

## Fiber and Monomer Recovery from an Amine-cured Epoxy Composite using Molten NaOH-KOH

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## 1. Literature review on the current state of the art of alkali and molten salt-based amine-epoxy recycling strategies

**Table S1.** Summary of alkali-based recycling strategies in literature.

| Ref                           | Reagents                                     |                  | Temperature (°C) | Pressurized | Treatment duration (hours) | Monomer recovery   |
|-------------------------------|--|------------------|------------------|-------------|----------------------------|--------------------|
| <b>This work</b>              | <b>NaOH/KOH eutectic salt</b>                |                  | <b>190-240</b>   | <b>N</b>    | <b>0.25-7</b>              | <b>BPA, amines</b> |
| Yan. et al <sup>1</sup>       | 1-propanol                                   | KOH              | 280-340          | Y           | 1-5                        | N/A                |
| Liu. et al <sup>2</sup>       | phenol                                       | KOH              | 300-325          | Y           | 0.5-3                      | N/A                |
| Jiang. et al <sup>3</sup>     | polyethylene glycol                          | KOH              | 160              | N           | 3.3                        | N/A                |
| Zhao. et al <sup>4</sup>      | MEA  | KOH              | 160              | N           | 1.5                        | N/A                |
| DiPucchio. et al <sup>5</sup> | Organic solvents                             | KOtBu/KOH        | 140-160          | Y           | 24-48                      | BPA, amines*       |
| Ahrens. et al <sup>6</sup>    | Organic solvents                             | triphos-Ru-TMM   | 160              | Y           | 24                         | BPA                |
| Sun. et al <sup>7</sup>       | Organic solvents                             | NaOH             | 190              | Y           | 24                         | BPA                |
| Takahashi. et al <sup>8</sup> | Molten NaOH salt                             |                  | 250-350          | N           | 1-1.5                      | N/A                |
| Wu. et al <sup>9</sup>        | KCl/ZnCl <sub>2</sub> eutectic salt          | TiO <sub>2</sub> | 350              | N           | 1.5                        | N/A                |
| Ren. et al <sup>10</sup>      | Molten Na <sub>2</sub> CO <sub>3</sub> /NaOH |                  | 350-400          | N           | 2                          | N/A                |
| Nie. et al <sup>11</sup>      | Molten KOH salt                              |                  | 285-350          | N           | 0.5-2                      | N/A                |

\*: Amine recovery reported from molecular models and thermoplastics

## 2. General Procedure

### a. Fabrication of Bi/DDS amine-epoxy CFRPs

Matrix of Bi/DDS amine-epoxy CFRPs was formulated using diglycidyl ether of bisphenol A (17.95 g, epoxy equivalent weight (EEW) = 187 g/eq; Araldite GY6010, Huntsman) and 3,3-diaminodiphenyl sulfone (6.05 g, amine hydrogen equivalent weight (AHEW) = 63 g/eq, ARADUR 9719-1, Huntsman). The mixing ratio of amine and epoxy was 100%, and the amounts of reagents (g) were calculated using EEW and AHEW. Amine and epoxy were hand-mixed in an aluminum jar for 10 min. Then, the mixture was heated and vacuum degassed in a vacuum oven at 120 °C until the mixture became clean and homogenous.

Resin films were then prepared on a Wabash hot press. Resin mixture (4.5 g) was spread on a 203 x 203 mm release film (Airtech, Release Ease 236 TFNP) preheated to 60 °C on the hot press. Then, two resin films were attached to pre-cut CF fabrics (203 x 203 mm, 2 x 2 twill, FiberGlast 1069). Then, the stack was heated and pressed by the hot press at 60 °C with 0.1 tons force (18.8 Kpa) for 2 mins.

Cured Bi/DDS CFRP panels were then laminated via a vacuum bag-only process (VBO) using 4 layers of prepreg. The curing cycle was (1) 1.5 °C /min to 120 °C, (2) hold at 120 °C for

3 hrs, (3) 1.5 °C/min to 180 °C and (4) hold at 180 °C for 3 hrs. Fully cured Bi/DDS CFRP panels were cut into 50.8 x 10.2 mm samples on a water-jet cutter (ProtoMax, OMAX).

**b. Fabrication of 5320-1 CFRPs**

5320-1 CFRPs were fabricated using commercial aerospace-grade resin film (Solvay CYCOM 5320-1). Resin films were cut into 203 x 203 mm. 2 layers of resin films were attached to pre-cut CF fabrics (203 x 203mm, 2 x 2 twill, FiberGlast 1069). Prepreg was fabricated by heating and pressing the stack on a hot press at 60 °C with 0.1 tons force (18.8 kPa) for 2 mins. After cooling down, release films of prepreg were peeled off, and an additional 2 layers of resin film were attached to the prepreg. Then, the stack was heated and pressed again on a hot press at 60 °C with 0.1 tons force (18.8 kPa) for 2 mins.

5320-1 CFRP panels were then laminated via the VBO process using 4 layers of prepreg. The curing cycle was (1) hold at 60 °C for 2 hrs, (2) 1 °C/min to 120 °C, (2) hold at 120 °C for 2 hrs, (3) 1.7 °C/min to 177 °C, and (4) hold at 177 at 120 °C for 2 hrs. Fully cured 5320-1 CFRP panels were cut into 50.8 x 38.1 mm samples on a water-jet cutter (ProtoMax, OMAX).

**c. Fabrication of Bi/MDEA neat resin samples**

Bi/MDEA neat resin samples were formulated using diglycidyl ether of bisphenol A (3.53 g, epoxy equivalent weight (EEW) = 187 g/eq; Araldite GY6010, Huntsman) and 4,4'-methylenebis (2,6-diethylaniline) (MDEA, 1.47 g, amine hydrogen equivalent weight (AHEW) = 78 g/eq, Sigma-Aldrich). The mixing ratio of amine and epoxy was 100%, and the amounts of reagents (g) were calculated using EEW and AHEW. Amine and epoxy were hand-mixed in an aluminum jar for 10 min. The mixture was then heated, and vacuum degassed in a vacuum oven at 120 °C until the mixture became clean and homogenous.

Resin mixture was then poured into cubic silicone molds (10 x 10 x10 mm), and transferred to a convection oven for curing. The curing cycle was (1) 1.5 °C /min to 120 °C, (2) hold at 120 °C for 3 hrs, (3) 1.5 °C/min to 180 °C and (4) hold at 180 °C for 3 hrs. Fully cured Bi/M-DEA neat resin cubes were ground using an electrical grinder.

**d. Matrix content weight ( $W_m$ ) percent measurement**

Matrix contents of CFRP samples were determined using the method described in ASTM D3171-22, Procedure B. Weights of CFRPs were first weighted to the nearest 0.0001 gram ( $M_i$ ). Then, CFRPs were placed into a 100 mL 2-neck round bottom flask with sulfuric acid (50 mL, 95-98%). The flask was heated to 100 °C in an oil bath with magnetic stirring and a reflux condenser connected. Hydrogen peroxide (10 mL, 30%) was added to the flask each time at 1, 2, and 3 hrs of reactions. After 4 hrs, the flask was removed from the oil bath to cool to room temperature. CFs were filtered out and washed with DI water and acetone. Cleaned CFs were dried in a convection oven at 125 °C overnight and weighed again to the nearest 0.0001 gram ( $M_f$ ).  $W_m$  is calculated using the following equation. Six measurements were taken on Bi/DDS CFRP, as shown in the Table S2.

$$W_m = \frac{(M_i - M_f)}{M_i} \times 100\%$$

**Table S2.** Summary of matrix content weight measurement of Bi/DDS CFRP.

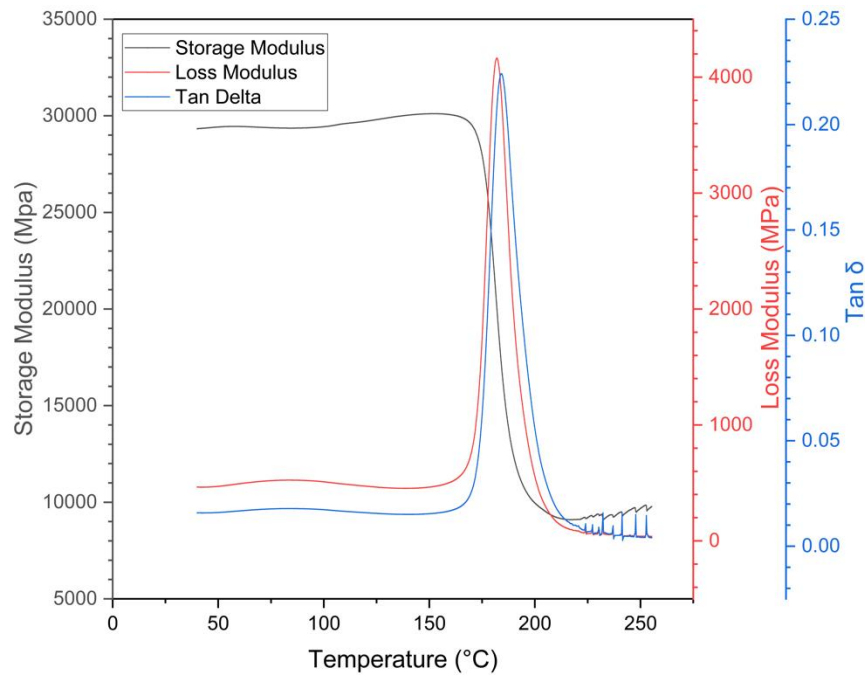
| Entry | M <sub>i</sub> (g) | M <sub>f</sub> (g) | W <sub>m</sub> (%) |
|-------|--------------------|--------------------|--------------------|
| 1     | 0.5825             | 0.2906             | 50.11              |
| 2     | 0.5591             | 0.2953             | 47.18              |
| 3     | 0.5746             | 0.2924             | 49.11              |
| 4     | 0.6379             | 0.3173             | 50.26              |
| 5     | 0.5567             | 0.3015             | 45.85              |
| 6     | 0.5541             | 0.3015             | 45.59              |
| Avg   |                    |                    | 48.02              |

**e.  $T_g$  measurement of Bi/DDS and 5320-1 CFRPs**

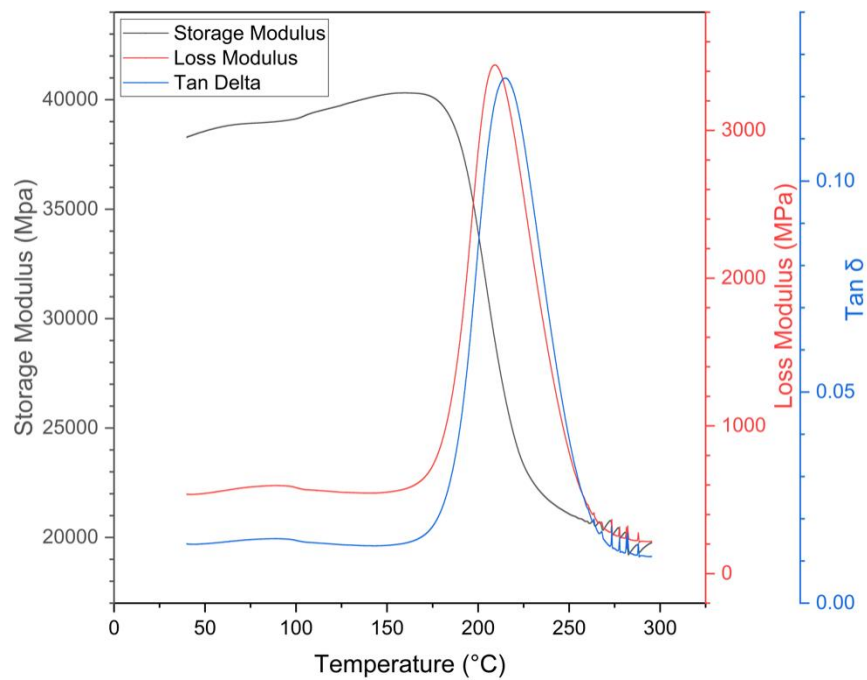
Glass transition temperatures ( $T_g$ ) were measured on TA instrument Q800 DMA using a dual cantilever fixture following ASTM D 7028-07 standard. DMA samples (60 x 10 mm) were cut from fully cured Bi/DDS and 5320-1 CFRP panels on a water jet cutter (ProtoMax, OMAX). Heating rate, frequency, and strain amplitude were set as 5 °C /min, 1 Hz, and 0.05%, respectively. Measurement started at 40 °C and ended at 250 °C (Bi/DDS CFRP) or 300 °C (5320-1 CFRPs), as shown in Figures S1-S2.  $T_g$  (onset of storage modulus),  $T_{loss}$ , and  $T_{tan\delta}$  (temperatures at loss modulus and  $\tan\delta$  peaks) are summarized in Table S3.

**Table S3.** DMA  $T_g$  measurements of Bi/DDS and 5320-1 CFRPs.

| CFRPs  | $T_g$ (°C) | $T_{loss}$ (°C) | $T_{tan\delta}$ (°C) |
|--------|------------|-----------------|----------------------|
| Bi/DDS | 174.84     | 182.00          | 184.01               |
| 5320-1 | 188.49     | 209.24          | 214.74               |



**Figure S1.** DMA  $T_g$  measurements of Bi/DDS CFRP



**Figure S2.** DMA  $T_g$  measurements of 5320-1 CFRP.

## **f. Chemical synthesis and analysis**

All synthetic procedures were conducted in a chemical fume hood with exposure to air, unless otherwise indicated. Deuterated NMR solvents were purchased from Cambridge Isotopes Laboratories. Unless otherwise specified, sodium and potassium hydroxide salts and the other reagents are commonly available from major commercial suppliers (Macron Fine Chemicals and VWR chemicals). Other chemicals included benzyl alcohol (TCI America), dichloromethane (EMD Millipore), ethyl acetate (EMD Millipore), bisphenol A diglycidyl ether (Huntsman), 3,3'-diaminodiphenyl sulfone (Huntsman), sodium hydride (Acros Organics), 1-butanol (BeanTown Chemical), sulfuric acid (Supelco), hydrochloric acid (Sigma-Aldrich), hydrogen peroxide (EMD Millipore), iodomethane (Sigma-Aldrich), and glycidyl phenyl ether (Thermo Fisher Scientific Chemicals). These chemicals were used without further purification. Deionized water was purified in-house using a deionizer cartridge (Philadelphia Scientific).

NMR spectra were recorded on a Varian Mercury 400, Varian VNMRS 500, or VNMRS 600, spectrometers and processed using MestReNova. All chemical shifts are reported in units of ppm and referenced to the residual  $^1\text{H}$  or  $^{13}\text{C}$  solvent peak and line-listed according to (s) singlet, (bs) broad singlet, (d) doublet, (t) triplet, (dd) double doublet, etc.  $^{13}\text{C}$  spectra are delimited by carbon peaks, not carbon count. Mass spectral data were acquired on an Agilent 6545 LC-QTOF instrument with electrospray set to positive ionization and a Bruker AutoFlex MALDI. IR data were acquired on a Thermo Fisher Nicolet 4700 FT-IR spectrometer.

Silica sulfuric acid catalysts were made in our labs. These catalysts were prepared by soaking 20 grams of silica-gel with 25 mL of concentrated sulfuric acid in a 100 mL beaker. Afterwards, the beaker was placed in a heated oil bath (130 °C) for one hour and was carefully monitored. After one hour had elapsed, the contents were transferred to a 250 mL round bottom flask and placed in a heated rotary evaporator (95 °C) for 30 minutes under reduced pressure. Finally, the contents of the flask were transferred to a glass jar and stored in a desiccator.



### 3. Molten hydroxide digestion of CFRPs

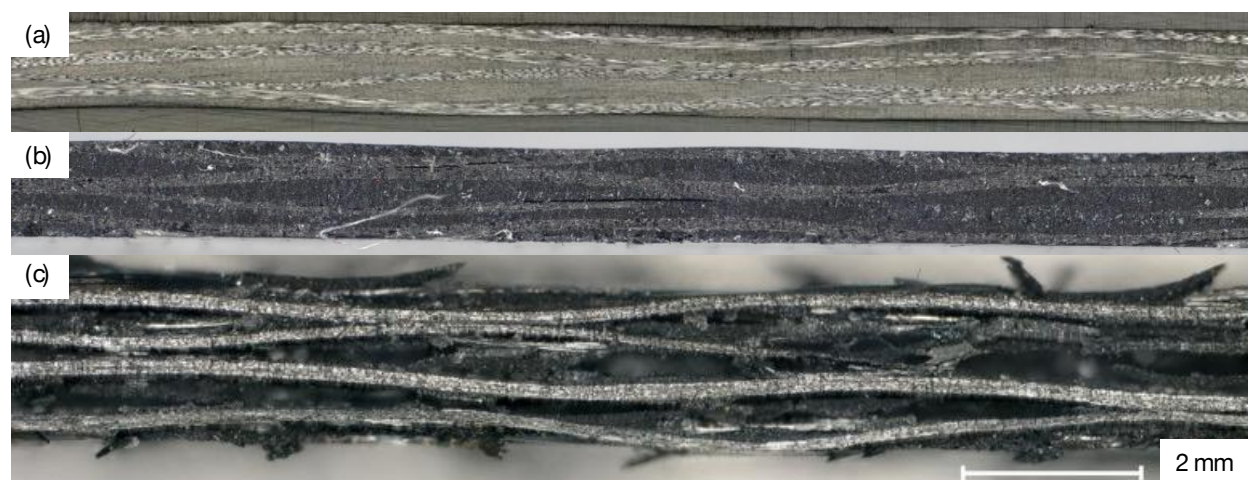
#### a. Pre-treatment of Bi/DDS CFRPs

A pre-treatment step was applied to Bi/DDS CFRP before molten hydroxide digestion. This process involved the use of organic solvents to swell the CFRP matrix to reduce the rate-limiting factor of diffusion. To effectively swell the matrix, pre-treatment temperatures need to be higher than  $T_g$  of CFRP, which requires pre-treatment agents with a boiling point  $> 190^\circ\text{C}$ . Solvents with appropriate boiling points are summarized in Table S4, alongside their safety, health, and environmental assessments from the CHEM 21 solvent selection guide. *N*-Methyl-2-pyrrolidone (NMP) was excluded due to its significant health risks. Ethylene glycol and benzyl alcohol were selected and tested for pre-treating Bi/DDS CFRPs.

**Table S4.** Selection of solvents with desirable boiling point from CHEM 21 solvent guide.

| Solvent         | BP ( $^\circ\text{C}$ ) | Safety Score | Health score | Env. score | Ranking     |
|-----------------|-------------------------|--------------|--------------|------------|-------------|
| Benzyl alcohol  | 206                     | 1            | 2            | 7          | Problematic |
| Ethylene glycol | 198                     | 1            | 2            | 5          | Recommended |
| NMP             | 202                     | 1            | 9            | 7          | Hazardous   |

A Bi/DDS CFRP sample (50.8 x 10.2 mm) was first weighted to the nearest 0.0001 gram ( $M_i$ ) and then placed in a 50 mL single-neck round bottom flask with benzyl alcohol or ethylene glycol (25 mL). The flask was then heated to  $185^\circ\text{C}$  with a reflux condenser connected. After 5 hours, the pre-treated Bi/DDS CFRP was retrieved and then prepared for hydroxide eutectic digestion. Light microscope images of pre-treated Bi/DDS CFRP samples in ethylene glycol and benzyl alcohol were shown in Figure S3 (b) and (c) respectively. Benzyl alcohol was selected as pre-treatment solvent as it clearly outperformed ethylene glycol, inducing more significant swelling of the CFRP matrix, as shown in the Figure S3.

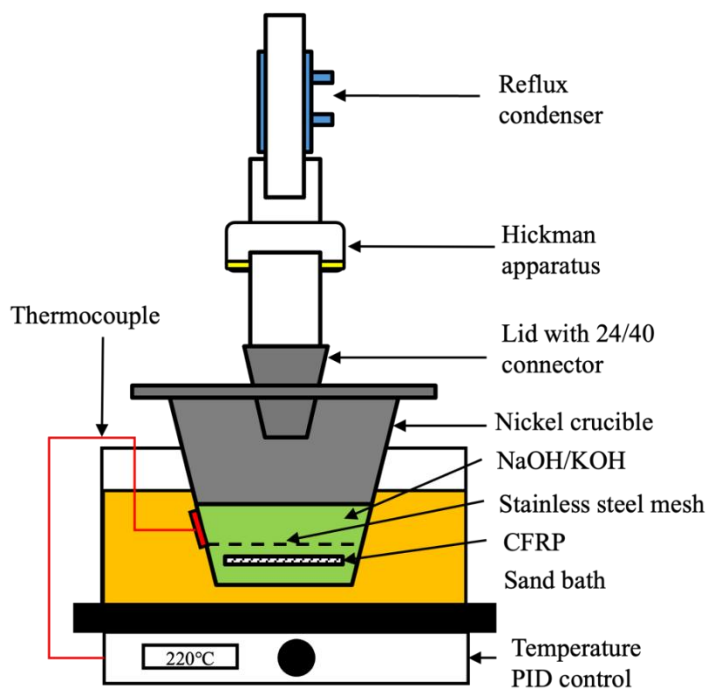


**Figure S3.** Cross-sectional images of Bi/DDS CFRPs before (a), after ethylene glycol (b) and benzyl alcohol (c) pre-treatment

### b. Molten hydroxide digestion of Bi/DDS CFRP

NaOH and KOH pellets were ground to a fine powder using a pill grinder. A mixture of NaOH and KOH (40 g, 1:1 molar) was placed in a 100 mL nickel crucible. A thermocouple was attached to the crucible using high-temperature tapes (flash breaker, Airtech). The crucible and hydroxide mixtures were then heated to pre-set temperatures in a sand bath. A pre-treated Bi/DDS CFRP was placed into the crucible. A stainless-steel mesh (304 stainless steel, 5 mesh, 46 mm diameter) was placed on top of the CFRP to ensure the sample was fully submerged in hydroxide mixtures. A customized stainless-steel lid with a 24/40 female joint was placed on top to connect a Hickman distillation apparatus to collect volatiles, as shown in Figure S4.

After a reaction period, the recycled CFs were taken out from molten hydroxide mixture and cooled to room temperature. The recycled CFs were washed with DI water and acetone three times and dried in a convection oven at 125 °C overnight. Dried CFs were weighted to the nearest 0.0001 gram. Hydroxide melts were subjected to extraction following the procedure described in Section S5. Table S5 summarizes the digestion experiments performed on Bi/DDS CFRPs.



**Figure S4.** Schematic diagram of Bi/DDS CFRP decomposition setup

**Table S5.** Summary of Bi/DDS CFRP digestion experiments.

| Entry | Substrate                | $M_{\text{eutectic}}$ (g) | Temperature ( $^{\circ}\text{C}$ ) | Duration (hr) |
|-------|--------------------------|---------------------------|------------------------------------|---------------|
| 1     | Bi/DDS CFRPs             | 40                        | 190                                | 3             |
| 2     |                          |                           | 200                                |               |
| 3     |                          |                           | 220                                |               |
| 4     |                          |                           | 240                                |               |
| 5     | Pre-treated Bi/DDS CFRPs | 40                        | 190                                | 3             |
| 6     |                          |                           | 200                                |               |
| 7     |                          |                           | 220                                |               |
| 8     |                          |                           | 240                                |               |
| 9     | Pre-treated Bi/DDS CFRPs | 40                        | 240                                | 0.25          |
| 10    |                          |                           | 220                                | 0.25          |
| 11    |                          |                           | 220                                | 0.5           |
| 12    |                          |                           | 220                                | 0.5           |
| 13    | Pre-treated Bi/DDS CFRPs | 40                        | 200                                | 1             |
| 14    |                          |                           | 200                                | 4             |
| 15    |                          |                           | 200                                | 5             |
| 16    |                          |                           | 200                                | 1             |
| 17    | Pre-treated Bi/DDS CFRPs | 40                        | 190                                | 4             |
| 18    |                          |                           | 190                                | 5             |
| 19    |                          |                           | 190                                | 6             |
| 20    |                          |                           | 190                                | 7             |

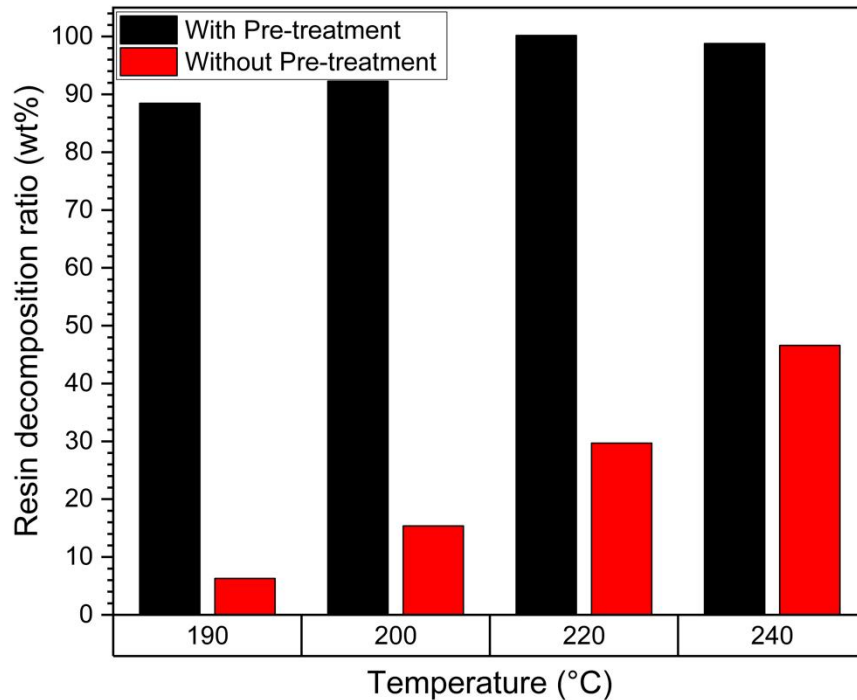
**c. Effect of pre-treatment on digestion efficiency**

The purpose of pre-treatment is to use organic solvent to swell the cross-linked network of the matrix, inducing delamination, as shown in Figure S3, thus reducing the rate-limiting effect of diffusion. Entry 1-4 and 5-8 in Table S6 study the effect of the pre-treatment on resin decomposition ratio at different temperatures after 3hrs of digestion. Resin decomposition ratios ( $R_d$ ) are calculated using matrix content weight ( $W_m$ ), Bi/DDS CFRP weight ( $M_i$ ), and recovered CF weight ( $M_d$ ) as summarized in Table S6 and Figure S5:

$$R_d = \frac{M_i - M_d}{M_i \times W_m} \times 100\%$$

**Table S6.** Summary of the effect of pre-treatment on digestion efficiency.

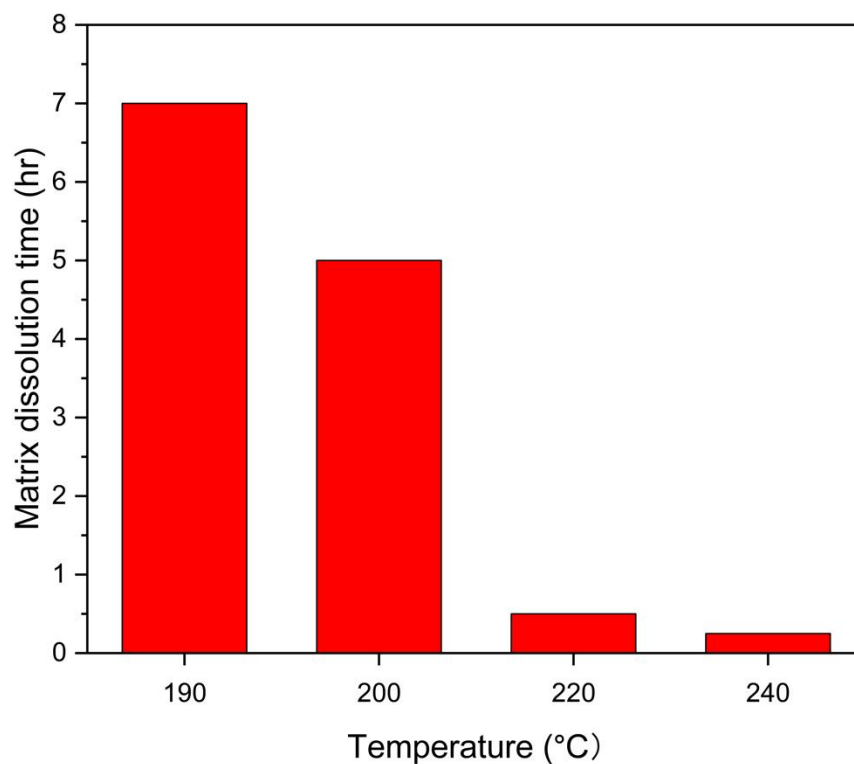
| Entry | $M_i$ (g) | $M_d$ (g) | $R_d$ (%) |
|-------|-----------|-----------|-----------|
| 1     | 0.5798    | 0.5623    | 6.3       |
| 2     | 0.5849    | 0.5459    | 13.9      |
| 3     | 0.5672    | 0.4863    | 29.7      |
| 4     | 0.5616    | 0.4359    | 46.6      |
| 5     | 0.594     | 0.3417    | 88.5      |
| 6     | 0.5797    | 0.3228    | 92.3      |
| 7     | 0.5882    | 0.3062    | 99.8      |
| 8     | 0.5869    | 0.3075    | 99.1      |



**Figure S5.** Effect of pre-treatment on matrix dissolution rate at different temperatures.

**d. Digestion duration at different temperatures**

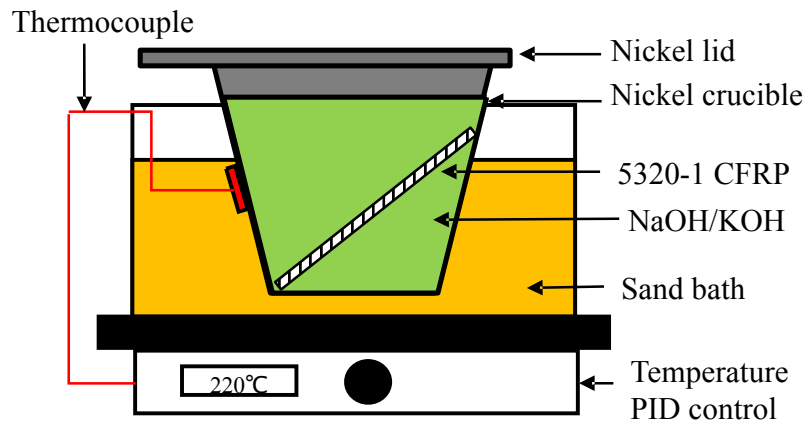
Matrix dissolution time of Bi/DDS CFRPs were determined by performing reaction with incremental treatment duration as shown in Table S3, Entry 5-19. After each digestion, rCFs were cleaned, dried, and visually inspected for resin residues. The 240 and 220 °C reactions were monitored incrementally every 15 minutes. The 200 and 190 °C reactions were monitored incrementally every 60 minutes. The matrix dissolution times at selected temperatures are summarized in Figure S6.



**Figure S6.** Matrix dissolution time of Pre-treated Bi/DDS CFRP at different temperatures.

#### e. Digestion of 5320-1 CFRPs

A mixture of NaOH and KOH (140 g, 1:1 molar) powders were placed in a 100 mL nickel crucible. The crucible, with a nickel lid on top and a thermocouple attached, was then heated to 220 °C in a sand bath. A 5320-1 CFRP sample (50.8 x 38.1 mm) was placed into the crucible for digestion, as shown in Figure S7. After 20 hrs, recycled CFs were retrieved from the crucible and cooled to room temperature. Then, recycled CFs were washed with DI water, acetone, and dimethyl sulfoxide (DMSO) three times. Cleaned CFs were dried in a convection oven at 125 °C overnight. Dried CFs were used with fresh resin films to manufacture 2<sup>nd</sup>-Generation 5320-1 CFRPs, as described in Section S3 part (a).



