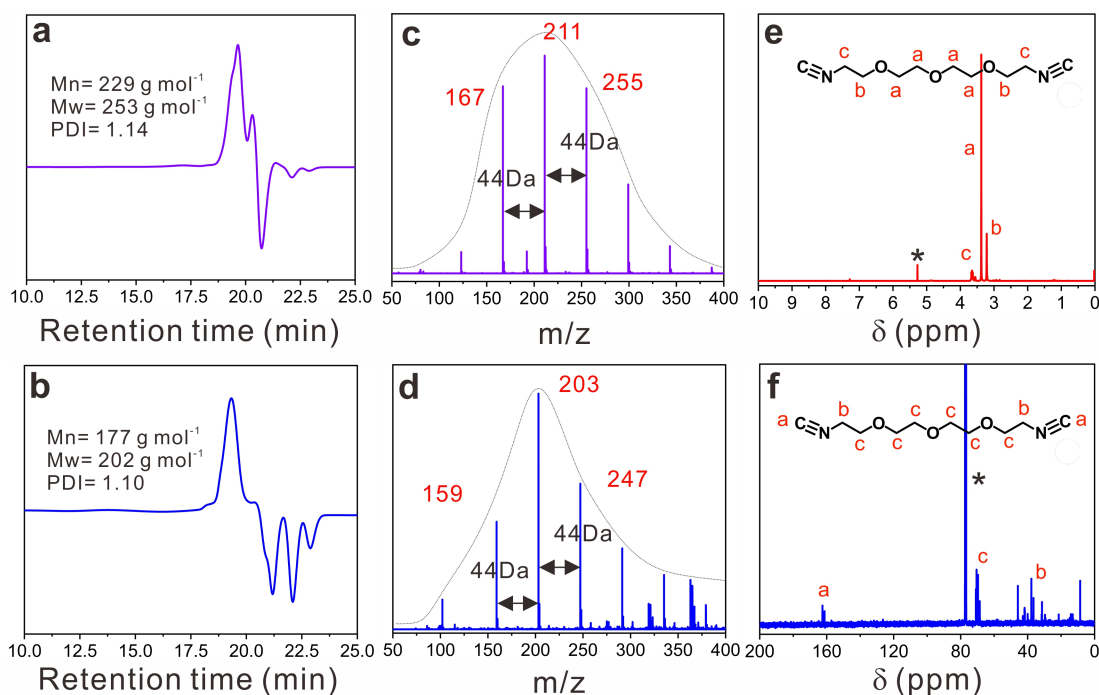
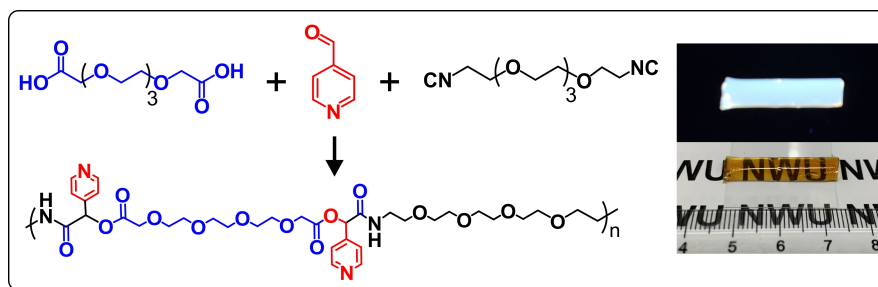


## Supporting Information for

# New Soft-matter Material with Old Chemistry: Passerini Multicomponent Polymerization-induced Assembly of An AIE-active Double-helical Polymer with Rapid Visible-light Degradability



**Figure S1.** GPC curves and mass spectra of (a, c) commercialized HOOC-PEG-COOH and (b, d) as-synthesized NC-PEG-CN. (e)  $^1\text{H}$ -NMR and (f)  $^{13}\text{C}$ -NMR spectra of as-synthesized NC-PEG-CN.

