

***Supporting Information***

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## Supporting Information

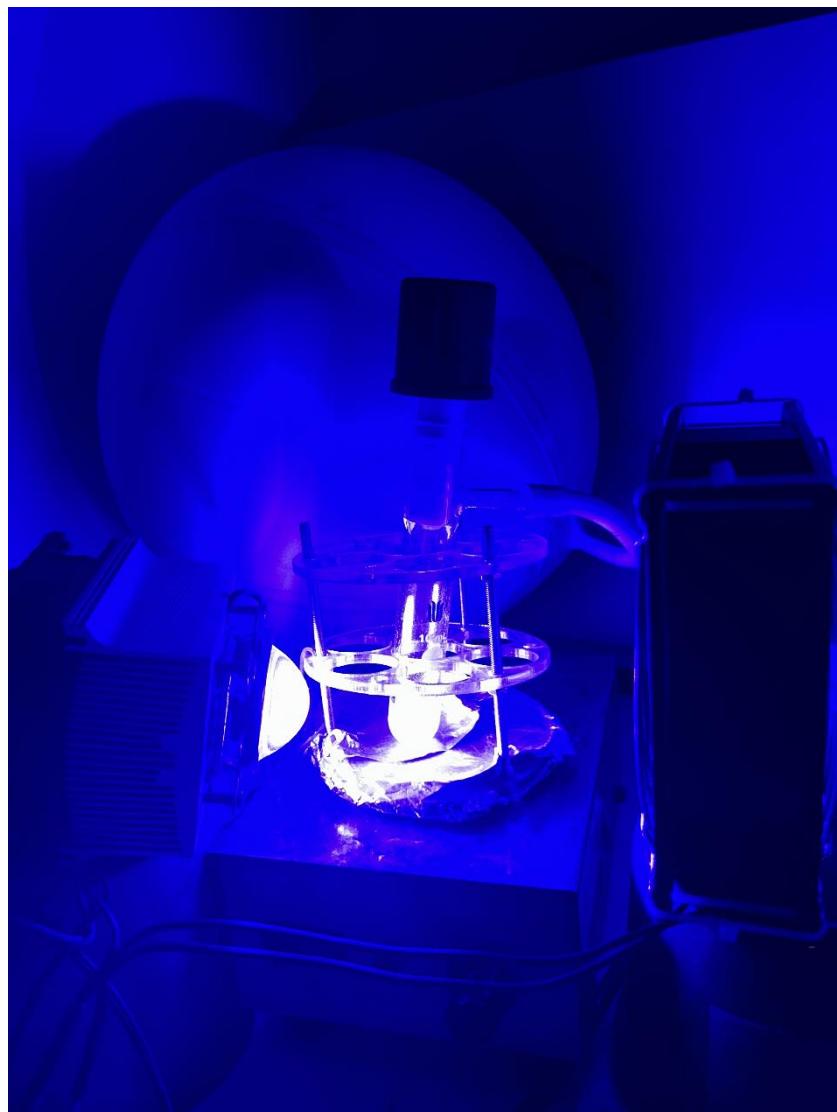
### ***Table of Contents***

<b>1. General Information .....</b>	<b>3</b>
<b>2. Sample Preparation.....</b>	<b>4</b>
<b>3. Optimization of the Reaction Conditions.....</b>	<b>5</b>
Table S1. Screening of catalysts <sup>[a]</sup> .....	5
Table S2. Screening of wavelength of light <sup>[a]</sup> .....	6
Table S3. Screening of the catalyst loading <sup>[a]</sup> .....	6
Table S4. Screening of the amount of HCl <sup>[a]</sup> .....	7
Table S5. Screening of reaction time <sup>[a]</sup> .....	7
<b>4. Comparison with Previous Works .....</b>	<b>8</b>
<b>5. Oxidative Depolymerization of PET materials to PTA.....</b>	<b>8</b>
5.1 Depolymerization of common PET-based consumer products.....	10
5.2 Depolymerization of blended fabrics.....	12
<b>6. Scale-up reaction in continuous-flow reactors.....</b>	<b>14</b>
Table S6. Optimization of Continuous Flow Reaction Conditions <sup>[a]</sup> .....	16
6.1 Large-Scale Reaction for PET Depolymerization in continuous-flow reactors .....	16
<b>7. Mechanistic Studies .....</b>	<b>18</b>
7.1 Formic Acid Capture .....	18
7.2 TEMPO trapping reaction .....	18
7.3 BHT inhibition reaction.....	19
7.4 Water contact angle assessments.....	19
7.5 FT-IR spectra.....	21
<b>8. Determination of Cerium (Ce) Content in PTA by ICP-MS .....</b>	<b>21</b>
<b>9. NMR spectra .....</b>	<b>22</b>

## 1. General Information

The  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR spectra were recorded on 400 MHz Bruker FT-NMR spectrometers (400 MHz or 100 MHz, respectively). Unless otherwise noted, all spectra were acquired in  $\text{CDCl}_3$  or  $\text{DMSO}-d_6$ . Chemical shifts are reported in parts per million (ppm,  $\delta$ ), downfield from tetramethylsilane (TMS,  $\delta = 0.00$  ppm) and are referenced to residual solvent ( $\text{CDCl}_3$ ,  $\delta = 7.26$  ppm ( $^1\text{H}$ ) and 77.00 ppm ( $^{13}\text{C}$ ) or  $\text{DMSO}-d_6$ ,  $\delta = 2.50$  ppm ( $^1\text{H}$ ) and 39.5 ppm ( $^{13}\text{C}$ ). Coupling constants were reported in Hertz (Hz). Data for NMR spectra were reported as follows: s = singlet, coupling constant (Hz), and integration. High resolution mass spectroscopy data of the product were collected on a Waters Micromass GCT instrument (EI) or an Agilent Technologies 6540 UHD Accurate-Mass Q-TOF LC/MS (ESI).

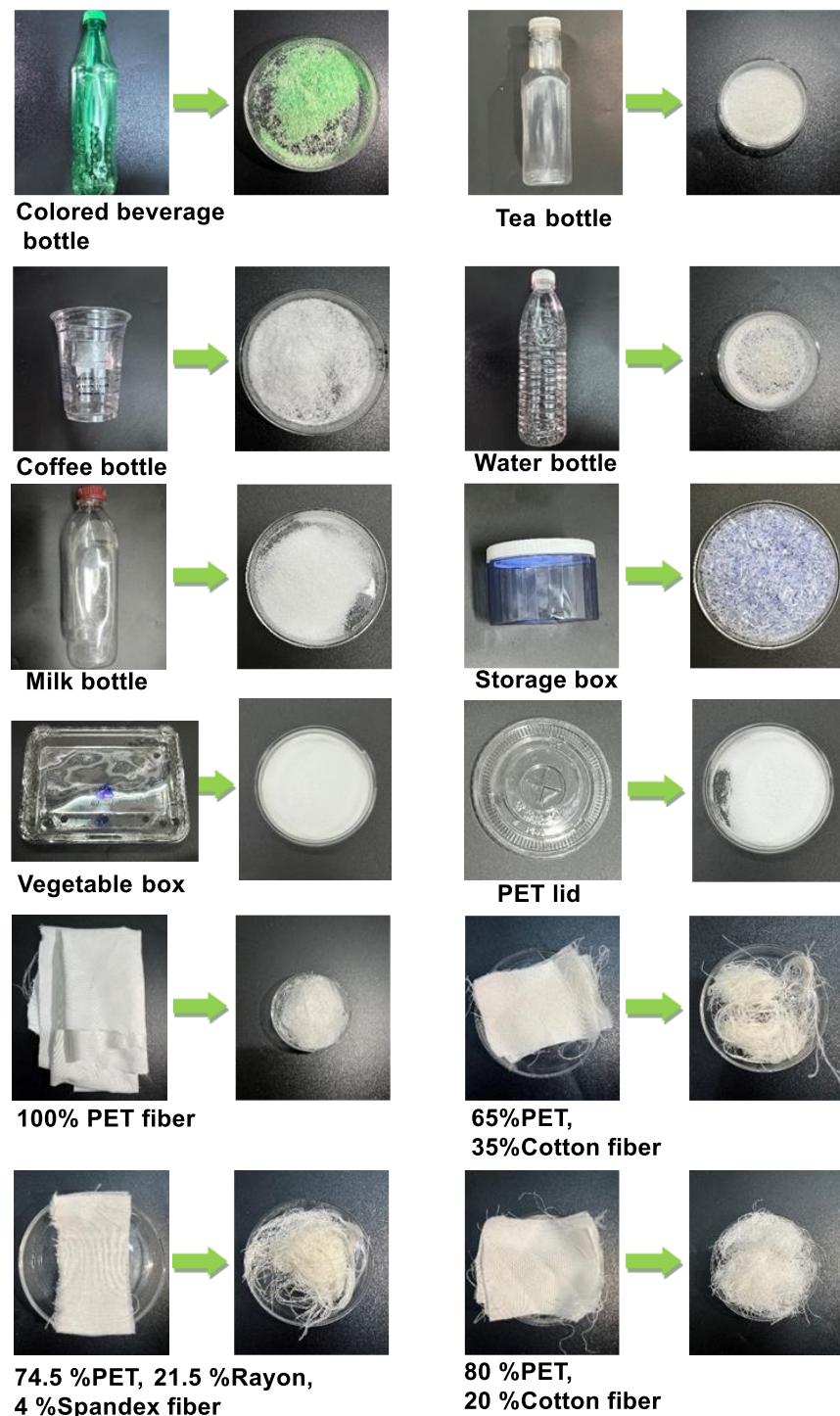
Polyethylene Terephthalate (PET) pellets were supplied by SINOPEC YIZHENG CHEMICAL FIBRE CO. LTD. Before use in reactions, PET pellets need to be crushed and sieved to ensure that their particle size is 30-50 mesh (300-500  $\mu\text{m}$ ). Terephthalic acid (PTA) was obtained as a white crystalline solid through post-treatment of the reaction mixture. Analytical grade Hexafluoroisopropanol (HFIP, >99%), Cerium trichloride ( $\text{CeCl}_3$ , 99.99%), Cerium dioxide ( $\text{CeO}_2$ , 99.99%), Cerium disulfate ( $\text{Ce}(\text{SO}_4)_2$ , 99.99%), Cerium fluoride ( $\text{CeF}_3$ , 99.99%) and Ferric trichloride ( $\text{FeCl}_3$ , 99.99%) were purchased from Aldrich and used without further purification.



**Figure S1: The setting-up reactions: a 50 W 450 nm LED lamp and a reaction tube.**

## 2. Sample Preparation

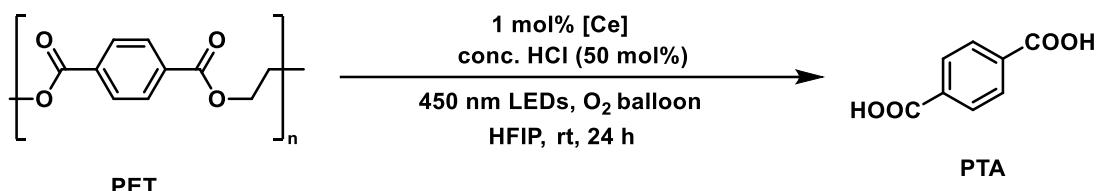
To facilitate higher efficiencies for oxidative depolymerization of plastic products, all PET materials are uniformly shredded into powder using a high-speed shredder in advance.



**Figure S2: Sample preparation**

### 3. Optimization of the Reaction Conditions

**Table S1. Screening of catalysts<sup>[a]</sup>**



Entry	[Ce] (1 mol%)	Yield (%)
1	-	8
2	CeCl <sub>3</sub>	<b>91</b>
3	CeO <sub>2</sub>	58
4	Ce(SO <sub>4</sub> ) <sub>2</sub>	49
5	CeF <sub>3</sub>	51
6	FeCl <sub>3</sub>	58

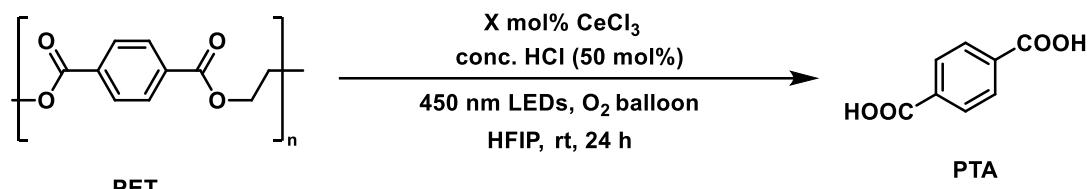
<sup>[a]</sup> Reaction conditions: PET (0.25 mmol), [Ce] or [Fe] (1 mol%), conc. HCl (50 mol%), HFIP (2.0 mL) at room temperature (25 °C), Blue LED (450 nm, 50 W) for 24 h, isolated yield.