**Figure S7.** Schematic diagram of 5320-1 CFRP decomposition setup.

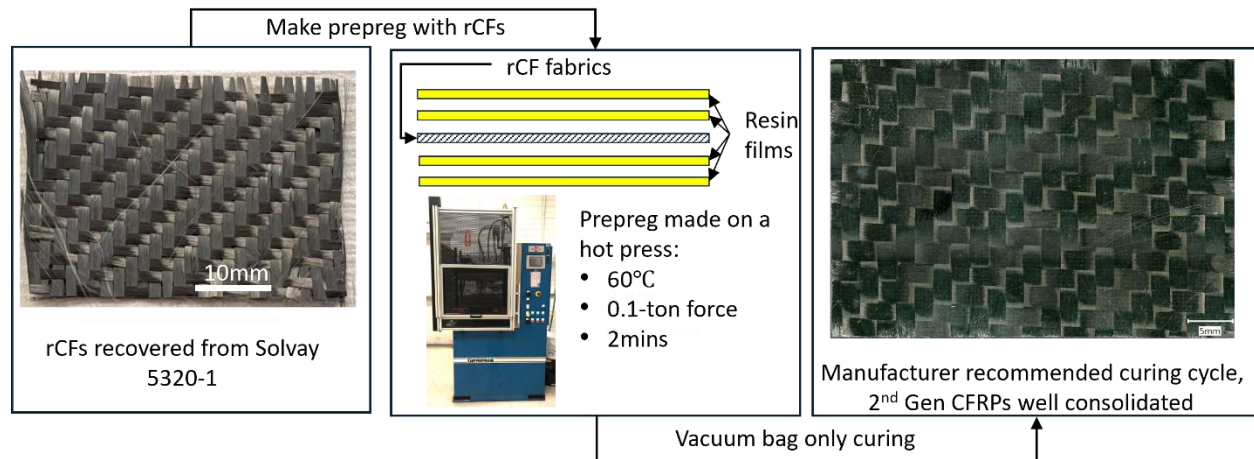
#### f. Molten hydroxide digestion of Bi/M-DEA neat resin

Digestion of unpretreated Bi/M-DEA neat resin was performed using a 15 g mixture of NaOH and KOH (40 g, 1:1 molar) placed in a 100 mL nickel crucible. The crucible and hydroxide mixture were heated to 260 °C in a sand bath. Pre-grounded Bi/M-DEA neat resin (0.2018 g) was placed into the crucible. A customized stainless-steel lid with a 24/40 female joint was placed on top to connect a Hickman distillation apparatus to collect volatiles, as shown in Figure S4. The resin was completely dissolved after 6 hours of reaction at 260 °C.

### 4. Manufacture 2<sup>nd</sup> Gen 5320-1 CFRPs using recovered CFs

#### a. Fabrication of 2<sup>nd</sup> Gen 5320-1 CFRPs

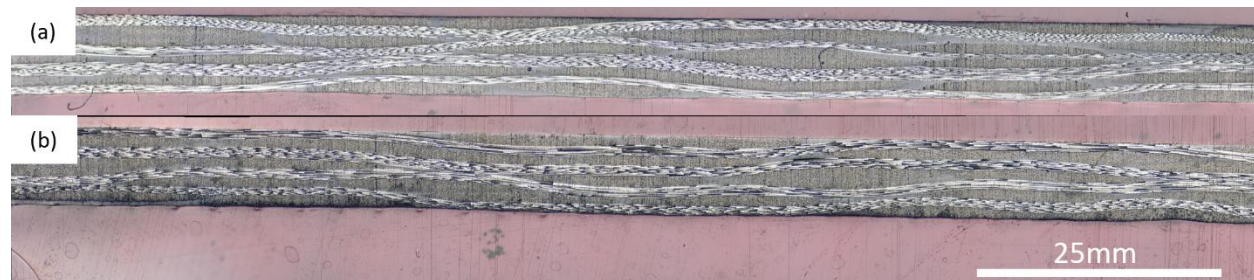
As shown in Figure S8, recycled CFs (~50.8 x 38.1 mm) were combined with fresh resin film (50.8 x 38.1 mm, Solvay CYCOM 5320-1) to fabricate prepreg and 2<sup>nd</sup> generation 5320-1 CFRPs following the same procedure described in Section S1(b). A reference 5320-1 CFRP was made with fresh CFs (50.8 x 38.1 mm) as a benchmark for characterizing 2<sup>nd</sup> CFRPs.



**Figure S8.** Process to remanufacture recovered CFs into 2<sup>nd</sup> Gen CFRPs.

**b. Cross-section of 2nd Gen 5320-1 CFRP**

Strips were cut from 2<sup>nd</sup> generation 5320-1, and its reference using a diamond saw. Strips were mounted with transparent resin (CitoPress-1, Struers) for cross-section polishing. Mounted samples were then polished using silicon carbide papers and aluminum oxide slurry. Polished cross-sections were imaged using a light microscope (X200, VHX5000, Kenyence), as shown in Figure S9.



**Figure S9.** Cross section of reference 5320-1 CFRP (a) and 2nd Gen 5320-1 CFRP (b) at 200X.

**c. Short beam shear strength measurement of 2<sup>nd</sup> 5320-1 gen CFRPs**

Short beam shear strength (SBS) samples of reference and 2nd Gen CFRPs were manufactured using 10 layers of fresh or recycled CF fabrics following the procedure described in Section S3 part a. SBS samples (13.7 x 0.46 x 0.23 mm) were cut from fully cured 10-layers 5320-1 reference and 2<sup>nd</sup> generation CFRP panels on a water jet cutter.

SBS was measured following ASTM D 2344 standard on an INSTRON 5544 load frame equipped with a 2 kN load cell. The speed of crosshead movement was set as 1.0 mm/min. Eight measurements were taken on reference 10-layer 5320-1 CFRP and 8 measurements were taken on 10-layer 2<sup>nd</sup> generation 5320 CFRPs.

**Table S7.** SBS measurements of 10-layers ref 5320-1 and 10-layers 2<sup>nd</sup> Gen 5320-1 CFRPs.

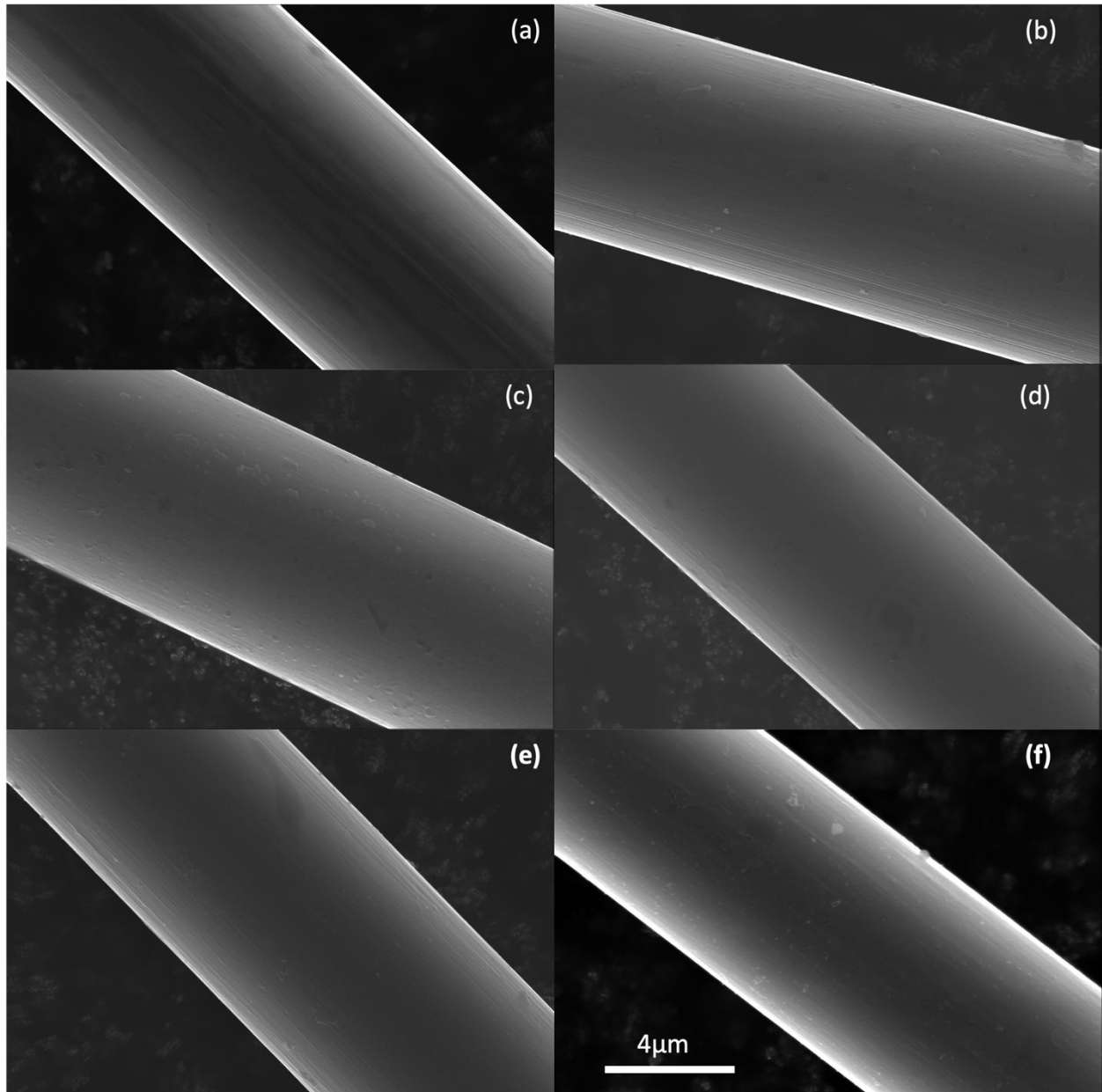
| Sample                          | SBS strength (Mpa) |
|---------------------------------|--------------------|
| Ref 5320-1 CFRP                 | 72.67±5.41         |
| 2 <sup>nd</sup> Gen 5320-1 CFRP | 55.43±3.16         |
| Retention (%)                   | 76.28%             |



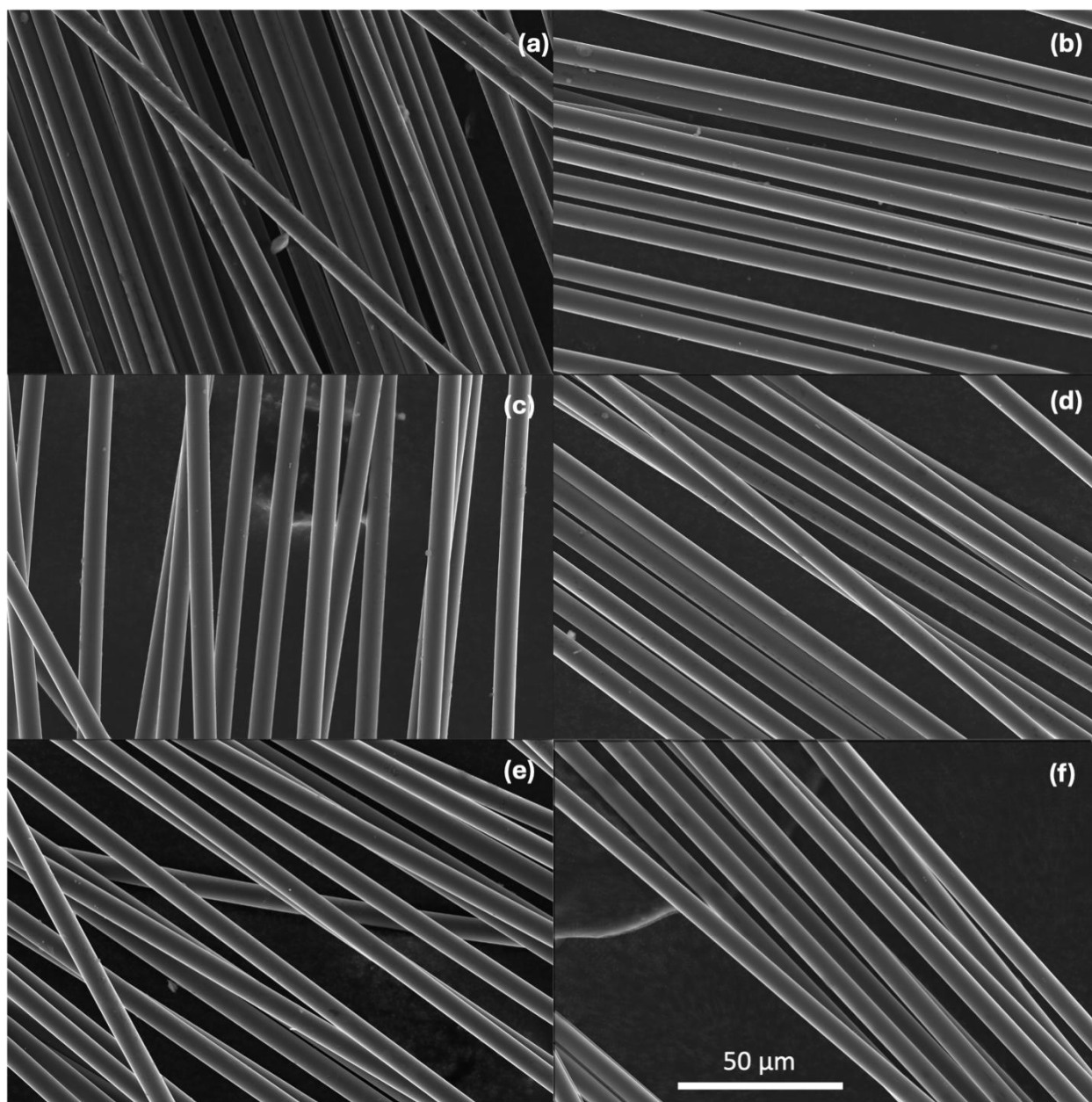
## 5. Recycled carbon fiber characterization

### a. SEM

SEM images were taken on a Helios G4 FIB/SEM (Thermo Scientific). The acceleration voltage and working distance were set as 15 Kv and around 5.5 mm respectively.



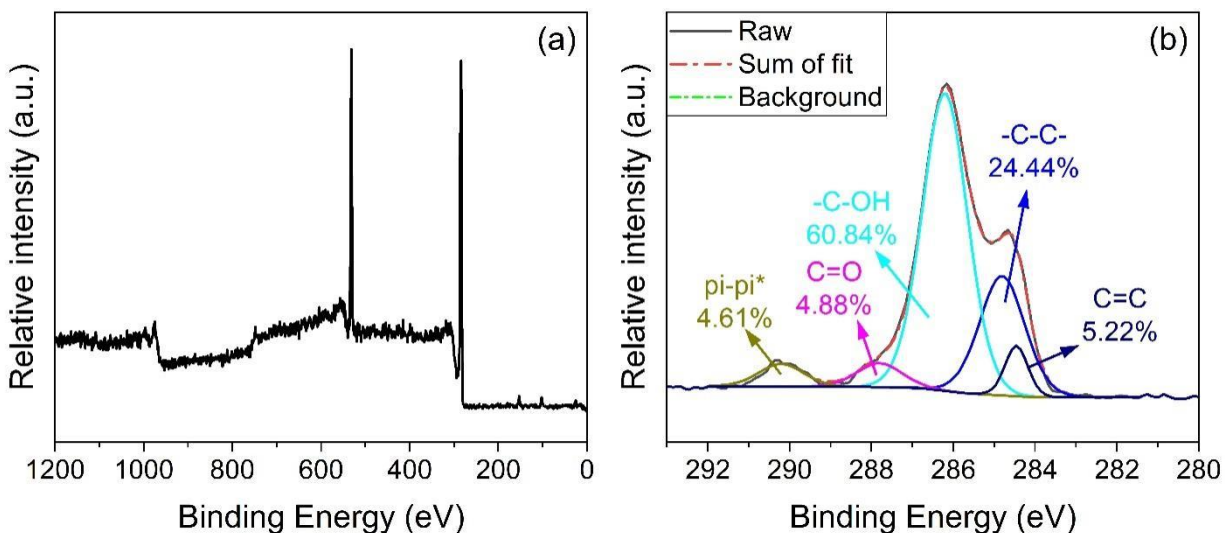
**Figure S10.** SEM images of fresh CF (a), recycled CFs from Bi/DDS CFRPs at 190 °C (b), 200 °C (c), 220 °C (d), 240 °C (e) and 5320-1 CFRP (f) at 2,5000 X.



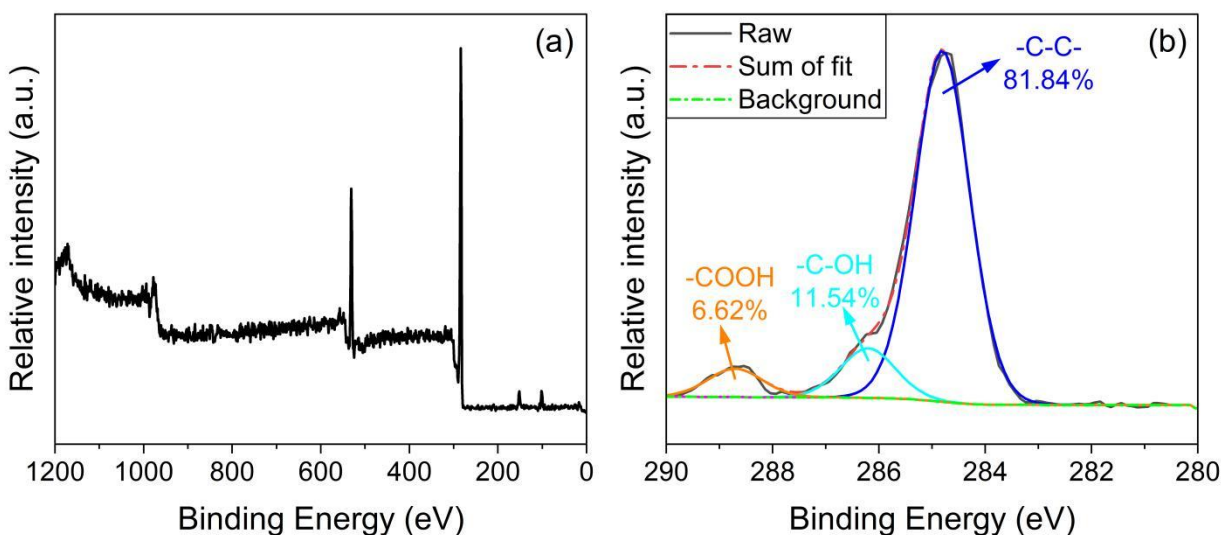
**Figure S11.** SEM images of fresh CF(a), recycled CFs from Bi/DDS CFRPs at 190 °C (b), 200°C (c), 220°C (d), 240°C (e) and 5320-1 CFRP (f) at 2500 X.

## b. XPS

X-ray photoelectron spectroscopy (XPS, Kratos Axis Ultra DLD) was used to analyze elements and functional groups on CF surface. A survey scan (0-1200 eV) was first acquired on each sample, followed by a high-resolution C1s scan. X-ray source was mono aluminum, and the incident angle was 90°. Curve-fitting of the C1s spectra was performed in CasaXPS software using Shirley baseline and Gaussian-Lorentzian functions, as shown in Figure S12-S13.



**Figure S 12.** XPS survey (a) and C1s high-resolution spectra (b) of fresh CF



**Figure S 13.** XPS survey (a) and C1s high-resolution spectra (b) of CFs recovered from a 220 °C reaction

XPS survey spectra show four peaks assigned to C, O, N and Si. The detailed element compositions of fresh and recycled CFs are summarized in Table S6. The results show the content ratio of O/C decreased after recycling at 220 °C for 30 minutes, indicating the recycled CFs were not oxidized during the recycling.<sup>9</sup>

**Table S8.** XPS analysis summary of fresh CFs and recycled CFs recovered from 220 °C.

| Sample    | Surface element (%) |      |      |       |       | C1s Components (%) |       |      |       |      |                 |
|-----------|---------------------|------|------|-------|-------|--------------------|-------|------|-------|------|-----------------|
|           | C                   | N    | Si   | O     | O/C   | C-C                | C-OH  | C=O  | O-C=O | C=C  | $\pi$ - $\pi^*$ |
| Fresh CF* | 78.86               | 1.25 | 3.35 | 16.55 | 20.99 | 24.44              | 60.84 | 4.88 | N/A   | 5.22 | 4.61            |
| rCF-220   | 82.19               | 0.73 | 4.13 | 12.95 | 15.76 | 81.84              | 11.54 | N/A  | 6.62  | N/A  | N/A             |

\*: with sizing

The C1s high-resolution spectra can be curve-fitted into five peaks: C-C (284.8ev), C-OH (286.2 ev), C=O (287.8 ev), O-C=O (288.8 ev) and  $\pi$ - $\pi^*$  satellite (290.2ev).<sup>12</sup> After recycling, C-O concentration greatly decreased. This indicates the sizing agents of the CFs were removed during hydroxide degradation.

### c. Single fiber tensile tests of fresh and recycled CFs

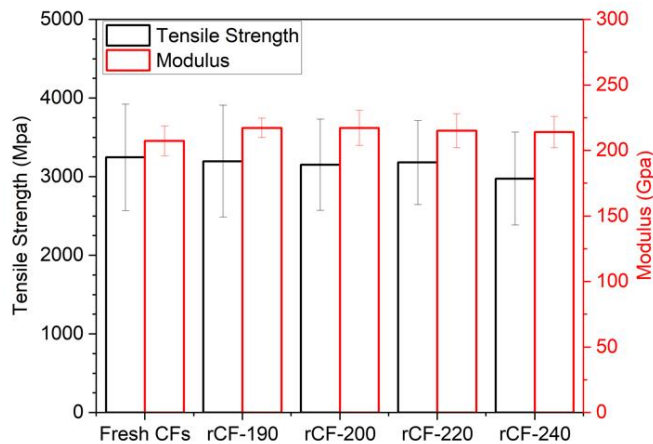
The tensile properties of both fresh and recycled CFs were tested in accordance with the ISO 11566 standard. To prepare the samples, individual fibers were separated from tows and mounted on paper strips measuring 81.3 x 25.4 mm, with a 25.4 x 12.7 mm window in the center. Fibers were mount across the center window. Both ends of fiber were affixed using double side tape and epoxy adhesive (Henkel E-20HP) to secure the fibers in place.

Once the epoxy adhesive had cured, the mounted samples were examined using a light microscope (Keyence VHX-5000) using a 1500X lens to measure fiber diameter. The reference length of pixel is calibrated using reference scale (Keyence OP-87426). The diameter of each sample was measured three times, and measured values were averaged to obtain the final measurement. The CF cross-sections were assumed to be circular, and the areas were calculated using the averaged diameters.

After mounting the sample onto the load frame, a cut was made in the center of the mounting sheet to free the fiber for testing. The samples were tested at 2mm/min crosshead speed till break. A total of 200 tests were conducted, with 40 tests performed on fresh CFs and another 160 tests on recycled CFs. The tensile strength was then calculated and plotted against strain (%) using these calculated CF cross-section areas. The slope of stress-strain curves was taken as tensile modulus. Table S9-14 and Figure S14 summarize all tensile test results.

**Table S9.** Single fiber tensile test summary of fresh and recycled CFs.

| Samples   | Diameter ( $\mu\text{m}$ ) | Strain at Break (%) | Tensile Strength (MPa) | Modulus (Gpa)      |
|-----------|----------------------------|---------------------|------------------------|--------------------|
| Fresh CFs | 7.47 $\pm$ 0.13            | 1.49 $\pm$ 0.30     | 3244.96 $\pm$ 676.10   | 207.28 $\pm$ 11.44 |
| rCF-190   | 7.51 $\pm$ 0.12            | 1.42 $\pm$ 0.31     | 3196.92 $\pm$ 712.37   | 217.23 $\pm$ 7.47  |
| rCF-200   | 7.43 $\pm$ 0.13            | 1.43 $\pm$ 0.25     | 3151.45 $\pm$ 579.72   | 217.20 $\pm$ 13.42 |
| rCF-220   | 7.43 $\pm$ 0.12            | 1.49 $\pm$ 0.25     | 3179.89 $\pm$ 535.30   | 215.06 $\pm$ 13.03 |
| rCF-240   | 7.41 $\pm$ 0.06            | 1.40 $\pm$ 0.30     | 2975.99 $\pm$ 591.12   | 214.03 $\pm$ 11.97 |



**Figure S14.** Single fiber tensile strength study of fresh and recycled CFs.

**Table S10.** Single fiber tensile strength test summary of fresh CFs.

| Test#   | Diameter ( $\mu\text{m}$ ) | Strain at break (%) | Force at break(N) | Strength (Mpa) | Modulus (Gpa) |
|---------|----------------------------|---------------------|-------------------|----------------|---------------|
| 1       | 7.44                       | 1.55                | 0.1474            | 3391.05        | 214.67        |
| 2       | 7.52                       | 1.80                | 0.1856            | 4179.80        | 217.67        |
| 3       | 7.50                       | 1.78                | 0.1732            | 3918.05        | 207.57        |
| 4       | 7.53                       | 1.58                | 0.1560            | 3507.47        | 208.47        |
| 5       | 7.22                       | 1.66                | 0.1393            | 3404.38        | 193.65        |
| 6       | 7.48                       | 1.12                | 0.1092            | 2484.29        | 206.74        |
| 7       | 7.63                       | 1.17                | 0.1186            | 2594.73        | 212.20        |
| 8       | 7.45                       | 1.90                | 0.1773            | 4069.13        | 209.42        |
| 9       | 7.50                       | 1.21                | 0.1188            | 2687.83        | 213.47        |
| 10      | 7.68                       | 1.22                | 0.1255            | 2707.95        | 210.64        |
| 11      | 7.51                       | 1.56                | 0.1550            | 3500.47        | 217.59        |
| 12      | 7.61                       | 1.52                | 0.1487            | 3271.82        | 208.97        |
| 13      | 7.39                       | 1.56                | 0.1614            | 3765.06        | 211.09        |
| 14      | 7.46                       | 0.80                | 0.0760            | 1738.11        | 199.79        |
| 15      | 7.36                       | 0.81                | 0.0989            | 2328.14        | 202.64        |
| 16      | 7.57                       | 1.48                | 0.1450            | 3224.23        | 211.45        |
| 17      | 7.64                       | 1.69                | 0.1650            | 3598.33        | 208.01        |
| 18      | 7.44                       | 1.66                | 0.1450            | 3336.05        | 192.33        |
| 19      | 7.46                       | 1.49                | 0.1378            | 3151.25        | 203.84        |
| 20      | 7.49                       | 1.05                | 0.0959            | 2176.95        | 197.60        |
| 21      | 7.68                       | 1.58                | 0.1534            | 3310.00        | 203.66        |
| 22      | 7.49                       | 1.54                | 0.1475            | 3345.22        | 211.79        |
| 23      | 7.56                       | 1.59                | 0.1650            | 3674.86        | 224.13        |
| 24      | 7.03                       | 1.83                | 0.1466            | 3777.51        | 200.82        |
| 25      | 7.48                       | 1.03                | 0.0985            | 2239.98        | 207.54        |
| 26      | 7.56                       | 2.01                | 0.2002            | 4465.49        | 219.32        |
| 27      | 7.43                       | 1.28                | 0.1064            | 2455.93        | 187.05        |
| 28      | 7.43                       | 1.38                | 0.1293            | 2983.21        | 209.32        |
| 29      | 7.47                       | 1.61                | 0.1509            | 3444.00        | 208.93        |
| 30      | 7.39                       | 1.31                | 0.1068            | 2492.16        | 184.99        |
| 31      | 7.58                       | 1.94                | 0.1801            | 3997.02        | 198.44        |
| 32      | 7.47                       | 1.45                | 0.1306            | 2981.25        | 197.87        |
| 33      | 7.07                       | 1.36                | 0.1433            | 3652.06        | 232.83        |
| 34      | 7.57                       | 0.95                | 0.0779            | 1730.63        | 177.73        |
| 35      | 7.41                       | 1.88                | 0.1869            | 4336.84        | 228.31        |
| 36      | 7.48                       | 1.84                | 0.1691            | 3847.34        | 201.90        |
| 37      | 7.50                       | 1.38                | 0.1388            | 3140.59        | 201.90        |
| 38      | 7.35                       | 1.60                | 0.1627            | 3837.28        | 217.49        |
| 39      | 7.33                       | 1.71                | 0.1672            | 3963.75        | 229.31        |
| 40      | 7.53                       | 1.52                | 0.1375            | 3088.06        | 199.87        |
| Average | 7.47                       | 1.49                | 0.1420            | 3244.96        | 207.28        |
| STDV.P  | 0.13                       | 0.30                | 0.0294            | 676.10         | 11.44         |

**Table S11.** Single fiber tensile strength test summary of recovered CFs at 190 °C.

| Test# | Diameter (μm) | Strain at break (%) | Force at break(N) | Strength (Mpa) | Modulus (Gpa) |
|-------|---------------|---------------------|-------------------|----------------|---------------|
| 1     | 7.65          | 1.31                | 0.1323            | 2881.91        | 218.49        |
| 2     | 7.54          | 1.16                | 0.1091            | 2445.07        | 217.92        |
| 3     | 7.48          | 1.16                | 0.1721            | 3919.08        | 215.52        |
| 4     | 7.48          | 1.12                | 0.1088            | 2474.96        | 219.49        |
| 5     | 7.66          | 1.12                | 0.1882            | 4090.37        | 220.76        |
| 6     | 7.50          | 1.73                | 0.1695            | 3839.09        | 220.07        |
| 7     | 7.48          | 1.52                | 0.1449            | 3299.10        | 215.66        |
| 8     | 7.45          | 1.76                | 0.1695            | 3891.03        | 224.20        |
| 9     | 7.71          | 1.76                | 0.1798            | 3853.11        | 221.28        |
| 10    | 7.51          | 1.49                | 0.1392            | 3143.60        | 211.42        |
| 11    | 7.31          | 1.00                | 0.0863            | 2057.10        | 203.30        |
| 12    | 7.44          | 0.70                | 0.0674            | 1552.97        | 217.50        |
| 13    | 7.45          | 1.49                | 0.1504            | 3452.65        | 211.42        |
| 14    | 7.39          | 1.70                | 0.1494            | 3484.21        | 205.02        |
| 15    | 7.66          | 1.49                | 0.1494            | 3240.97        | 217.54        |
| 16    | 7.39          | 1.40                | 0.1297            | 3026.32        | 217.48        |
| 17    | 7.41          | 1.60                | 0.1406            | 3264.20        | 198.92        |
| 18    | 7.82          | 1.54                | 0.1563            | 3256.35        | 211.33        |
| 19    | 7.45          | 1.48                | 0.1484            | 3404.91        | 228.02        |
| 20    | 7.59          | 1.85                | 0.1832            | 4050.21        | 221.15        |
| 21    | 7.37          | 1.15                | 0.1070            | 2508.75        | 214.93        |
| 22    | 7.51          | 0.99                | 0.0992            | 2241.49        | 222.93        |
| 23    | 7.53          | 1.61                | 0.1628            | 3656.91        | 228.73        |
| 24    | 7.52          | 1.77                | 0.1794            | 4038.13        | 229.61        |
| 25    | 7.41          | 0.86                | 0.0814            | 1886.11        | 217.62        |
| 26    | 7.72          | 1.28                | 0.1316            | 2810.24        | 216.45        |
| 27    | 7.74          | 1.50                | 0.1598            | 3395.51        | 223.96        |
| 28    | 7.41          | 1.74                | 0.1552            | 3600.46        | 205.63        |
| 29    | 7.45          | 1.71                | 0.1617            | 3712.23        | 215.59        |
| 30    | 7.43          | 1.87                | 0.1866            | 4304.98        | 224.97        |
| 31    | 7.37          | 1.02                | 0.0948            | 2224.03        | 214.54        |
| 32    | 7.51          | 1.38                | 0.1300            | 2932.52        | 211.82        |
| 33    | 7.50          | 1.35                | 0.1266            | 2869.64        | 211.84        |
| 34    | 7.53          | 1.28                | 0.1317            | 2957.97        | 230.13        |
| 35    | 7.27          | 1.73                | 0.1667            | 4018.85        | 210.14        |
| 36    | 7.57          | 1.90                | 0.1847            | 4109.27        | 225.14        |
| 37    | 7.55          | 1.61                | 0.1649            | 3688.20        | 225.53        |
| 38    | 7.38          | 0.92                | 0.0813            | 1900.30        | 205.76        |
| 39    | 7.65          | 1.57                | 0.16              | 3483.21        | 224.17        |
| 40    | 7.59          | 1.23                | 0.12              | 2658.76        | 213.40        |
| Avg   | 7.51          | 1.42                | 0.1416            | 3196.92        | 217.23        |
| STDV  | 0.12          | 0.31                | 0.0325            | 712.37         | 7.47          |