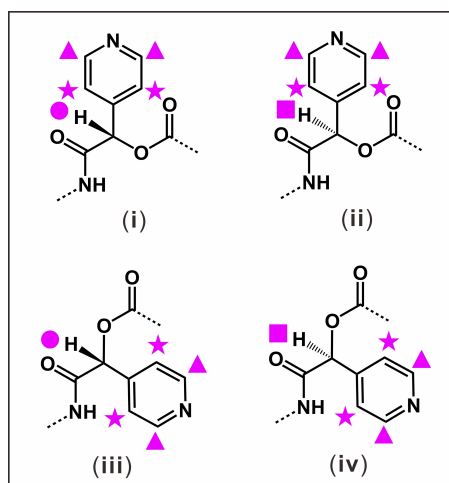
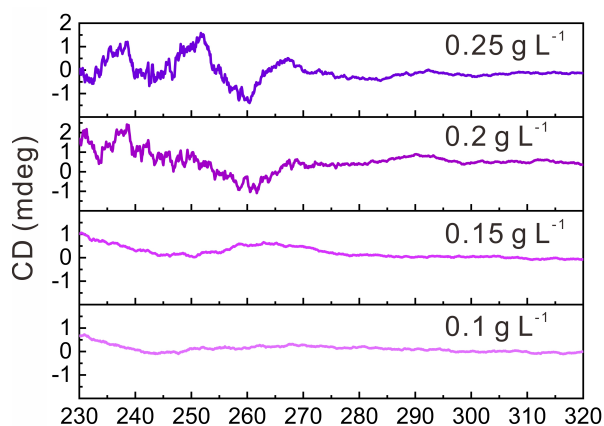


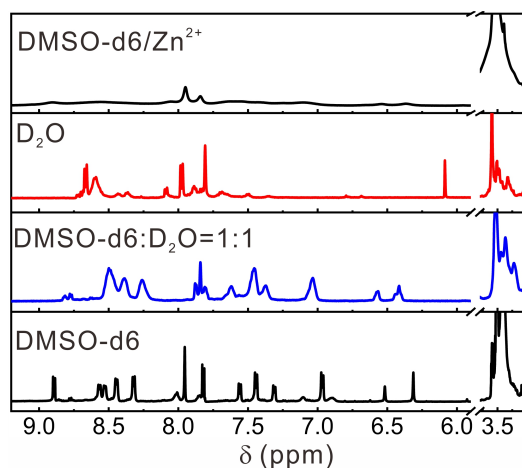
**Figure S2.** Synthesis of WADP via the PMPIA strategy in DMF or THF, and the dried sample under white and UV light irradiation.

**Table 1** Synthesis conditions listed of WADP

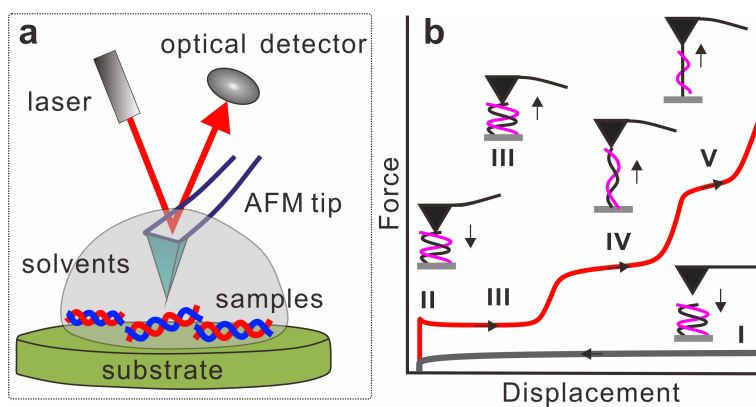
| polymer   | solvent    | molar ratio  | conc. (M)  | time (d) | Mn ( $\times 10^4$ ) | PDI        |
|-----------|------------|--------------|------------|----------|----------------------|------------|
| P1        | THF        | 1:2:1        | 1.6        | 2.5      | 1.5                  | 2.3        |
| P2        | THF        | 1:2:1        | 0.8        | 4        | 1.3                  | 2.5        |
| P3        | THF        | 1:2:1        | 0.8        | 2.5      | 1.7                  | 2.0        |
| P4        | DMF        | 1:2:1        | 0.8        | 4        | 1.2                  | 2.1        |
| P5        | THF        | 1:3:1        | 0.8        | 2.5      | 0.9                  | 1.7        |
| <b>P6</b> | <b>THF</b> | <b>1:2:1</b> | <b>1.6</b> | <b>3</b> | <b>1.8</b>           | <b>2.0</b> |

Note: The molar concentration listed in each case is the total concentration of HOOC-PEG-COOH, PyCHO and CN-PEG-NC. The molar ratio is  $n_{\text{HOOC-PEG-COOH}} : n_{\text{PyCHO}} : n_{\text{CN-PEG-NC}}$ .