**Table S2. Screening of wavelength of light<sup>[a]</sup>**

Entry	Wavelength of light (nm)	Yield (%)
1	390	83
2	420	86
<b>3</b>	<b>450</b>	<b>91</b>

<sup>[a]</sup> Reaction conditions: PET (0.25 mmol), CeCl<sub>3</sub> (1 mol%), conc. HCl (50 mol%), HFIP (2.0 mL) at room temperature (25 °C), LED ( $\lambda$  nm, 50 W) for 24 h, isolated yield.

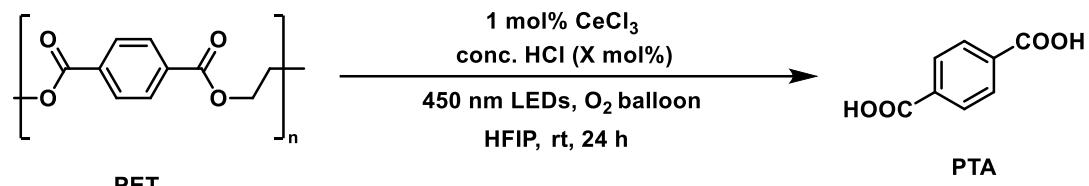
**Table S3. Screening of the catalyst loading<sup>[a]</sup>**



PET	X mol% CeCl <sub>3</sub>	PTA
Entry	CeCl <sub>3</sub> (X mol%)	Yield (%)
1	0.5	56
2	1	<b>91</b>
3	2.5	92
4	5	88

<sup>[a]</sup> Reaction conditions: PET (0.25 mmol), CeCl<sub>3</sub> (X mol%), conc. HCl (50 mol%), HFIP (2.0 mL) at room temperature (25 °C), Blue LED (450 nm, 50 W) for 24 h, isolated yield.

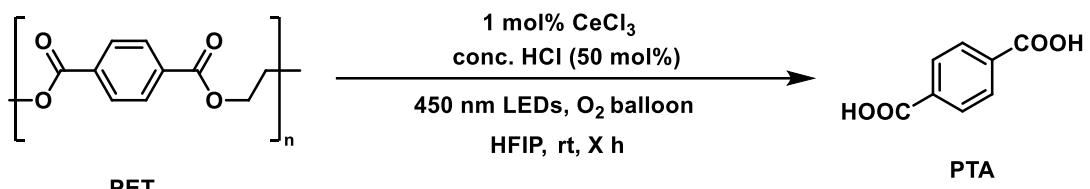
**Table S4. Screening of the amount of HCl<sup>[a]</sup>**



PET	1 mol% CeCl <sub>3</sub>	PTA
Entry	HCl (X mol%)	Yield (%)
1	25	81
2	<b>50</b>	<b>91</b>
3	75	86
4	100	83

<sup>[a]</sup> Reaction conditions: PET (0.25 mmol), CeCl<sub>3</sub> (1 mol%), conc. HCl (X mol%), HFIP (2.0 mL) at room temperature (25 °C), Blue LED (450 nm, 50 W) for 24 h, isolated yield.

**Table S5. Screening of reaction time<sup>[a]</sup>**



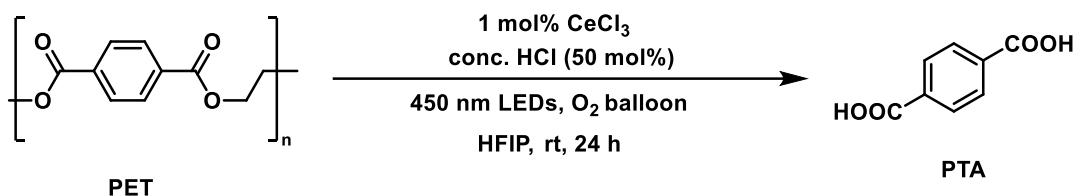
PET	Time (h)	Yield (%)
1	8	21
2	12	46
3	16	65
4	20	82
<b>5</b>	<b>24</b>	<b>91</b>
6	36	92

<sup>[a]</sup> Reaction conditions: PET (0.25 mmol), CeCl<sub>3</sub> (1 mol%), conc. HCl (50 mol%), HFIP (2.0 mL) at room temperature (25 °C), Blue LED (450 nm, 50 W) for X h, isolated yield.

#### 4. Comparison with Previous Works

Entry	T (°C)	wavelengths	t(h)	Pressure	Yield of	Catalyst	Reference
					PTA (%)		
1	85	390 nm	24	1 atm O <sub>2</sub>	97	FeCl <sub>3</sub>	<i>Chin. J. Chem.</i> 2024, <b>42</b> , 2431-2437
2	25	460 nm	72	1 atm O <sub>2</sub>	86	UO <sub>2</sub> (NO <sub>3</sub> ) <sub>2</sub> ·6H <sub>2</sub> O	<i>Science Bulletin</i> 2023, <b>68</b> , 1522-1530
3	260	no	24	1 atm H <sub>2</sub>	87	C/MoO <sub>2</sub>	<i>Angew. Chem. Int. Ed.</i> 2020, <b>59</b> , 19857-19861
4	265	no	24	1 atm H <sub>2</sub>	98	Hf(OTf) <sub>4</sub> /Pd/C	<i>Angew. Chem. Int. Ed.</i> 2022, <b>61</b> , e202112576
5	260	no	24	1 atm H <sub>2</sub>	72	UiO-66	<i>Angew. Chem. Int. Ed.</i> 2022, <b>61</b> , e202117528
6	180	no	24	5 MPa H <sub>2</sub>	85	[BMMI m]Br-Pd/C	<i>Nature Communications</i> , 2024, <b>15</b> , 160
7	25	450 nm	24	1 atm O <sub>2</sub>	95	CeCl <sub>3</sub>	This work

#### 5. Oxidative Depolymerization of PET materials to PTA



**Typical Procedure 1:** PET powder (Mw = 15 kDa) (0.25 mmol, 48 mg) was added to 2 mL HFIP and mixed with CeCl<sub>3</sub> (1 mol%, 0.0025 mmol, 0.62 mg), and concentrated HCl (12 N, 0.125 mmol, 10.0  $\mu$ L). The mixture was stirred under an oxygen atmosphere (1 atm, balloon) at room temperature for 24 hours using a parallel reactor illuminated with blue light (450 nm, 50 W). The solvent (HFIP) was removed in vacuo and recovery and the pH of the residue was adjusted to 10-11 by aq. NaOH (1 N). Subsequently, the mixture was stirred for 1 h at room temperature and the precipitate was separated by filtration. The pH of the filtrate was adjusted to 3–4 with concentrated HCl, resulting in the crystallization of a white solid. The solid was filtered, washed with deionized water (2–3 times), collected, and dried at 80 °C for 12 hours to afford terephthalic acid as a pure white powder (37.8 mg, 91 % yield). In addition, the use of recycled HFIP solvents in the reaction obtain terephthalic acid (35.4 mg, 85% yield).

The steps of filtering: The Büchner funnel vacuum filtration was performed using an 8 cm inner diameter funnel (60  $\mu$ m pore size) inserted into a 50 mL filter flask sealed with a rubber stopper. The flask's sidearm was connected to an SHB-IV circulating water vacuum pump via rubber tubing. A 7 cm outer diameter qualitative filter paper (Cytiva<sup>TM</sup>) was used to fully cover the funnel's pores, wetted with solvent, and adhered by brief vacuum activation. After turning off the pump, the mixture was slowly poured along a glass rod into the funnel center. The pump was restarted, and suction pressure was maintained at 0.02 – 0.04 MPa to rapidly separate the liquid (collected in the flask) from the retained solids.

The yield calculation for PTA obtained from PET depolymerization is systematically determined as follows:

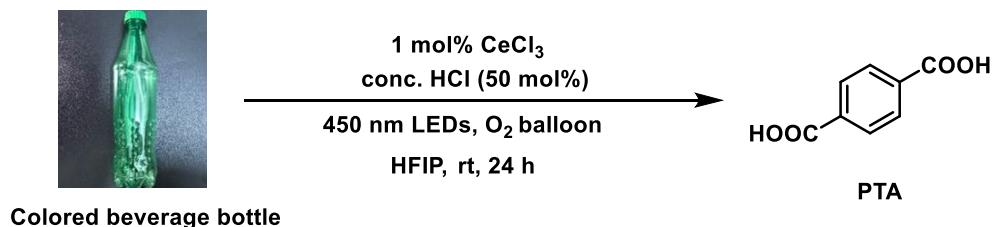
Theoretical PTA mass = (Initial PET mass  $\times$  166.13 g/mol) / (PET repeating unit molar mass 192.13 g/mol)

Rationale: Each PET monomer unit (C<sub>10</sub>H<sub>8</sub>O<sub>4</sub>) yields one PTA molecule (C<sub>8</sub>H<sub>6</sub>O<sub>4</sub>)

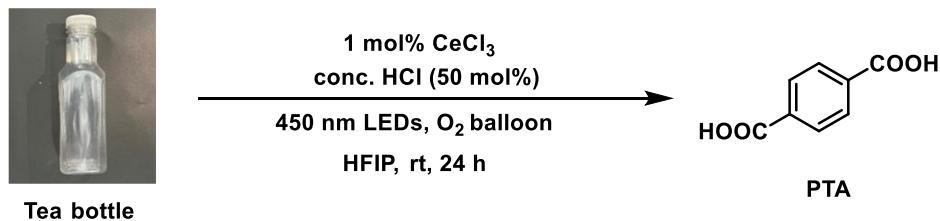
upon complete depolymerization

$$\text{Yield (\%)} = \frac{\text{Actual PTA mass}}{\text{Theoretical PTA mass}} * 100$$

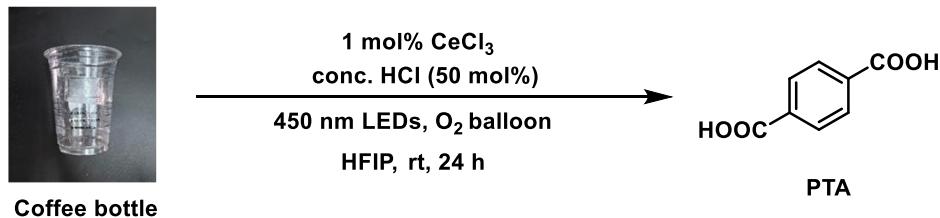
### 5.1 Depolymerization of common PET-based consumer products



Following Typical Procedure 1, the reaction of colored beverage powder (48 mg, 0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (39.46 mg, 95 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).

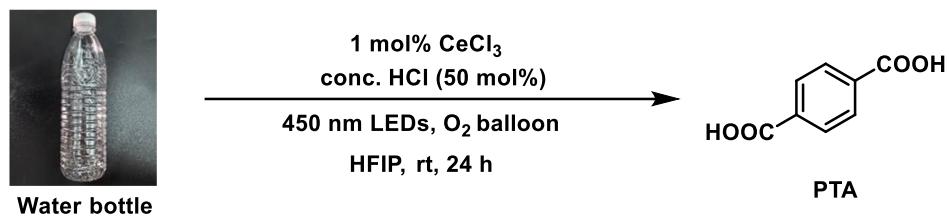


Following Typical Procedure 1, the reaction of tea bottle powder (48 mg, 0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (36.96 mg, 89 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).

