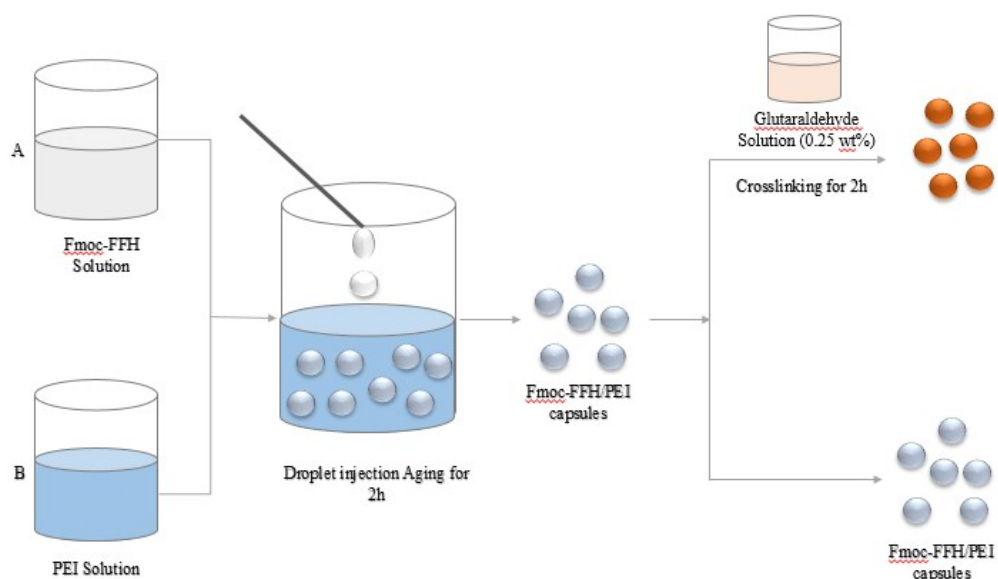
## Supplementary Figures



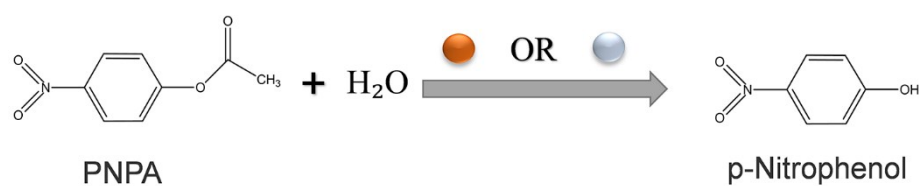
**Fig. S1** a-b) Photograph of Fmoc-FFH solution and Fmoc-FFH hydrogels, respectively. c-d) SEM and TEM image of Fmoc-FFH nanofibers.



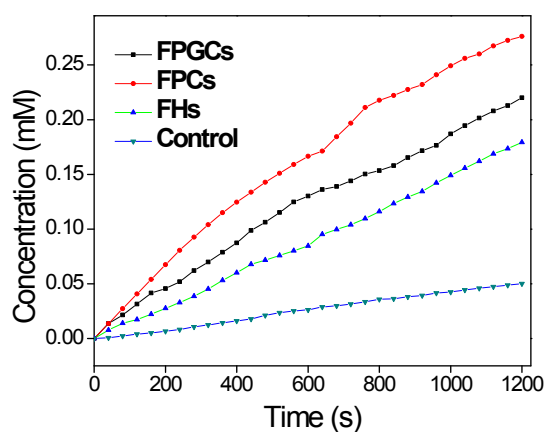
**Fig. S2** Photograph of Fmoc-FFH/PEI capsules formed in 1.0wt% PEI solutions with different pH values ranging from 3.0 to 12.0.



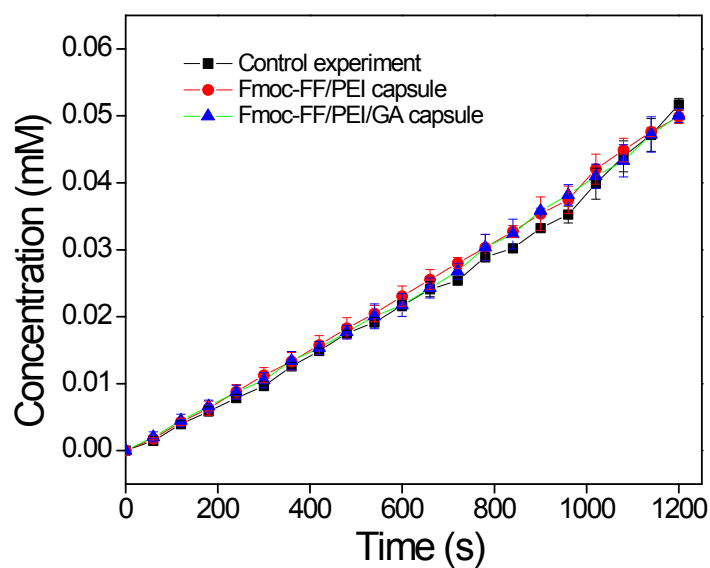
**Fig. S3** Schematic illustration for the formation of Fmoc-FFH//PEI hybrid capsules and Fmoc-FFH//PEI/GA hybrid capsules. The default conditions: Fmoc-FFH: 4 mg mL<sup>-1</sup>, PEI: 1.0wt%, pH of solution A: 9.0, pH of solution B: 8.0.



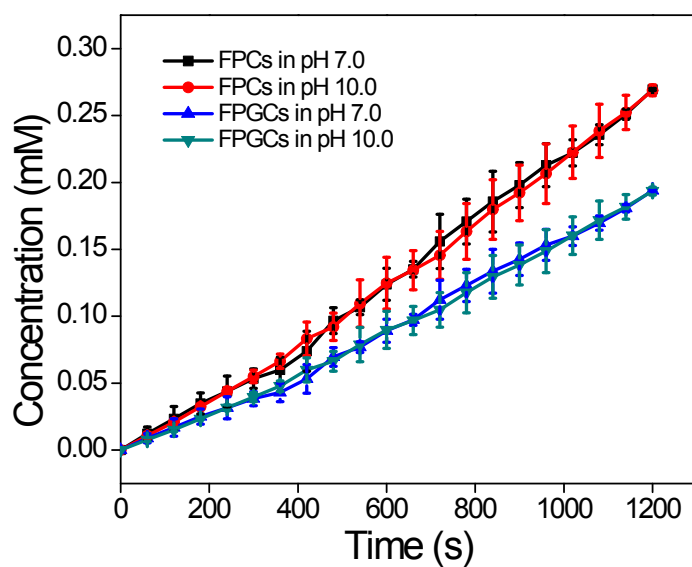
**Fig. S4** The hydrolysis process of PNPA in the presence of hybrid capsules.



**Fig. S5** The hydrolysis activity of PNPA determined in a SpectraMax microplate reader (Molecular Devices, Sunnyvale, CA, USA).



**Fig. S6** The hydrolysis of PNPA in the absence and presence of Fmoc-FF/PEI capsules or Fmoc-FF/PEI/GA capsules.



**Fig. S7** The hydrolysis activity of PNPA using different capsules as catalyst.