Supporting Information

Synergistic Co-Upcycling of Polycarbonate and Organophosphate Ester Wastes via Chemically Complementary Reactivity

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S1. Chemicals and computational methods

Analytical grade solvents and commercially available reagents were purchased from commercial sources and used directly without further purification unless otherwise stated. PC Powder (~200 mesh, supplied by Crestron Polymers Ltd), trimethyl phosphate (≥98%, Aladdin), ethyl dimethylphosphonoacetate (＞98%, Adamas), benzyl dimethyl phosphonoacetate (98%, Adamas), tetramethyl methylenediphosphonate (＞98%, Adamas), trimethyl phosphite (98%, Macklin), dimethyl phosphonate (98%, Energy Chemical), triethyl phosphate (99%, Adamas), tripropyl phosphate (99%, Heowns), tributyl phosphate (98%, Adamas), triallyl phosphate (＞96%, TCI), tris(2-butoxyethyl) phosphate (98%, Bidepharm), tris(2-ethylhexyl) phosphate (97%, Energy Chemical), caesium carbonate (≥99%, Aladdin), cesium chloride (99%, J&K Scientific), sodium acetate anhydrous (99%, Macklin), cesium acetate (99.9%, Bidepharm), sodium hydrogen carbonate (≥99.5%, Shanghai Linfeng Chemical Reagent CO. LTD), cesium bicarbonate (99%, Bidepharm), lithium carbonate (99.42%, Bidepharm), sodium carbonate anhydrous(≥99.5%, Macklin), potassium carbonate (99%, Aladdin), sodium hydroxide (≥96%, Shanghai Linfeng Chemical Reagent CO. LTD), potassium hydroxide (99%, Aladdin), cesium hydroxide (99.9%, Aladdin), DMF (≥99.8%, Aladdin), *p*-Xylene(≥99.0%, Macklin), Toluene (≥99.5%, Shanghai Linfeng Chemical Reagent CO. LTD), 1,4-Dioxane (≥99.5%, Shanghai Linfeng Chemical Reagent CO. LTD), Cyclohexanone (≥99.5%, Shanghai Linfeng Chemical Reagent CO. LTD), acetone (≥99.5%, Shanghai Linfeng Chemical Reagent CO. LTD), CH_3CN (≥99.5%, Shanghai Linfeng Chemical Reagent CO. LTD), THF (≥99.5%, Shanghai Linfeng Chemical Reagent CO. LTD), DMSO (≥99.5%, Shanghai Linfeng Chemical Reagent CO. LTD), NMP (99.9%, Macklin), mesitylene (≥98%, Shanghai Linfeng Chemical Reagent CO. LTD), ethyl acetate (99.5%, Hangzhou Fangping Chemical Co. LTD), BPA (＞99%, Macklin), $CDCl_3$ (99%, Sigma-Aldrich), DMSO-*d*6 (99%, Adamas), and deionized water were used herein.

S2. Reaction conditions reported in the literature for PC hydrolysis and alcoholysis.

Table S1. Reaction conditions and catalysts for PC hydrolysis in literature.

Entry	Catalysts	Temp. (°C)	T (h)	BPA Yield (%)	Ref.
1	CTAB+NaOH	160	10 min	Conv. > 80%	[1]
2	NaOH	100	8	100%	[2]
3	$Fe_3O_4/SiO_2/NH_2$	160	93 min	100%	[3]
4	ZnO-NPs/Nbu	100	7	99%	[4]
5	[Bmim][Cl]	165	3	95%	[5]
6	[Bmim][Ac]	140	3	96%	[6]
7	CeO_2	200	5	90%	[7]
8	$La(O_3SCF_3)_3$	160	6	97%	[8]
9	[HDBU][LAc]	140	4	97%	[9]
10	Natural chlorite	140	6	98%	[10]
11	Concentrated Sulfuric Acid	150	10	90%	[11]
12	TiO_2+NaOH	reflux	83 min	70%	[12]
13	Sub-critical water	280	46 min	100%	[13]

Table S2. Reaction conditions and catalysts for PC methanolysis in literature.

Entry	Catalyst	Temp. (℃)	T (h)	BPA Yield (%)	DMC Yield (%)	Ref.
1	CeO_2-CaO-ZrO_2	100	2	99.1	/	[14]
2	Mg/Al-LDOs	110	1	>98	/	[15]
3	$NaAlO_2$	60	2	96.8	75	[16]
4	DBU	70	0.6	99	99	[17]
5	TBD	50	12	98	98	[18]
6	urea	140	2	93.4	74.7	[19]
7	TBD:MSA	50	12	>98	/	[20]
8	Si-TBD	65	2	96	/	[21]
9	[BMIM][Ac]	90	2.5	>95	/	[22]
10	[HDBU][Suc]	70	2	99	/	[23]
11	[HDBU][LAc]	120	1	99	/	[24]
12	SBA-15-Pr-MIM-OH	120	1	>98	/	[25]
13	ChCl-2Urea	130	2.5	>99	/	[26]
14	[EMIMOH]Cl-2Urea	120	2	98	/	[27]
15	Zn^{II}-complexes	75	1	95	/	[28]
16	lanthanum β-diketonate complexes	80	2	87	93	[29]
17	/	100	6	99	/	[30]
18	/	140	6	91	/	[31]
19	CO_2	140	6	89	/	[32]