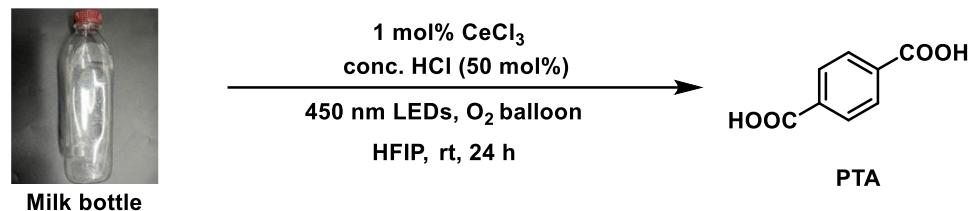


Following Typical Procedure 1, the reaction of coffee bottle powder (48 mg, 0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (32.40 mg, 78 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s,

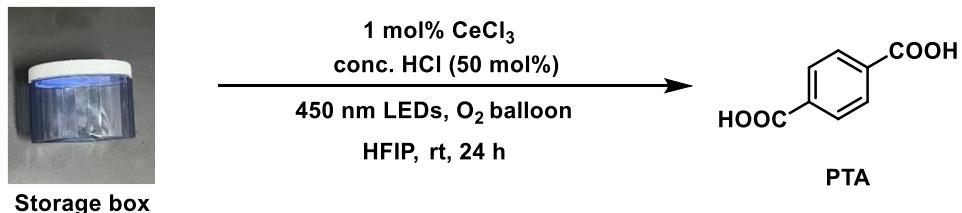
4H).



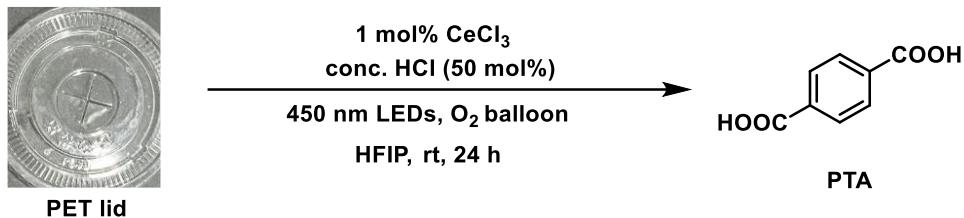
Following Typical Procedure 1, the reaction of water bottle powder (48 mg, 0.25 mmol), CeCl<sub>3</sub> (1 mol%, 0.0025 mmol, 0.62 mg) and conc. HCl (12 N, 0.125 mmol, 10.0  $\mu$ L) in HFIP (2.0 mL) under O<sub>2</sub> atmosphere afforded terephthalic acid (36.55 mg, 88 % yield): white solid. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>)  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).



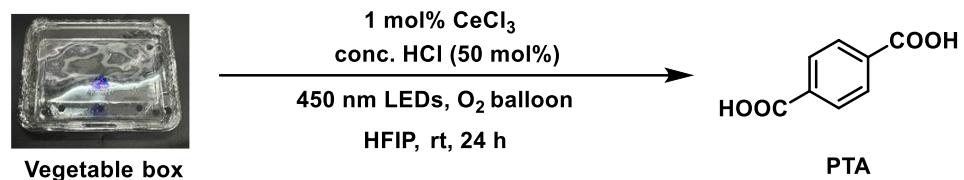
Following Typical Procedure 1, the reaction of milk bottle powder (48 mg, 0.25 mmol), CeCl<sub>3</sub> (1 mol%, 0.0025 mmol, 0.62 mg) and conc. HCl (12 N, 0.125 mmol, 10.0  $\mu$ L) in HFIP (2.0 mL) under O<sub>2</sub> atmosphere afforded terephthalic acid (38.63 mg, 93 % yield): white solid. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>)  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).



Following Typical Procedure 1, the reaction of storage box powder (48 mg, 0.25 mmol), CeCl<sub>3</sub> (1 mol%, 0.0025 mmol, 0.62 mg) and conc. HCl (12 N, 0.125 mmol, 10.0  $\mu$ L) in HFIP (2.0 mL) under O<sub>2</sub> atmosphere afforded terephthalic acid (34.06 mg, 82 % yield): white solid. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>)  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).



Following Typical Procedure 1, the reaction of PET lid powder (48 mg, 0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (33.64 mg, 81 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).



Following Typical Procedure 1, the reaction of Vegetable box powder (48 mg, 0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (35.72 mg, 86 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).

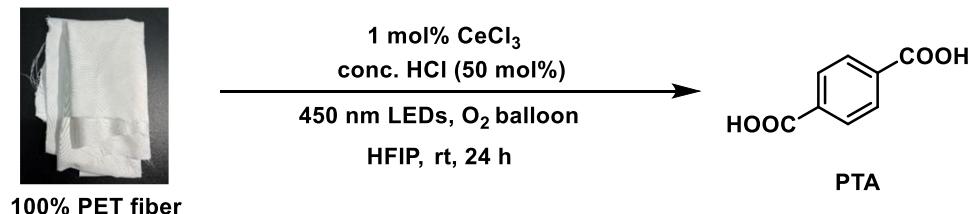
## 5.2 Depolymerization of blended fabrics

Theoretical PTA mass = (Initial PET mass  $\times$  166.13 g/mol) / (PET repeating unit molar mass 192.13 g/mol)

Rationale: Each PET monomer unit ( $\text{C}_{10}\text{H}_8\text{O}_4$ ) yields one PTA molecule ( $\text{C}_8\text{H}_6\text{O}_4$ ) upon complete depolymerization.

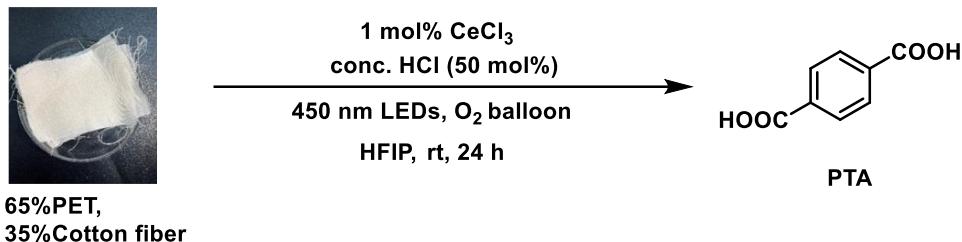
Mass fraction of PET (wt%) =  $(m_{\text{PET}} / m_{\text{total}}) * 100\%$

$$\text{Yield (\%)} = \frac{\text{Actual PTA mass}}{\text{Theoretical PTA mass} * \text{wt\%}} * 100$$

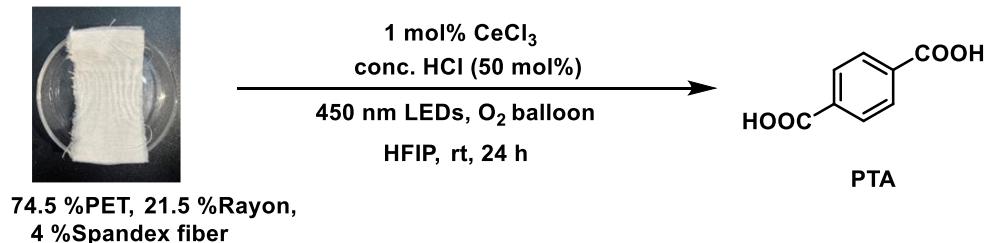


Following Typical Procedure 1, the reaction of 100 % PET fiber (48 mg, 0.25 mmol),

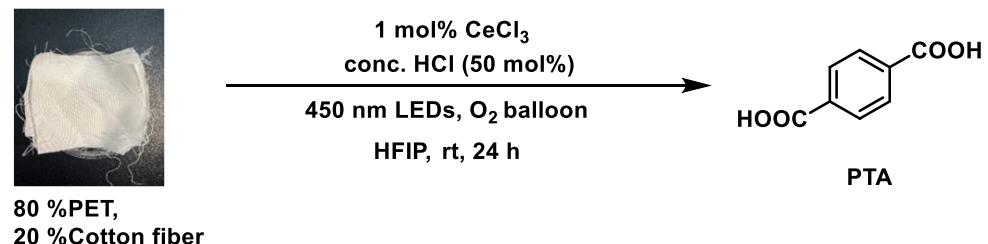
$\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (34.06 mg, 82 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).



Following Typical Procedure 1, the reaction of 65 % PET, 35 % Cotton fiber (48 mg, 0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (22.41 mg, 83 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.30 (s, 2H),  $\delta$  8.04 (s, 4H).

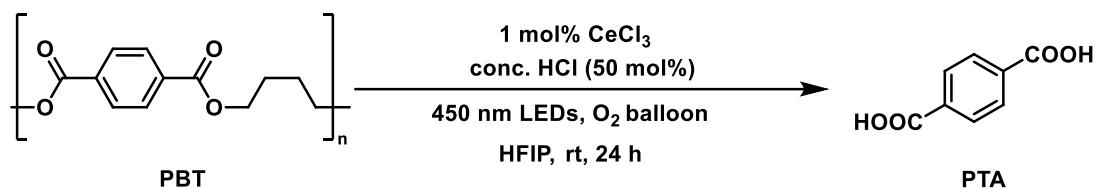


Following Typical Procedure 1, the reaction of 74.5 % PET, 21.5 % Rayon, 4 % Spandex fiber (48 mg, 0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (26.92 mg, 87 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.31 (s, 2H),  $\delta$  8.04 (s, 4H).



Following Typical Procedure 1, the reaction of 80 % PET, 20 % Cotton fiber (48 mg,

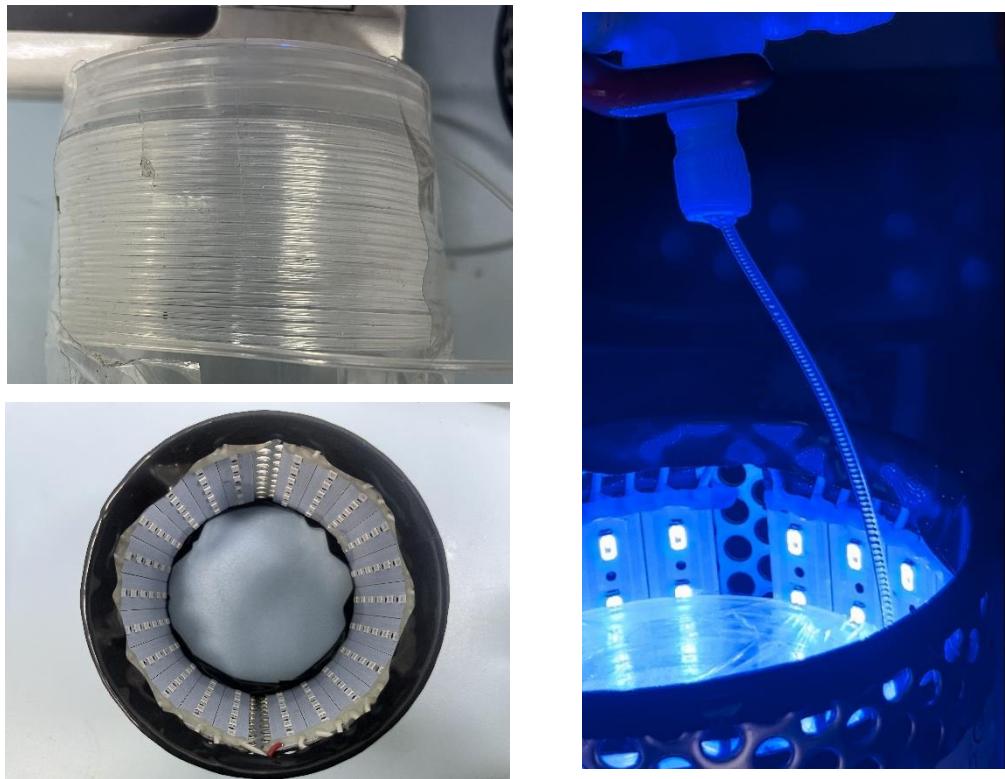
0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (28.57 mg, 86 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.28 (s, 2H),  $\delta$  8.04 (s, 4H).



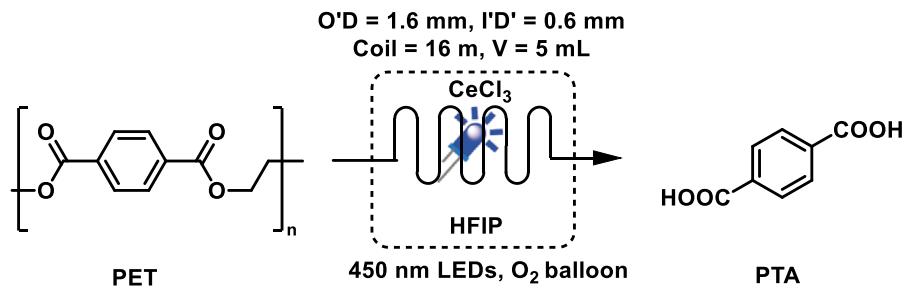
Following Typical Procedure 1, the reaction of PBT powder (55.1 mg, 0.25 mmol),  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg) and conc.  $\text{HCl}$  (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ) in HFIP (2.0 mL) under  $\text{O}_2$  atmosphere afforded terephthalic acid (24.09 mg, 58 % yield): white solid.  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.26 (s, 2H),  $\delta$  8.04 (s, 4 H).