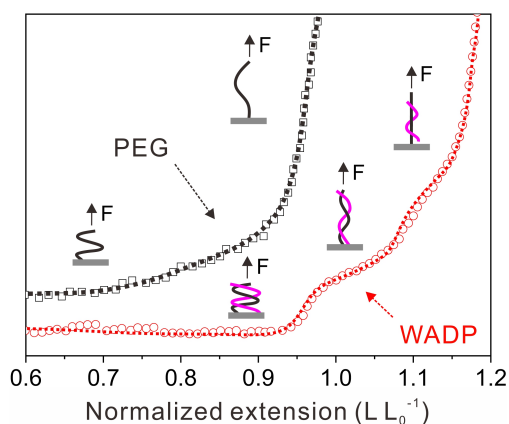
**Figure S3.** The chiral structures of pyridine units in the as-prepared WADP polymers.**Figure S4.** Circular dichroism characterization of WADP with different concentrations in DMSO.



**Figure S5.**  $^1\text{H}$ -NMR spectra of WADP in DMSO- $d_6$ , DMSO- $d_6/\text{D}_2\text{O}$ ,  $\text{D}_2\text{O}$ , and DMSO- $d_6/\text{Zn}^{2+}$ .



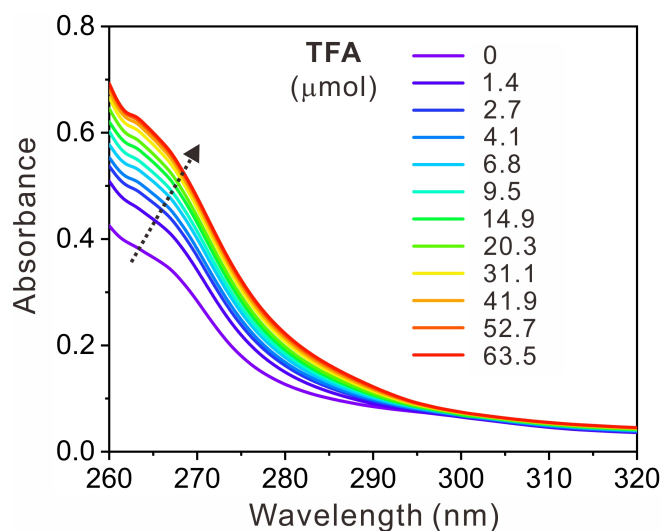
**Figure S6.** (a) The schematic illustration of single-molecule force spectroscopy for in situ studying the conformational transitions in helical polymers. (b) The corresponding conformations of polymers during the characterization process.



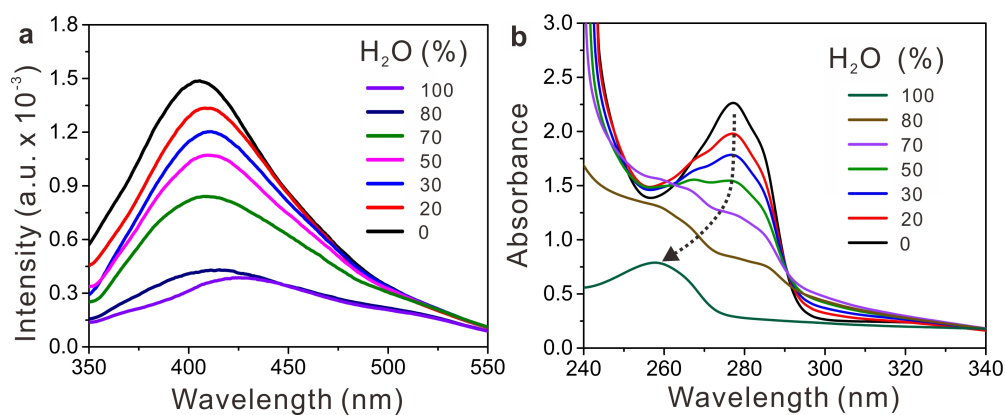
**Figure S7.** Single-molecule force spectroscopy characterization of PEG and WADP in water.