**Table S12.** Single fiber tensile strength test summary of recovered CFs at 200 °C.

| Test#   | Diameter ( $\mu\text{m}$ ) | Strain at break (%) | Force at break(N) | Strength (Mpa) | Modulus (Gpa) |
|---------|----------------------------|---------------------|-------------------|----------------|---------------|
| 1       | 7.37                       | 1.13                | 0.1112            | 2608.89        | 225.29        |
| 2       | 7.38                       | 1.13                | 0.1345            | 3142.56        | 219.66        |
| 3       | 7.21                       | 1.36                | 0.1161            | 2844.33        | 209.79        |
| 4       | 7.21                       | 1.48                | 0.1341            | 3287.97        | 225.88        |
| 5       | 7.33                       | 0.85                | 0.0691            | 1636.84        | 188.32        |
| 6       | 7.50                       | 1.54                | 0.1375            | 3110.72        | 207.56        |
| 7       | 7.55                       | 1.20                | 0.1094            | 2443.74        | 202.36        |
| 8       | 7.51                       | 1.36                | 0.1303            | 2943.26        | 215.67        |
| 9       | 7.31                       | 1.62                | 0.1464            | 3489.85        | 215.64        |
| 10      | 7.43                       | 1.73                | 0.1687            | 3892.16        | 224.19        |
| 11      | 7.43                       | 1.71                | 0.1554            | 3585.95        | 217.15        |
| 12      | 7.53                       | 1.37                | 0.1314            | 2949.74        | 214.76        |
| 13      | 7.44                       | 1.59                | 0.1453            | 3341.57        | 216.98        |
| 14      | 7.41                       | 1.37                | 0.1309            | 3037.39        | 222.33        |
| 15      | 7.45                       | 1.53                | 0.1532            | 3516.91        | 230.97        |
| 16      | 7.23                       | 1.85                | 0.1465            | 3569.95        | 200.63        |
| 17      | 7.47                       | 0.95                | 0.0938            | 2142.83        | 224.07        |
| 18      | 7.50                       | 1.48                | 0.1485            | 3363.06        | 227.15        |
| 19      | 7.54                       | 1.16                | 0.1292            | 2895.23        | 247.07        |
| 20      | 7.45                       | 1.62                | 0.1621            | 3720.50        | 234.63        |
| 21      | 7.21                       | 1.23                | 0.1497            | 3667.22        | 237.26        |
| 22      | 7.25                       | 1.23                | 0.1154            | 2796.55        | 227.63        |
| 23      | 7.41                       | 1.47                | 0.1428            | 3310.49        | 235.96        |
| 24      | 7.33                       | 1.67                | 0.1601            | 3794.94        | 228.83        |
| 25      | 7.41                       | 1.54                | 0.1499            | 3476.80        | 198.12        |
| 26      | 7.29                       | 1.67                | 0.1442            | 3457.49        | 208.85        |
| 27      | 7.49                       | 1.46                | 0.1414            | 3208.19        | 226.51        |
| 28      | 7.54                       | 1.77                | 0.1827            | 4091.07        | 239.54        |
| 29      | 7.45                       | 1.69                | 0.1540            | 3533.44        | 207.36        |
| 30      | 7.48                       | 1.66                | 0.1550            | 3525.01        | 214.80        |
| 31      | 7.13                       | 1.66                | 0.1675            | 4197.76        | 225.18        |
| 32      | 7.53                       | 1.60                | 0.1505            | 3381.25        | 211.55        |
| 33      | 7.46                       | 1.57                | 0.1495            | 3425.17        | 218.73        |
| 34      | 7.43                       | 1.38                | 0.1152            | 2654.77        | 201.83        |
| 35      | 7.70                       | 1.83                | 0.1701            | 3651.76        | 203.76        |
| 36      | 7.71                       | 1.03                | 0.0978            | 2098.09        | 203.76        |
| 37      | 7.71                       | 1.33                | 0.1407            | 3013.23        | 222.70        |
| 38      | 7.43                       | 1.41                | 0.1346            | 3105.05        | 217.67        |
| 39      | 7.37                       | 0.93                | 0.0782            | 1832.84        | 190.07        |
| 40      | 7.51                       | 1.15                | 0.1024            | 2313.54        | 197.67        |
| Average | 7.43                       | 1.43                | 0.1364            | 3151.45        | 217.20        |
| STDV.P  | 0.13                       | 0.25                | 0.0247            | 579.72         | 13.42         |



**Table S13.** Single fiber tensile strength test summary of recovered CFs at 220 °C .

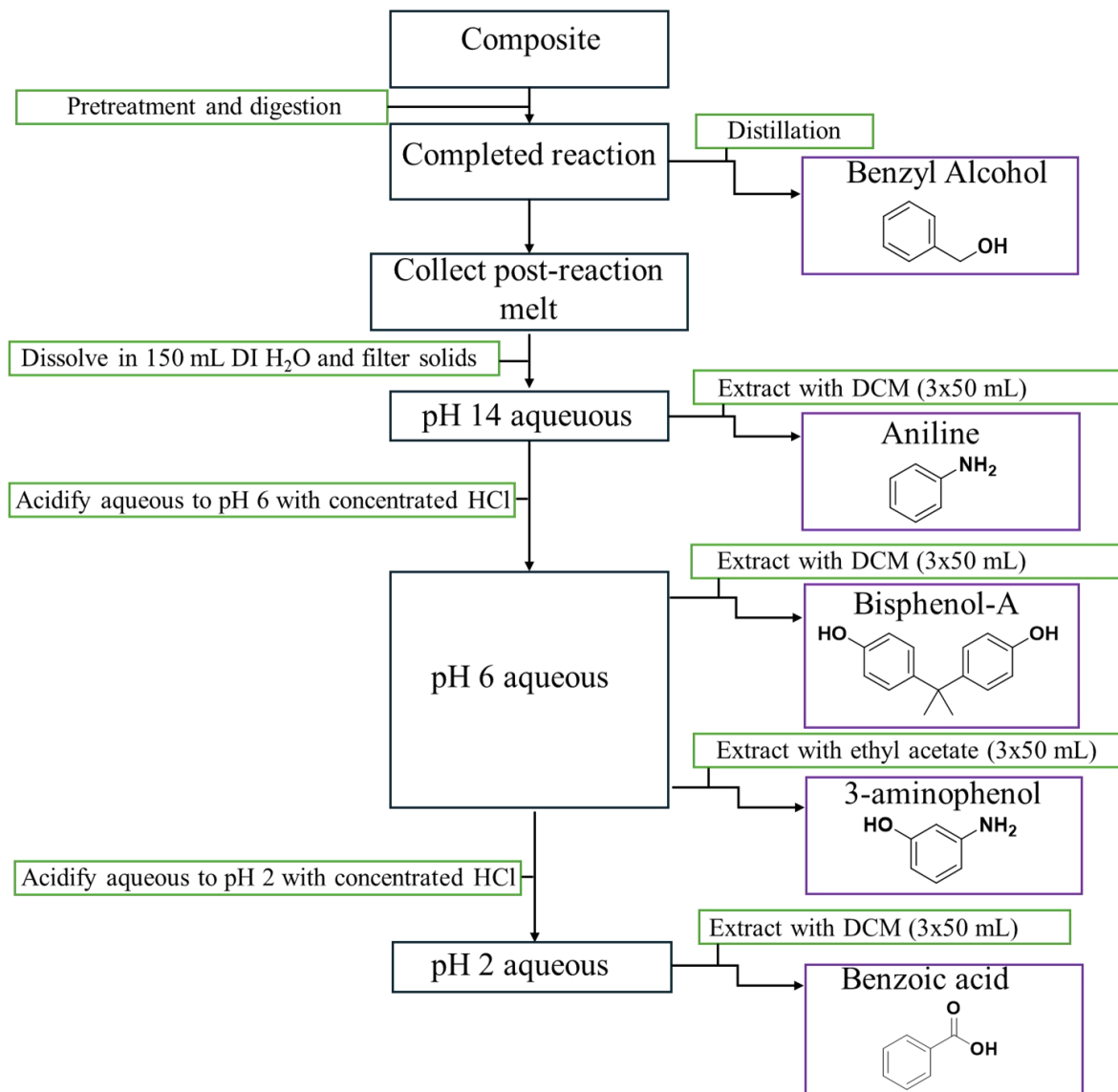
| Test# | Diameter ( $\mu\text{m}$ ) | Strain at break (%) | Force at break(N) | Strength (Mpa) | Modulus (Gpa) |
|-------|----------------------------|---------------------|-------------------|----------------|---------------|
| 1     | 7.45                       | 1.63                | 0.1481            | 3395.90        | 217.22        |
| 2     | 7.47                       | 1.34                | 0.1334            | 3044.49        | 226.74        |
| 3     | 7.45                       | 1.26                | 0.1266            | 2902.88        | 225.41        |
| 4     | 7.38                       | 1.32                | 0.1347            | 3147.46        | 236.28        |
| 5     | 7.41                       | 1.39                | 0.1378            | 3193.20        | 226.46        |
| 6     | 7.41                       | 1.38                | 0.1347            | 3124.39        | 225.27        |
| 7     | 7.41                       | 1.30                | 0.1139            | 2641.36        | 200.91        |
| 8     | 7.72                       | 1.48                | 0.1493            | 3187.82        | 214.05        |
| 9     | 7.40                       | 1.21                | 0.1147            | 2670.21        | 219.47        |
| 10    | 7.47                       | 1.18                | 0.1151            | 2627.86        | 226.88        |
| 11    | 7.19                       | 1.62                | 0.1517            | 3737.92        | 229.16        |
| 12    | 7.49                       | 1.45                | 0.1416            | 2902.30        | 200.20        |
| 13    | 7.39                       | 1.88                | 0.1651            | 3846.50        | 213.98        |
| 14    | 7.35                       | 1.86                | 0.1789            | 3667.29        | 197.64        |
| 15    | 7.27                       | 1.42                | 0.1187            | 2860.00        | 208.61        |
| 16    | 7.25                       | 1.74                | 0.1582            | 3242.98        | 187.17        |
| 17    | 7.51                       | 1.26                | 0.1270            | 2603.85        | 206.70        |
| 18    | 7.36                       | 1.52                | 0.1593            | 3265.53        | 213.38        |
| 19    | 7.51                       | 1.22                | 0.1249            | 2560.81        | 213.33        |
| 20    | 7.29                       | 1.60                | 0.1613            | 3305.50        | 205.75        |
| 21    | 7.43                       | 0.87                | 0.0811            | 1661.77        | 191.47        |
| 22    | 7.75                       | 1.50                | 0.1505            | 3194.54        | 212.79        |
| 23    | 7.53                       | 1.63                | 0.1709            | 3838.67        | 235.33        |
| 24    | 7.51                       | 1.71                | 0.1735            | 3918.32        | 237.27        |
| 25    | 7.39                       | 2.05                | 0.1894            | 4417.96        | 220.87        |
| 26    | 7.29                       | 1.20                | 0.1069            | 2562.68        | 214.59        |
| 27    | 7.43                       | 1.63                | 0.1628            | 3759.85        | 230.46        |
| 28    | 7.43                       | 1.60                | 0.1382            | 3189.28        | 199.55        |
| 29    | 7.37                       | 1.46                | 0.1308            | 3064.86        | 209.39        |
| 30    | 7.29                       | 0.83                | 0.0735            | 1762.78        | 208.78        |
| 31    | 7.35                       | 1.70                | 0.1355            | 3194.23        | 187.30        |
| 32    | 7.69                       | 1.30                | 0.1317            | 2840.35        | 219.21        |
| 33    | 7.47                       | 1.44                | 0.1420            | 3242.65        | 226.40        |
| 34    | 7.65                       | 1.66                | 0.1683            | 3664.11        | 223.05        |
| 35    | 7.47                       | 1.75                | 0.1533            | 3499.70        | 204.65        |
| 36    | 7.52                       | 1.87                | 0.1617            | 3645.32        | 196.93        |
| 37    | 7.39                       | 1.55                | 0.1456            | 3396.04        | 218.06        |
| 38    | 7.49                       | 1.56                | 0.1487            | 3372.67        | 216.35        |
| 39    | 7.39                       | 1.54                | 0.1499            | 3496.11        | 227.87        |
| 40    | 7.25                       | 1.56                | 0.1464            | 3545.56        | 227.30        |
| Avg   | 7.43                       | 1.49                | 0.1414            | 3179.89        | 215.06        |
| STDV  | 0.12                       | 0.25                | 0.0238            | 535.30         | 13.03         |

**Table S14.** Single fiber tensile strength test summary of recovered CFs at 240 °C .

| Test#   | diameter ( $\mu\text{m}$ ) | Strain at break (%) | Force at break(N) | Strength (Mpa) | Modulus (Gpa) |
|---------|----------------------------|---------------------|-------------------|----------------|---------------|
| 1       | 7.42                       | 1.29                | 0.1229            | 2845.96        | 219.30        |
| 2       | 7.40                       | 0.89                | 0.0821            | 1907.48        | 212.23        |
| 3       | 7.43                       | 1.26                | 0.1247            | 2875.64        | 229.90        |
| 4       | 7.39                       | 1.63                | 0.1595            | 3717.39        | 235.51        |
| 5       | 7.45                       | 1.63                | 0.1566            | 3598.17        | 219.68        |
| 6       | 7.34                       | 1.32                | 0.1229            | 2903.80        | 218.50        |
| 7       | 7.51                       | 1.23                | 0.1162            | 2625.24        | 211.52        |
| 8       | 7.40                       | 0.99                | 0.0905            | 2106.74        | 210.43        |
| 9       | 7.40                       | 0.95                | 0.0871            | 2024.16        | 210.34        |
| 10      | 7.55                       | 1.61                | 0.1624            | 3626.09        | 228.14        |
| 11      | 7.33                       | 1.62                | 0.1423            | 3376.93        | 208.04        |
| 12      | 7.28                       | 1.84                | 0.1459            | 3505.93        | 204.24        |
| 13      | 7.53                       | 2.03                | 0.1728            | 3881.81        | 196.02        |
| 14      | 7.40                       | 1.12                | 0.1004            | 2338.65        | 206.77        |
| 15      | 7.37                       | 1.47                | 0.1185            | 2776.18        | 189.30        |
| 16      | 7.33                       | 1.05                | 0.0967            | 2295.03        | 190.14        |
| 17      | 7.4                        | 1.33                | 0.1133            | 2621.98        | 199.15        |
| 18      | 7.30                       | 0.99                | 0.0866            | 2067.07        | 203.58        |
| 19      | 7.41                       | 1.78                | 0.1517            | 3515.63        | 206.67        |
| 20      | 7.53                       | 1.55                | 0.1589            | 3572.46        | 235.02        |
| 21      | 7.49                       | 1.15                | 0.1161            | 2637.23        | 228.65        |
| 22      | 7.38                       | 1.58                | 0.1387            | 3244.10        | 203.17        |
| 23      | 7.42                       | 1.22                | 0.1139            | 2633.26        | 215.86        |
| 24      | 7.46                       | 1.89                | 0.1492            | 3418.98        | 186.09        |
| 25      | 7.36                       | 1.45                | 0.1290            | 3032.94        | 208.17        |
| 26      | 7.45                       | 1.41                | 0.1391            | 3195.23        | 226.69        |
| 27      | 7.40                       | 1.84                | 0.1660            | 3864.45        | 218.58        |
| 28      | 7.43                       | 1.84                | 0.1744            | 4028.46        | 223.70        |
| 29      | 7.49                       | 1.83                | 0.1771            | 4017.90        | 220.78        |
| 30      | 7.46                       | 1.22                | 0.1097            | 2511.99        | 207.06        |
| 31      | 7.34                       | 1.47                | 0.1392            | 3287.21        | 223.78        |
| 32      | 7.43                       | 1.01                | 0.0959            | 2215.63        | 219.64        |
| 33      | 7.35                       | 1.24                | 0.1188            | 2801.38        | 223.71        |
| 34      | 7.45                       | 1.18                | 0.1074            | 2465.72        | 209.92        |
| 35      | 7.40                       | 1.68                | 0.1518            | 3534.75        | 213.14        |
| 36      | 7.36                       | 1.21                | 0.1096            | 2578.36        | 208.39        |
| 37      | 7.31                       | 1.01                | 0.0946            | 2255.69        | 221.15        |
| 38      | 7.42                       | 1.44                | 0.1321            | 3056.28        | 214.32        |
| 39      | 7.43                       | 1.52                | 0.1443            | 3333.49        | 224.50        |
| 40      | 7.38                       | 1.19                | 0.1172            | 2744.17        | 229.50        |
| Average | 7.41                       | 1.40                | 0.1284            | 2975.99        | 214.03        |
| STDV.P  | 0.06                       | 0.30                | 0.0263            | 591.20         | 11.97         |

## 6. Work-up procedure

The work up procedure used to isolate components from a pretreated composite degradation reaction is shown below.



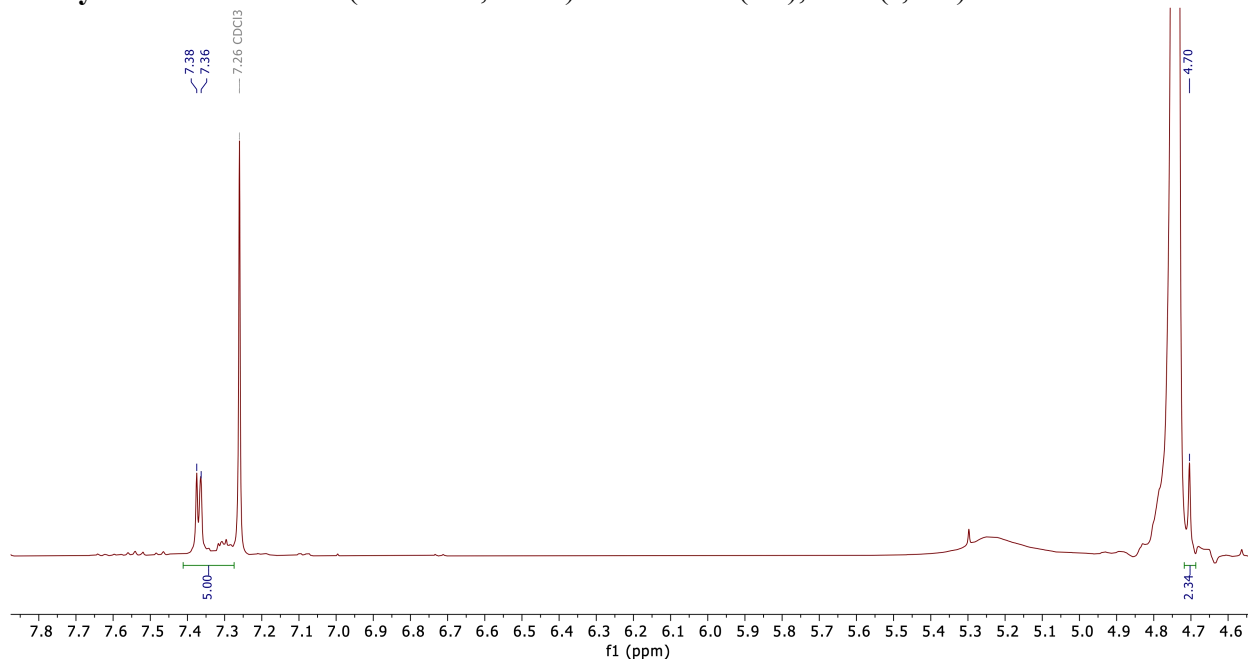
**Figure S15.** Work up procedure involving pH-based isolation.

After work-up the aqueous was then distilled and the leftover solid material was analyzed by NMR and showed no organic material.

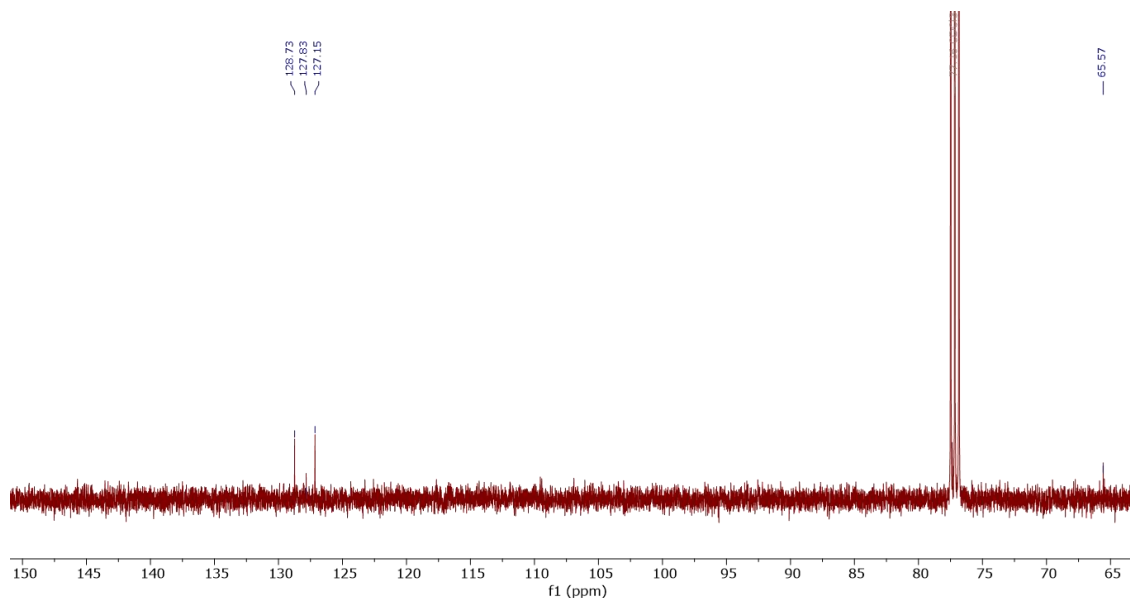
### a. Distillate collection

When the reaction was complete, it was allowed to cool down to 160 °C. Afterwards, the Hickman distillation head set up was removed and placed above a 100 mL beaker. Then it was flushed with 20 mL of dichloromethane (DCM) and combined with the collected distillate. The solution was then transferred to a 50 mL round bottom flask and the solvent was removed under reduced pressure to yield a light brown liquid. The  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra can be found in Figures S16 and S17 respectively. It comprises mostly of benzyl alcohol.

**Benzyl alcohol.**  $^1\text{H}$  NMR (400 MHz,  $\text{cdcl}_3$ )  $\delta$  7.41-7.27 (5H), 4.70 (s, 2H).



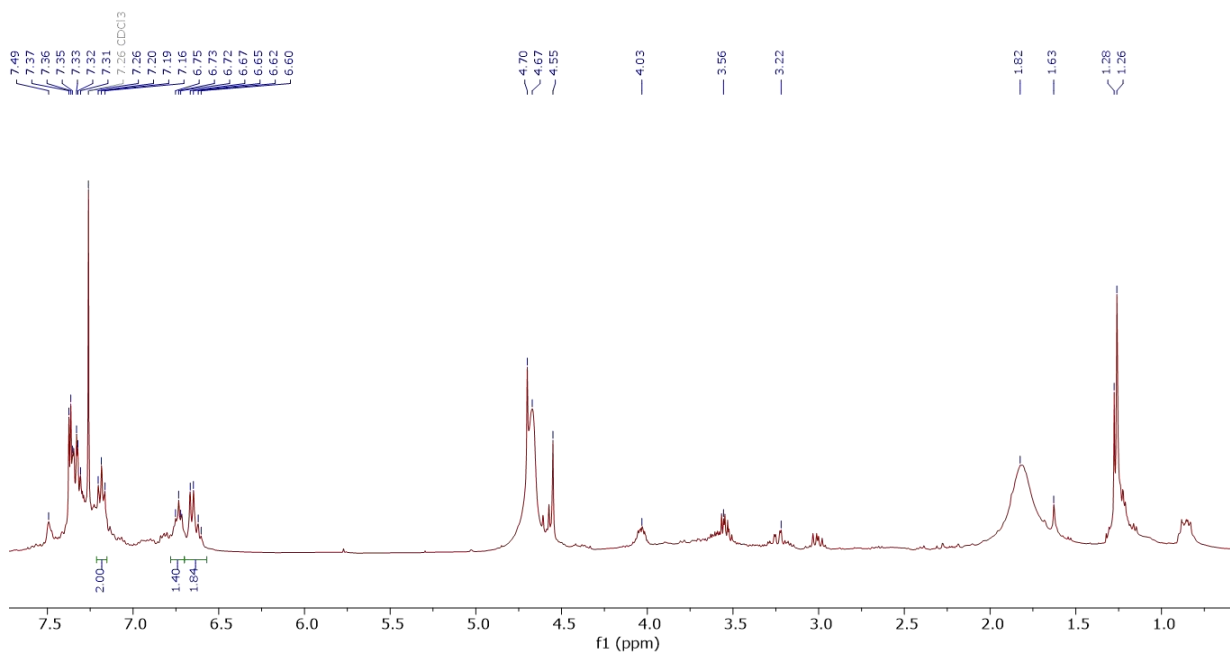
**Figure S16.**  $^1\text{H}$  NMR of distillate in  $\text{CDCl}_3$ .



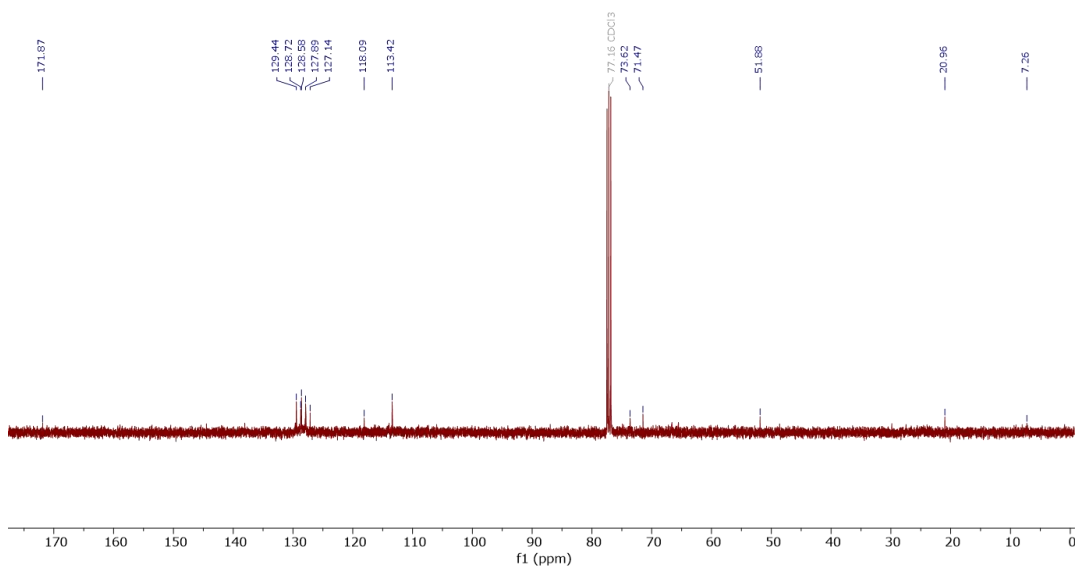
**Figure S17.**  $^{13}\text{C}\{^1\text{H}\}$  NMR of distillate in  $\text{CDCl}_3$ .

### b. Organic base extract

After the reaction was cooled down to  $160\text{ }^\circ\text{C}$ , it was scooped into a 500 mL beaker with a scoopula. The solid melt was then dissolved with  $\text{H}_2\text{O}$  (deionized, 150 mL). The mixture was solubilized and passed through a Buchner filter. The filtrate was collected, and the aqueous layer was extracted with DCM (3 x 50 mL). Afterwards, the organic layer was dried over sodium sulfate. Once dried, the solvent was removed under reduced pressure. The  $^1\text{H}$   $^{13}\text{C}$  NMR spectra of the crude organic base extract can be found in Figures S18 and S19 respectively. The extracts show a mixture but the signals at 6.65, 6.74, and 7.18 in the HNMR resemble aniline.