## 6. Scale-up reaction in continuous-flow reactors





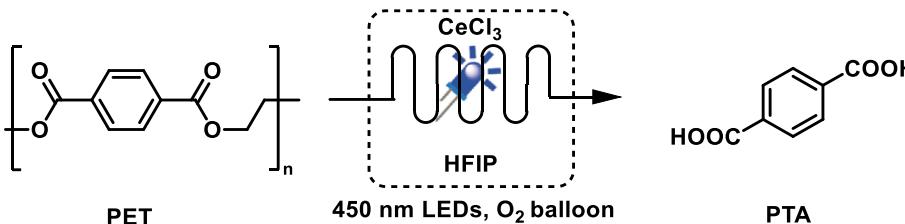
**Figure S3: Photocatalytic-microfluidic field reaction device**



**Typical Procedure 2:** We built a photocatalytic-microfluidic field reaction device consisting of a feeder, microreactor, and collector, and irradiated it with blue LEDs at about 2-3 cm from the reactor to ensure that the reactor received uniform irradiation. And PET (Mw = 15 kDa) (0.5 mmol, 96 mg) was then added to 10 mL of HFIP and mixed with CeCl<sub>3</sub> (1 mol%, 0.005 mmol, 1.24 mg), and concentrated HCl (12 N, 0.25 mmol, 20.0  $\mu$ L). We mixed the reactants and injected them into the microreactor using a syringe pump at a flow rate of 0.1 mL/min under O<sub>2</sub> atmosphere. The residence time of the reaction solution was 45 min. The solvent (HFIP) was removed in vacuo and recovery and the pH of the residue was adjusted

to 10-11 by aq. NaOH (1 N). Subsequently, the mixture was stirred for 1 h at room temperature and the precipitate was separated by filtration. The pH of the filter was adjusted to 3-4 by conc. HCl, with white solid crystallized, was filtered and dried to obtain terephthalic acid (58.98 mg, 71 % yield).  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ )  $\delta$  13.28 (s, 2H),  $\delta$  8.04 (s, 4H).  $^{13}\text{C}$  NMR (101 MHz,  $\text{DMSO}-d_6$ )  $\delta$  166.80, 134.53, 129.58.

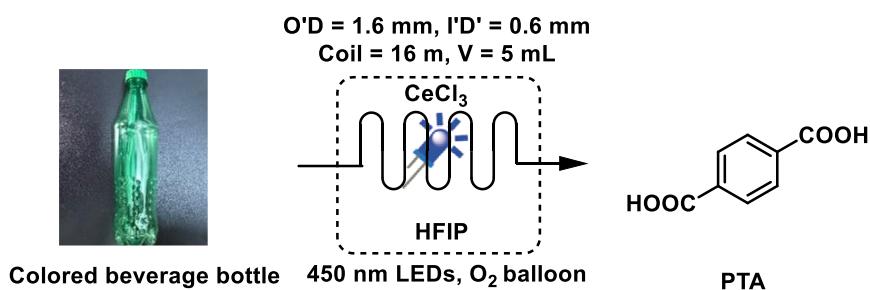
**Table S6. Optimization of Continuous Flow Reaction Conditions <sup>[a]</sup>**



Entry	Rate (mL/min)	Residue time	Yield (%)
1	0.1	45	71
2	0.15	36	54
3	0.2	30	48
4	0.25	26	45

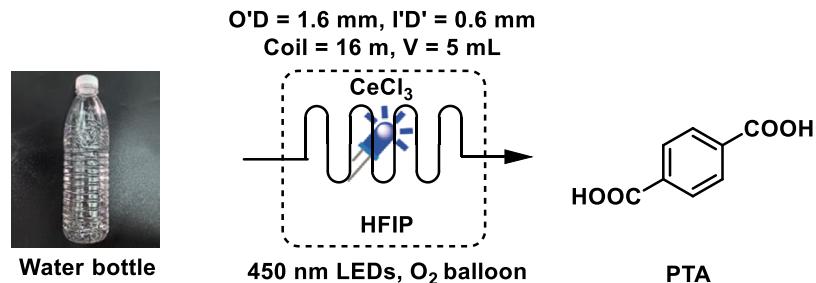
[a] Reaction conditions: PET (0.5 mmol),  $\text{CeCl}_3$  (1 mol%), conc. HCl (50 mol%), HFIP (10.0 mL) at room temperature (25 °C), Blue LED (450 nm, 50 W), isolated yield.

## 6.1 Large-Scale Reaction for PET Depolymerization in continuous-flow reactors

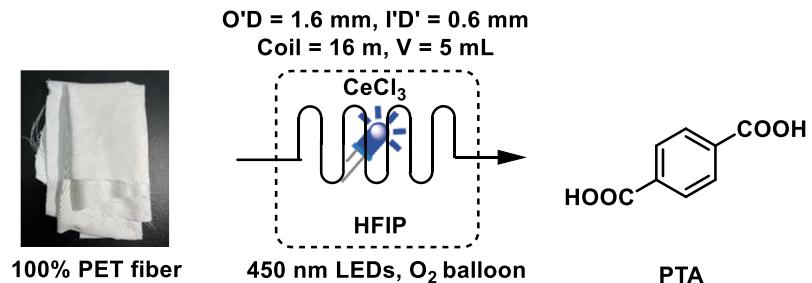


Following Typical Procedure 2, the reaction of colored beverage powder (96 mg, 0.5 mmol),  $\text{CeCl}_3$  (1 mol%, 0.005 mmol, 1.24 mg) and conc. HCl (12 N, 0.125 mmol, 20.0  $\mu\text{L}$ ) in HFIP (10.0 mL). We mixed the reactants and injected them into the microreactor

using a syringe pump at a flow rate of 0.1 mL/min under O<sub>2</sub> atmosphere. The residence time of the reaction solution was 45 min and finally afforded terephthalic acid (44.02 mg, 53 % yield): white solid. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 13.31 (s, 2H), δ 8.04 (s, 4H).



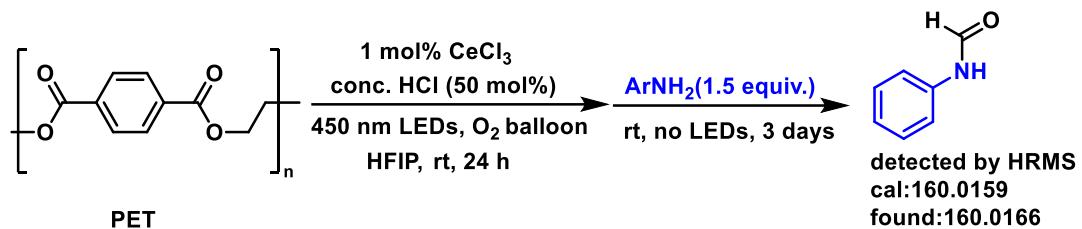
Following Typical Procedure 2, the reaction of water bottle powder (96 mg, 0.5 mmol), CeCl<sub>3</sub> (1 mol%, 0.005 mmol, 1.24 mg) and conc. HCl (12 N, 0.125 mmol, 20.0 μL) in HFIP (10.0 mL). We mixed the reactants and injected them into the microreactor using a syringe pump at a flow rate of 0.1 mL/min under O<sub>2</sub> atmosphere. The residence time of the reaction solution was 45 min and finally afforded terephthalic acid (47.35 mg, 57 % yield): white solid. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 13.31 (s, 2H), δ 8.04 (s, 4H).



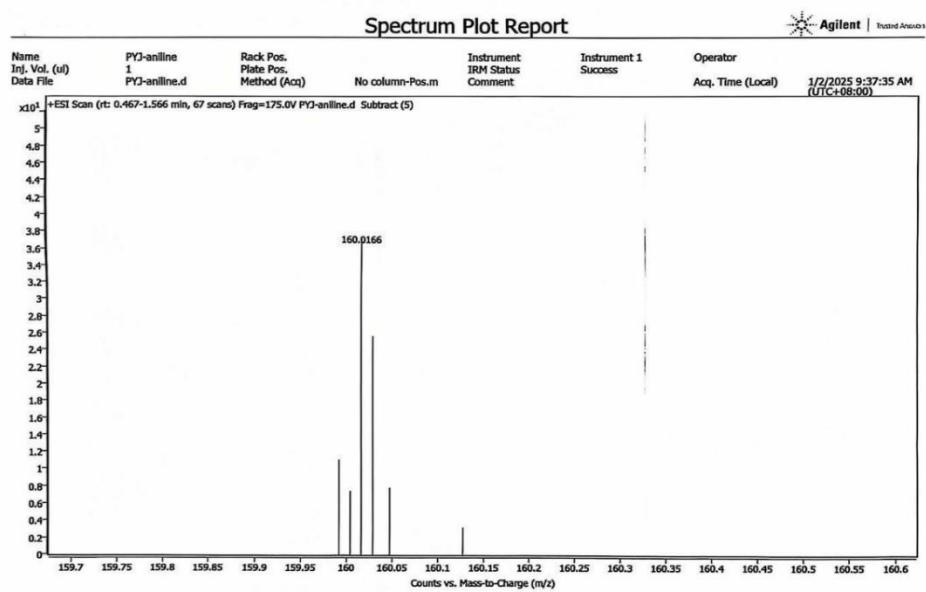
Following Typical Procedure 2, the reaction of 100 % PET fiber (96 mg, 0.5 mmol), CeCl<sub>3</sub> (1 mol%, 0.005 mmol, 1.24 mg) and conc. HCl (12 N, 0.125 mmol, 20.0 μL) in HFIP (10.0 mL). We mixed the reactants and injected them into the microreactor using a syringe pump at a flow rate of 0.1 mL/min under O<sub>2</sub> atmosphere. The residence time of the reaction solution was 45 min and finally afforded terephthalic acid (53.99 mg, 65 % yield): white solid. <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 13.31 (s, 2H), δ 8.04 (s, 4H).

## 7. Mechanistic Studies

### 7.1 Formic Acid Capture

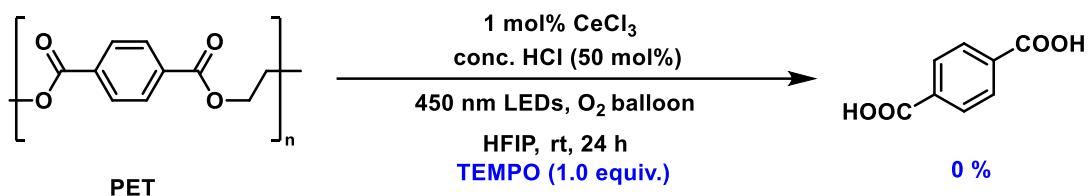


PET powder (Mw = 15 kDa) (0.25 mmol, 48 mg) was added to 2 mL of HFIP and mixed with  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg), and concentrated HCl (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ). The mixture was stirred under an oxygen atmosphere (1 atm, balloon) at room temperature for 24 hours using a parallel reactor illuminated with blue light (450 nm, 50 W). After reaction completing, aniline (34  $\mu\text{L}$ , 34.92 mg, 0.375 mmol) was added, then switched  $\text{O}_2$  to  $\text{N}_2$  atmosphere (1 atm), the resulting mixture was stirred for 72 hours at room temperature. Formanilide was detected by HRMS (ESI) m/z [M + K]<sup>+</sup> Calcd for  $\text{C}_7\text{H}_7\text{NOK}$  160.0159; found 160.0166.