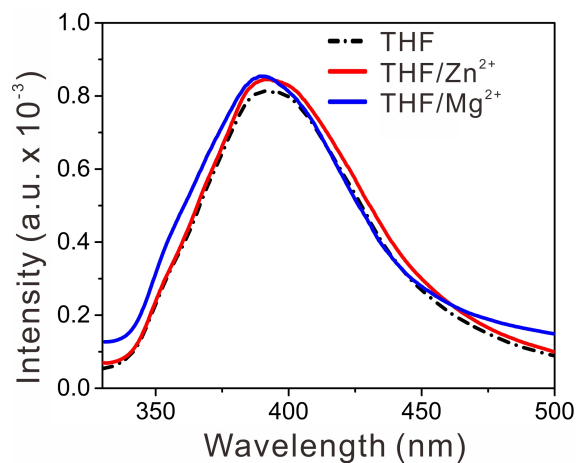




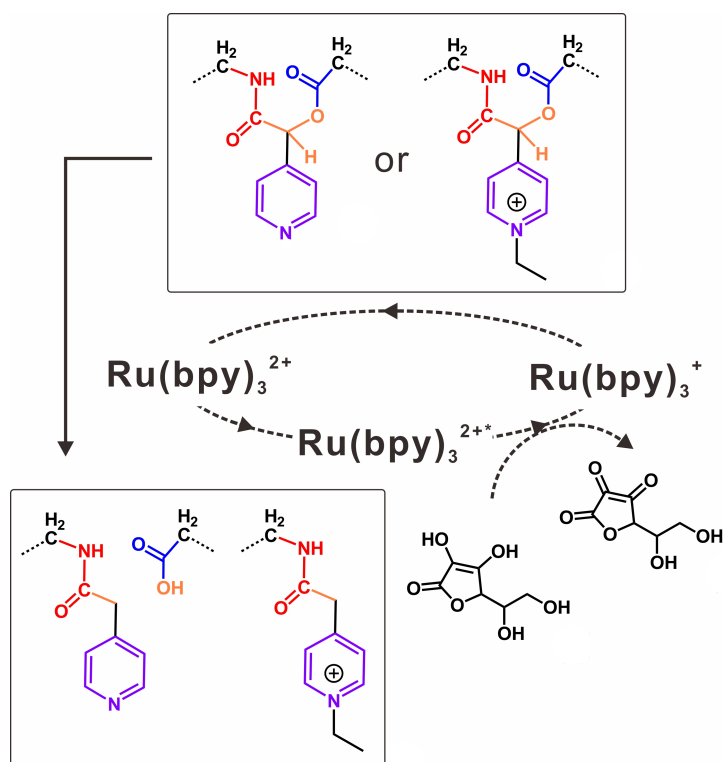
**Figure S10.** UV-vis spectra of WADP in DMSO ( $0.125 \text{ g L}^{-1}$ ) with different contents of trifluoroacetic acid (TFA).



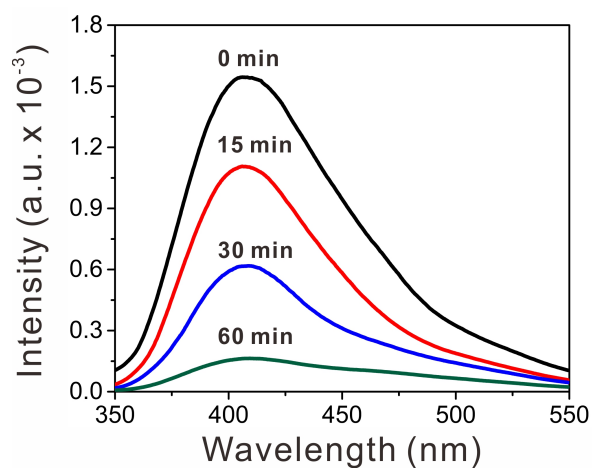
**Figure S11.** (a) The fluorescence and (b) UV spectra of WADP in THF with different weight ratios of water.



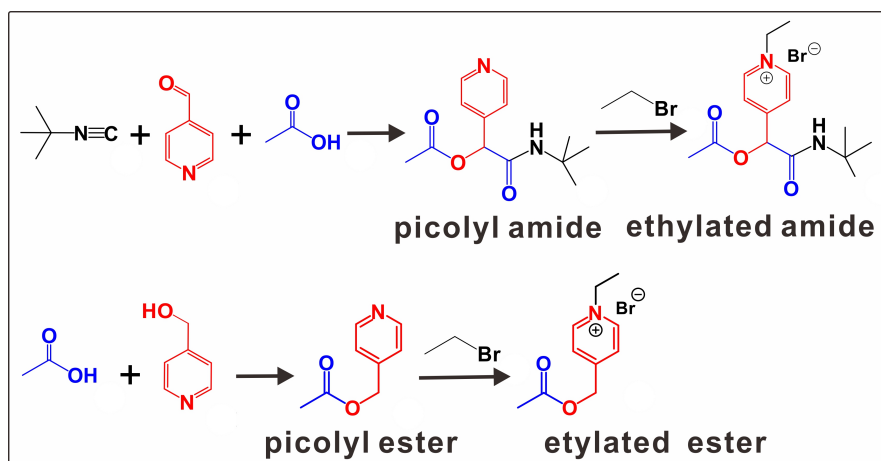
**Figure S12.** Fluorescence spectra of WADP in pure and  $\text{Zn}^{2+}$  (or  $\text{Mg}^{2+}$ )-containing THF.