S3. Optimization of reaction conditions for the PC-TMP system

To achieve the depolymerization of PC to produce bisphenol A (BPA) while simultaneously enabling the etherification of BPA with TMP to generate bisphenol A methyl ether (BPAME), we evaluated the catalytic performance of various basic catalysts (Fig. S1a). In the absence of catalyst, PC remained intact with no detectable products, confirming the necessity of catalytic activation for initiating this process. Among tested catalysts, cesium carbonate (Cs_2CO_3) exhibited optimal performance, achieving 99% BPAME yield in the model PC-TMP system. This superior activity stems from two critical factors: (i) moderate basicity that activates PC without decomposing TMP, and (ii) strong ionization in polar aprotic solvents that generates active carbonate anions. Other cesium salts (CsCl, $CsHCO_3$, CsOAc) showed diminished activity (<3% yields) due to weaker basicity, while other carbonates (Na_2CO_3, K_2CO_3) suffered from poor ionization (<2% yields). Conversely, strong bases (NaOH, KOH, CsOH) preferentially reacted with TMP, consuming the base and preventing PC depolymerization (<50% yields).

Solvent selection is particularly critical for this system, as industrial OPEs waste streams exist in various organic solvents depending on their source and application. We systematically screened aprotic solvents and observed a positive correlation between solvent polarity (π^*)[33] and BPAME yield, with DMF (π^* = 88) delivering optimal performance (99% yield, Fig. S1b and Fig. S2). Notably, all tested organic solvents exhibited appreciable activity, demonstrating the versatility of this system for processing OPEs waste in diverse organic media. Even for OPEs waste streams in less polar solvents (e.g., *p*-xylene), where lower reactivity might be encountered, the reaction efficiency can be effectively enhanced by increasing the reaction temperature and time without the complete solvent replacement (Fig. S3). Moreover, reducing solvent volume (which increases phosphate ester concentration) showed negligible impact on BPAME yield, further confirming the system's robustness (Fig. S4). This flexibility is particularly advantageous for industrial implementation, as it enables direct utilization of OPEs waste in native solvent matrices with minimal pretreatment, significantly reducing processing costs and environmental burden.

Subsequent parametric studies demonstrated that increasing Cs_2CO_3 loading, TMP loading, reaction temperature, and reaction time all contributed to improved BPAME yields (Fig. S1c–f). Notably, substoichiometric TMP loading (<1 equiv) yielded combined BPA derivatives and unreacted BPA totaling below 100%, indicating incomplete PC conversion due to unfavorable equilibrium. This observation reveals TMP's dual role in this system: it functions simultaneously as an alkylating agent for BPA and as an equilibrium driver that consumes BPA as it forms, enabling quantitative PC depolymerization. These optimized conditions, established with the PC-TMP model system, provide the foundation for extending the system to diverse OPEs waste streams.