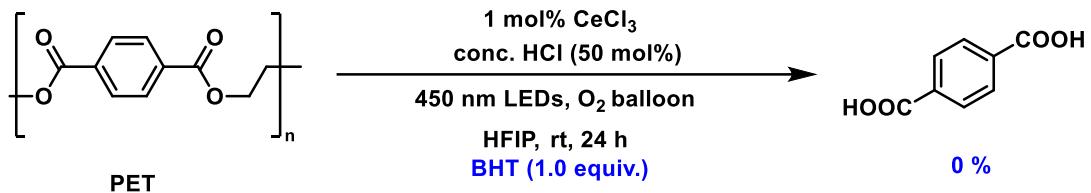
**Figure S4: HRMS detection of formanilide.**

### 7.2 TEMPO trapping reaction



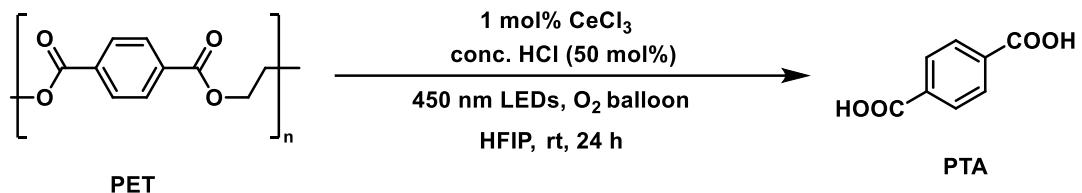
PET powder (Mw = 15 kDa) (0.25 mmol, 48 mg) was added to 2 mL of HFIP and mixed with  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg), and TEMPO (39.06 mg, 0.25 mmol), and concentrated HCl (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ). The mixture was stirred under an oxygen atmosphere (1 atm, balloon) at room temperature for 24 hours using a parallel reactor illuminated with blue light (450 nm, 50 W). The solvent (HFIP) was removed in vacuo and the pH of the residue was adjusted to 10-11 by aq. NaOH (1 N). Subsequently, the mixture was stirred for 1 h at room temperature and the precipitate was separated by filtration. The pH of the filter was adjusted to 3-4 by conc. HCl, no terephthalic acid was afforded.

### 7.3 BHT inhibition reaction



PET powder (Mw = 15 kDa) (0.25 mmol, 48 mg) was added to 2 mL of HFIP and mixed with  $\text{CeCl}_3$  (1 mol%, 0.0025 mmol, 0.62 mg), and BHT (110.18 mg, 0.5 mmol), and concentrated HCl (12 N, 0.125 mmol, 10.0  $\mu\text{L}$ ). The mixture was stirred under an oxygen atmosphere (1 atm, balloon) at room temperature for 24 hours using a parallel reactor illuminated with blue light (450 nm, 50 W). The solvent (HFIP) was removed in vacuo and the pH of the residue was adjusted to 10-11 by aq. NaOH (1 N). Subsequently, the mixture was stirred for 1 h at room temperature and the precipitate was separated by filtration. The pH of the filter was adjusted to 3-4 by conc. HCl, no terephthalic acid was afforded.

### 7.4 Water contact angle assessments



PET powder (Mw = 15 kDa) (0.25 mmol, 48 mg) was added to 2 mL of HFIP and mixed with CeCl<sub>3</sub> (1 mol%, 0.0025 mmol, 0.62 mg), and concentrated HCl (12 N, 0.125 mmol, 10.0  $\mu$ L). The mixture was stirred under an oxygen atmosphere (1 atm, balloon) at room temperature for 24 hours using a parallel reactor illuminated with blue light (450 nm, 50 W). After reaction completing, PET and the resulting products after 24 hours depolymerization undergone the water contact angle assessments. The results revealed a noteworthy reduction in the water contact angle, transitioning from  $\theta_{avg} = 78.6^\circ$  to  $\theta_{avg} = 61.9^\circ$ . This outcome suggests the substantial generation of hydrophilic groups, including hydroxyl and carboxyl groups during the reaction process.

$$\theta_{avg} = (\theta_1 + \theta_2 / 2)$$

Preparation of samples for water contact angle measurements:

Prior to characterization, all substrates underwent standardized pretreatment: sequential ultrasonic cleaning in acetone (HPLC grade), neutral detergent, deionized water, and isopropanol ( $\geq 99.8\%$ ), each for 15 min, followed by nitrogen drying and oxygen plasma activation. For PET powder analysis, raw pellets were cryogenically ground in liquid nitrogen using a shredder and sieved through 30-50 mesh (300-500  $\mu$ m) standard screens. Depolymerization products were isolated by rotary evaporation (Büchi R-300, 45°C, 200 mbar) to remove solvents, then vacuum-dried 45 °C for 12 h under controlled atmospheric conditions.

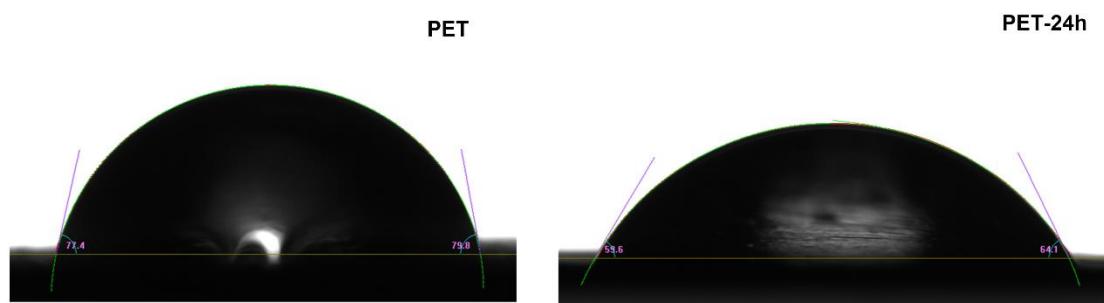
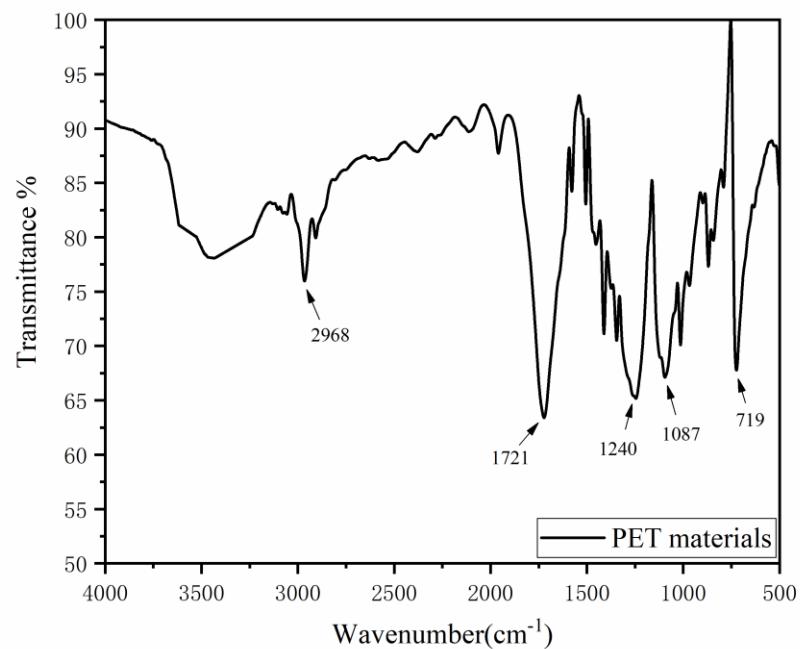
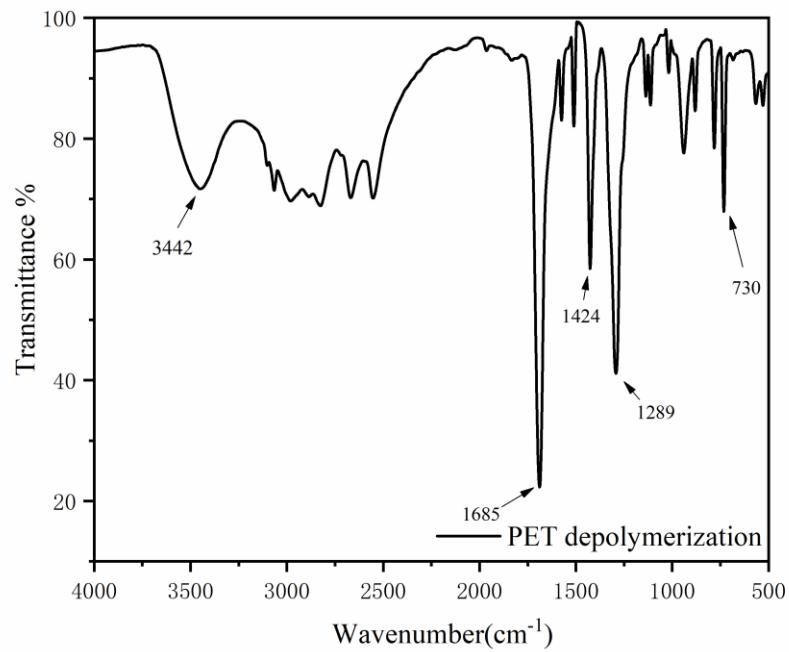


Figure S5: Water contact angles of PET and depolymerization product.

## 7.5 FT-IR spectra



**Figure S6: FT-IR spectra of PET materials (Mw = 15 kDa).**



**Figure S7: FT-IR spectra of PET after depolymerization reaction.**

## 8. Determination of Cerium (Ce) Content in PTA by ICP-MS

Element	$m_0$ (g)	$V_0$ (mL)	$C_0$ (mg/L)	f	$C_1$ (mg/L)	$C_x$ (mg/kg)	W(%)
Ce	0.0338	25	0.562	1	0.56	415.68	0.04
	0.0338	25	0.587	1	0.59	434.17	0.04
	0.0338	25	0.585	1	0.59	432.69	0.04

$$C_1(\text{mg/L}) = C_0(\text{mg/L}) * f$$

$$C_x(\text{mg/kg}) = \frac{C_0(\text{mg/L}) * f * V_0(\text{mL}) * 10^{-3}}{m_0(\text{g}) * 10^{-3}} = \frac{C_1(\text{mg/L}) * V_0(\text{mL}) * 10^{-3}}{m_0(\text{g}) * 10^{-3}}$$

$$W (\%) = \frac{C_x(\text{mg/kg})}{10^6} * 100\%$$

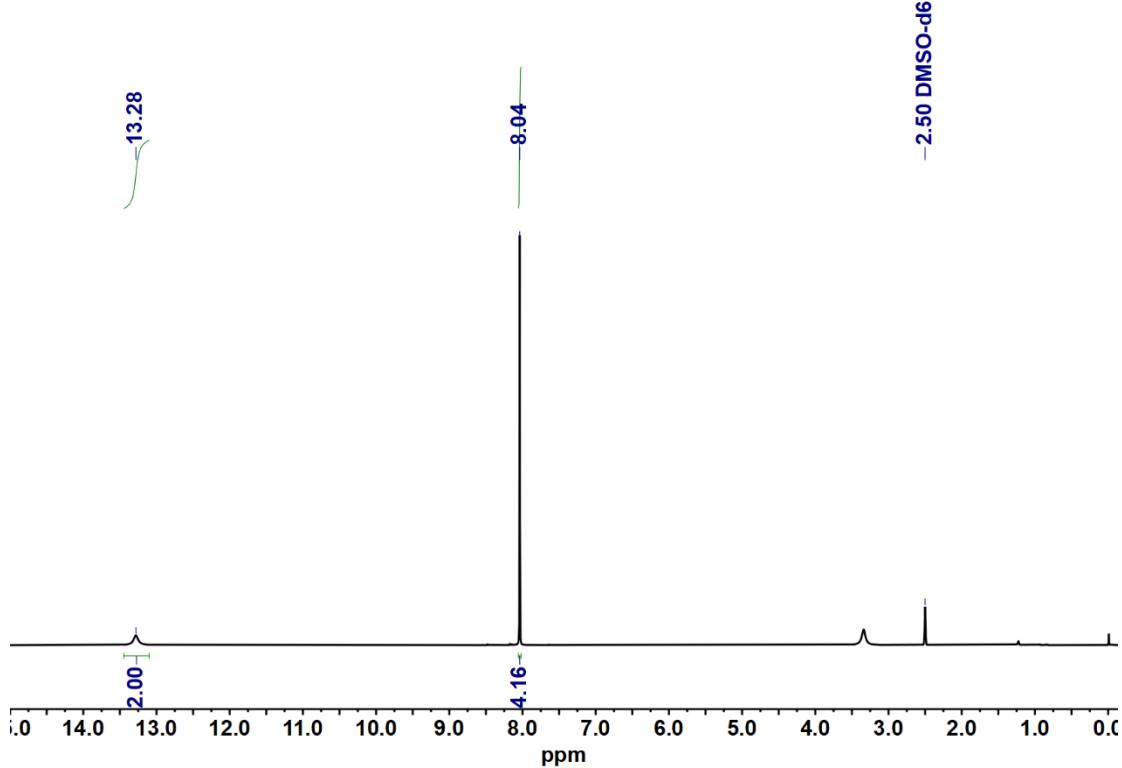
$m_0$ : mass of the test sample (g)