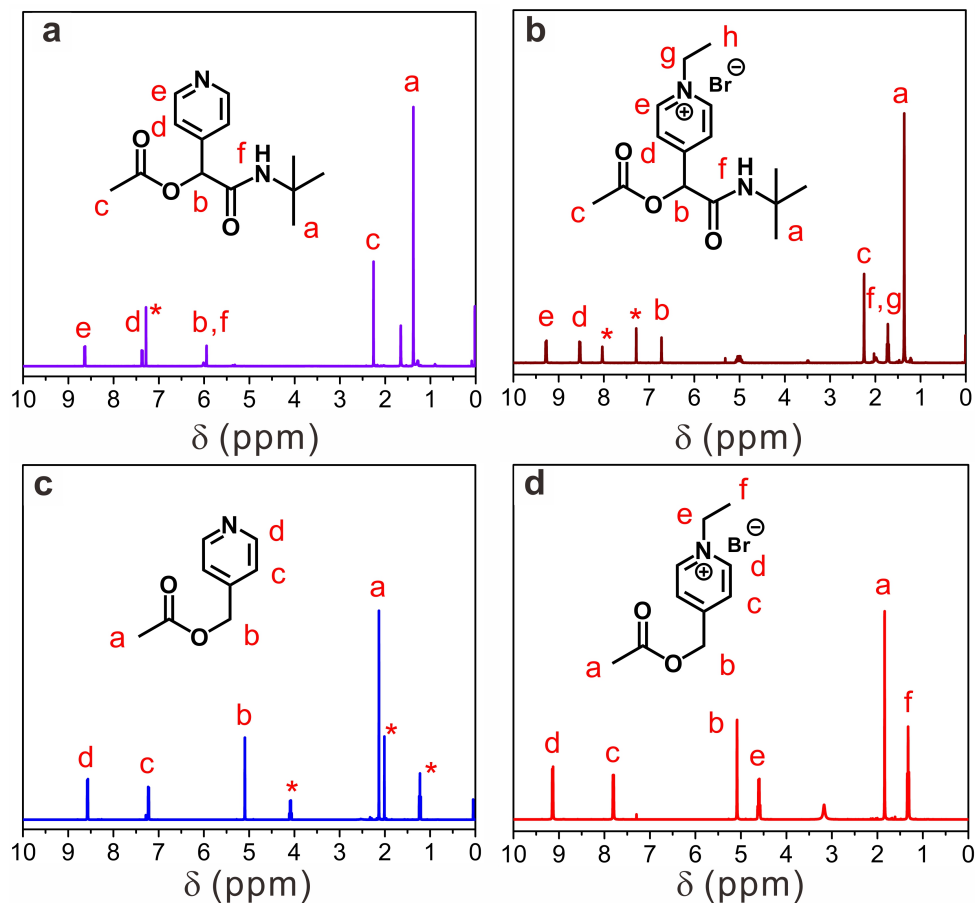
**Figure S13.** The photodegradation mechanism of small molecules and polymers with the catalysis of tris(bipyridine) ruthenium(II) chloride ( $\text{Ru(II)}$ ) and ascorbic acid under blue light irradiation.



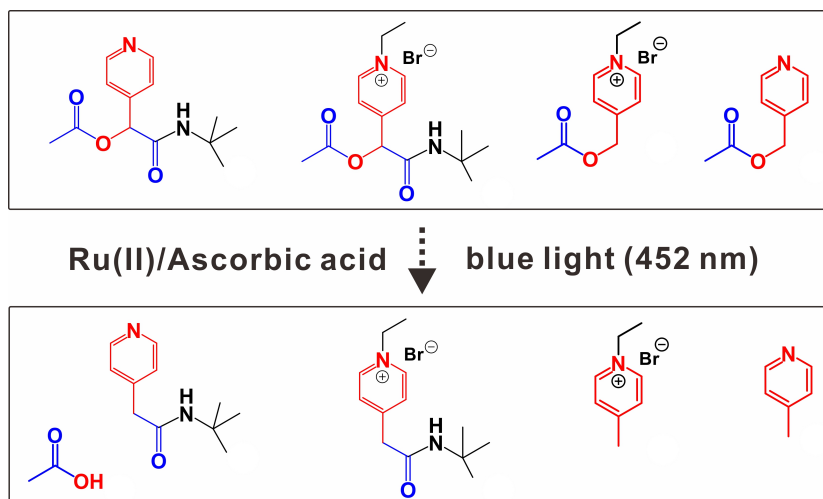
**Figure S14.** Fluorescence spectra of degradation products in THF with different reaction times.