**Figure S18.**  $^1\text{H}$  NMR of crude organic base extract in  $\text{CDCl}_3$ .

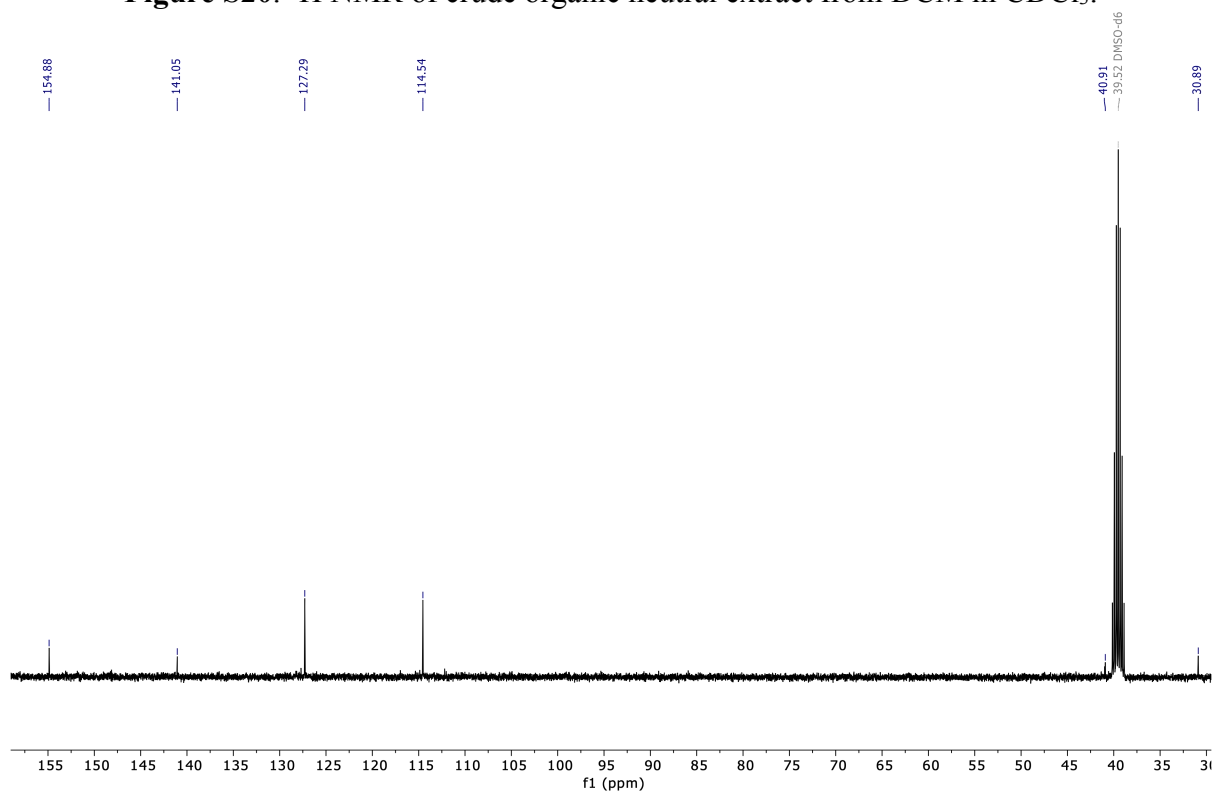
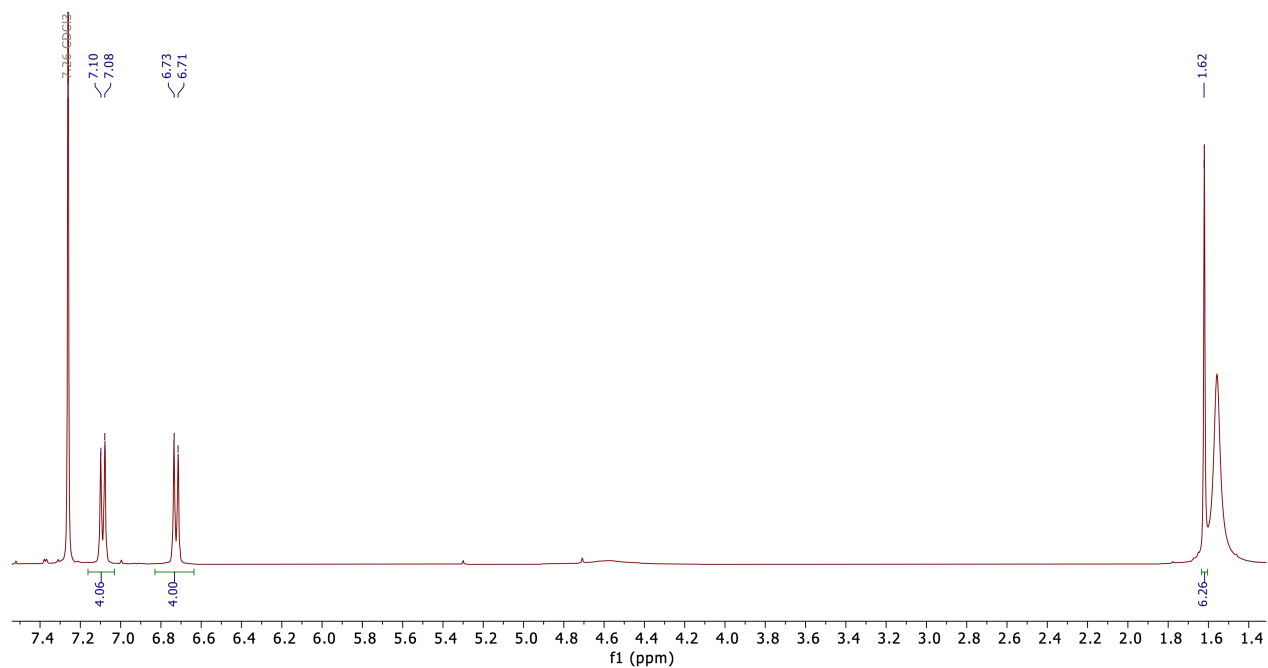


**Figure S19.**  $^{13}\text{C}\{^1\text{H}\}$  NMR of crude organic base extract in  $\text{CDCl}_3$ .

**c. Organic neutral extract from DCM**

The previously extracted aqueous layer was then acidified to pH 6 using concentrated hydrochloric acid (HCl). Once the solution was neutralized and cooled down, the organic material was extracted with DCM (3 x 50 mL). The organic layer was then dried over sodium sulfate and the solvent was removed under reduced pressure. The  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of the crude organic neutral extract can be found in Figures S20 and S21 respectively. The extract is primarily bisphenol A (BPA).

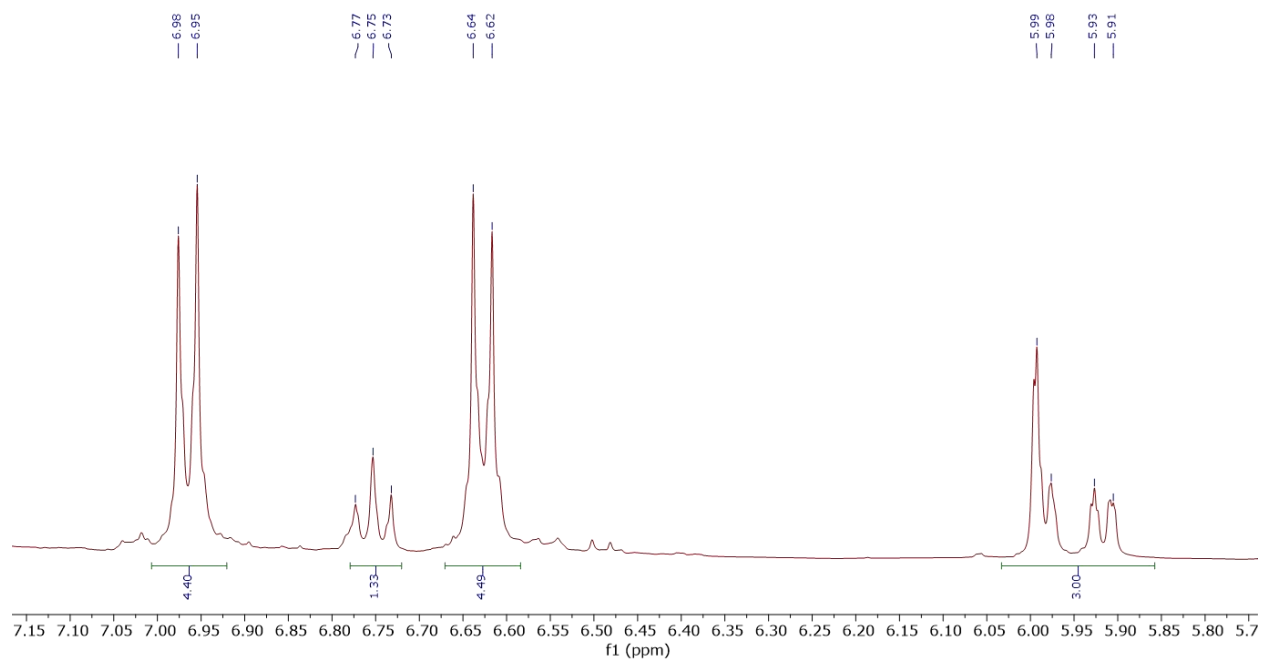
**Bisphenol-A:**  $^1\text{H}$  NMR (400 MHz,  $\text{cdcl}_3$ )  $\delta$  7.09 (d,  $J = 7.7$  Hz, 4H), 6.72 (d,  $J = 7.8$  Hz, 4H), 1.62 (s, 6H).  $^{13}\text{C}$  NMR (100 MHz,  $\text{dms}$ )  $\delta$  154.88, 141.05, 127.29, 114.54, 40.91, 30.89.



#### d. Organic neutral from ethyl acetate

The aqueous layer was then extracted with ethyl acetate (3 x 50 mL). The organic layers were then collected, combined and dried over sodium sulfate. Once dried, the solvent was removed under reduced pressure and left behind a brown-yellow oily residue. The  $^1\text{H}$  NMR spectrum of this extract can be found in Figure S22. The extract contained both BPA and 3-aminophenol.

**3-aminophenol.**  $^1\text{H}$  NMR (400 MHz, dmsO)  $\delta$  6.75 (t,  $J = 8.2$  Hz, 1H), 6.03 – 5.87 (m, 3H).



**Figure S22.**  $^1\text{H}$  NMR of crude organic neutral extract from ethyl acetate in  $\text{DMSO-}d_6$ .



### e. Organic acid extract

The previously extracted aqueous layer was then acidified to pH 2 using concentrated HCl. Once the solution was acidified, it was allowed to cool, and the organic material was extracted with DCM (3 x 50 mL). The organic layers were then combined and dried over sodium sulfate. Afterwards, the solvent was removed under reduced pressure. The  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra of the crude organic acid extract can be found in Figures S23 and S24. It contains benzoic acid.

**Benzoic acid.**  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  8.12 (d,  $J = 8.4$  Hz, 2H), 7.62 (t,  $J = 7.4$  Hz, 1H), 7.48 (t,  $J = 7.7$  Hz, 2H).  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ )  $\delta$  170.73, 133.88, 130.35, 129.30, 128.65.

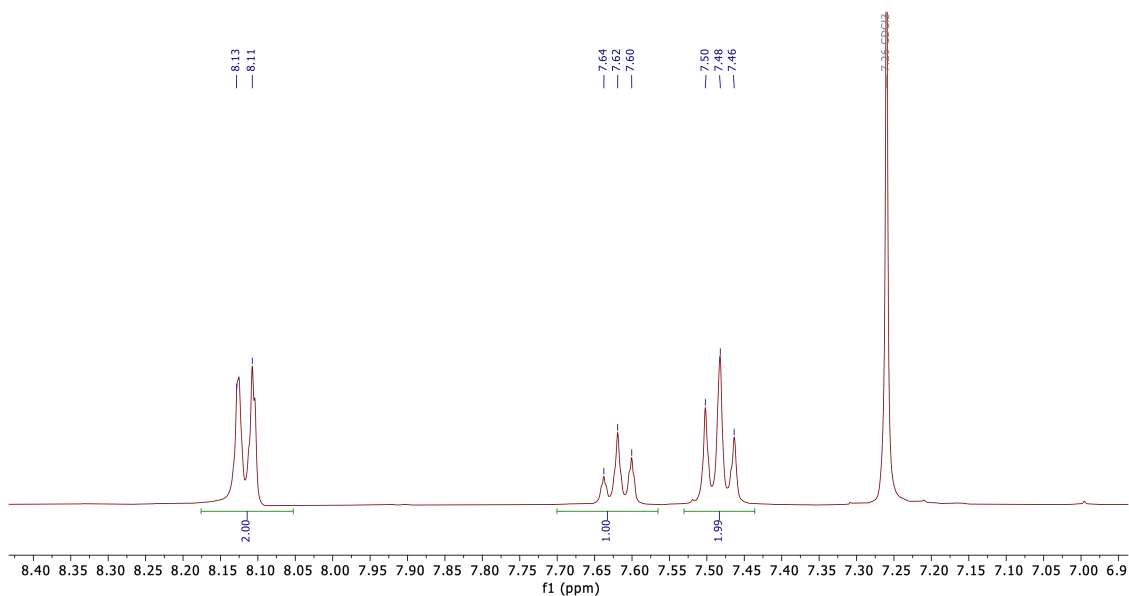


Figure S23.  $^1\text{H}$  NMR of crude organic acid extract in  $\text{CDCl}_3$ .

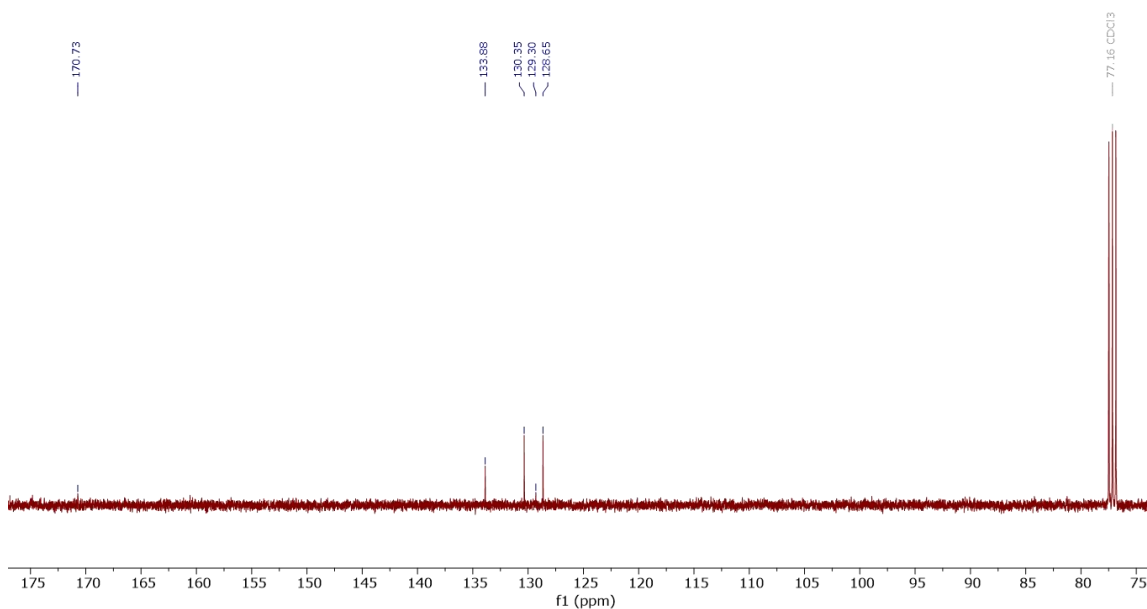
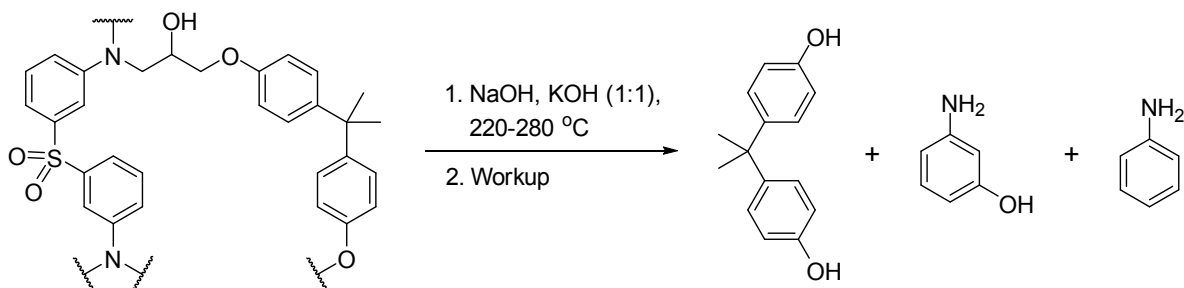


Figure S24.  $^{13}\text{C}\{^1\text{H}\}$  NMR of crude organic acid extract in  $\text{CDCl}_3$ .

## 7. Unpretreated composite degradation extracts

**Scheme S1.** Unpretreated degradation of Bi/DDS neat resin and CFRPs.



Resin degradation trials are summarized in Table S15, 20. Ground resin ( $d < 5$  mm) or pieces of resin/carbon fiber composite (13 x 10 mm) were tested. The reaction was conducted either in a round-bottom flask (25 or 50 mL, borosilicate glass) or in a regular glass test tube (entry 3). Heat sources were either a sand bath or an alcohol burner (entries 3 and 5). The reaction temperature was measured by a thermocouple.

A source of resin, NaOH and KOH· $\frac{1}{2}$ H<sub>2</sub>O (1:1 molar ratio) were placed in a reactor and the degradation was conducted at 220–280 °C for 0.3–3 hours according to Table S15. Manual stirring was applied for entries 3–5.

When a distillation head was used (Table S20, entries 1, 2, and 5), a colorless emulsion was collected (ca. 1–2 mL) as a distillate. It was extracted with DCM (1 mL x 3). The extract was dried with sodium sulfate and evaporated to give a yellow oil (crude aniline). Once cooled, the reaction melt was dissolved in a minimal amount of DI water to form a clear beige-brown solution. Carbon fibers and/or unreacted resin were filtered, washed with water, and air-dried.

The basic solution was extracted with DCM (5 mL x 3). The extract was dried with sodium sulfate and evaporated to give a brown residue (unreacted oligomers).

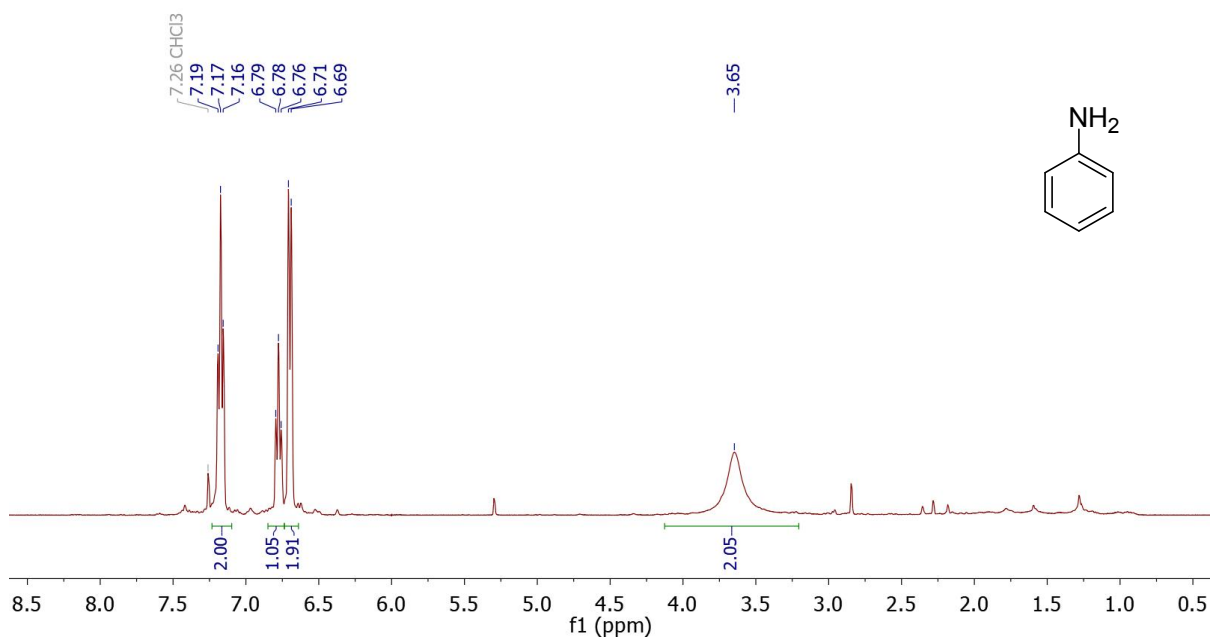
Then, the aqueous solution was brought to pH 0–1 by adding 6 M HCl carefully at stirring (Caution: exothermic reaction and evolution of SO<sub>2</sub> gas). Once at 25 °C, the acidic solution was extracted with DCM (5 mL x 4). The extract was dried with sodium sulfate and evaporated to give crude BPA as yellowish crystalline solid.

The resulting aqueous solution was filtered (to remove SiO<sub>2</sub>· $n$ H<sub>2</sub>O) and brought to pH 7–8 by adding solid sodium bicarbonate in small portions at stirring. The neutral solution was extracted with ethyl acetate (5 mL x 4). The extract was dried with sodium sulfate and evaporated to give a yellow oil (mostly 3-aminophenol). All the isolated materials were characterized by <sup>1</sup>H and <sup>13</sup>C NMR spectroscopy (Figures S25-30).

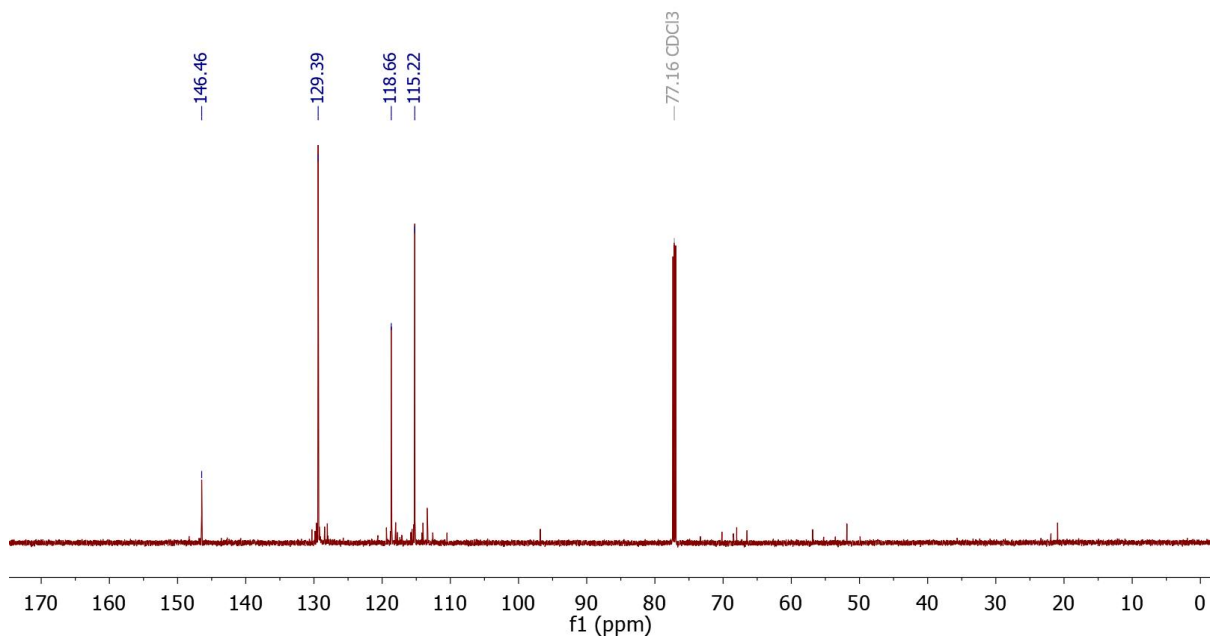
**Table S15.** Summary of reaction conditions of untreated Bi/DDS neat resin and CFRP.

| Entry | Substrate         | M <sub>I</sub> (g) | NaOH-KOH mixture (g) | Temperature (°C) | Time(hr) |
|-------|-------------------|--------------------|----------------------|------------------|----------|
| 1     | Bi/DDS neat resin | 2.00               | 10                   | 220-240          | 2        |
| 2     | Bi/DDS neat resin | 2.00               | 20                   | 230-250          | 3        |
| 3     | Bi/DDS neat resin | 0.50               | 5                    | 250-280          | 0.5      |
| 4     | Bi/DDS CFRP       | 3.00               | 20                   | 230-250          | 3        |
| 5     | Bi/DDS CFRP       | 2.00               | 9.6                  | >270             | 0.3      |

**Aniline.** <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.17 (t, *J* = 7.0 Hz, 2H, *m*-CH), 6.78 (t, *J* = 7.3 Hz, 1H, *p*-CH), 6.70 (d, *J* = 7.7 Hz, 2H, *o*-CH), 3.65 (br s, 2H, NH<sub>2</sub>). <sup>13</sup>C{<sup>1</sup>H} NMR (151 MHz, CDCl<sub>3</sub>): δ 146.46, 129.39, 118.66, 115.22.

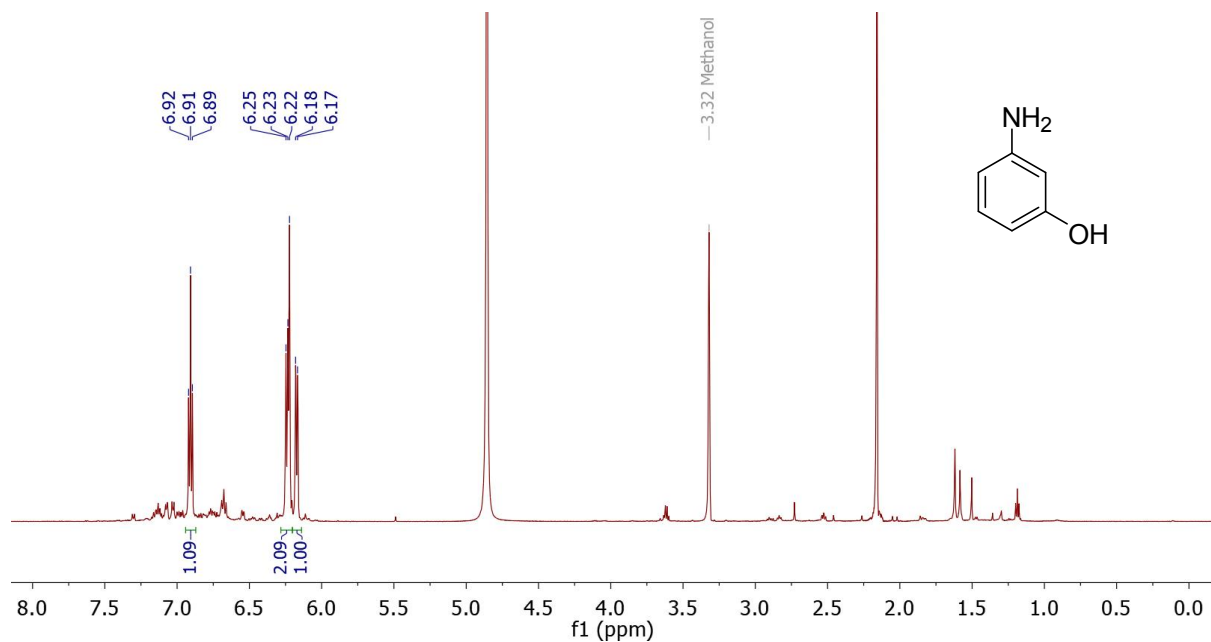


**Figure S25.** <sup>1</sup>H NMR spectrum of the crude aniline in CDCl<sub>3</sub>.

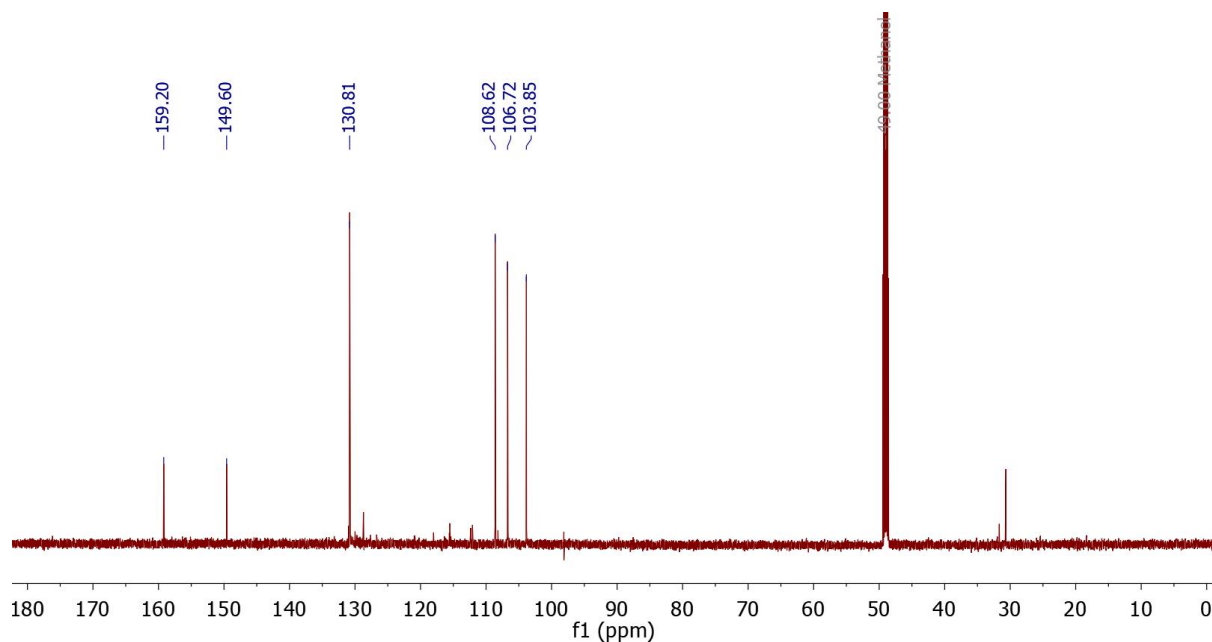


**Figure S26.**  $^{13}\text{C}\{^1\text{H}\}$  NMR spectrum of the crude aniline in  $\text{CDCl}_3$ .

**3-Aminophenol.**  $^1\text{H}$  NMR (600 MHz,  $\text{CD}_3\text{OD}$ ):  $\delta$  6.91 (t,  $J = 7.9$  Hz, 1H, CH), 6.24 (d,  $J = 7.9$  Hz, 1H, CH), 6.22 (s, 1H, CH), 6.17 (d,  $J = 8.0$  Hz, 1H, CH).  $^{13}\text{C}\{^1\text{H}\}$  NMR (151 MHz,  $\text{CD}_3\text{OD}$ ):  $\delta$  159.20, 149.60, 130.81, 108.62, 106.72, 103.85.