$V_0$ : final volume of the digested sample after dilution (mL)

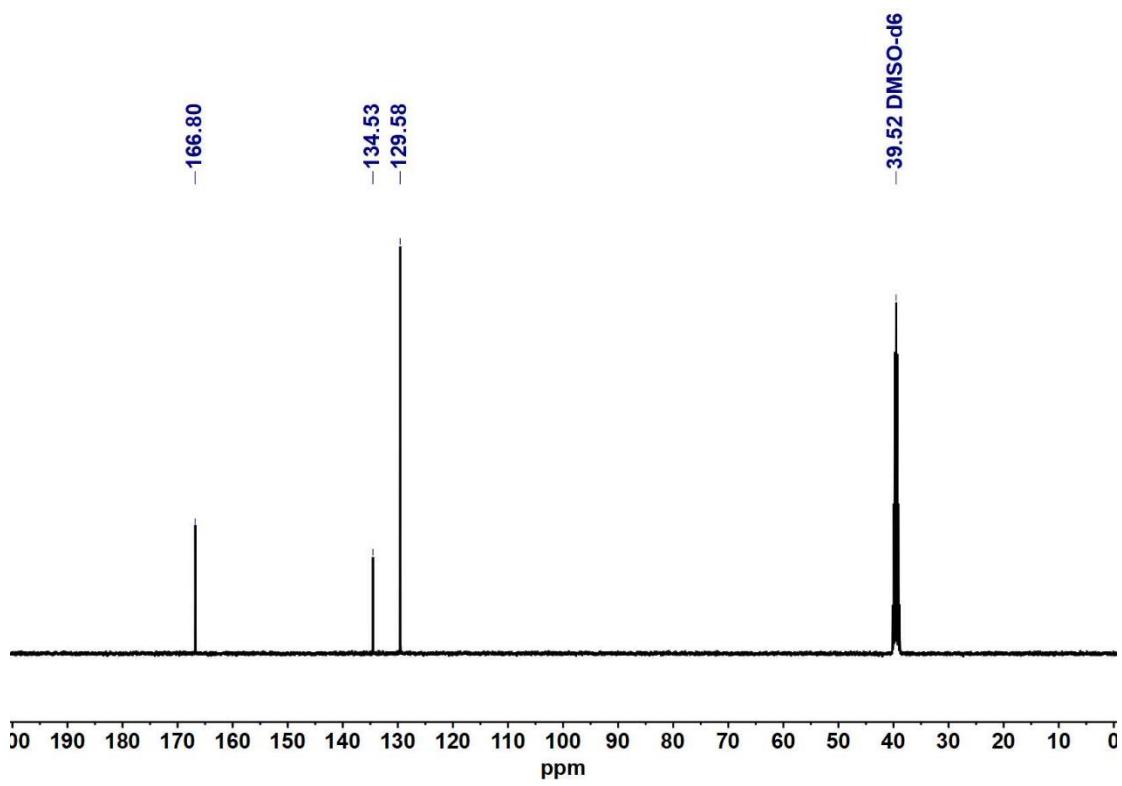
$C_0$ : concentration of target elements in the analytical solution (mg/L)

f: fold dilution

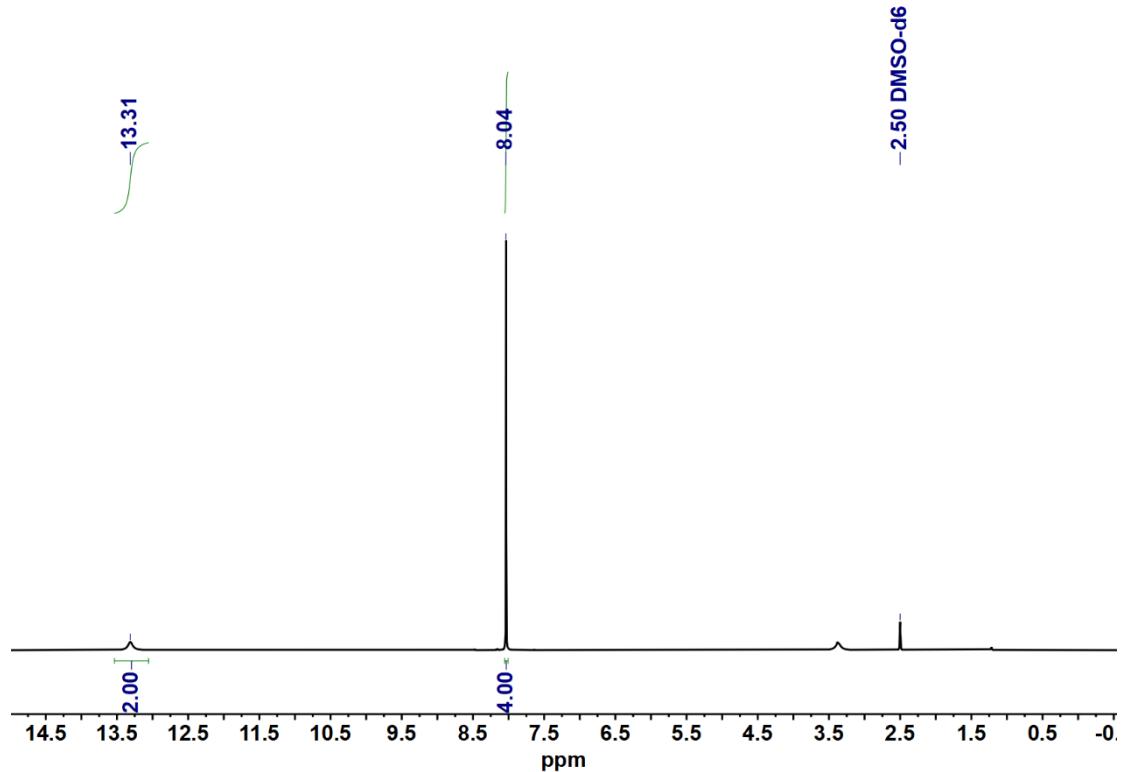
## 9. NMR spectra



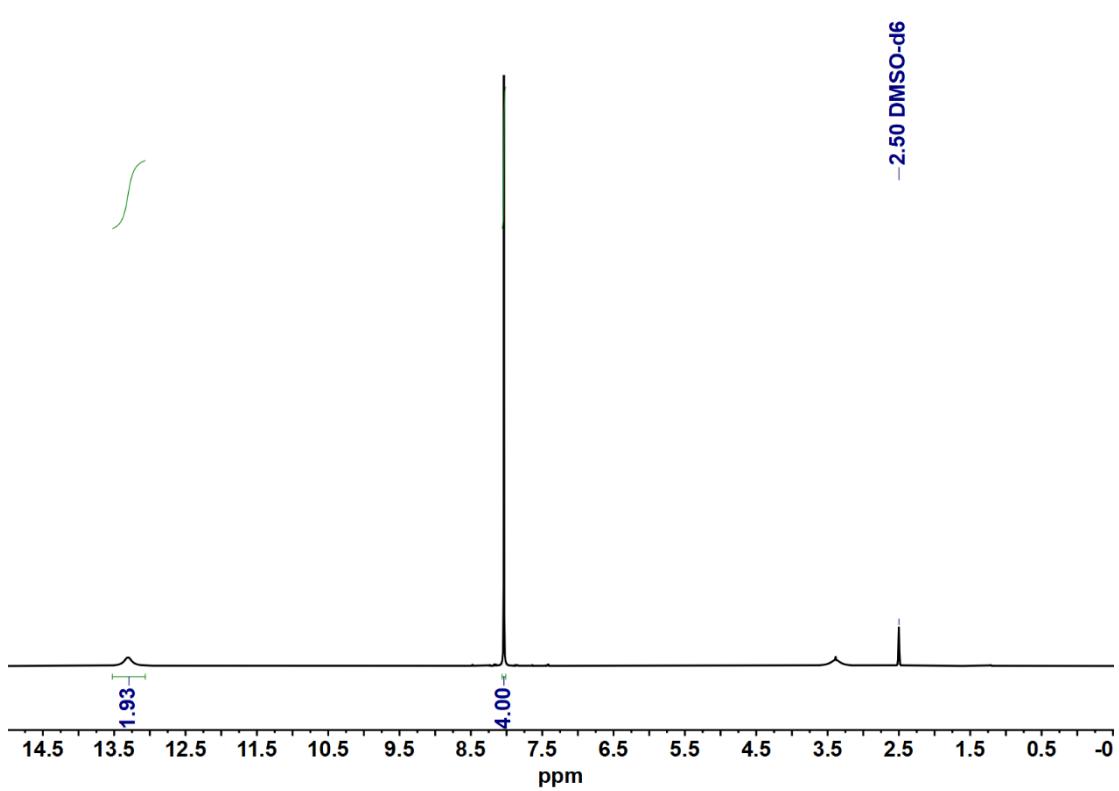
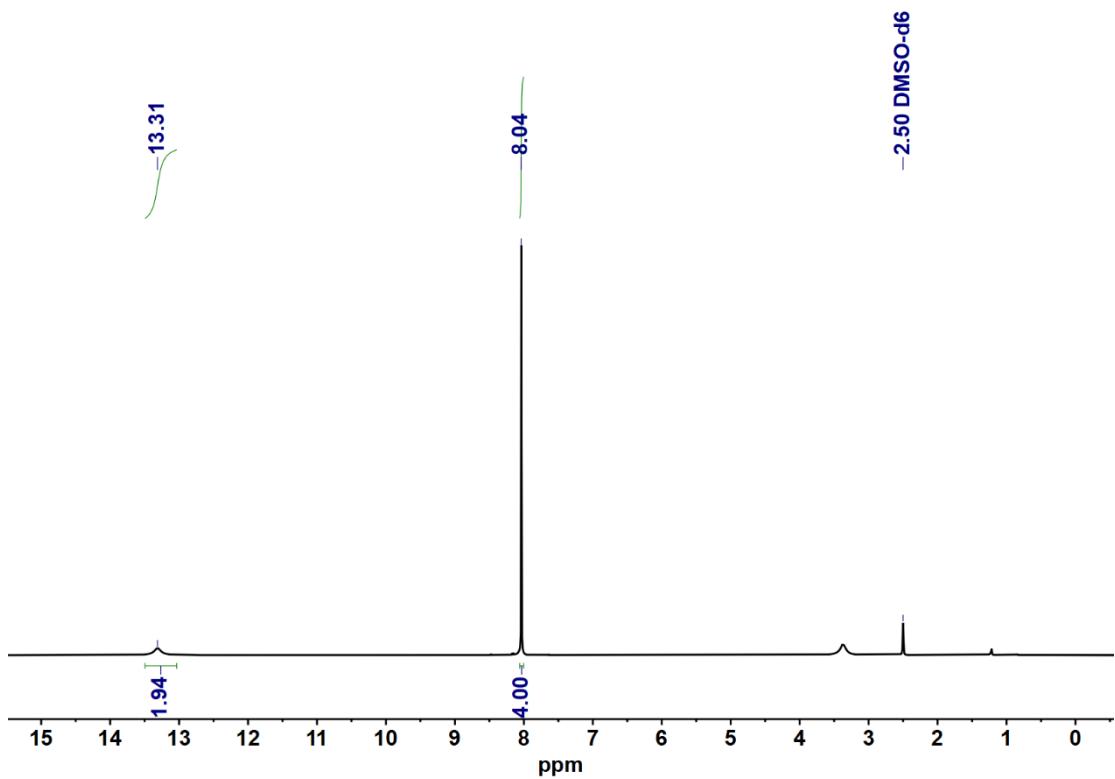
$^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) of compound PTA.