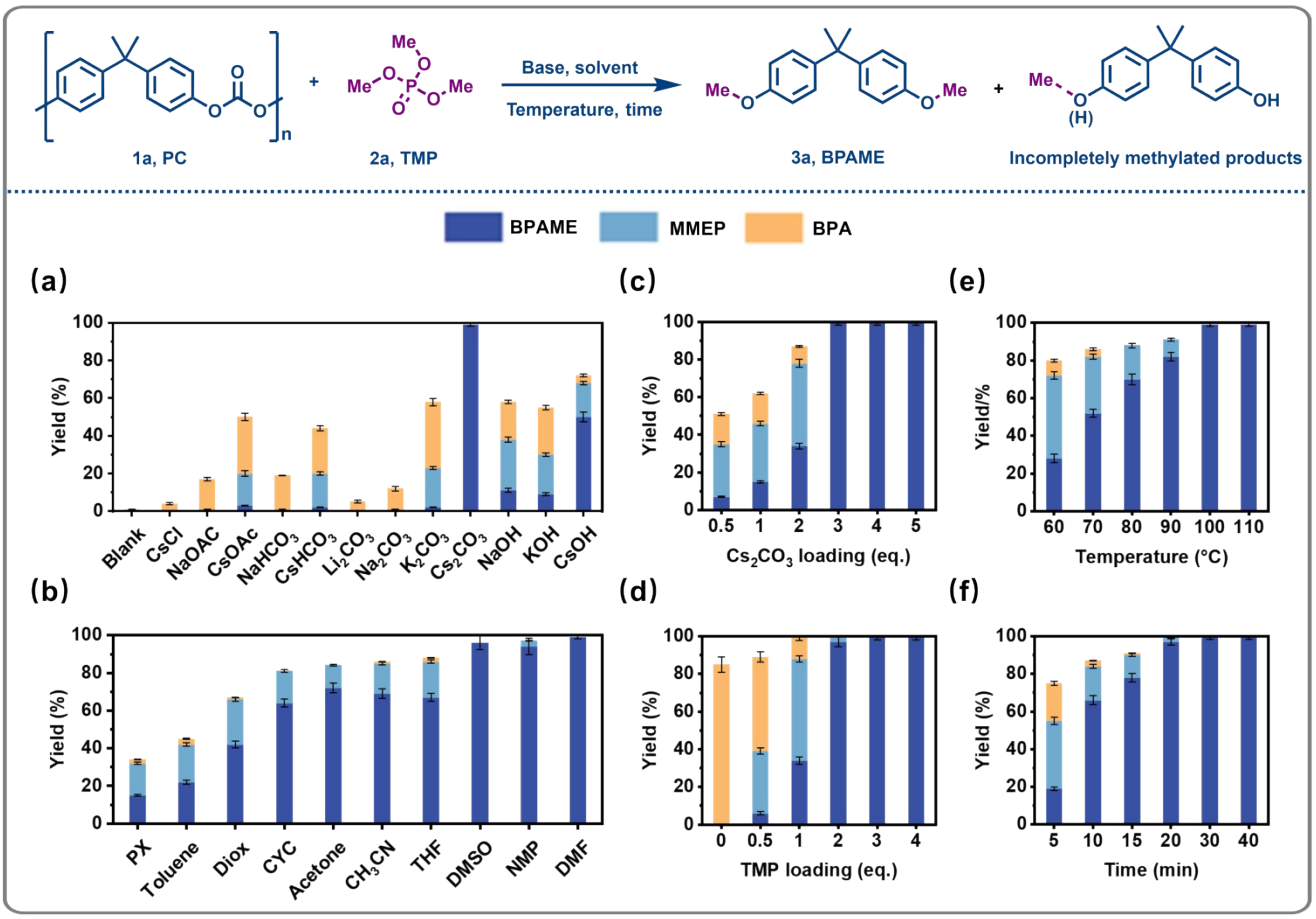


Fig. S1. Optimization of reaction conditions. Effect of (a) catalyst, (b) solvent, (c) Cs_2CO_3 loading, (d) TMP loading, (e) reaction temperature, and (f) reaction time. Standard conditions: PC (1 mmol PC unit), Cs_2CO_3 (3 equiv), TMP (3 mmol), DMF (2 mL), 100 °C, 30 min, unless otherwise noted. Error bars represent standard deviation (n = 3).

S4. Role of the solvent in the PC-TMP system

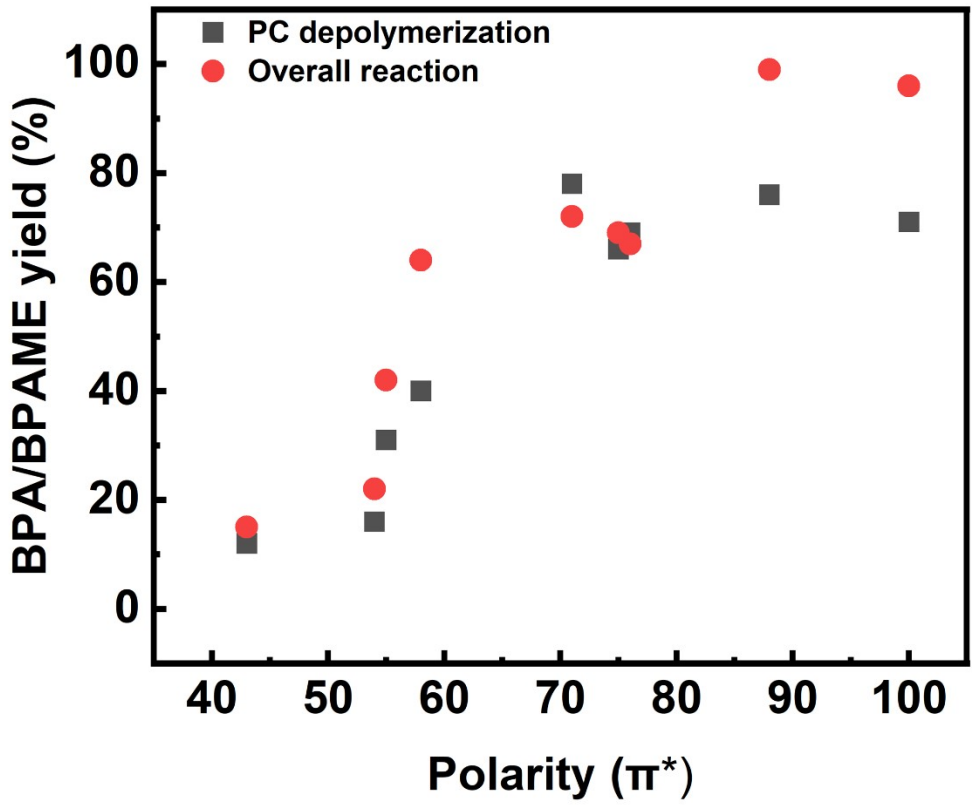


Fig. S2. BPAME yield in PC-TMP system and PC depolymerization as a function of solvent polarity.