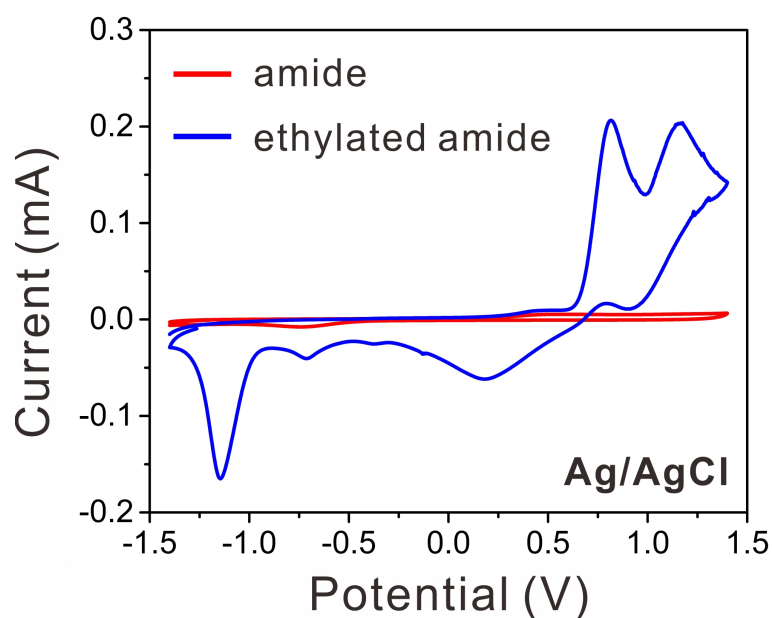
**Figure S15.** Synthesis of four small model molecules, including the amide, ethylated amide, picolyl ester, and ethylated ester.



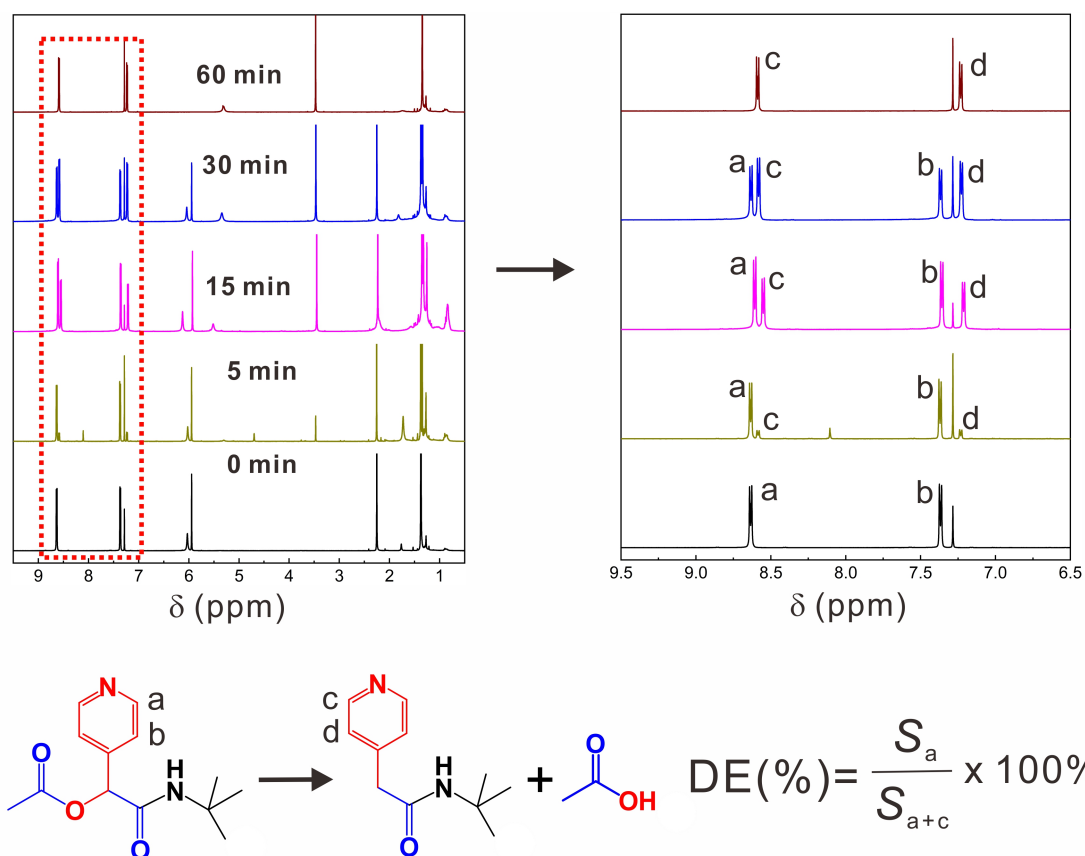
**Figure S16.**  $^1\text{H}$ -NMR spectra of small model molecules including (a) amide, (b) ethylated amide, (c) picolyl ester, and (d) ethylated picolyl ester in DMSO- $d_6$ .