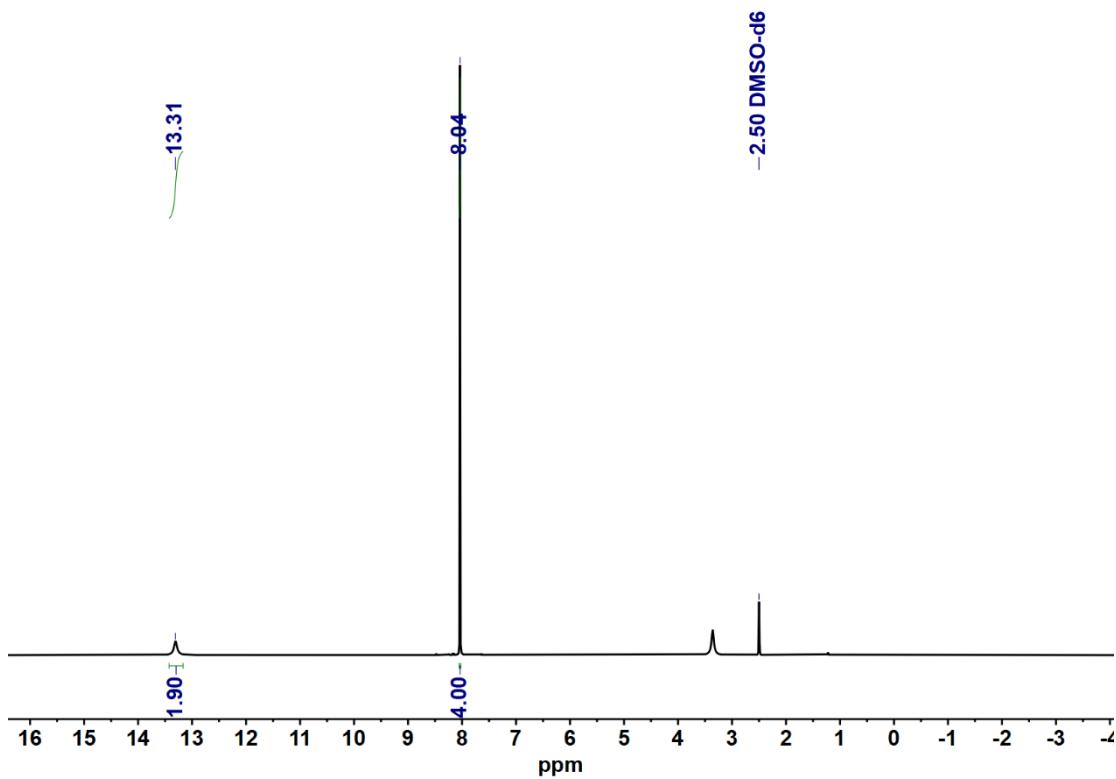


<sup>13</sup>C NMR (101 MHz, DMSO-*d*<sub>6</sub>) of compound PTA.

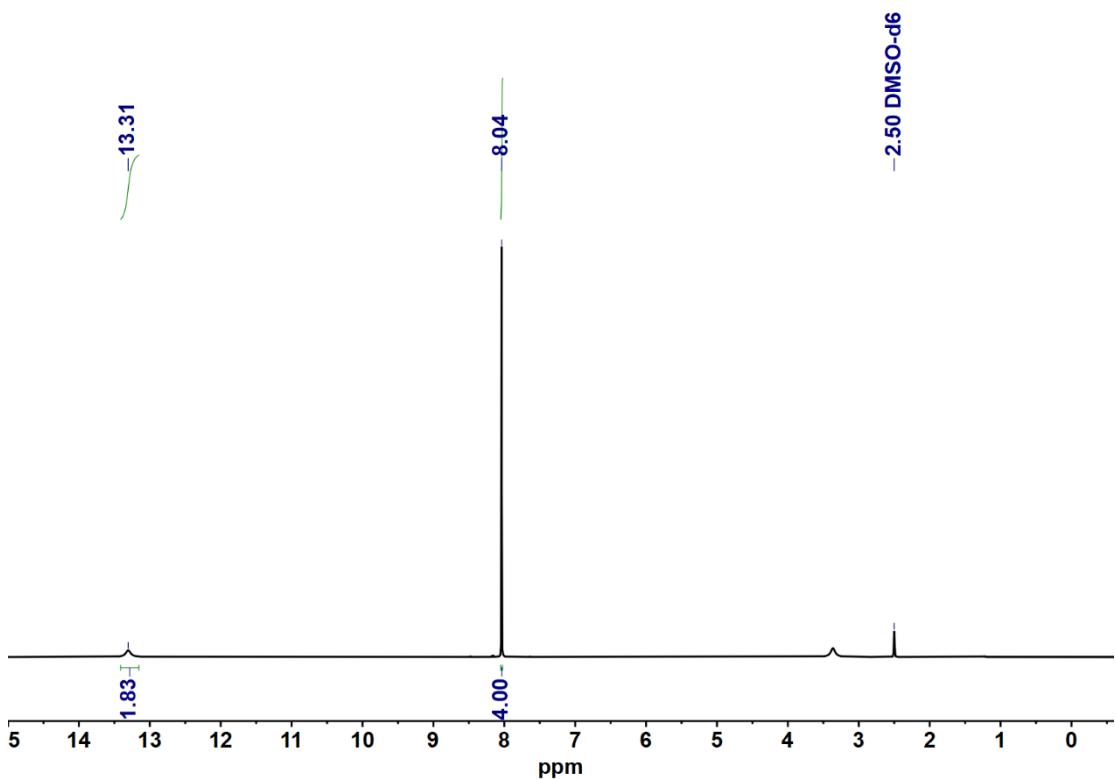


<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) of depolymerization products of colored bottles.

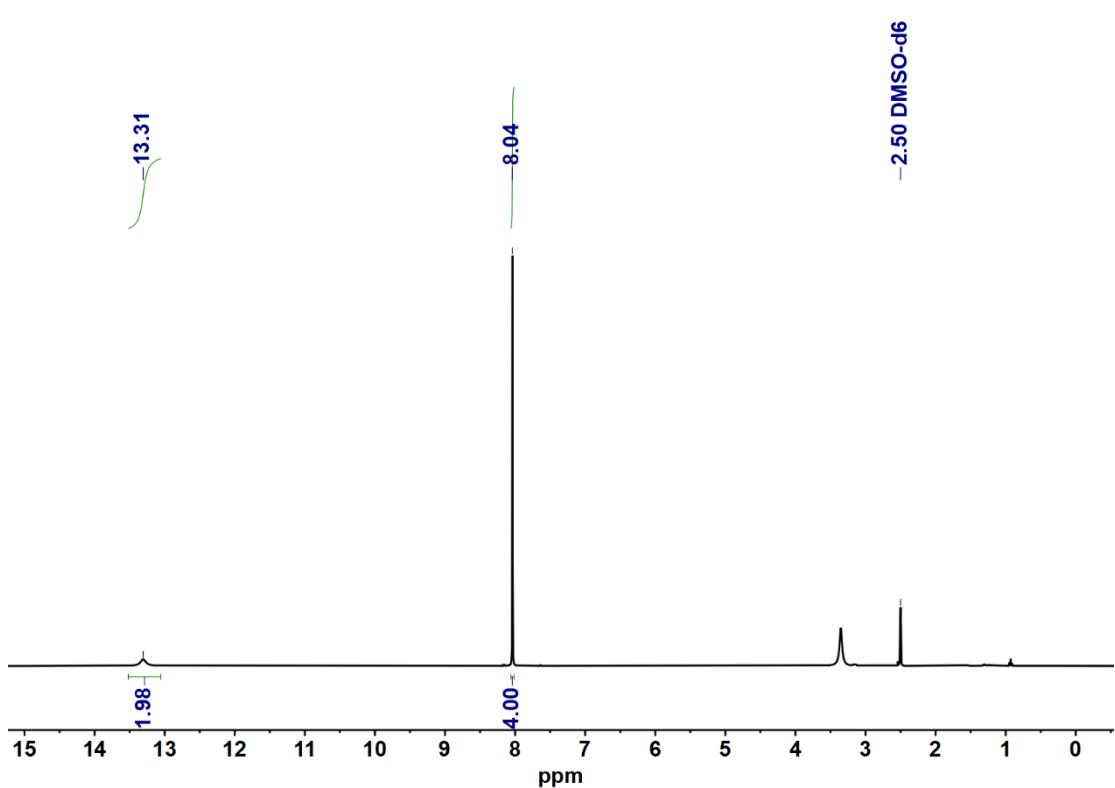
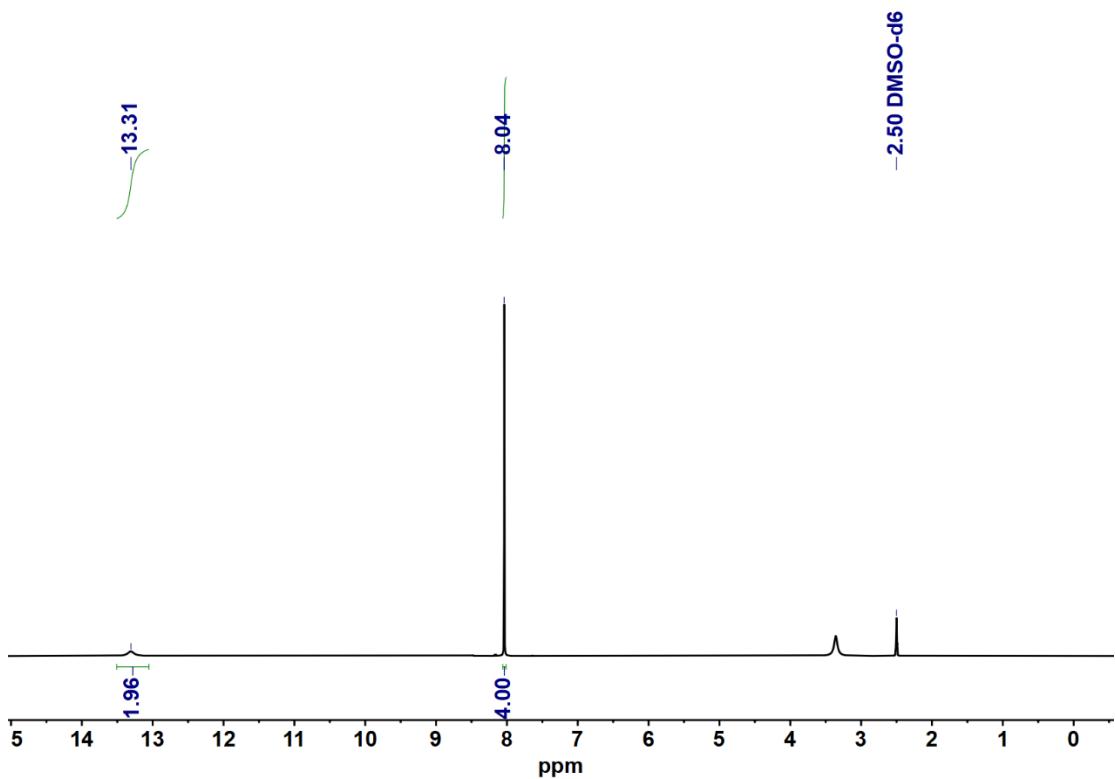


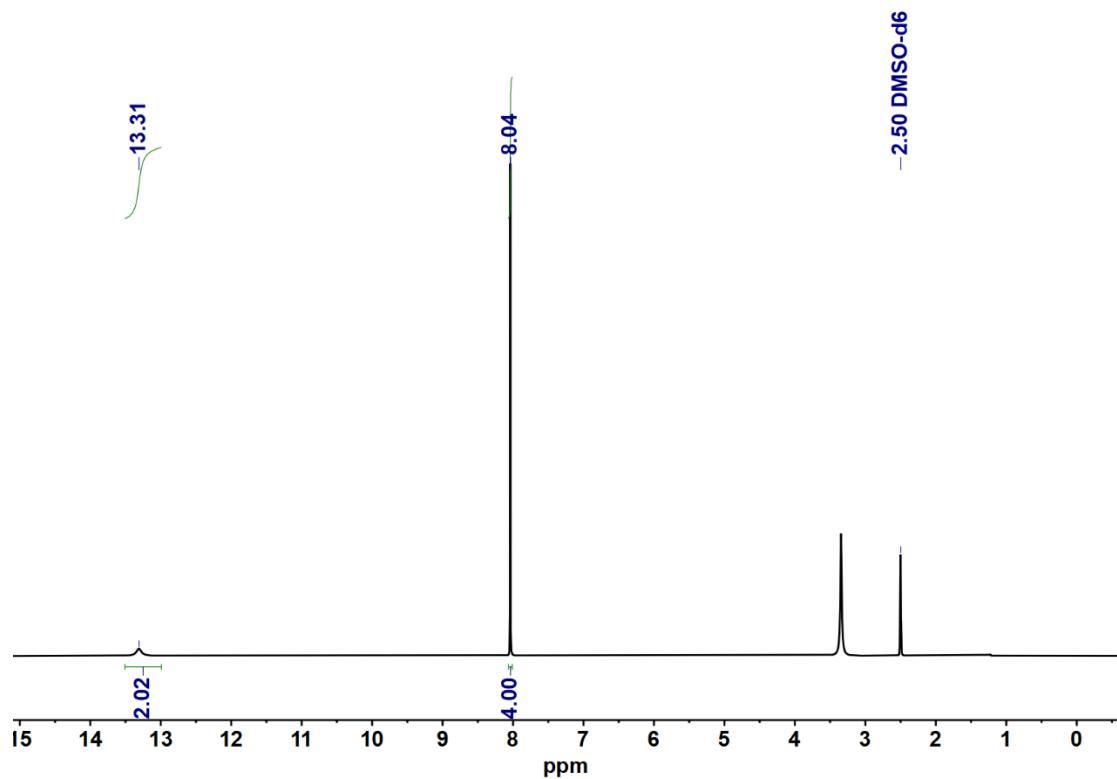


<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) of depolymerization products of water bottle.