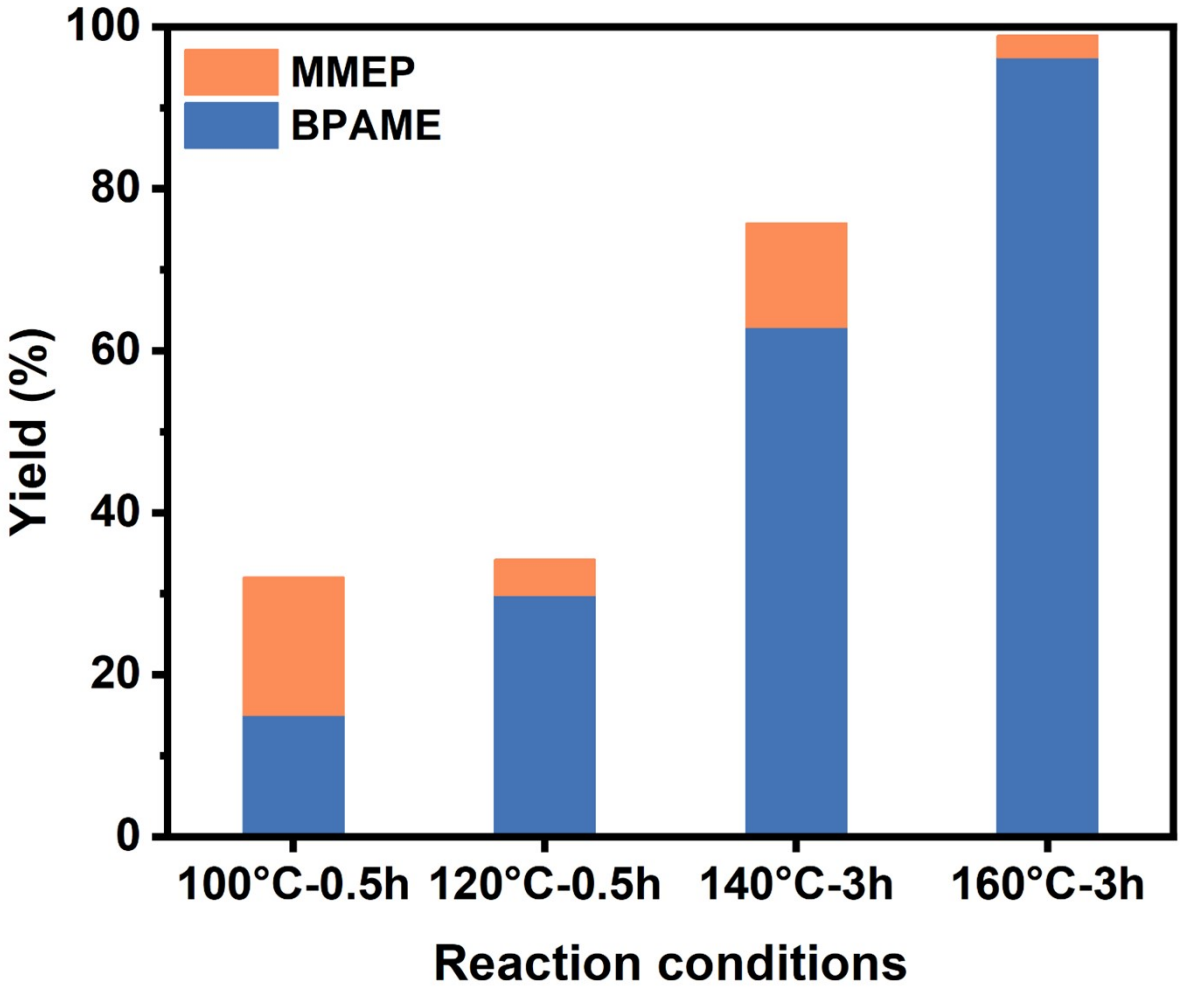


Fig. S3. Effect of reaction temperature and time on BPAME yield using *p*-xylene as solvent.