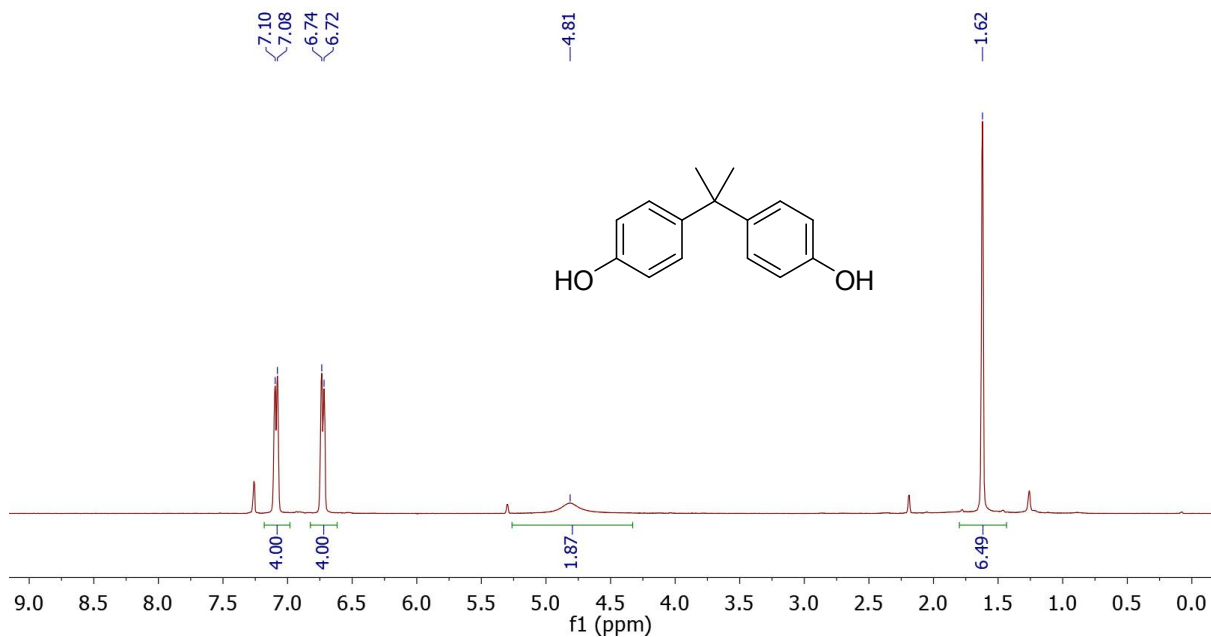


**Figure S27.**  $^1\text{H}$  NMR spectrum of the crude 3-aminophenol in  $\text{CD}_3\text{OD}$ .

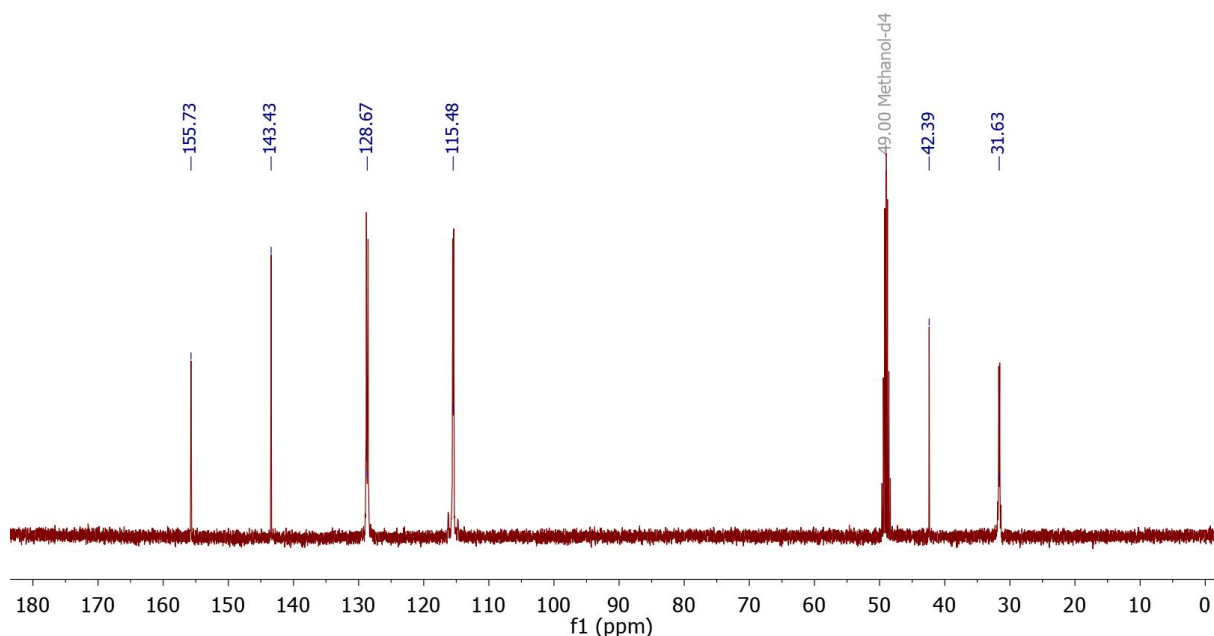


**Figure S28.**  $^{13}\text{C}\{^1\text{H}\}$  NMR spectrum of the crude 3-aminophenol in  $\text{CD}_3\text{OD}$ .

**2,2-Bis(4-hydroxyphenyl)propane (bisphenol A, BPA).**  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.09 (d,  $J = 6.8$  Hz, 4H, CH), 6.73 (d,  $J = 7.1$  Hz, 4H, CH), 4.81 (s, 2H, OH), 1.62 (s, 6H, 2 $\text{CH}_3$ ).  $^{13}\text{C}\{^1\text{H}\}$  NMR (101 MHz,  $\text{CD}_3\text{OD}$ ):  $\delta$  155.73, 143.43, 128.67, 115.48, 42.39, 31.63. The peaks of non-quaternary carbon atoms appear as multiplets, probably, due to a rapid acid-catalyzed H/D exchange with  $\text{CD}_3\text{OD}$ .



**Figure S29.**  $^1\text{H}$  NMR spectrum of the crude 2,2-bis(4-hydroxyphenyl)propane in  $\text{CDCl}_3$ .



**Figure S30.**  $^{13}\text{C}\{^1\text{H}\}$  NMR spectrum of the crude 2,2-bis(4-hydroxyphenyl)propane in  $\text{CD}_3\text{OD}$ .

## 8. Ion exchange chromatography

In addition to the pH-based separation strategy, we also offer a more sustainable separation strategy by using ion exchange resins. This technique requires less water, uses no organic solvent, and enables the recovery of hydroxide (91 mol%). The experiment went as described:

A CFRP composite (560 mg) is digested by NaOH (5.99 g, 0.1497 mol) and  $\text{KOH}\times 0.5\text{H}_2\text{O}$  (9.00 g, 0.1384 mol) in a steel crucible at 260 °C within 2 h. The cooled melt is dissolved in water (50 mL) and the brown solution is filtered through a PTFE filter (0.45  $\mu\text{m}$ ). CFs are recovered (309 mg).

A glass column (230 $\times$ 20 mm) is then filled with an anion-exchange resin (Amberlite, IRN-78), which is then prepared for the experiment by washing with aqueous NaOH (0.9 M) and DI water until pH 9.0–9.5. The resin is charged with the aqueous degradation solution. Then, pure  $\text{OH}^-$  is eluted by DI water. The collected fractions are listed in Table S17. Fractions 1–3 are combined ( $V_F = 136$  mL) and  $\text{OH}^-$  is quantified by the acid-base titration. A sample of the hydroxide solution ( $V_S = 1.00$  mL) is titrated with standard HCl ( $c_{\text{HCl}} = 0.1753$  M) in the presence of phenolphthalein indicator. Fractions 1–3 contain 0.263 mol of hydroxide ( $c_{\text{OH}} = 1.928$  M), which corresponds to 91% hydroxide recovery. The recovered solution shows no presence of organic compounds as suggested by  $^1\text{H}$  NMR after the  $\text{HCl}/\text{CH}_2\text{Cl}_2$  extraction (Figure S31).

$\text{BPA}^{2-}$  is not eluted by aqueous  $\text{OH}^-$ , so it was eluted as  $\text{H}_2\text{BPA}$  using HCl (6 M, 20 mL) in aqueous ethanol (1:1 v/v). The collected fractions are listed in Table S17. Fractions 5–8 are combined and evaporated under reduced pressure. A red oily residue is dried in a desiccator over KOH resulting in 100 mg of recovered organic material.

These collected fractions (5-8) were then analyzed by NMR and these spectra are shown in Figure S32.  $^1\text{H NMR}$  of crude combined fractions 5-8. Upon assignment, we observe that BPA is the major component in this mixture, however we do notice a number of impurities that are plasticizers from the plastic ethanol bottle we used to elute these fractions.

The crude mixture was then dissolved in 2 mL of DI  $\text{H}_2\text{O}$  in a vial and heated to  $90^\circ\text{C}$  using an oil bath for 5 minutes. Afterwards, 2 mL of toluene was added, and a second phase was observed. The mixture was then placed back over the heat for another 5 minutes with agitation. After 5 minutes have elapsed, the water layer was transferred to a 50 mL round bottom flask and another 2 mL of water was added back into the vial. Then it was heated again for another 5 minutes and when that time had elapsed, the water layer was combined with the previously extracted aqueous solution in the round bottom flask. Finally, the water was removed by rotary evaporation until the volume was 1 mL. At this point, the flask was allowed to cool to room temp and then the excess water was removed by pipette. The residue was then dissolved in deuterated chloroform and revealed BPA was purified as shown by NMR (Figure S38).

**Table S16.** Fraction of recovered hydroxide.

$$\text{Resin-NR}_3^+\text{OH}^- + \text{BPA}^{2-} \leftrightarrow \text{Resin-NR}_3^+\text{BPA}^{2-} + \text{OH}^-$$

| Fraction # | Color       | Volume, mL | pH    | $[\text{OH}^-]$ , M   | $n(\text{OH}^-)$ , mol |
|------------|-------------|------------|-------|-----------------------|------------------------|
| 1          | brown       | 53         | –     |                       |                        |
| 2          | light brown | 35         | –     | 1.928                 | 0.263                  |
| 3          | pale yellow | 42         | 13.39 |                       |                        |
| 4          | –           | 36         | 11.77 | $5.89 \times 10^{-3}$ |                        |
| 5          | –           | 44         | 10.64 | $4.37 \times 10^{-4}$ |                        |
| 6          | –           | 68         | 9.59  | $3.89 \times 10^{-5}$ | $2.34 \times 10^{-4}$  |
| 7          | –           | 22         | 9.36  | $2.29 \times 10^{-5}$ |                        |

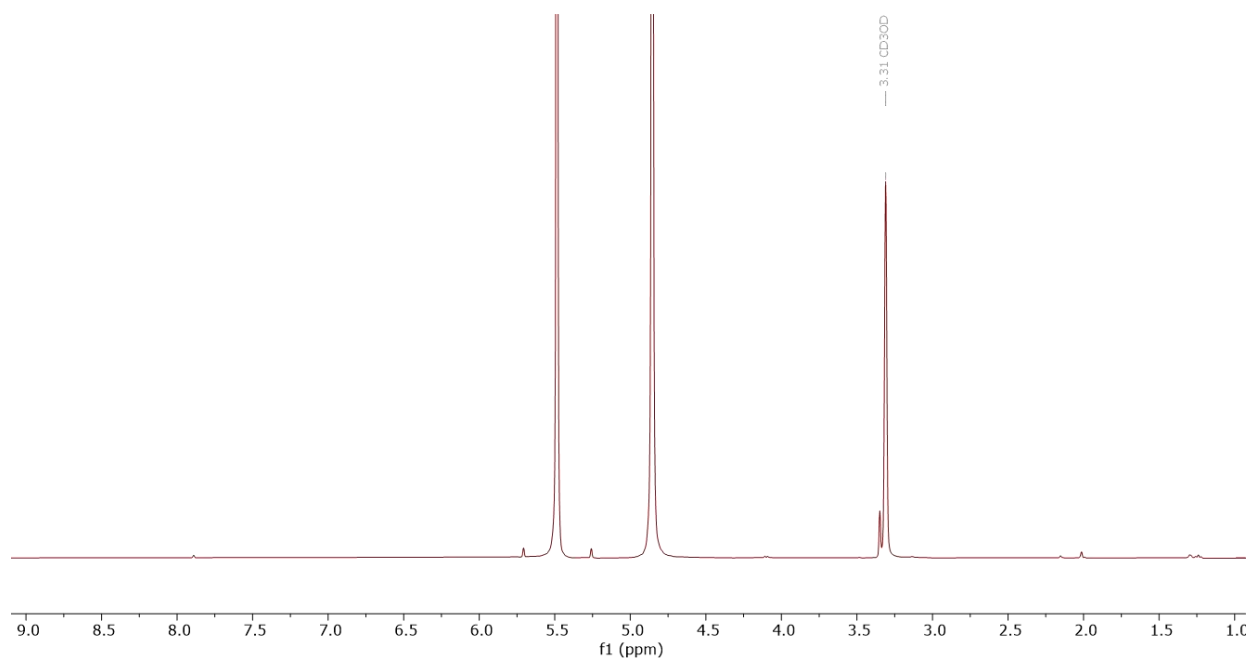
**Table S17.** Fractions of recovered organics acids.

$$\text{Resin-NR}_3^+\text{BPA}^{2-} + 2\text{H}^+ + \text{Cl}^- \leftrightarrow \text{Resin-NR}_3^+\text{Cl}^- + \text{H}_2\text{BPA}$$

| Fraction # | Color       | Volume, mL | pH  |
|------------|-------------|------------|-----|
| 1          | –           | 35         |     |
| 2          | –           | 50         |     |
| 3          | –           | 50         | 8–9 |
| 4          | –           | 20         |     |
| 5          | orange      | 50         |     |
| 6          | pale yellow | 50         |     |
| 7          | pale brown  | 40         | 1–3 |
| 8          | pale yellow | 45         |     |

**Table S18.** Summary of anionic exchange chromatography experiments

| CFRP (mg) | Recovered CFs (mg) | Isolated crude BPA (mg) | Crude BPA yield (%) | Initial hydroxide (g)                                 | Recovered hydroxide (mol) | Hydroxide recovery yield (mol %) |
|-----------|--------------------|-------------------------|---------------------|---|---------------------------|----------------------------------|
| 560       | 309                | 100                     | 81.2                | 5.99 g NaOH<br>+<br>9.00 g<br>KOH×0.5H <sub>2</sub> O | 0.263 mol                 | 91                               |



**Figure S31.** Extract from combined fractions 1-3 reveal no organic molecules in CD<sub>3</sub>OD.



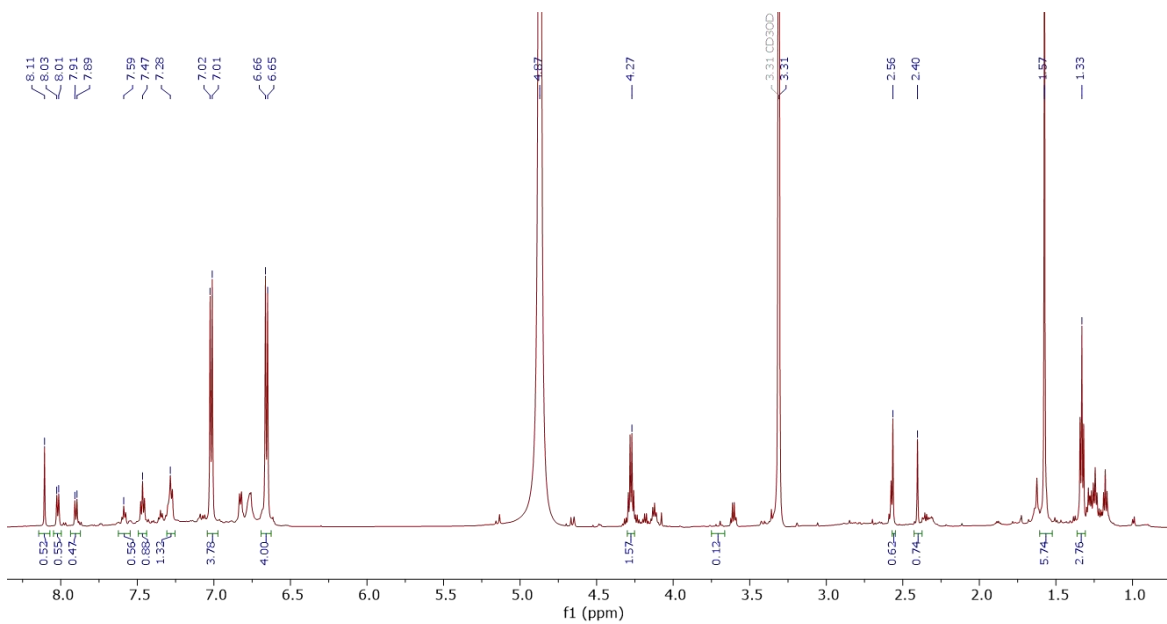


Figure S32.  $^1\text{H}$  NMR of crude combined fractions 5-8 in  $\text{CD}_3\text{OD}$ .

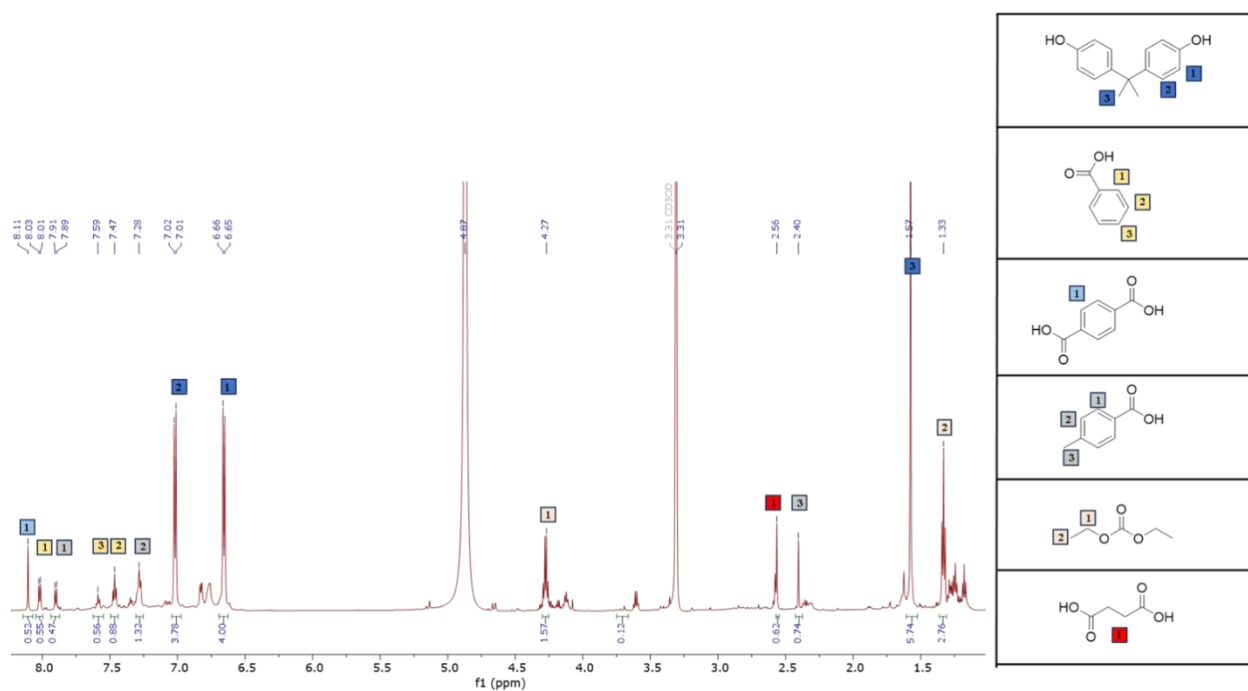
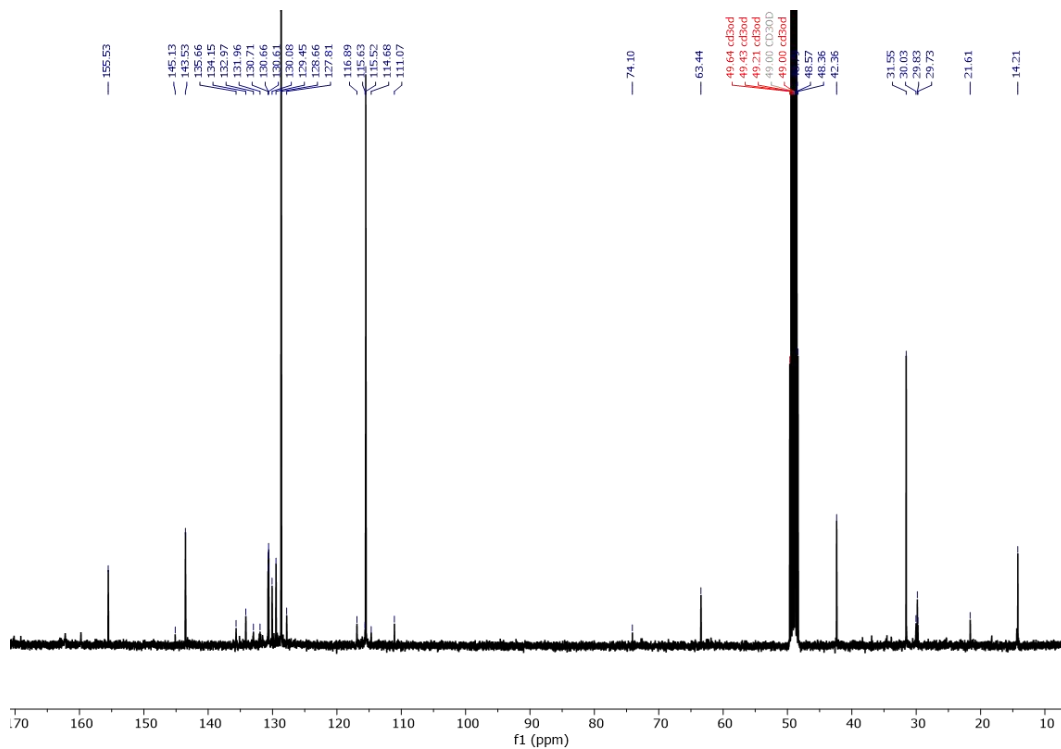
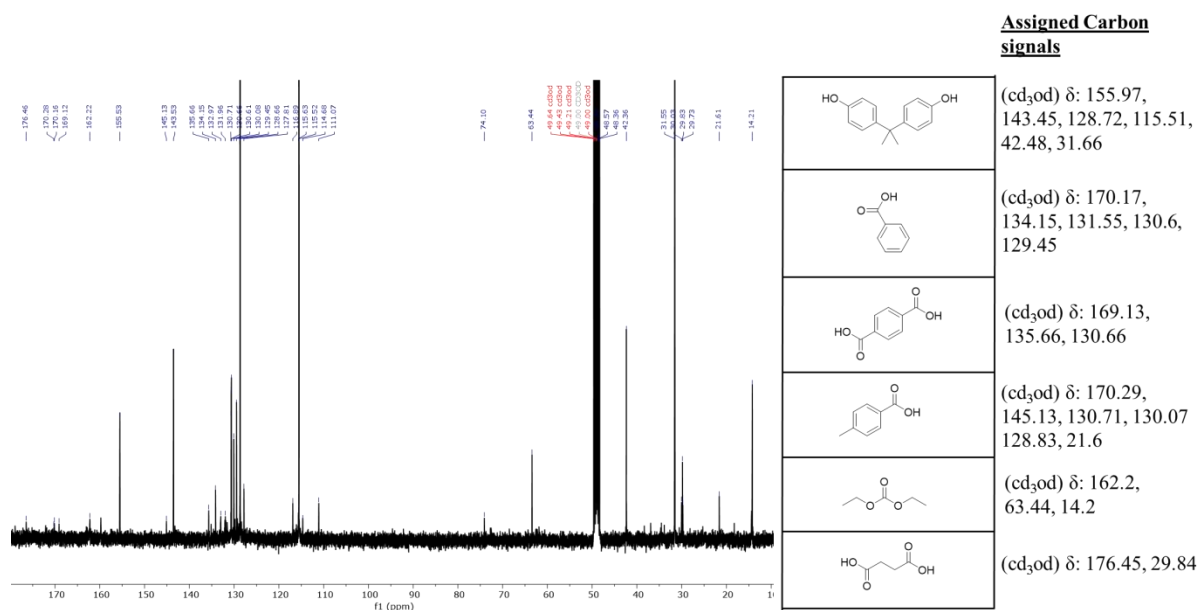


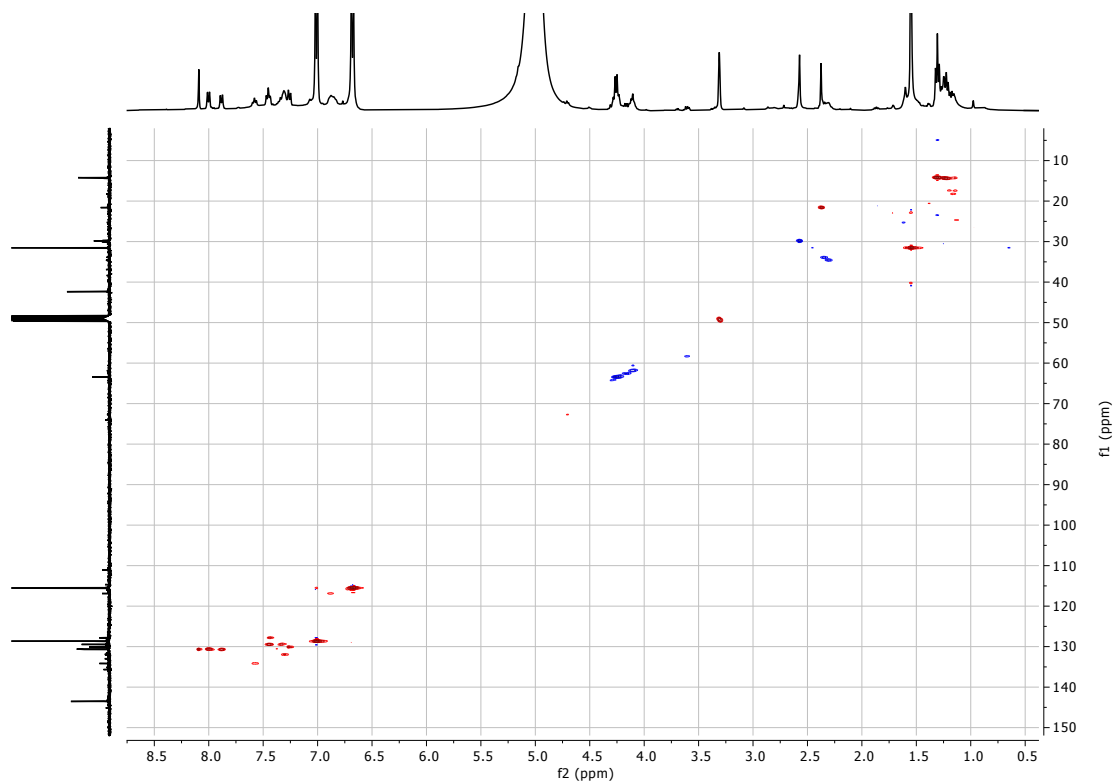
Figure S33. Annotated  $^1\text{H}$  NMR of crude combined fractions 5-8 in  $\text{CD}_3\text{OD}$ .



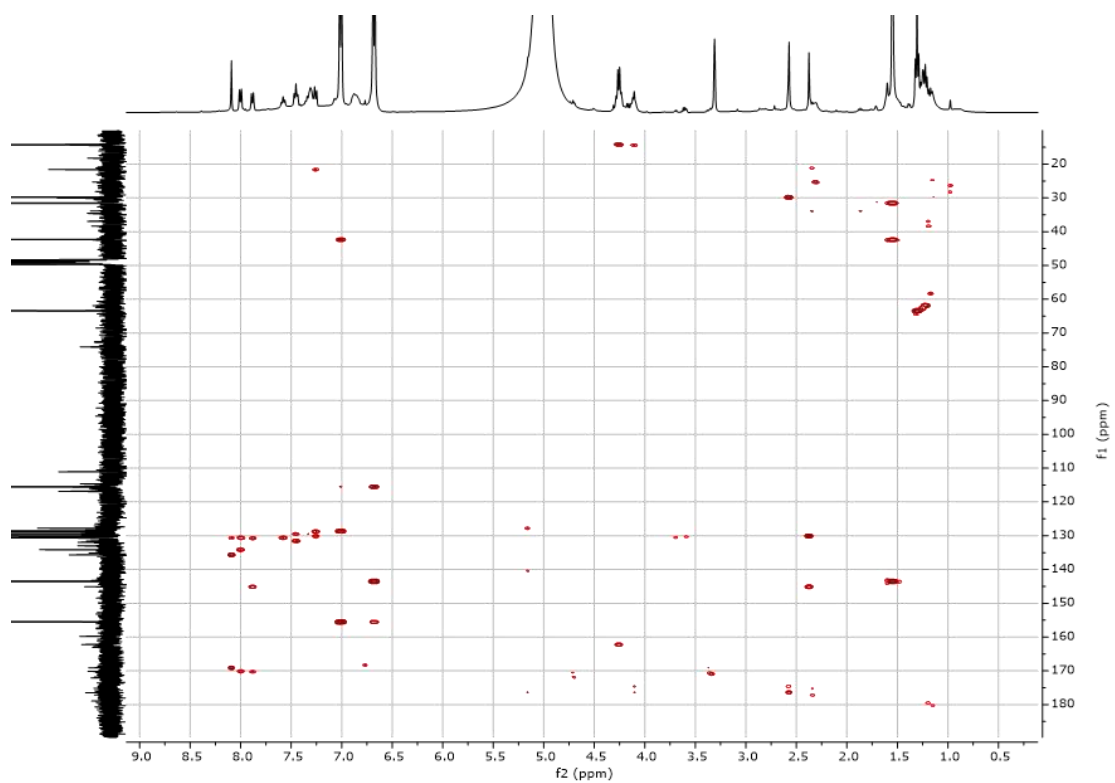
**Figure S34.**  $^{13}\text{C}\{^1\text{H}\}$  NMR of crude fractions 5-8 in  $\text{CD}_3\text{OD}$ .



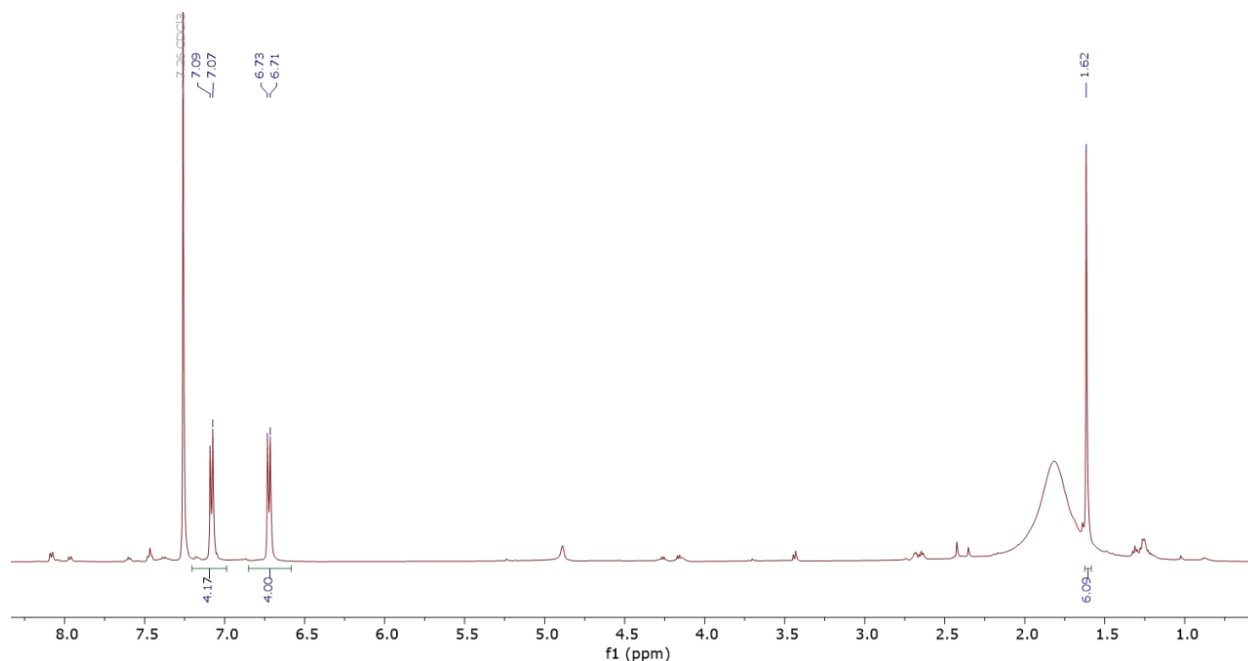
**Figure S35.** Annotated  $^{13}\text{C}\{^1\text{H}\}$  NMR of crude fractions 5-8 in  $\text{CD}_3\text{OD}$ .