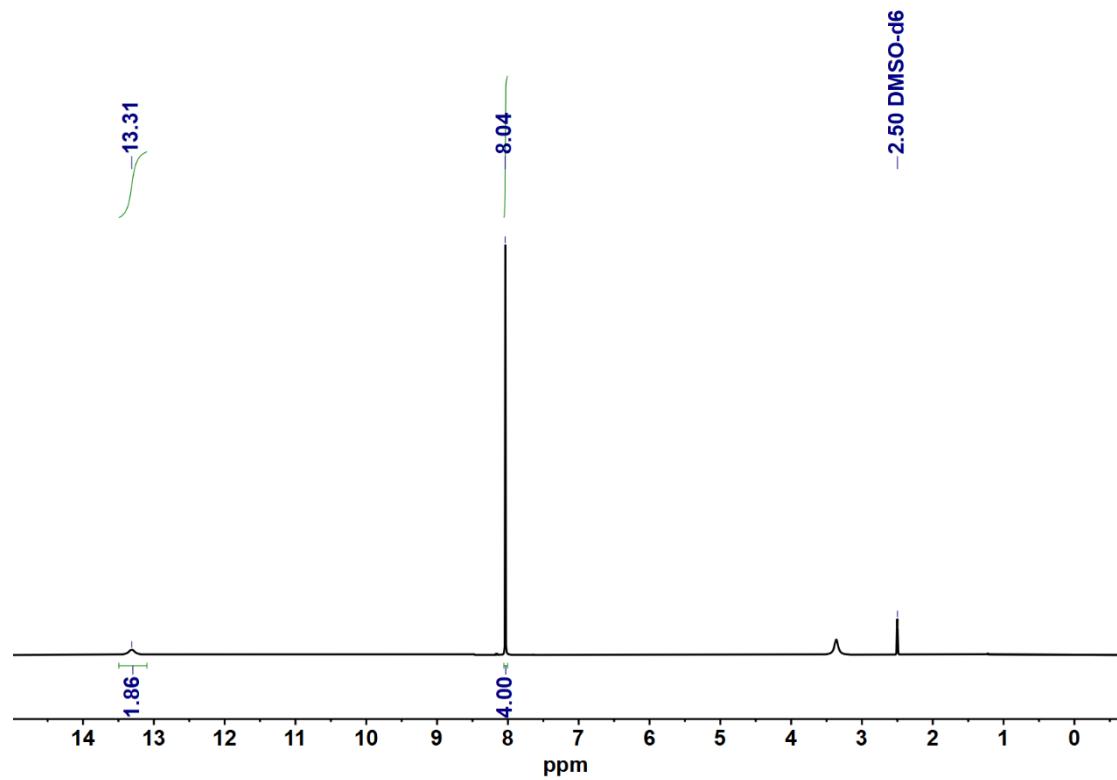


<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) of depolymerization products of milk bottle.

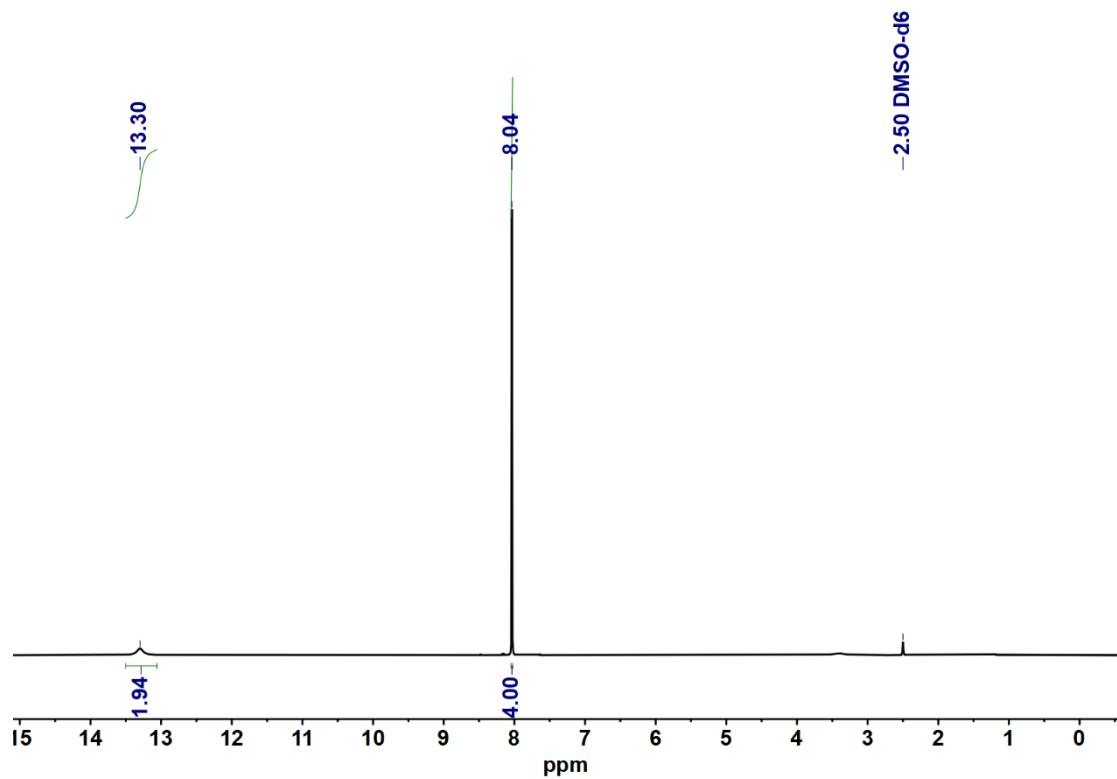




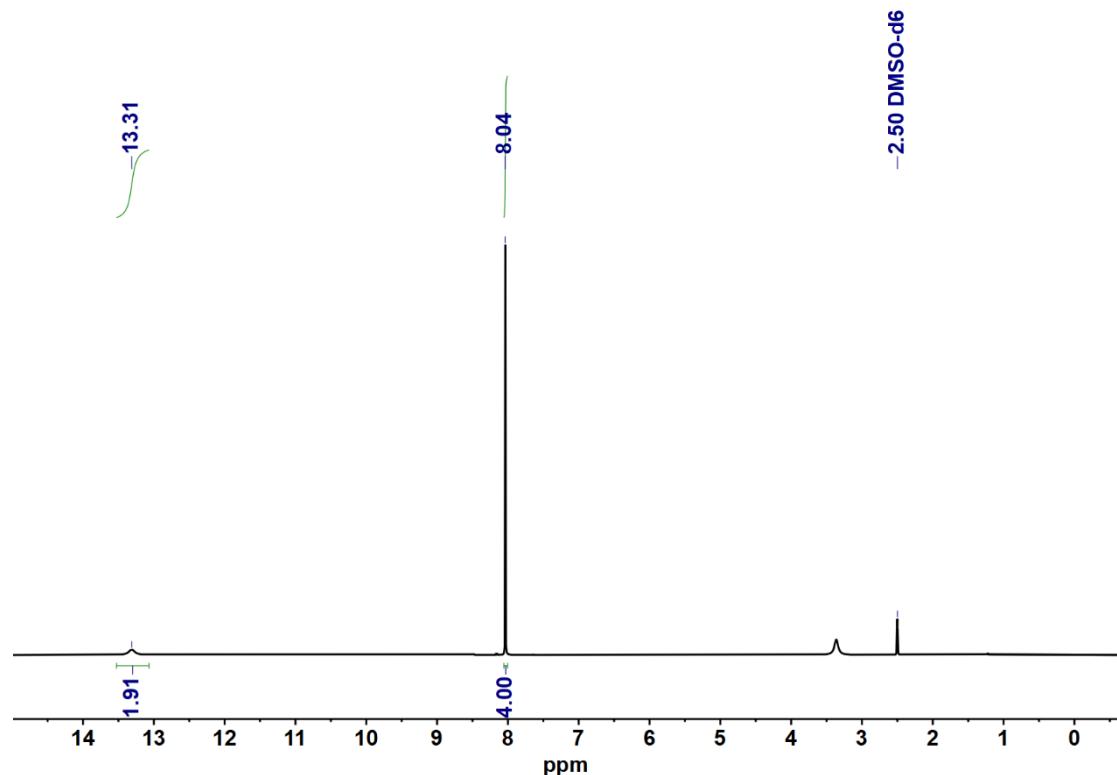
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) of depolymerization products of PET lid.



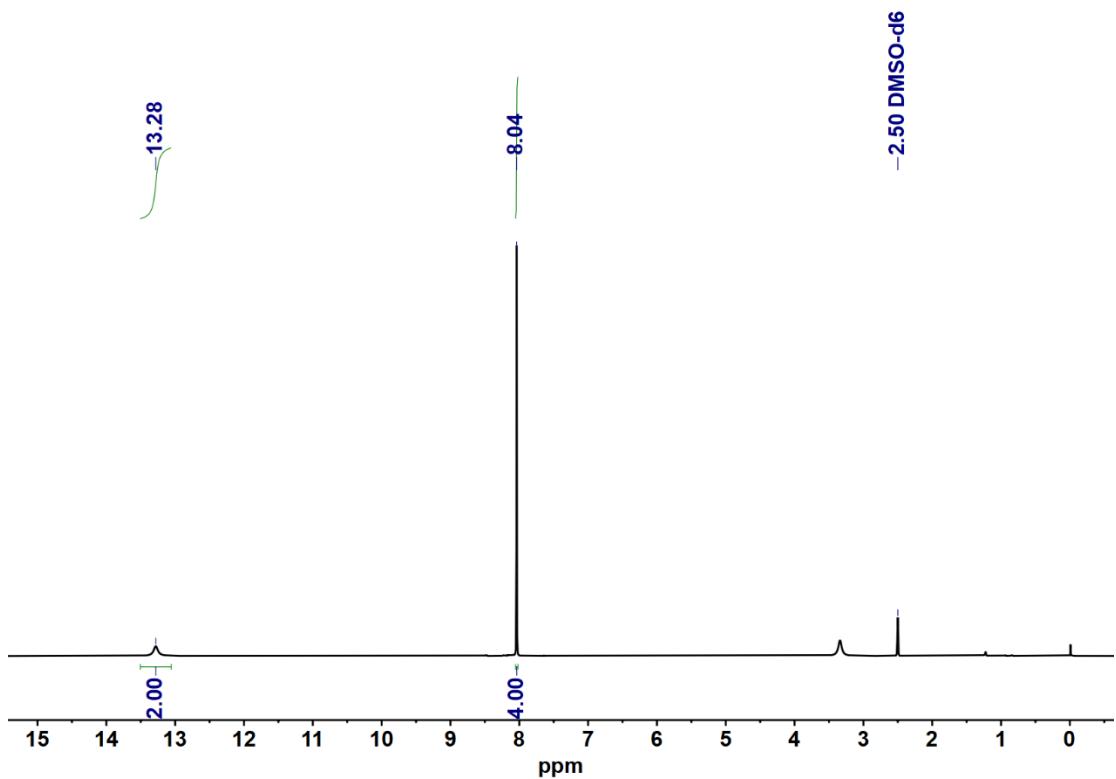
<sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) of depolymerization products of 100% PET fiber.



**<sup>1</sup>H NMR** (400 MHz, DMSO-*d*<sub>6</sub>) of depolymerization products of 65%PET,35%Cotton fiber.



**<sup>1</sup>H NMR** (400 MHz, DMSO-*d*<sub>6</sub>) of depolymerization products of 74.5 %PET, 21.5 %Rayon, 4 %Spandex fiber.



$^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ ) of depolymerization products of 80 %PET, 20 %Cotton fiber.