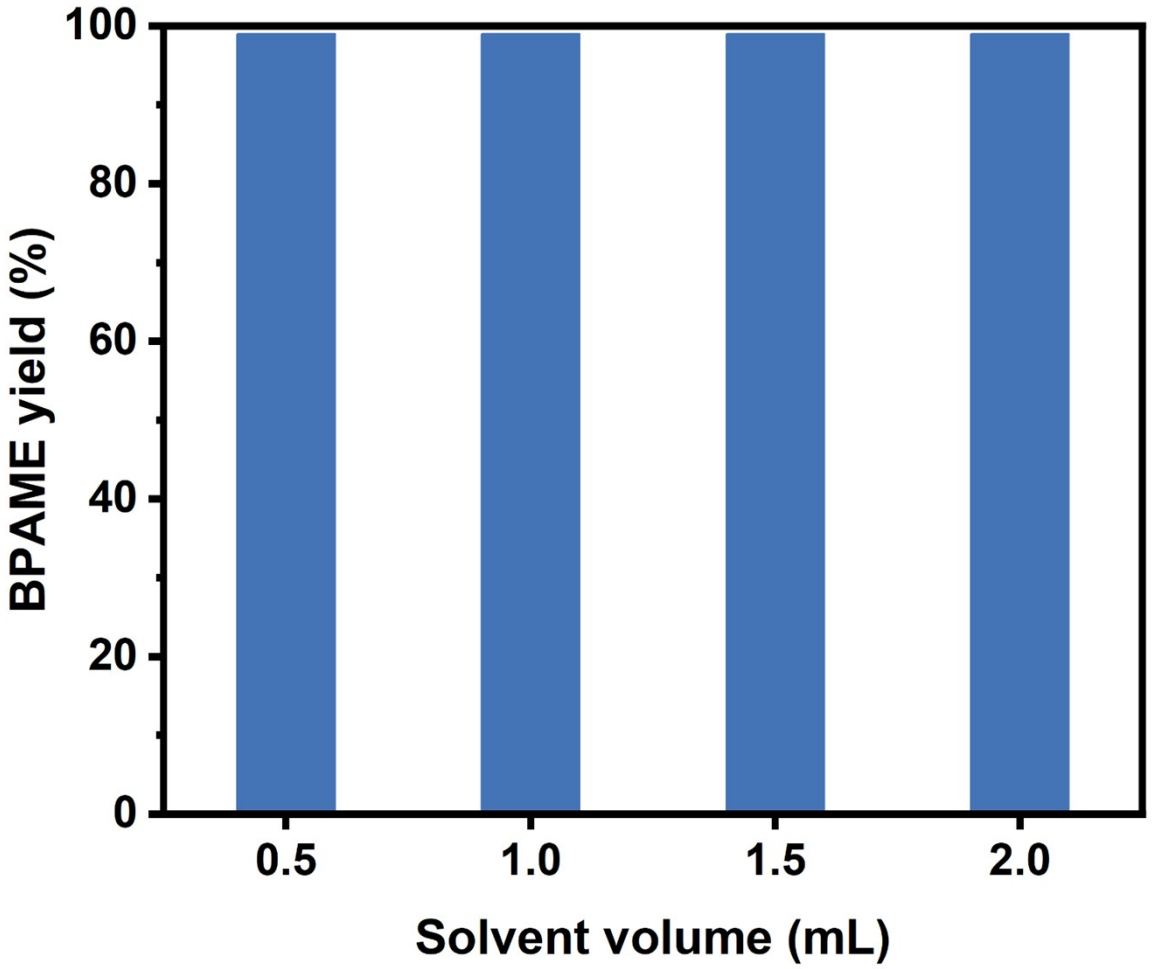
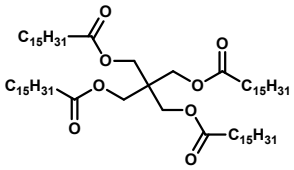
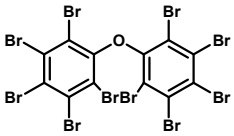
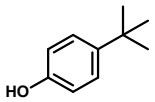


Fig. S4. Effect of solvent volume on BPAME yield.

Table S3. Effects of common PC additives on the catalytic performance of the PC-TMP system.

Entry	Additive name	Additive chemical formula	Yield BPAME (%)	Yield DMP (%)
1	/	/	99	99
2	Pentaerythrityl tetrastearate		99	99
3	Decabromodiphenyl ether		99	99
4	*p*-tert-Butylphenol		99	99

Standard reaction conditions: PC (1 mmol PC unit), Cs_2CO_3 (3 mmol), TMP (3 mmol), DMF (2 mL), additive (5 wt%) 100°C, 30 min.

Table S4. Effect of Cs_2CO_3 on polycarbonate depolymerization and BPA methylation

Entry	Reactant-1	Reactant-2	Catalyst	Reaction performance
1	PC	TMP	/	0% Conv. PC
2	PC	TMP	Cs_2CO_3	99% Yield BPAME, 99% Yield DMP
3	PC	/	/	0% Conv. PC
4	PC	/	Cs_2CO_3	92% Conv. PC
5	BPA	TMP	/	0% Conv. BPA
6	BPA	TMP	Cs_2CO_3	100% Conv. BPA

Standard reaction conditions: PC or BPA (1 mmol), Cs_2CO_3 (3 mmol, if applicable), TMP (3 mmol, if applicable), in DMF (2 mL) at 100 °C for 30 min.