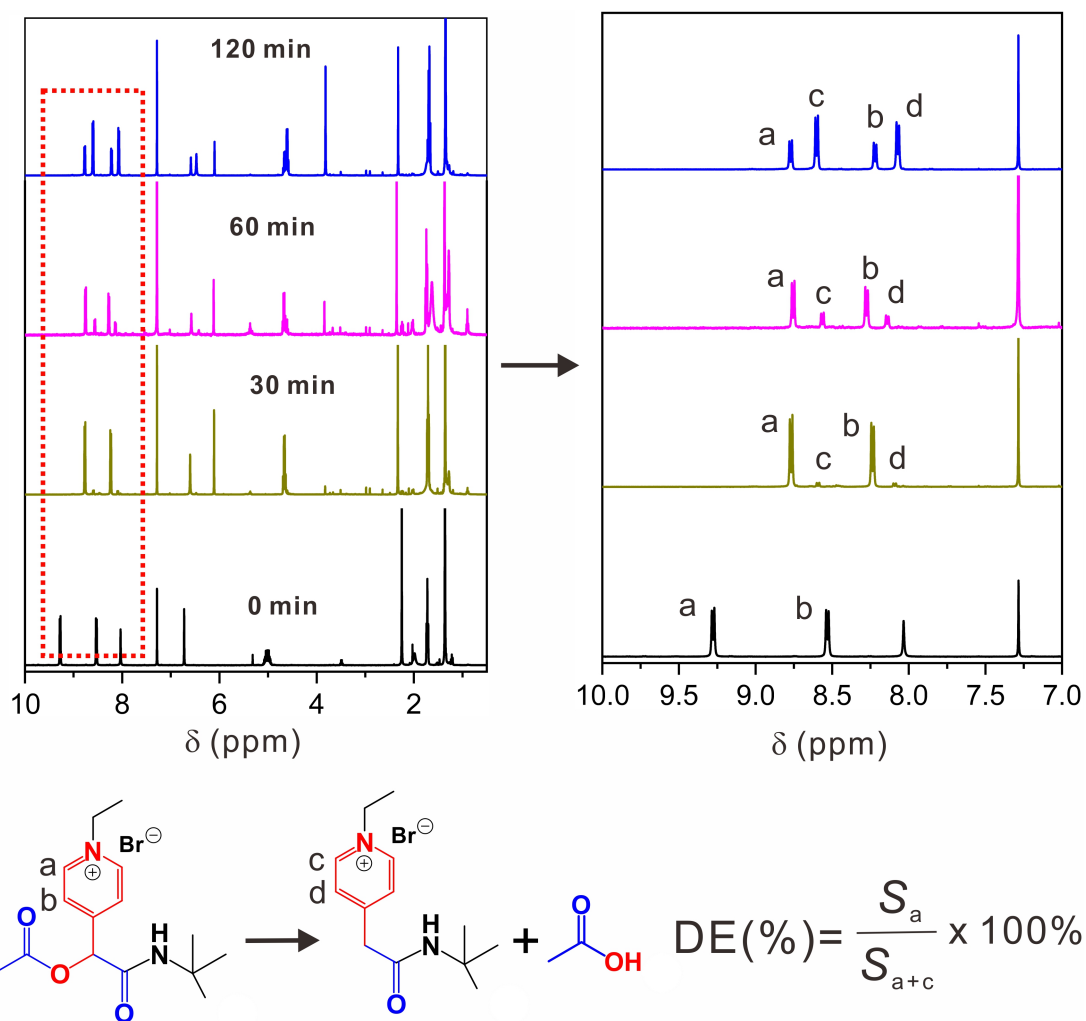
**Figure S17.** Degradation of small model molecules.