**Figure S36.** HSQC of crude fractions 5-8 in CD<sub>3</sub>OD.



**Figure S37.** HMBC of crude fraction 5-8 in CD<sub>3</sub>OD.



**Figure S38.**  $^1\text{H}$  NMR of purified BPA from fractions 5-8 in  $\text{CDCl}_3$ .

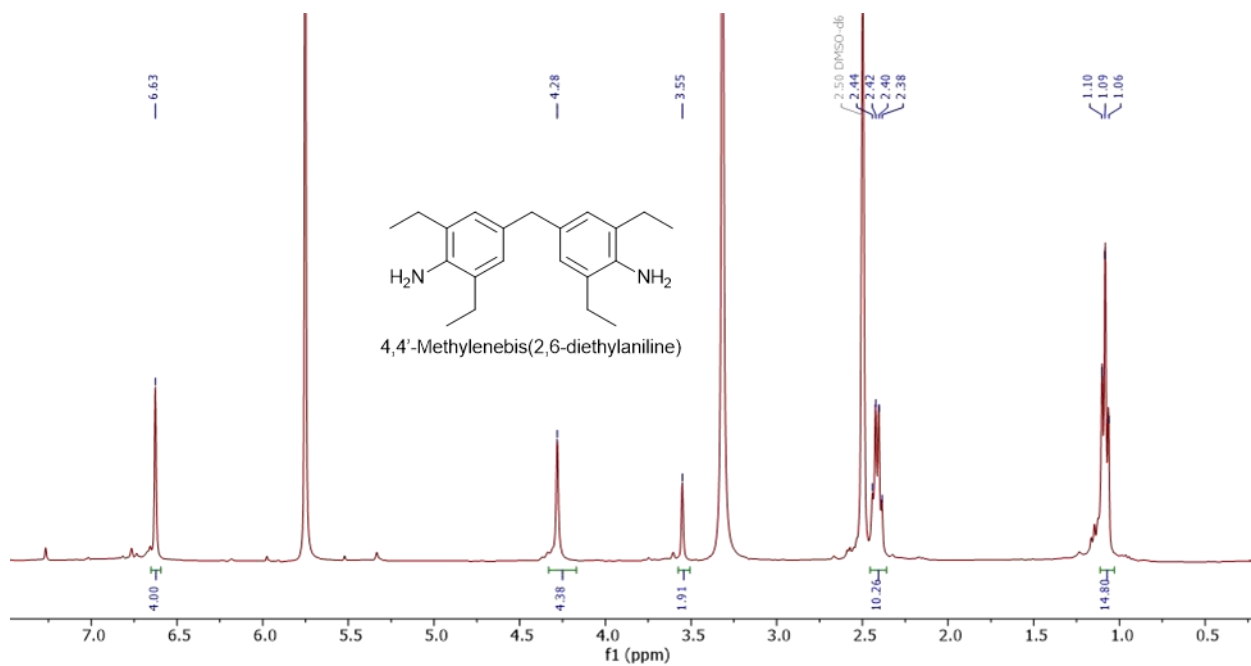
## 9. Bi/MDEA neat resin degradation component

After digestion and cooling, the reaction melt was dissolved using 15 mL DI water, forming a beige-brown basic solution. The stainless-steel lid and Hickman distillation apparatus were washed with 10 mL of DI water and dichloromethane (DCM). The wash solutions (DI water and DCM) were combined with the basic solution

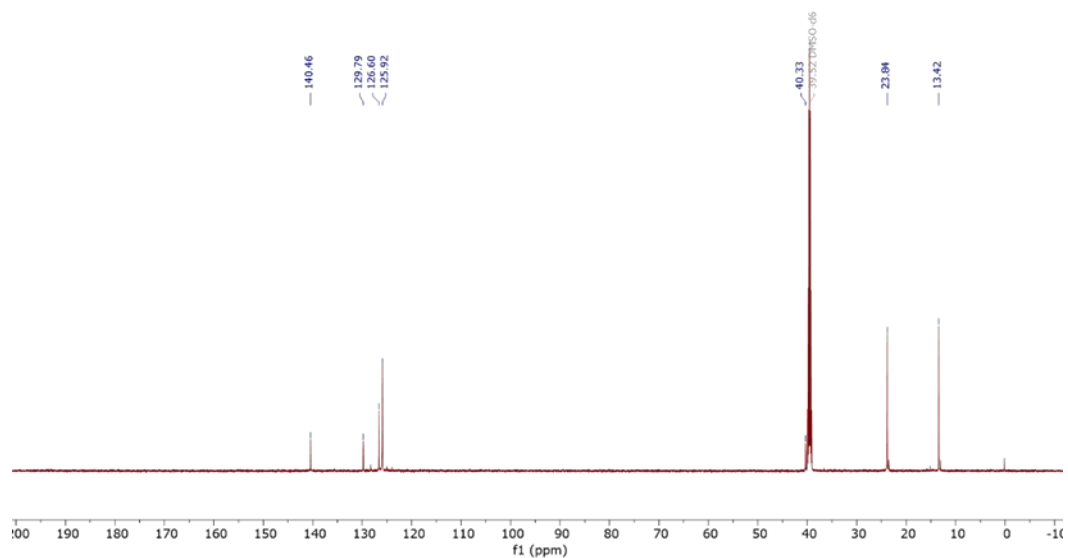
### a. Organic base extract from dichloromethane

The basic solution was extracted with DCM (15 mL x 3). The organic layer was washed using 1M NaOH aqueous solution (5 mL x 3), dried over sodium sulfate, and evaporated to give a black residue (recovered M-DEA). Isolated materials were characterized by  $^1\text{H}$ ,  $^{13}\text{C}$  NMR, and MALDI spectroscopy (Figure S39-S43).

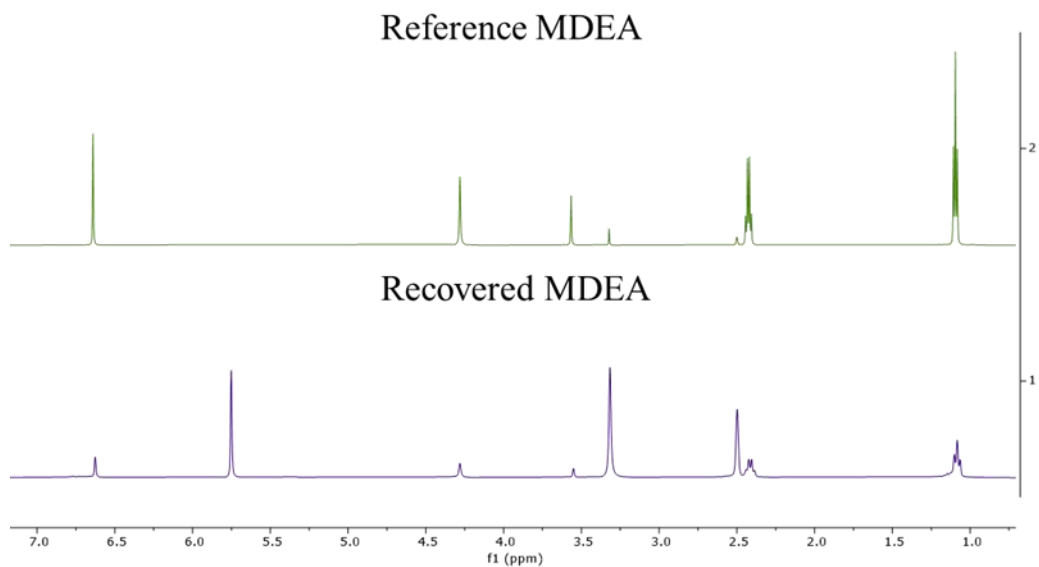
**Recovered 4,4'-Methylenebis(2,6-diethylaniline) (MDEA).**  $^1\text{H}$  NMR (400 MHz,  $\text{dms}_o$ )  $\delta$  6.63 (s, 4H), 4.28 (s), 3.55 (s, 2H), 2.49 – 2.32 (q, 10H), 1.08 (t,  $J = 7.5, 1.8$  Hz, 15H).  $^{13}\text{C}\{^1\text{H}\}$  NMR (151 MHz,  $\text{dms}_o$ )  $\delta$  140.46, 129.79, 126.60, 125.92, 40.33, 23.84, 13.42.



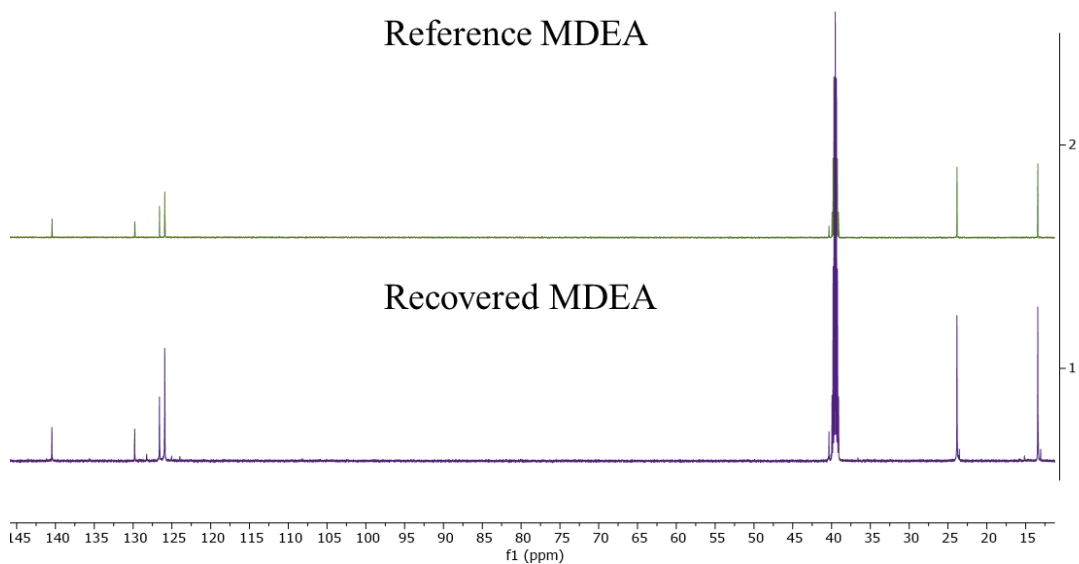
**Figure S39.**  $^1\text{H}$  NMR of recovered MDEA in  $\text{DMSO-}d_6$ .



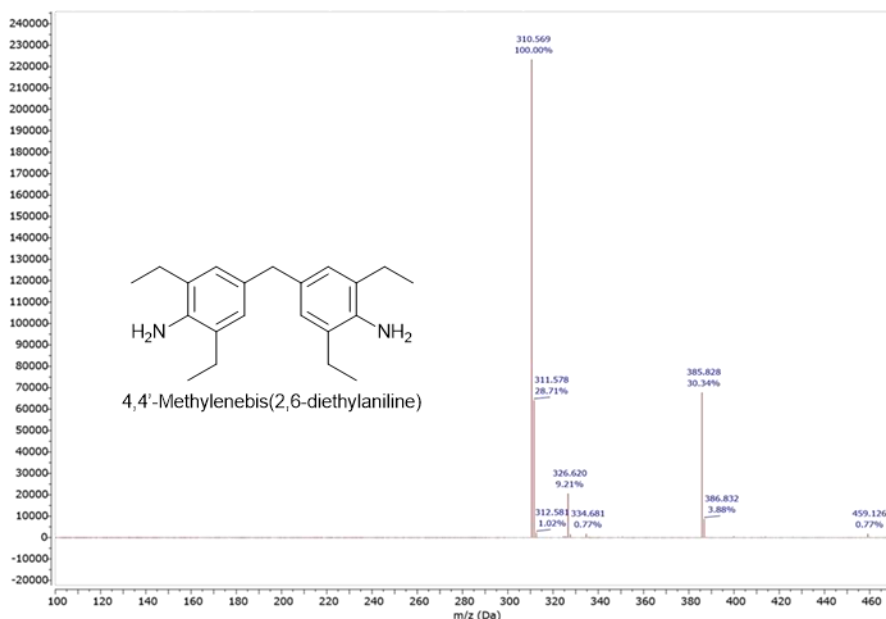
**Figure S40.**  $^{13}\text{C}\{^1\text{H}\}$  NMR of recovered MDEA in  $\text{DMSO-}d_6$ .



**Figure S41.** Stacked <sup>1</sup>H NMR of recovered MDEA and reference MDEA in DMSO-*d*<sub>6</sub>.



**Figure S42.** Stacked <sup>13</sup>C{<sup>1</sup>H} NMR of recovered MDEA and reference MDEA in DMSO-*d*<sub>6</sub>.

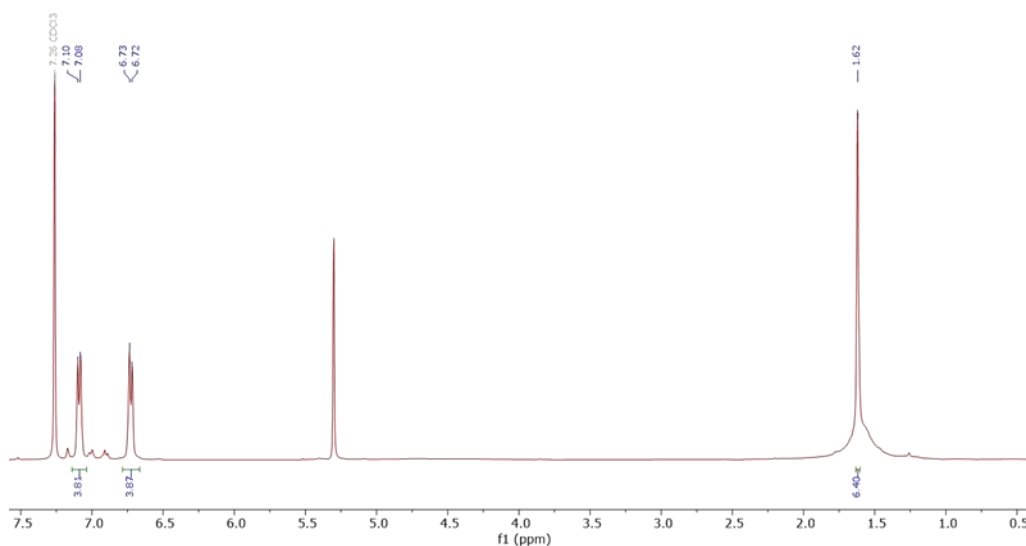


**Figure S43.** MALDI data of recovered MDEA, anthracene matrix.

#### b. Organic neutral extract from dichloromethane

Next, the aqueous phase was brought to pH 6 by adding concentrated HCl. After cooling, the neutral solution was extracted with DCM (15 mL x 3). The organic layer was cleaned using DI water (5 mL x 3), dried over sodium sulfate, and evaporated to give a yellowish solid (recovered BPA). Isolated materials were characterized by  $^1\text{H}$  NMR spectroscopy (Figure S44). A summary of the degradation component isolation is provided in Table S22.

**Recovered bisphenol-A (BPA).**  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  7.15 – 7.04 (m, 4H), 6.79 – 6.64 (m, 4H), 1.62 (s, 6H).

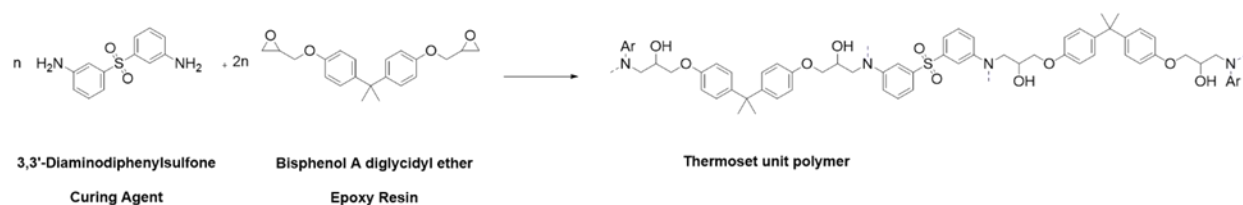


**Figure S44.**  $^1\text{H}$  NMR of recovered BPA from the MDEA-cured polymer degradation reaction in  $\text{CDCl}_3$ .

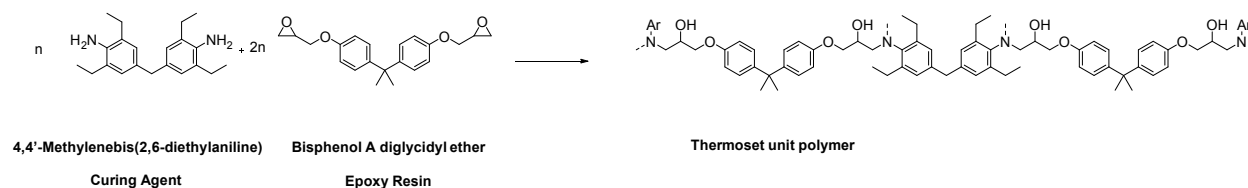
## 10. Reaction yields of degradation components

### a. Determination of unit polymer

To determine the moles of BPA recovered we first had to determine the number of moles of BPA in a given mass of polymer. Based on the formulation of the resin (SI section 1a), it is comprised of 2 molar equivalents of the epoxy monomer DGEBA and 1 molar equivalent of the curing agent 3,3'-DDS. Therefore, we can define the smallest unit of the polymer as (1 eq. 3,3'-DDS + 2 eq. DGEBA). The molecular weight of the Bi/DDS unit polymer is 928 g/mol ( $C_{54}H_{60}N_2O_{10}S$ ). With the same amine to epoxy mixing ratio, we assume that Bi/MDEA formulation has the same molar ratio between epoxy monomer DGEBA and curing agent MDEA. The molecular weight of the Bi/MDEA unit polymer is 990 g/mol ( $C_{63}H_{78}N_2O_8$ ).



**Figure S45.** Portrayal of the thermoset unit polymer ( $C_{54}H_{60}N_2O_{10}S$ ). The unit polymer has a molecular weight of 928 g/mol.



**Figure S46.** Portrayal of the thermoset unit polymer ( $C_{63}H_{78}N_2O_8$ ). The unit polymer has a molecular weight of 990 g/mol.

### b. Verification of proposed unit polymer using CHNS elemental analysis

To further verify that the proposed unit polymer is the smallest representative unit of this thermoset system we did an experiment by analyzing the polymer in the CHNS elemental analyzer (Flash 2000 Elemental Analyzer (CHNS)) and comparing its elemental percentages to the proposed unit polymer. The results are shown in Table S19.

The elemental percentages of the resin nearly match the elemental percentages of the proposed unit polymer. Therefore, the proposed unit polymer is an appropriate chemical formula to use to represent the polymer.



**Table S19.** CHNS elemental analysis results of the resin polymer. The elemental percentages are compared against the elemental percentages of the proposed unit polymer.

|  | C (%) | H (%) | N (%) | S (%) |
|--|-------|-------|-------|-------|
| C <sub>54</sub> H <sub>60</sub> N <sub>2</sub> O <sub>10</sub> S | 69.81 | 6.57  | 3.02  | 3.45  |
| Bi/DDS matrix  | 70.86 | 6.63  | 2.89  | 2.56  |

**c. Reaction yields of degradation components from Bi/DDS sample digestion**

Mass of degraded matrix of CFRP digestion:

$$\begin{aligned} & \text{Mass of degraded matrix of CFRP digestions} \\ & = \text{Mass of CFRP} - \text{Mass of recovered CFs} \end{aligned}$$

Moles of DGEBA in degraded matrix:

$$\text{Mol of DGEBA in degraded matrix} = \frac{\text{Mass of degraded matrix}}{(\text{Polymer Unit MW})} \cdot 2$$

Moles of 3,3'-DDS in degraded matrix:

$$\text{Mol of 3,3' DDS in degraded matrix} = \frac{\text{Mass of degraded matrix}}{(\text{Polymer Unit MW})}$$

Yield of BPA:

$$\text{BPA Yield} = \frac{\text{Mol of recovered BPA}}{\text{Mol of DGEBA in degraded matrix}} \cdot 100\%$$

Yield of aniline or 3-aminophenol:

$$\begin{aligned} & \text{Aniline or 3 aminophenol Yield} \\ & = \frac{\text{Mol of recovered aniline or 3 aminophenol}}{(\text{Mol of 3,3' DDS in degraded matrix}) \cdot 2} \cdot 100\% \end{aligned}$$

The following tables display the yield of isolated BPA from pre-treated and untreated Bi/DDS sample digestion experiments:

**Table S20.** BPA yield summary for pre-treated Bi/DDS CFRP digestion.

| Temperature (°C) | Composite initial mass (g) | Recovered CFs (g) | Isolated BPA (g) | BPA yield (%) |
|------------------|----------------------------|-------------------|------------------|---------------|
| 220              | 0.5739                     | 0.3514            | 0.0939           | 86            |

**Table S21.** Products isolation summary of unpretreated Bi/DDS neat resin and CFRP digestions.

| Entry | Mass change (g, wt%) | Extracted products (mg, yield%) |               |                    |  |
|-------|----------------------|---------------------------------|---------------|--------------------|--|
|       |                      | Distillate (aniline)            | Basic extract | Acid extract (BPA) | Neutral extract (C <sub>6</sub> H <sub>7</sub> NO) |
| 1     | 1.631 (82%)          | 46 (14%)                        | 27            | 626 (78%)          |  |
| 2     | 1.970 (99%)          | 75 (19%)                        |               |                    |  |
| 3     | 0.475 (95%)          |                                 | 15            | 213 (92%)          | 41 (<37%)  |
| 4     | 0.700 (23%)          |                                 | 13            | 343 (>99%)         | 43 (<26%)  |
| 5     | 0.720 (36%)          | 42 (29%)                        |               |                    |  |

**d. Reaction yields of degradation components from Bi/MDEA sample digestion**Moles of DGEBA in degraded matrix:

$$\text{Mol of DGEBA} = \frac{\text{Mass of degraded matrix}}{(\text{Polymer Unit MW})} \cdot 2$$

Moles of MDEA in degraded matrix:

$$\text{Mol of MDEA} = \frac{\text{Mass of degraded matrix}}{(\text{Polymer Unit MW})}$$

Yield of BPA:

$$\text{BPA Yield} = \frac{\text{Mol of recovered BPA}}{\text{Mol of DGEBA in degraded matrix}} \cdot 100\%$$

Yield of MDEA:

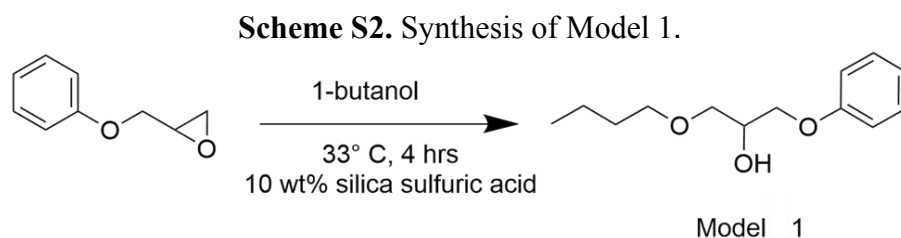
$$\text{MDEA Yield} = \frac{\text{Mol of recovered MDEA}}{\text{Mol of MDEA in degraded matrix}} \cdot 100\%$$

**Table S22.** Products isolation summary of Bi/MDEA neat resin and CFRP digestions.

| Temperature (°C) | Sample mass (g) | Isolated crude MDEA (g) | MDEA yield (%) | Isolated BPA (g) | BPA yield (%) |
|------------------|-----------------|-------------------------|----------------|------------------|---------------|
| 260              | 0.2018          | 0.0374                  | 59             | 0.0501           | 54            |

## 11. Model 1 synthesis and degradation reactions

### a. Synthesis of Model 1

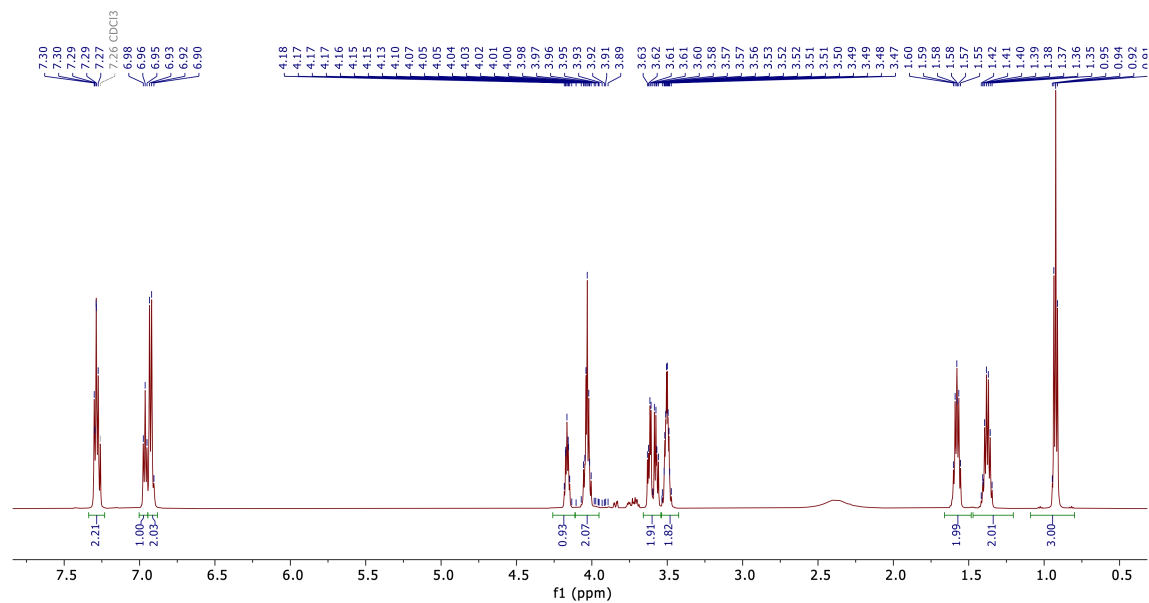


In a 100 mL round bottom flask 30 mL (24.3 g, 0.328 mol, 16.4 eq.) of 1-butanol was combined with 0.34 g silica sulfuric acid. Then 3.1 mL (3.45 g, 23 mmol) of the 2-(phenoxymethyl)oxirane was slowly added. Once all combined, the flask was placed in an oil bath and heated to 33°C for four hours.

After four hours have passed, 50 mL of dichloromethane was added. Then the reaction mixture was transferred to a 250 mL separatory funnel and washed with DI H<sub>2</sub>O (3 x 50 mL). The organic layer was collected, and the solvent was removed under reduced pressure. The oily liquid was then dry loaded on silica and the product was isolated by flash chromatography (15:85 Ethyl Acetate: Hexanes).

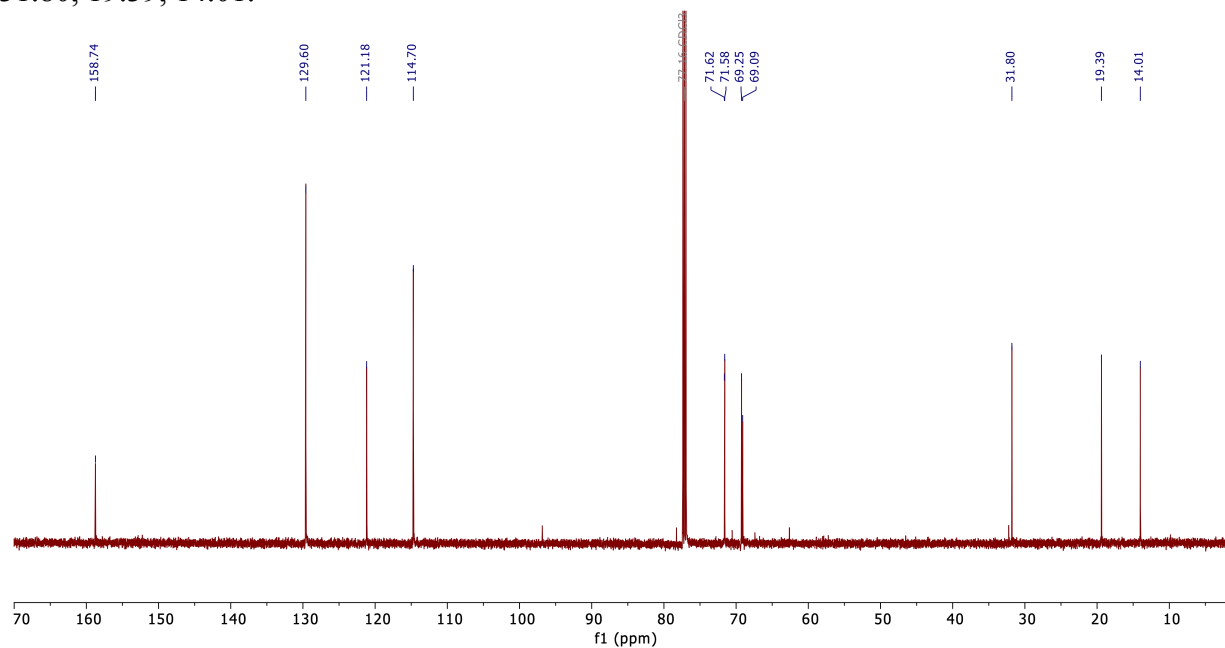
The <sup>1</sup>H and <sup>13</sup>C NMR spectra can be found in Figures S47 and S48. Literature reports of this compound exist and confirm we selectively synthesized the desired regioisomer (Yield = 63%)

<sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 7.35 – 7.22 (m, 2H), 6.95 (dd, J = 25.3, 7.8 Hz, 3H), 4.30 – 4.12 (m, 1H), 4.10 – 3.98 (m, 2H), 3.66 – 3.54 (m, 2H), 3.50 (tt, J = 6.6, 3.1 Hz, 2H), 1.66 – 1.46 (m, 2H), 1.38 (h, J = 7.4 Hz, 2H), 0.92 (t, J = 7.3 Hz, 3H).



**Figure S47.**  $^1\text{H}$ NMR of Model 1 in  $\text{CDCl}_3$

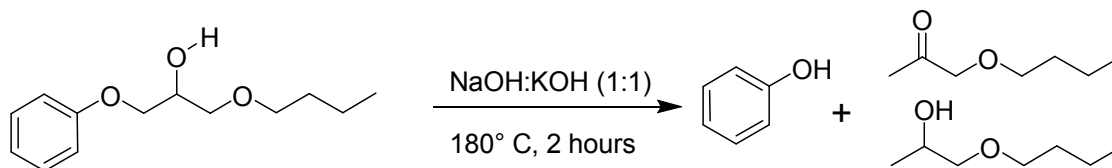
$^{13}\text{C}$  NMR (151 MHz,  $\text{CDCl}_3$ ):  $\delta$  158.74, 129.60, 121.18, 114.70, 71.62, 71.58, 69.25, 69.09, 31.80, 19.39, 14.01.