S5. Proposed reaction mechanism for the PC-TMP System

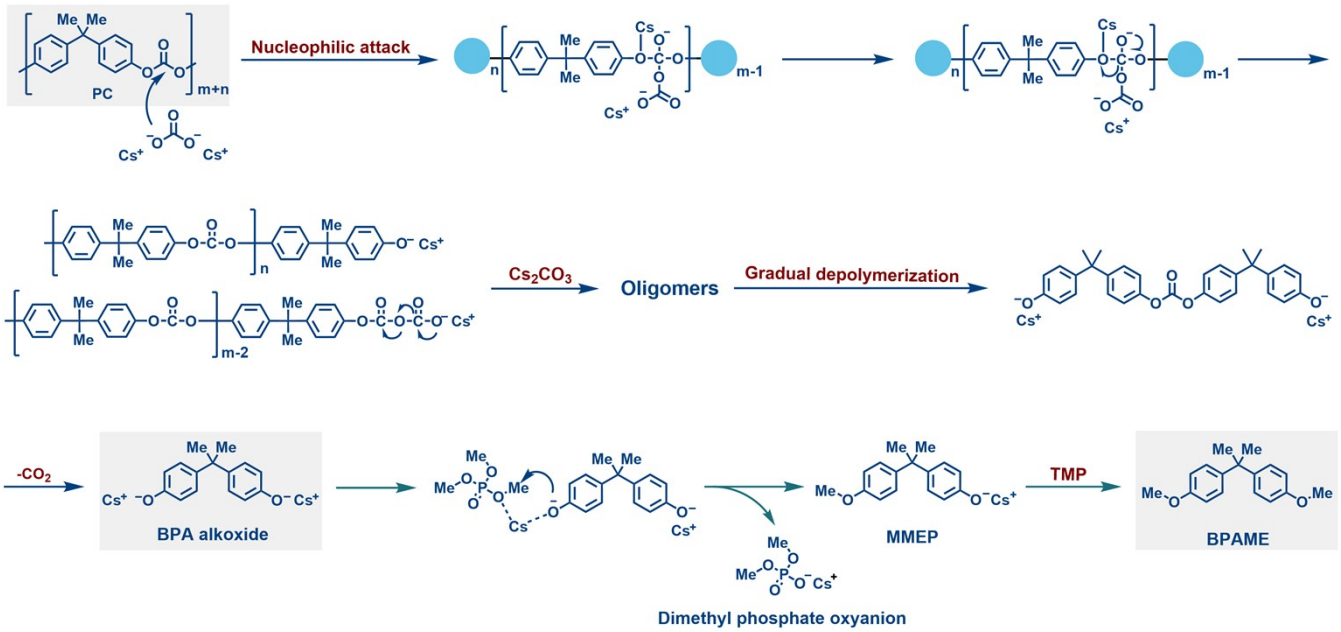
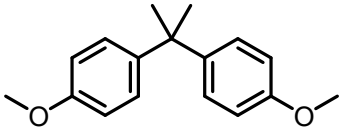
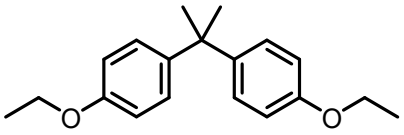


Fig. S5. Proposed two-stage mechanism for PC depolymerization and BPA alkylation.

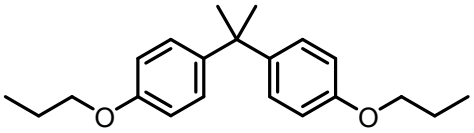
S6. ^{1}H NMR and ^{13}C NMR characterization date of all compounds



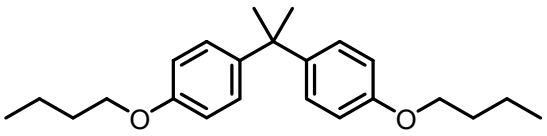
4,4'-(propane-2,2-diyl)bis(methoxybenzene) (BPAME) **3a**: white solid, ^{1}H NMR yield 99%; ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.14 (d, *J* = 10.8 Hz, 4H), 6.80 (d, *J* = 10.8 Hz, 4H), 3.77 (s, 6H), 1.64 (s, 6H); ^{13}C NMR (151 MHz, $CDCl_3$) δ = 157.3, 143.1, 127.7, 113.2, 55.2, 51.6, 31.0.



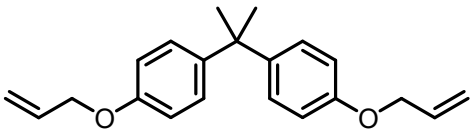
4,4'-(propane-2,2-diyl)bis(ethoxybenzene) **3b**: white solid, ^{1}H NMR yield 99%; ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.04 (d, *J* = 10.8 Hz, 4H), 6.70 (d, *J* = 10.2 Hz, 4H), 3.91 (q, *J* = 8.4, 16.8 Hz 4H), 1.55 (s, 6H), 1.31 (t, *J* = 8.4 Hz, 6H); ^{13}C NMR (151 MHz, $CDCl_3$) δ = 156.7, 127.7, 113.7, 63.3, 41.6, 31.0, 14.9.