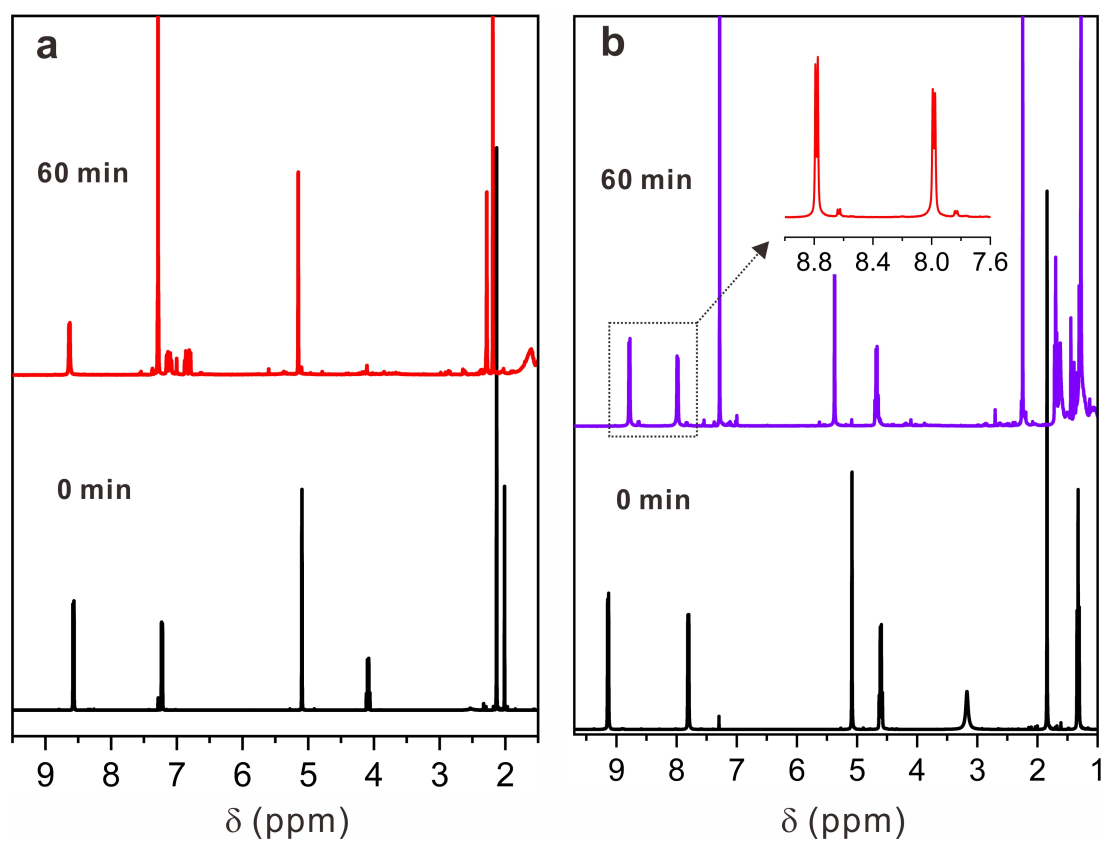
**Figure S18.** Cyclic voltammetry characterization of the model molecules of the amide and ethylated amide. The cyclic voltammetry was performed at room temperature by using a CHI-660E electrochemical workstation. A carbon electrode was applied as the working electrode. A Pt wire constituted the counter electrode, and an Ag/AgCl electrode served as the reference electrode. The supporting electrolyte was 0.1 M lithium perchlorate in dry acetonitrile. The solution was deoxygenated by bubbling argon gas through the solution for 15 min. The potential was swept from -1.5 to 1.5 V with the sweep rate of 100 mV/s to record the current-voltage curves.



**Figure S19.** Calculating the degradation efficiency of the model amide with different irradiation time.



**Figure S20.** Calculating the degradation efficiency of the ethylated amide with different irradiation time.



**Figure S21.**  $^1\text{H}$ -NMR spectra of the (a) picolyl ester and (b) ethylated ester before and after 60 min of visible-light irradiation. The degradation efficiency of ethylated ester in (b) is calculated from the area ratio of peaks between 8.6 and 8.8 ppm, or 7.8 and 8.0 ppm.