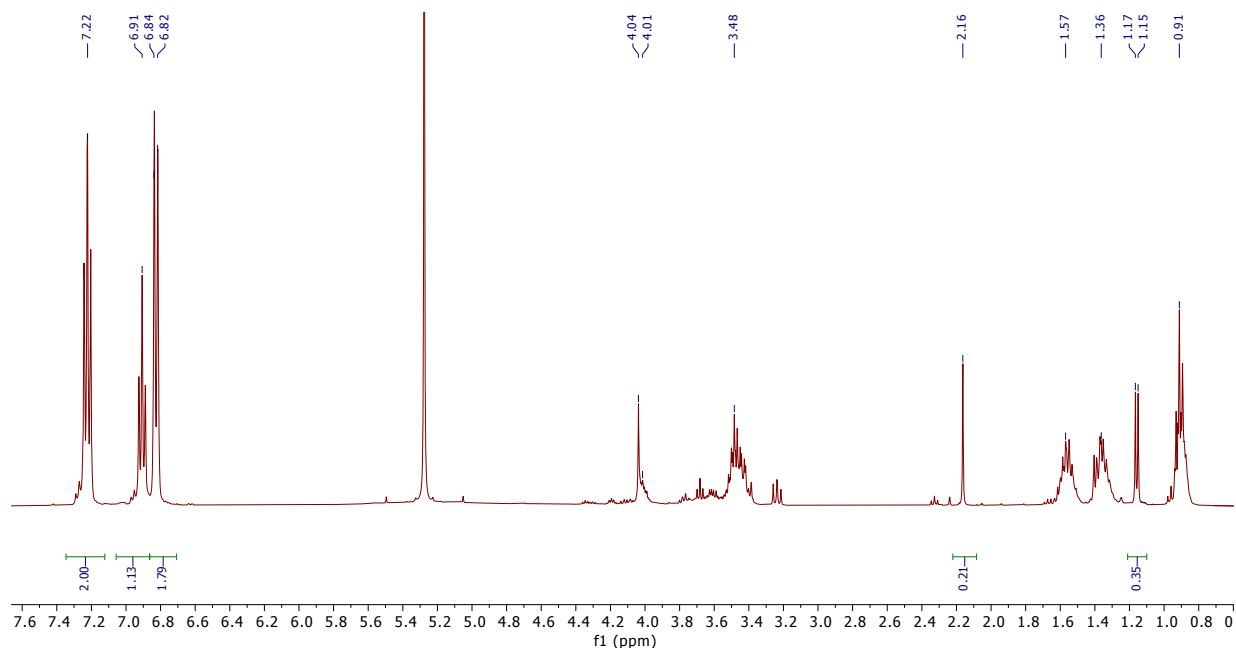
**Figure S48.**  $^{13}\text{C}\{^1\text{H}\}$  NMR of isolated Model 1 in  $\text{CDCl}_3$

**b. Model 1 degradation reaction at 180 °C**

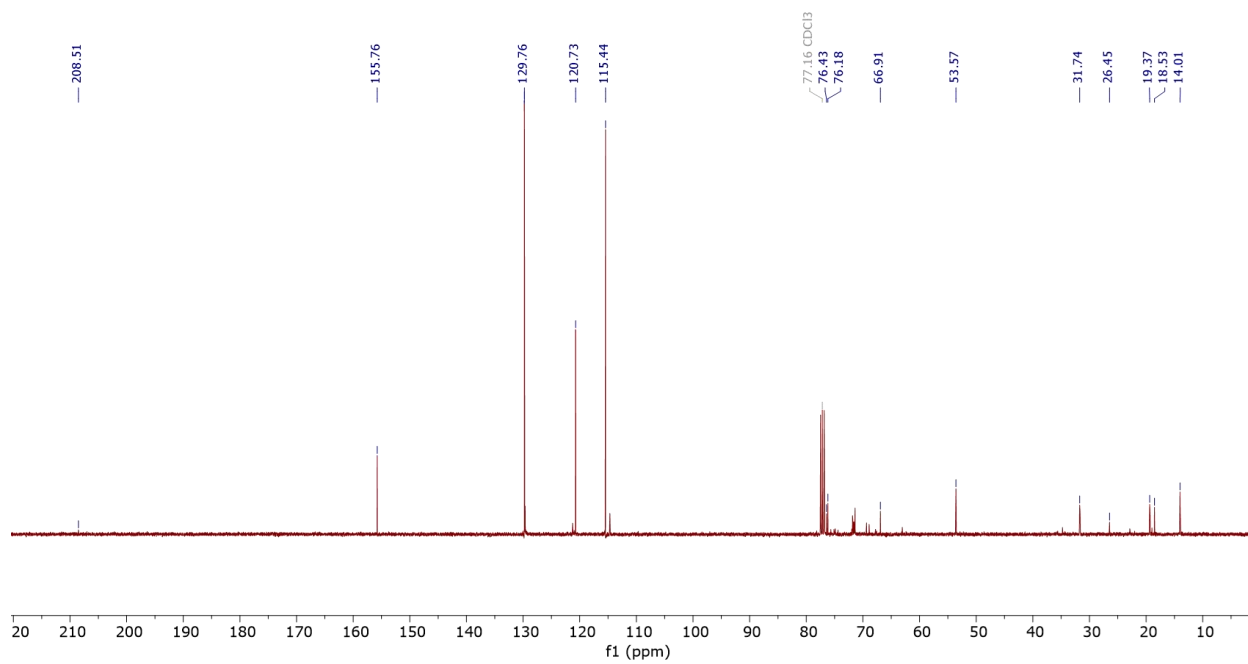
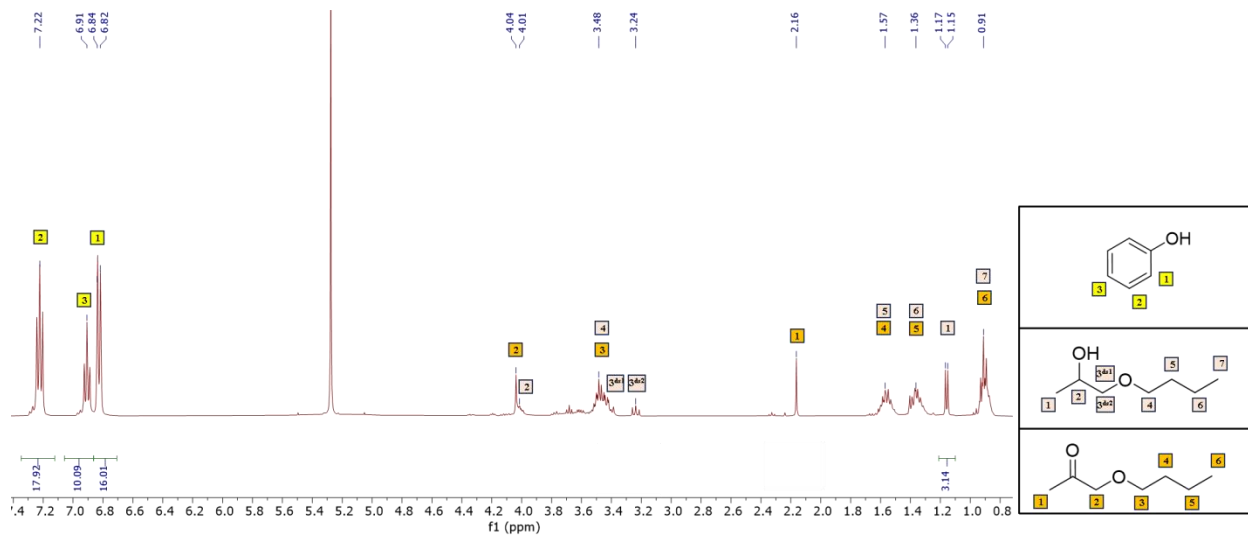
**Scheme S3.** Degradation reaction of Model 1 at 180 °C.

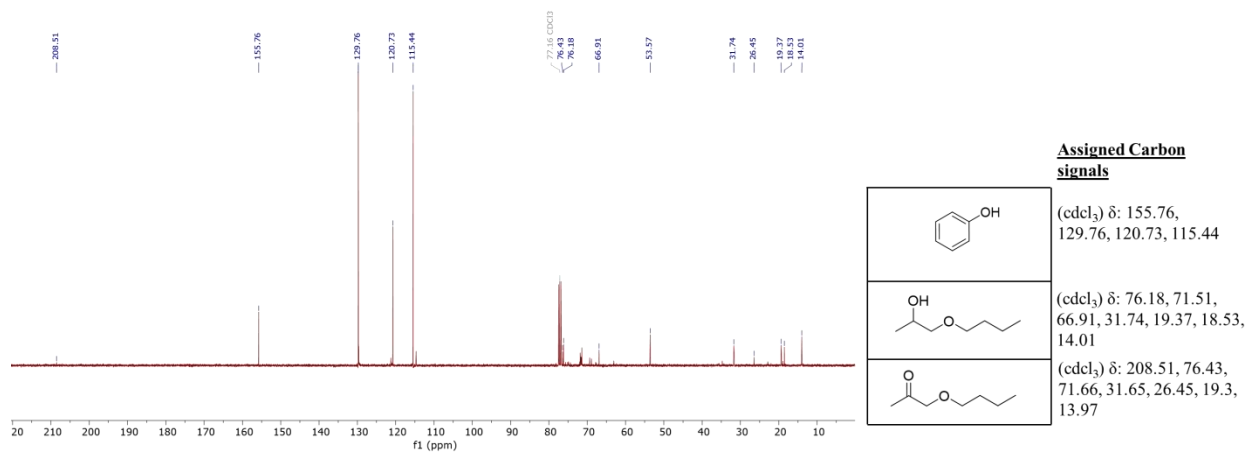


Finely ground NaOH (0.1 g, 2.5 mmol) and KOH (0.14 g, 2.5 mmol) were added to a 50 mL Schlenk flask. Model 1 (0.18 g, 0.8 mmol) was then transferred into the flask. Finally, the flask was flushed with nitrogen gas and placed in an oil bath set to 180 °C for two hours. After the reaction, aqueous sodium bicarbonate solution (saturated, 50 mL) was added to the Schlenk flask to dissolve the solid residue. The aqueous mixture was then transferred to a 250 mL separatory funnel. Dichloromethane (50 mL) was then added to the Schlenk flask and then transferred to the separatory funnel, extracted, and collected in a 250 mL round bottom flask. This was repeated twice. The organic layers were then combined, and the solvent was removed under reduced pressure. The  $^1\text{H}$ ,  $^{13}\text{C}$ , HMBC and HSQC spectra can be found in Figures S49-56 respectively.

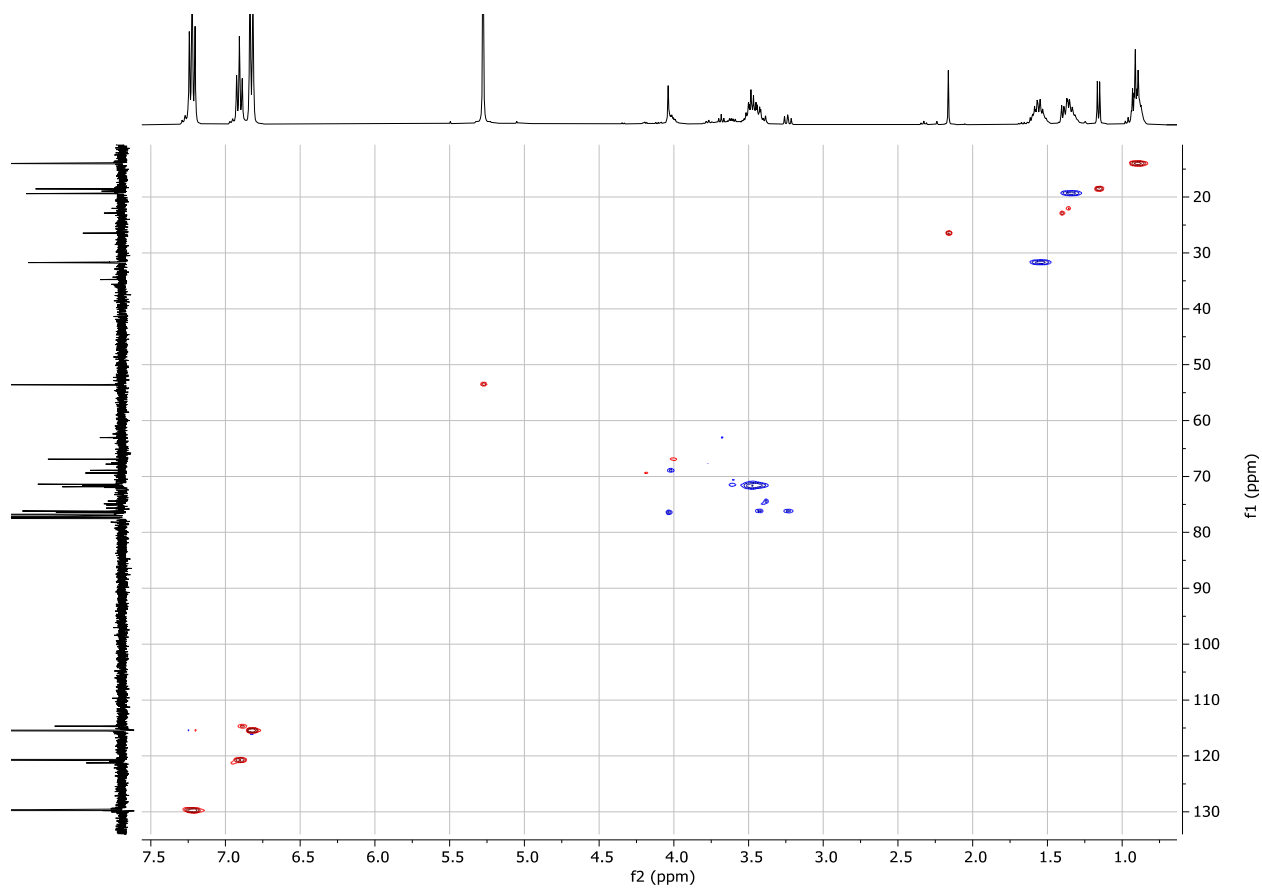


**Figure S49.**  $^1\text{H}$ NMR of 180 °C Model 1 degradation reaction extract in  $\text{CDCl}_3$ .

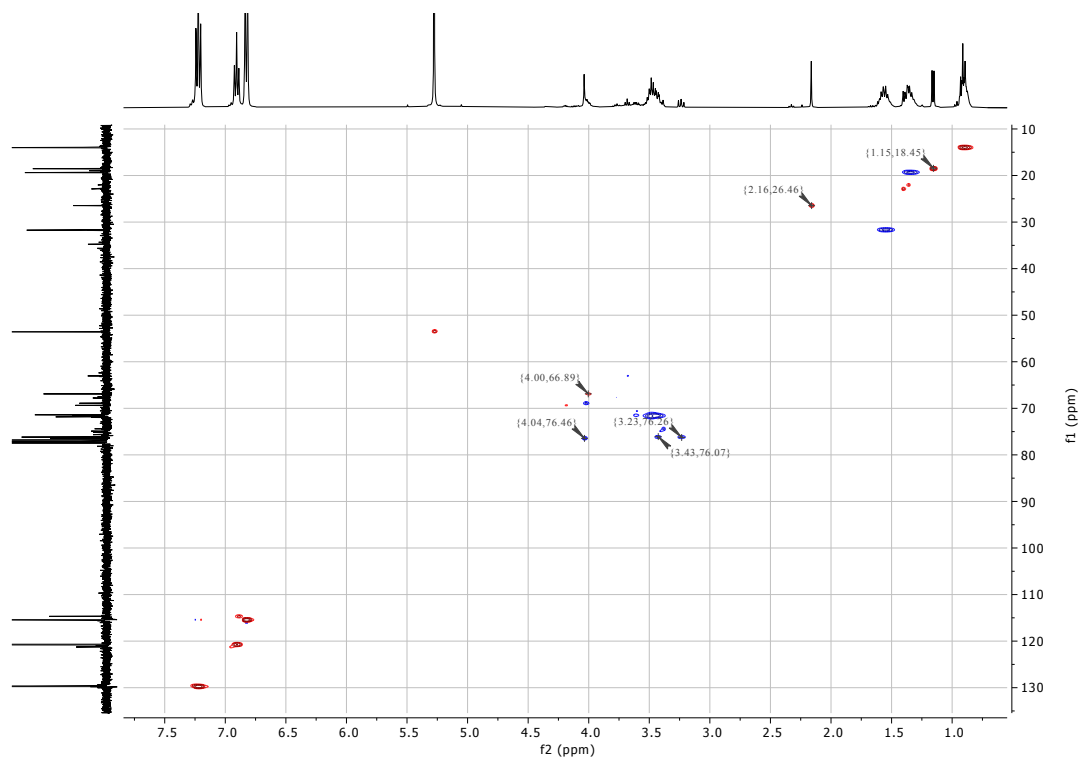




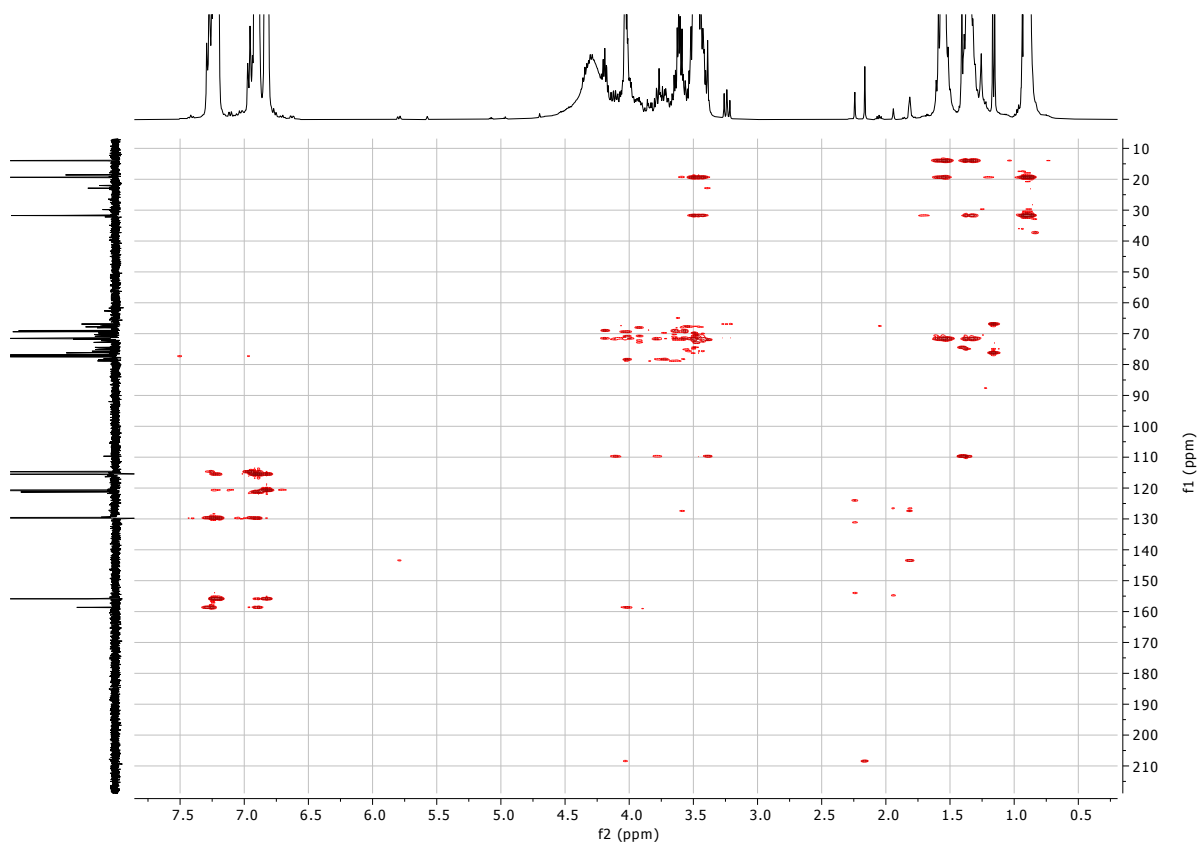
**Figure S52.** Annotated  $^{13}\text{C}\{^1\text{H}\}$  NMR of Model 1 180°C degradation reaction extract in  $\text{CDCl}_3$ .



**Figure S53.** HSQC of Model 1 180°C degradation reaction extract in  $\text{CDCl}_3$ .

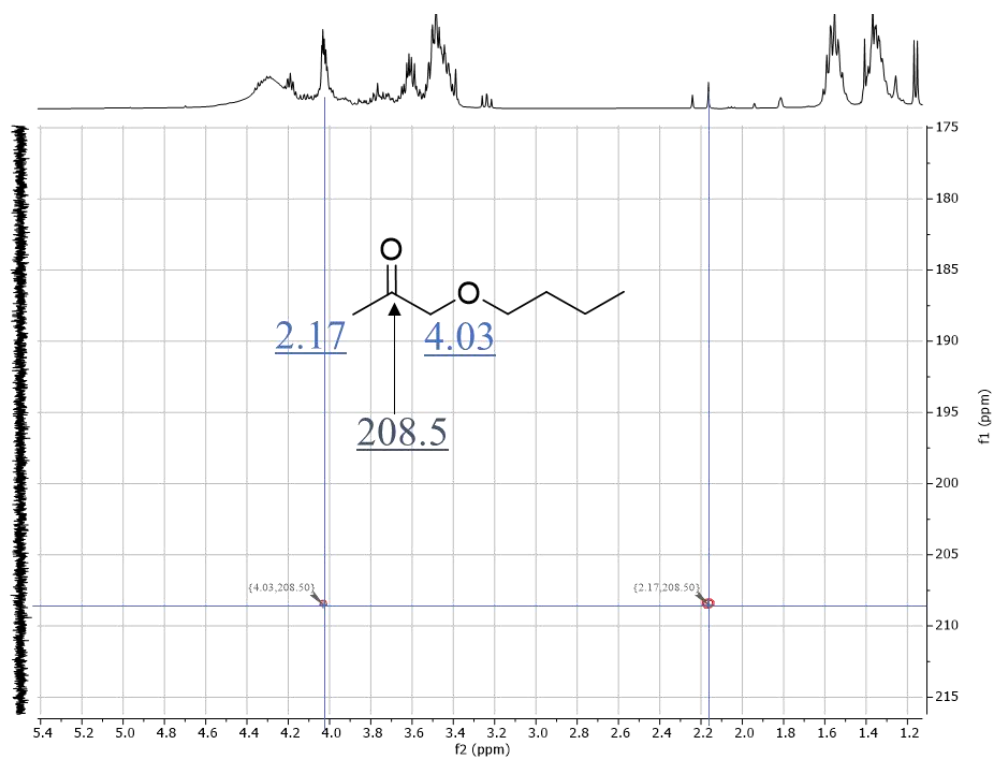


**Figure S54.** Annotated HSQC NMR of Model 1 180 °C degradation reaction extract in CDCl<sub>3</sub>.



**Figure S55.** HMBC of Model 1 180 °C degradation reaction extract in CDCl<sub>3</sub>.

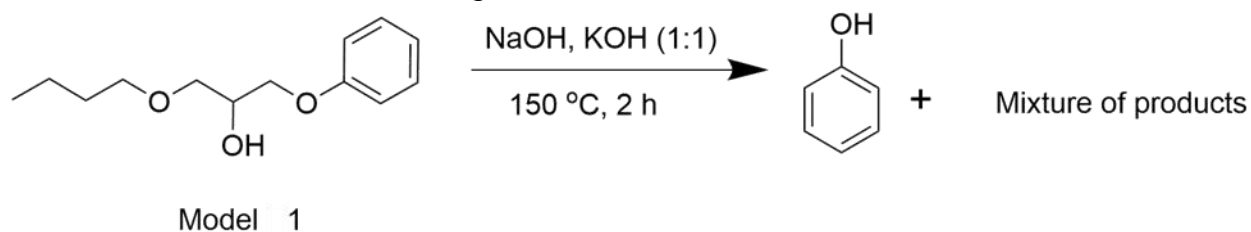




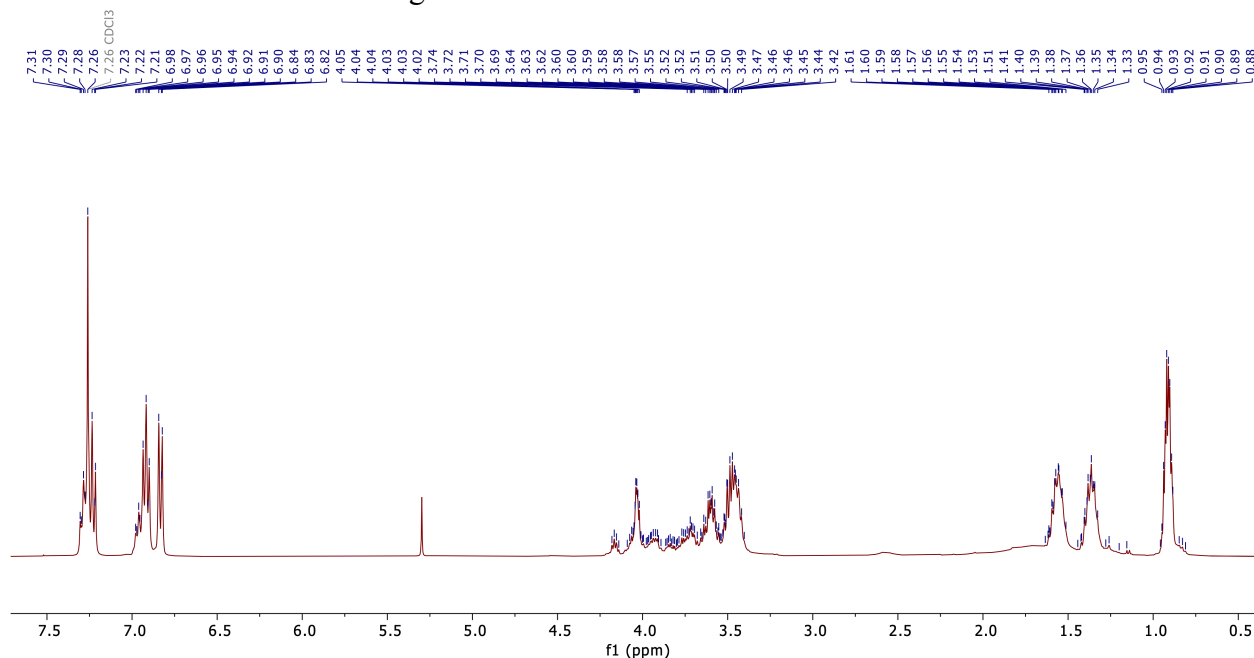
**Figure S56.** Annotated HMBC NMR of Model 1 180°C degradation reaction extract in CDCl<sub>3</sub>.

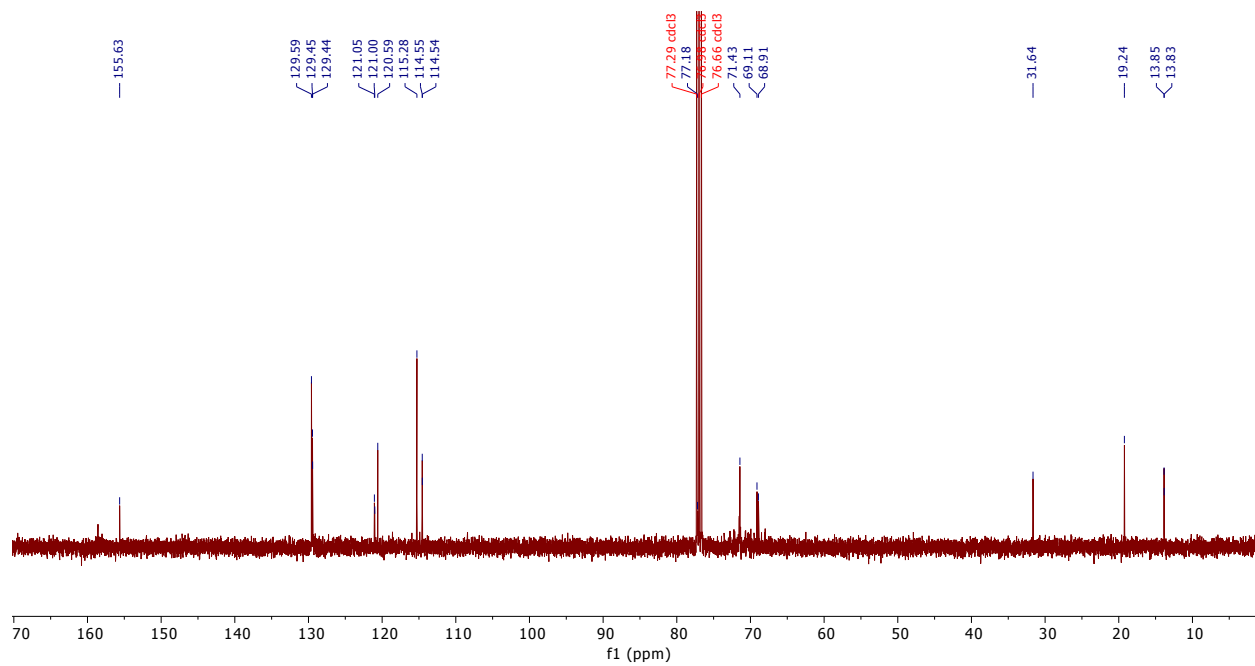
c. Model 1 Degradation Reaction at 150 °C

Scheme S4. Degradation reaction of Model 1 at 150 °C.



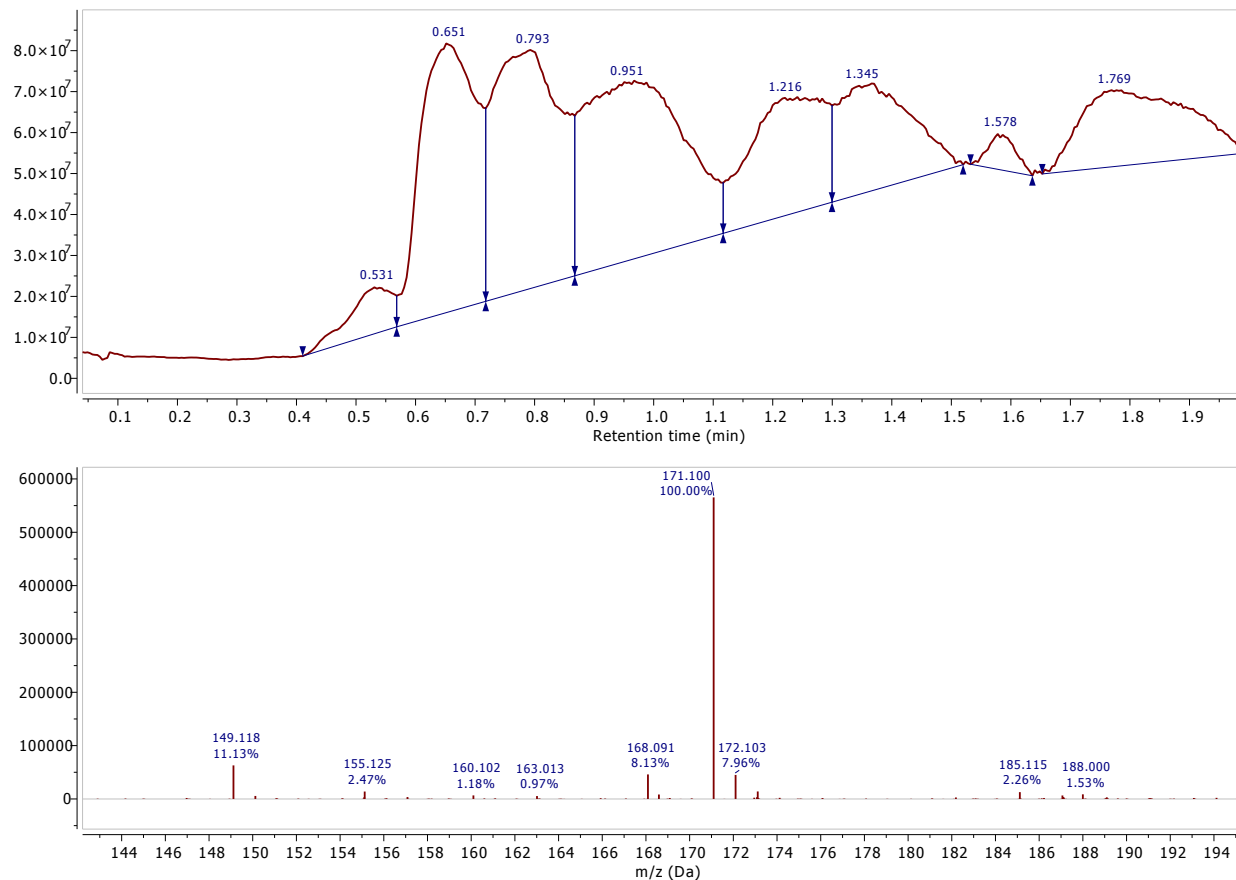
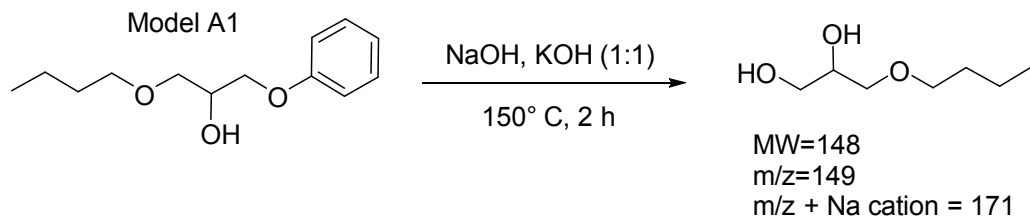
Finely grounded NaOH (0.1 g, 2.5 mmol) and KOH (0.14 g, 2.5 mmol) were added to a 50 mL Schlenk flask. Then Model 1 (0.18 g, 0.8 mmol) was transferred into the flask. Finally, the flask was carefully flushed nitrogen gas and placed in an oil bath set to 150 °C for two hours. After the reaction, aqueous sodium bicarbonate (saturated, 50 mL) was added to the Schlenk flask to dissolve the solid residue. The aqueous mixture was then transferred to a 250 mL separatory funnel. Dichloromethane (50 mL) was then added to the Schlenk flask and then transferred to the separatory funnel, extracted, and collected in a 250 mL round bottom flask. This was repeated twice. The organic layers were then combined, and the solvent was removed under reduced pressure. The  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra can be found in Figures S57-58. The LC/MS data can be found in Figure S59.