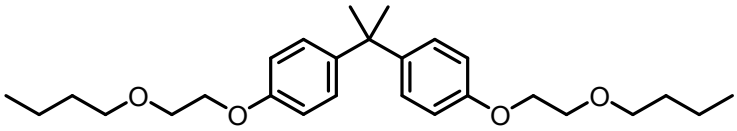
4,4'-(propane-2,2-diyl)bis(propoxybenzene) **3c**: white solid, ^{1}H NMR yield 77%; ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.12 (d, *J* = 10.8 Hz, 4H), 6.79 (d, *J* = 10.8 Hz, 4H), 3.89 (t, *J* = 7.8 Hz, 4H), 1.82-1.75 (m, 4H), 1.03 (s, 6H), 1.02(t, *J* = 9 Hz, 6H); ^{13}C NMR (151 MHz, $CDCl_3$) δ = 156.9, 143.0, 127.7, 113.8, 69.4, 41.6, 31.1, 22.6, 10.6.



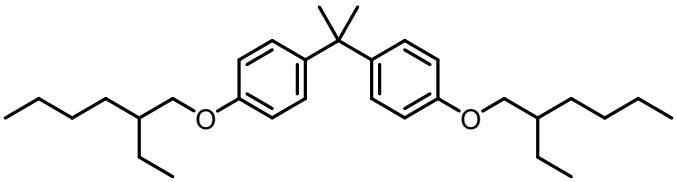
4,4'-(propane-2,2-diyl)bis(butoxybenzene) **3d**: white solid, ^{1}H NMR yield 79%; ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.12 (d, *J* = 10.8 Hz, 4H), 6.78 (d, *J* = 10.8 Hz, 4H), 3.92 (t, *J* = 7.8 Hz, 4H), 1.77-1.71 (m, 4H),1.63 (s, 6H), 1.50-1.45(m, 4H), 0.96 (t, *J* = 9.0 Hz, 6H); ^{13}C NMR (151 MHz, $CDCl_3$) δ = 156.9, 142.9, 127.6, 113.8, 67.5, 41.6, 31.4, 31.1, 19.3, 13.8.



4,4'-(propane-2,2-diyl)bis((allyloxy)benzene) **3e**: colorless liquid, ^{1}H NMR yield 99%; ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.13 (d, *J* = 10.2 Hz, 4H), 6.81 (d, *J* = 10.2 Hz, 4H), 6.09-6.01 (m, 2H), 5.40 (dd, *J* = 1.8, 22 Hz, 2H), 5.26 (dd, *J* = 1.8, 12.6 Hz, 2H), 4.50 (d, *J* = 6.6 Hz, 4H), 1.63 (s, 6H); ^{13}C NMR (151 MHz, $CDCl_3$) δ = 156.4, 143.3, 133.5, 127.7, 117.5, 114.0, 68.8, 41.7, 31.0.



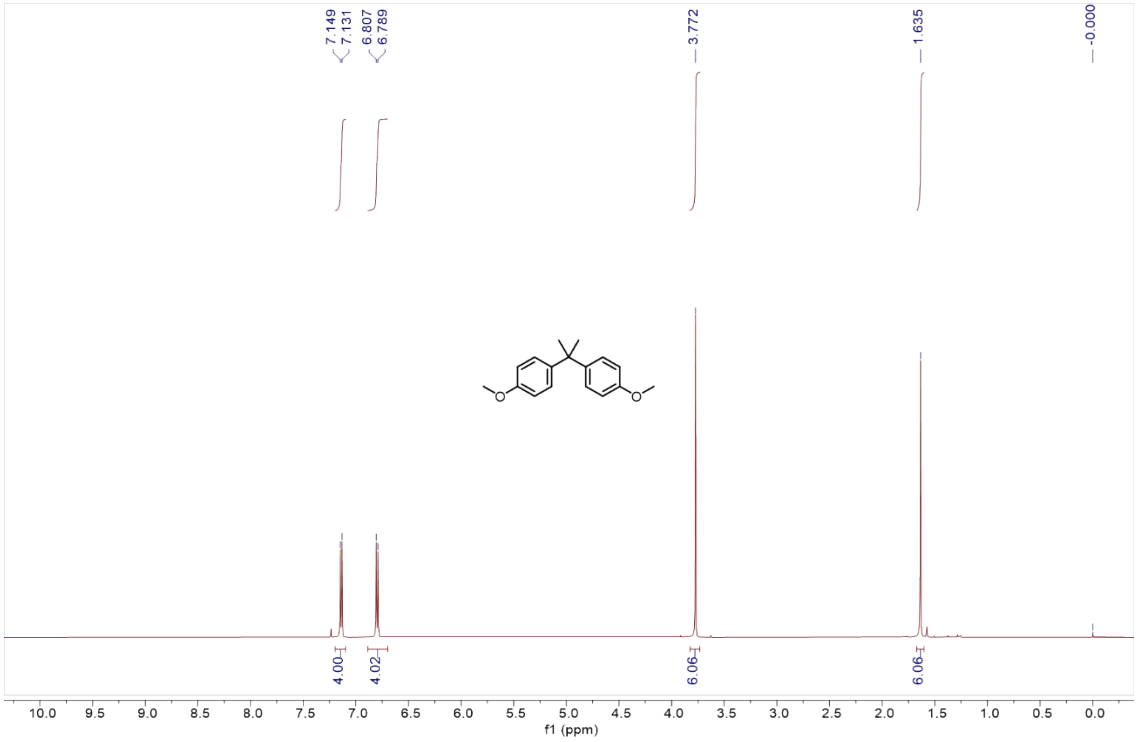
4,4'-(propane-2,2-diyl)bis((2-butoxyethoxy)benzene) **3f**: colorless liquid, ^{1}H NMR yield 93%; ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.11 (d, *J* = 8.4 Hz, 4H), 6.79 (d, *J* = 9.0 Hz, 4H), 4.08 (t, *J* = 4.8 Hz, 4H), 3.76 (t, *J* = 4.8 Hz, 4H), 3.52 (t, *J* = 6.6 Hz, 4H), 1.62 (s, 6H), 1.60-1.56 (m, 4H), 1.39-1.35 (m, 4H), 0.92 (t, *J* = 7.2 Hz, 6H); ^{13}C NMR (151 MHz, $CDCl_3$) δ = 156.7, 143.3, 127.7, 114.0, 71.4, 69.3, 67.4, 41.7, 31.8, 31.1, 19.3, 14.0.