**Figure S58.**  $^{13}\text{C}\{^1\text{H}\}$  of Model 1 150°C degradation reaction extract in  $\text{CDCl}_3$ .

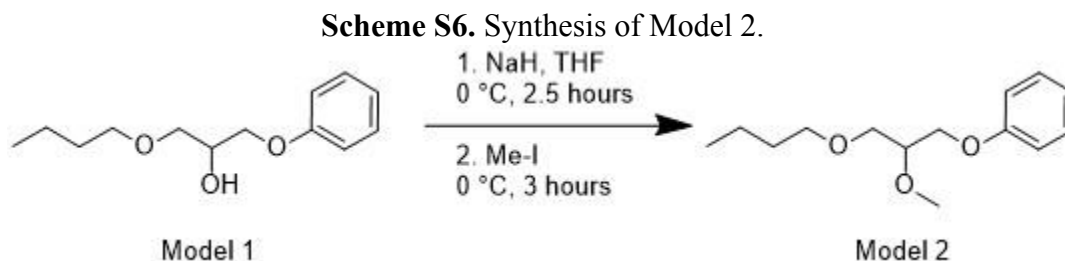
**Scheme S5.** Degradation of Model 1 at 150 °C examined on LC-QTOF.



**Figure S59.** LC-QTOF of Model 1 150 °C degradation reaction extract at retention time = 0.49 min.

## 12. Model 2 synthesis and degradation reactions

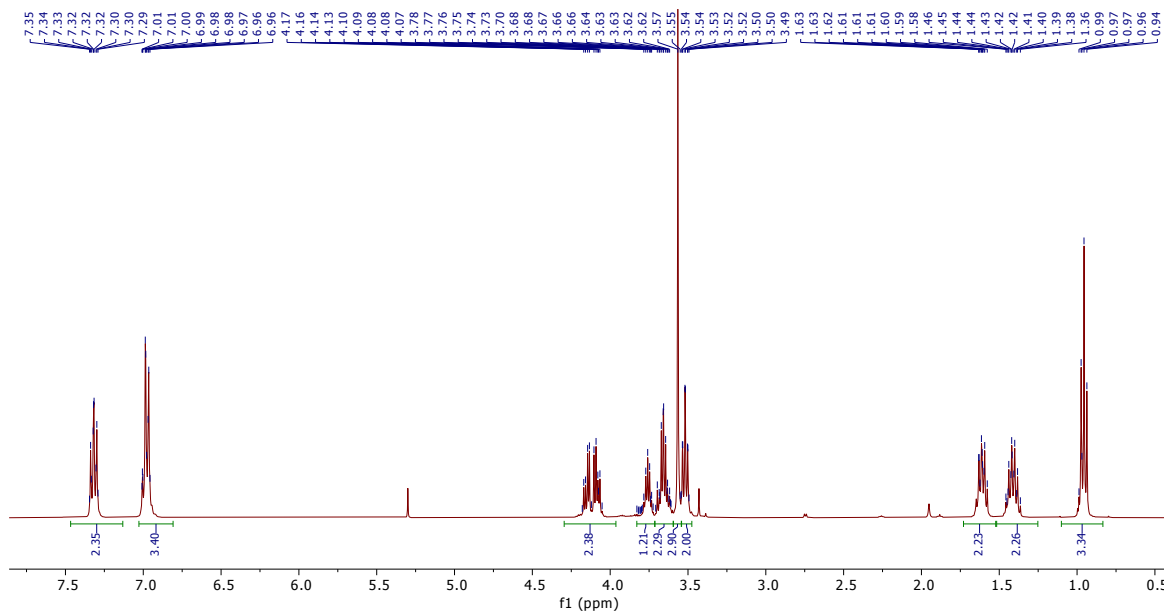
### a. Synthesis of Model 2



To a 250 mL round bottom flask was added tetrahydrofuran (THF, 80 mL, 71 g, 985 mmol), and the flask was placed in a 0 °C ice bath. Once cool, NaH (420 mg, 17.5 mmol, 1.56 eq.) was added to the flask to create a suspension. The ice bath was then moved over a stirrer and a magnetic stir bar was added inside the flask. Over the course of 15 minutes, Model 1 (2.5 g, 11.2 mmol, 1eq.) was added dropwise to the flask. After Model 1 was added, the vessel was flushed with nitrogen and a glass stopper was placed above the flask (without completely closing it) and the suspension was allowed to stir for 2.5 hours.

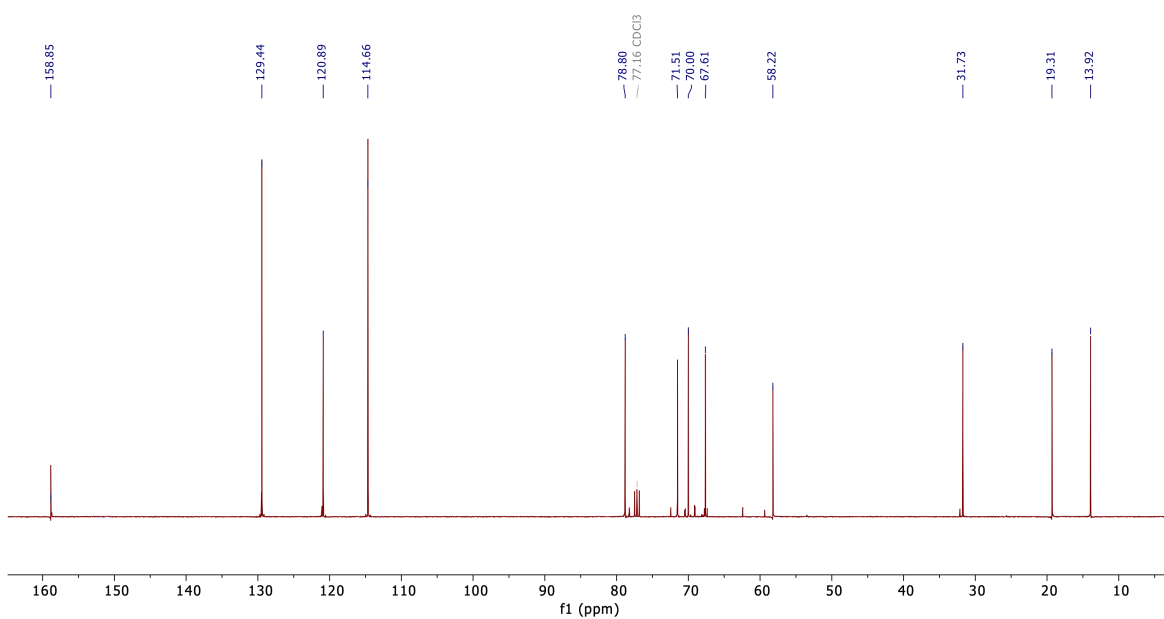
When 2.5 hours had elapsed, methyl iodide (1.88 mL, 4.29 g, 30.2 mmol, 2.7 eq.) was added into the flask and it was allowed to continue stirring for 3 hours. Thereafter, volatiles were removed under reduced pressure and the remaining contents of the flask were dissolved in H<sub>2</sub>O (deionized, 50 mL) and transferred to a separatory funnel. Then the liquid mixture was extracted with DCM (3 x 50 mL).

The organic layers were combined, and the solvent was removed under reduced pressure. The resulting oily liquid was then dry loaded on silica and the product was isolated by flash chromatography (15:85 ethyl acetate: hexanes). The <sup>1</sup>H NMR, <sup>13</sup>C NMR, FT-IR, and LC-QTOF spectra can be found in Figures S60-63.



**Figure S60.**  $^1\text{H}$ NMR of Model 2 in  $\text{CDCl}_3$ .

$^1\text{H}$  NMR (400 MHz,  $\text{cdcl}_3$ )  $\delta$  7.40 – 7.24 (m, 2H), 6.99 (td,  $J = 7.6, 1.4$  Hz, 3H), 4.22 – 4.03 (m, 2H), 3.76 (p,  $J = 5.0$  Hz, 1H), 3.71 – 3.60 (m, 2H), 3.57 (s, 3H), 3.52 (td,  $J = 6.6, 1.0$  Hz, 2H), 1.73 – 1.51 (m, 2H), 1.50 – 1.32 (m, 2H), 0.96 (t,  $J = 7.4$  Hz, 3H).



**Figure S61.**  $^{13}\text{C}$ NMR of Model 2 in  $\text{CDCl}_3$ .

$^{13}\text{C}$  NMR (100 MHz,  $\text{cdcl}_3$ )  $\delta$  158.86, 129.44, 120.89, 114.66, 78.80, 71.51, 70.00, 67.61, 58.22, 31.73, 19.31, 13.92.

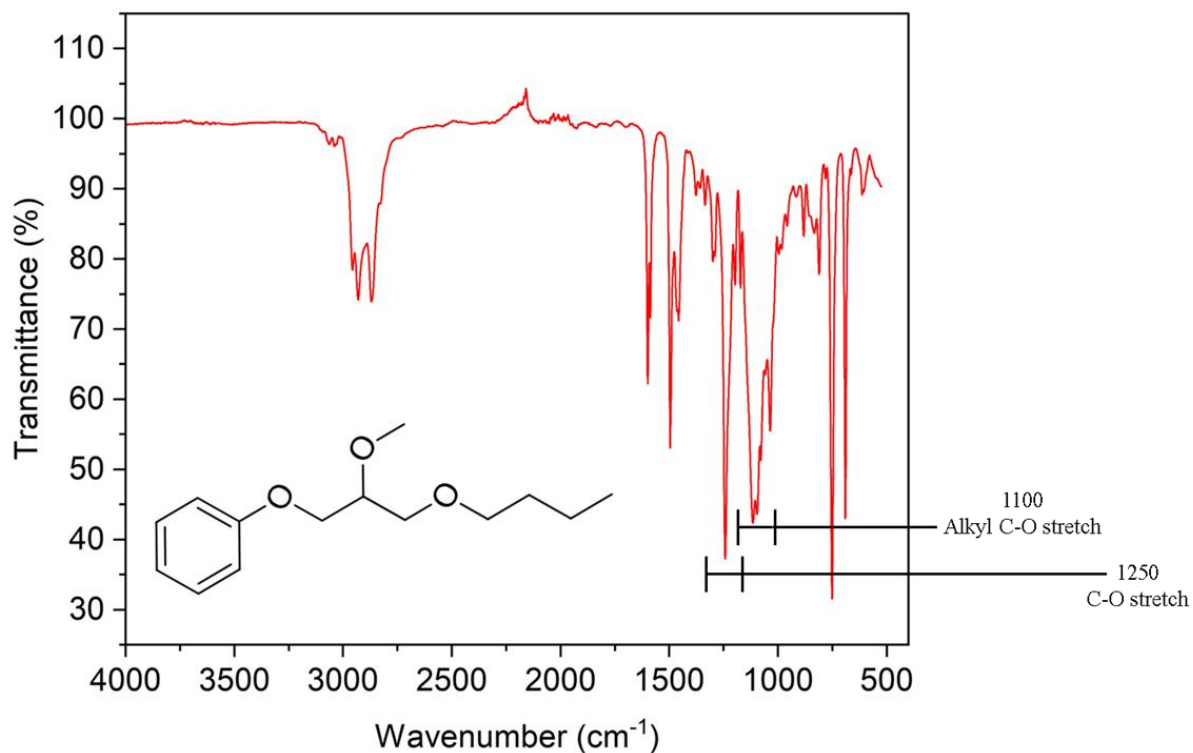


Figure S62. FTIR of Model 2.

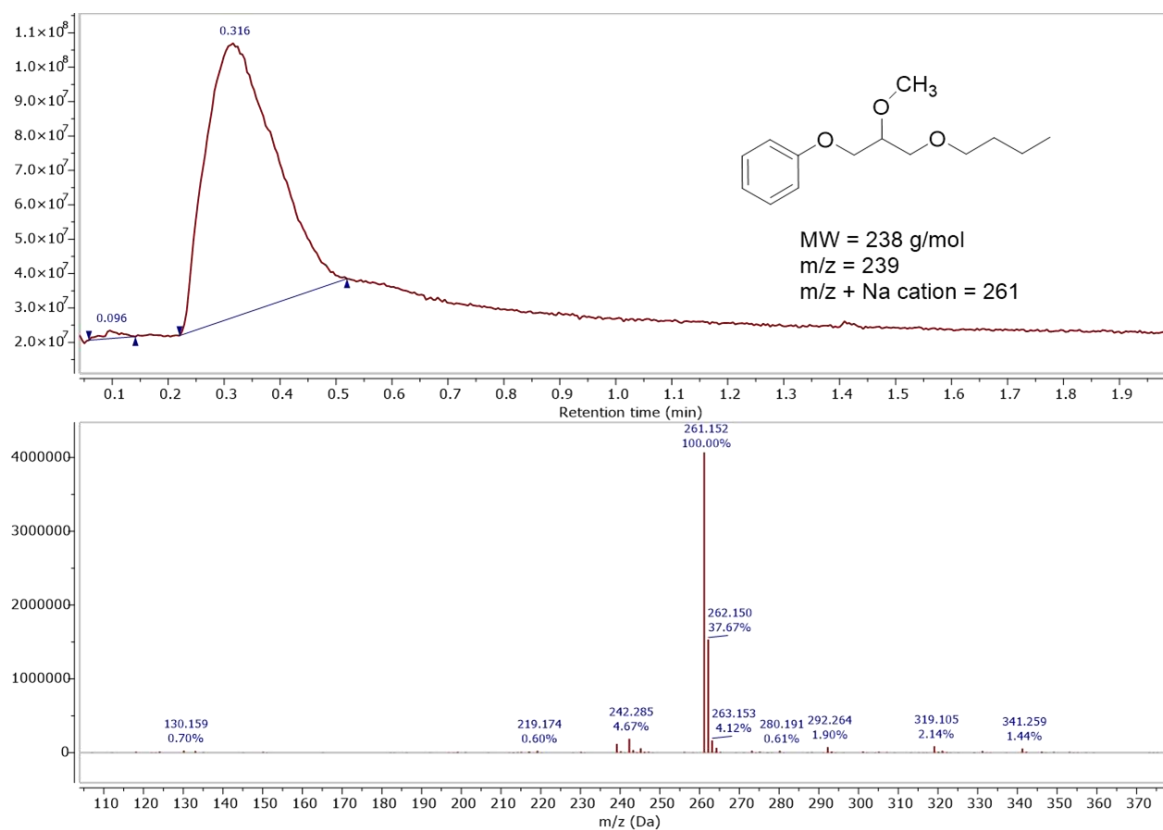
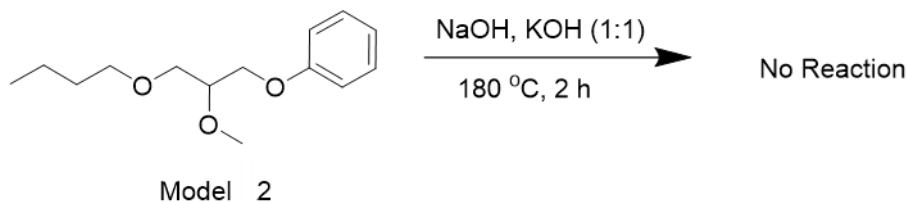


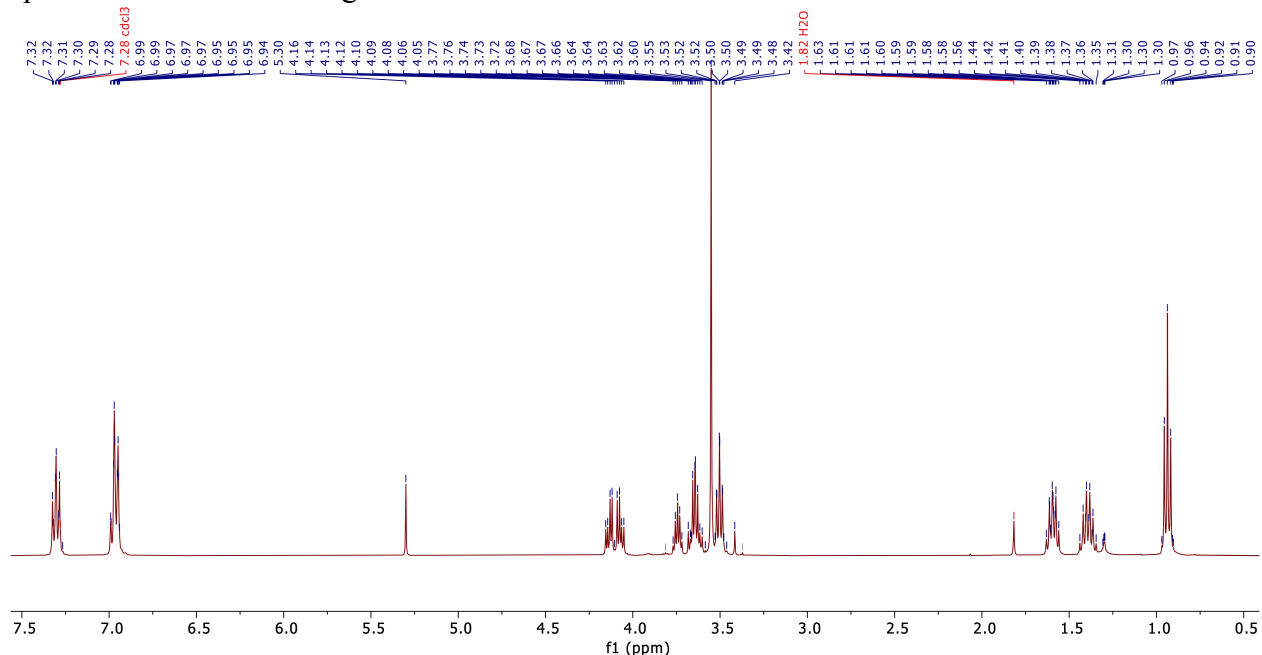
Figure S63. LC-QTOF of Model 2 at retention time = 0.32 min.

**b. Model 2 degradation reaction at 180 °C**

**Scheme S7.** Degradation reaction of Model 2 at 180 °C

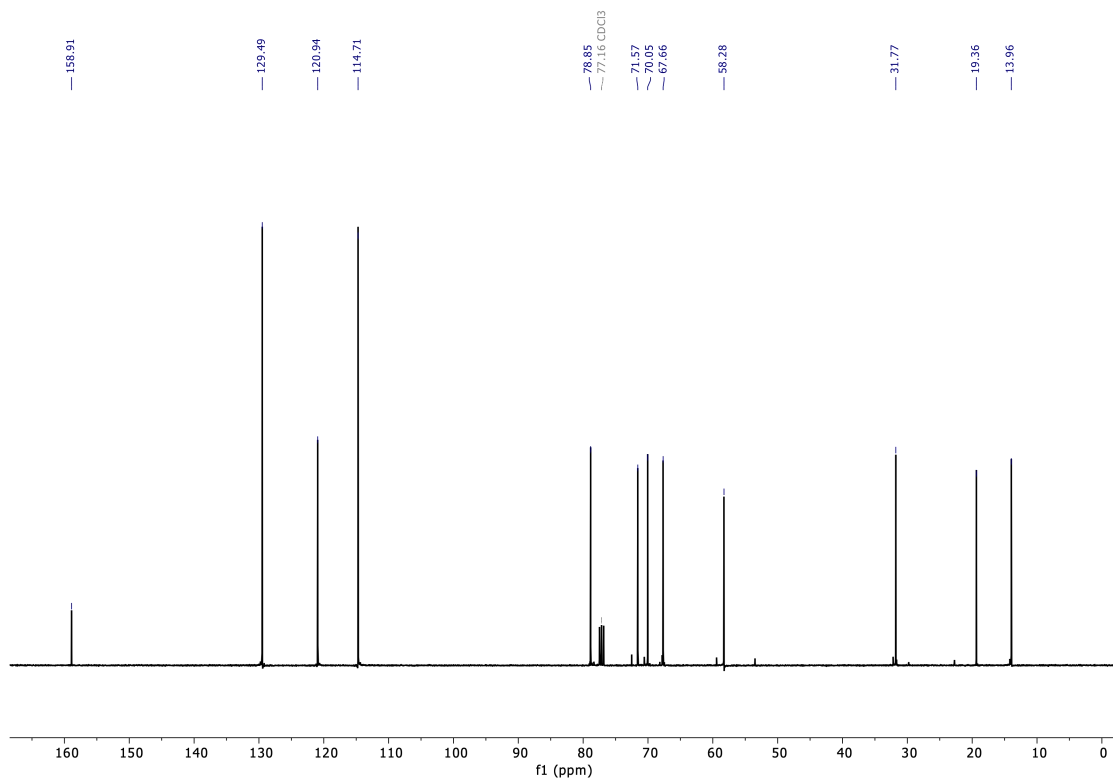


Finely ground NaOH (0.1 g, 2.5 mmol), and KOH (0.14 g, 2.5 mmol) was added to a 50 mL Schlenk flask. Then Model 2 (200 mg, 0.84 mmol) was transferred into the flask. Finally, the flask was carefully flushed with nitrogen gas and placed in an oil bath set to 180 °C for two hours. After the reaction, 50 mL of saturated sodium bicarbonate was added to the Schlenk flask to dissolve the solid residue. The aqueous mixture was then transferred to a 250 mL separatory funnel. The organic material was then extracted with DCM (3 x 30 mL). The organic layers were then combined, and the solvent was removed under reduced pressure. The <sup>1</sup>H and <sup>13</sup>C NMR spectra can be found in Figures S64 and S65.



**Figure S64.** <sup>1</sup>H NMR of Model 2 180 °C degradation reaction extract CDCl<sub>3</sub>.

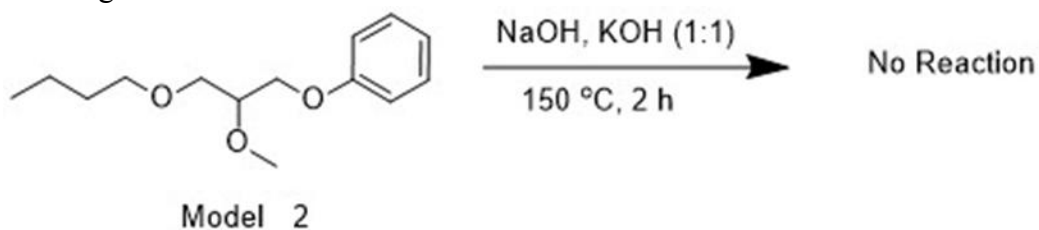




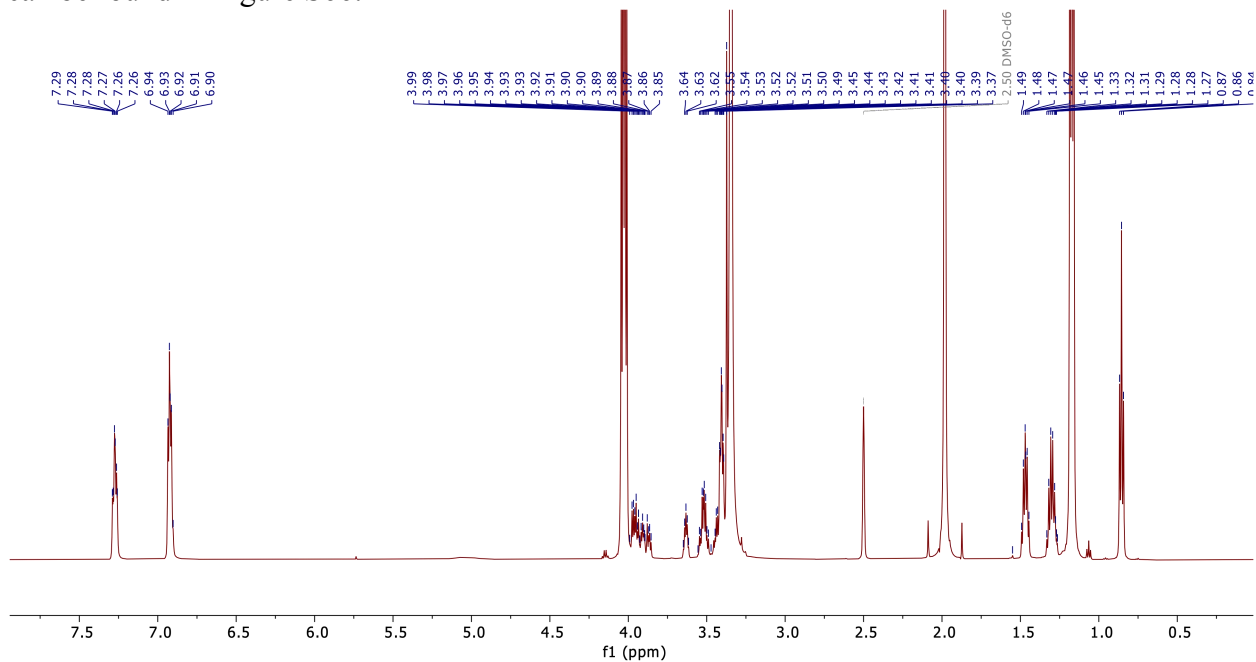
**Figure S65.**  $^{13}\text{C}\{^1\text{H}\}$  NMR of Model 2 180°C degradation reaction extract  $\text{CDCl}_3$ .

### c. Model 2 Degradation reaction at 150 °C

**Scheme S8.** Degradation reaction of Model 2 at 150 °C.



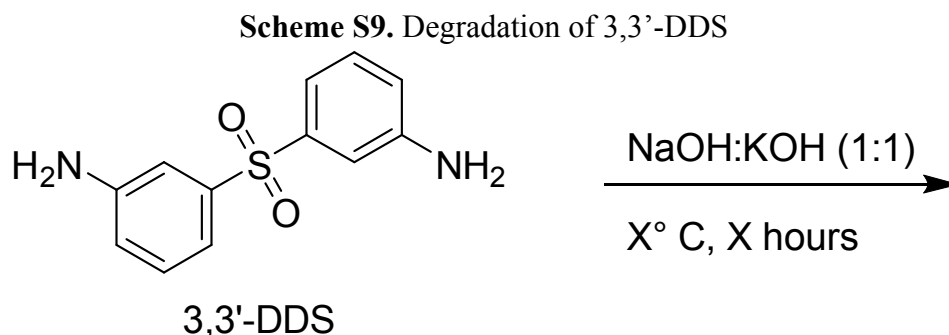
Finely ground NaOH (0.1 g, 2.5 mmol) and KOH (0.14 g, 2.5 mmol) were added to a 50 mL Schlenk flask. Then Model 2 (0.2 g, 0.84 mmol) was transferred into the flask. Finally, the flask was carefully flushed with nitrogen gas and placed in an oil bath set to 150 °C for two hours. After the reaction, aqueous sodium bicarbonate (saturated, 50 mL) was added to the Schlenk flask to dissolve the solid residue. The aqueous mixture was then transferred to a 250 mL separatory funnel. The organic material was then extracted with DCM (3 x 30 mL). The organic layers were then combined, and the solvent was removed under reduced pressure. The <sup>1</sup>HNMR can be found in Figure S66.



**Figure S66.** <sup>1</sup>HNMR of Model 2 150 °C degradation reaction extract in DMSO-*d*<sub>6</sub>.

### 13. 3,3'-DDS degradation experiments

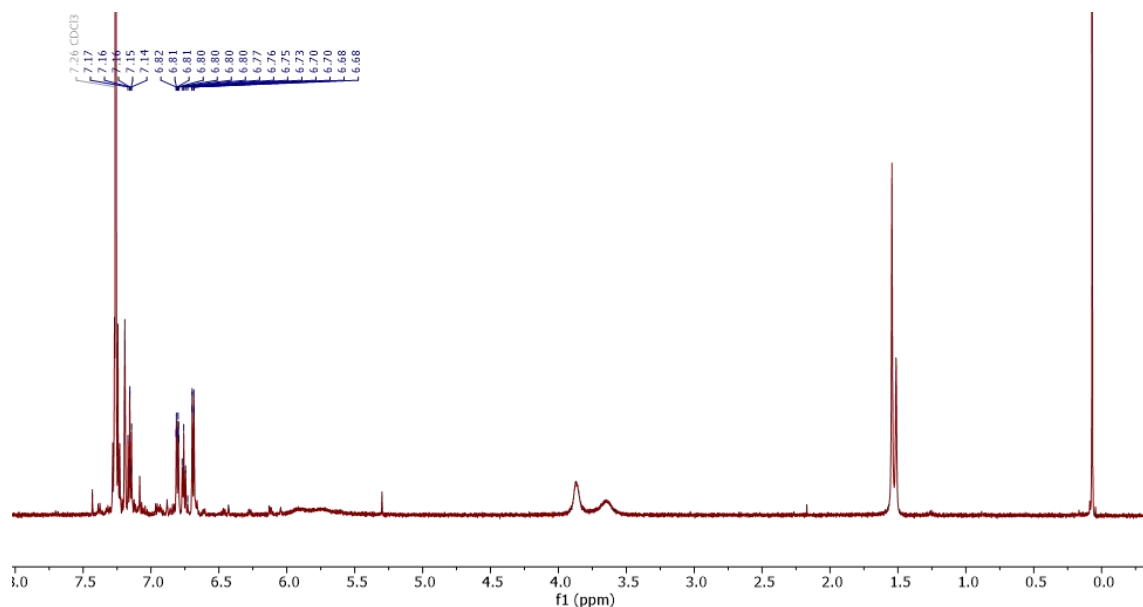
To evaluate the stability of sulfones under hot hydroxide conditions, we attempted to degrade 3,3'-DDS under various conditions shown below.



In a 50 mL Schlenk flask, a finely ground powder of NaOH (0.15 g, 3.75 mmol) and KOH (0.21 g, 3.75 mmol) were combined. Afterwards, 3,3'-DDS (0.1 g, 0.4 mmol) was added into the flask. Finally, the flask was carefully flushed with nitrogen gas and placed in a heated oil bath for X hours. Table S23 below shows the conditions used in each trial. Entries 1 and 2 were determined to have not reacted based on  $^1\text{H}$  NMR and  $^{13}\text{C}$  NMR. The  $^1\text{H}$  NMR spectra of entry 3 does show a reaction and is shown in Figure S 67. Due to the transformation of 3,3'-DDS in entry 3, we state that the sulfone is unstable under these conditions.

**Table S23.** Summary of 3,3'-DDS degradation experiment.

| Entry | Temperature (°C) | Time (hours) | Reaction    |
|-------|------------------|--------------|-------------|
| 1     | 150              | 2            | No reaction |
| 2     | 180              | 2            | No reaction |
| 3     | 190              | 6            | Reaction    |



**Figure S67.**  $^1\text{H}$ NMR of 3,3'-DDS degradation reaction extract in  $\text{CDCl}_3$ .

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