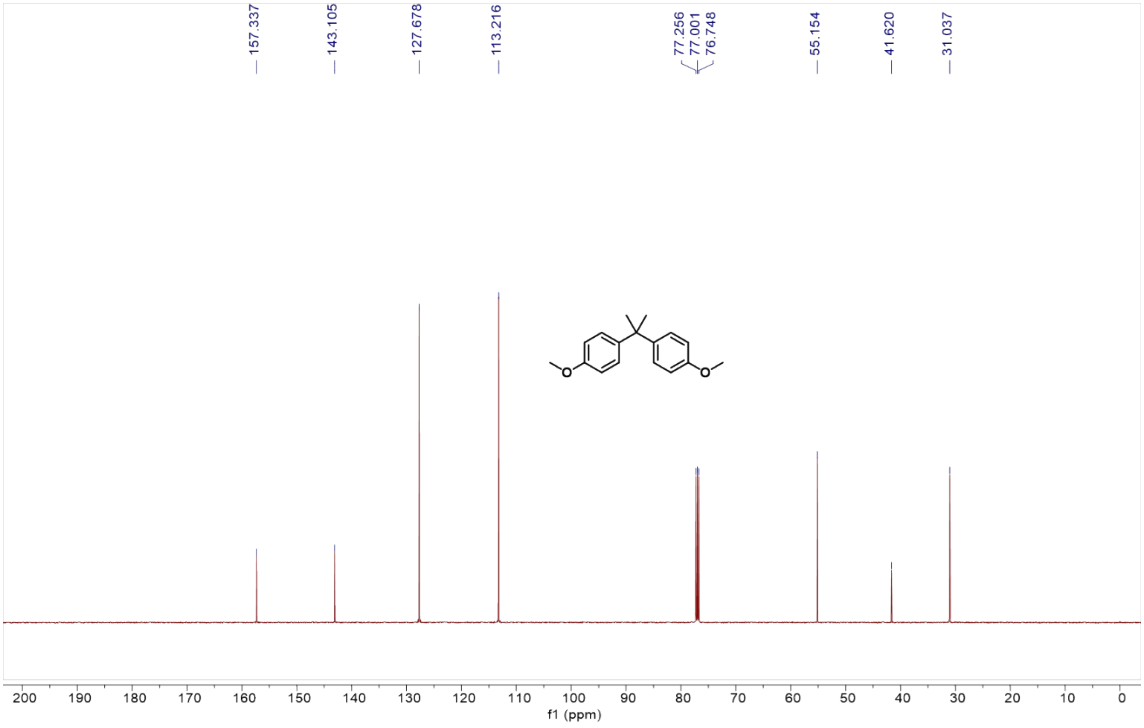


4,4'-(propane-2,2-diyl)bis(((2-ethylhexyl)oxy)benzene) **3g**: white solid, ^{1}H NMR yield 53%; ^{1}H NMR (600 MHz, $CDCl_3$) δ = 7.12 (d, *J* = 9.0 Hz, 4H), 6.79 (d, *J* = 9.0 Hz, 4H), 3.83 (dd, *J* = 2.4, 5.4 Hz, 4H), 1.72-1.67 (m, 2H), 1.63 (s, 6H), 1.49-1.29 (m, 16H), 0.93-0.88 (m, 12H); ^{13}C NMR (151 MHz, $CDCl_3$) δ = 157.2, 142.9, 127.7, 113.8, 70.3, 41.6, 39.5, 31.1, 30.6, 29.1, 23.9, 23.1, 14.1, 11.1.

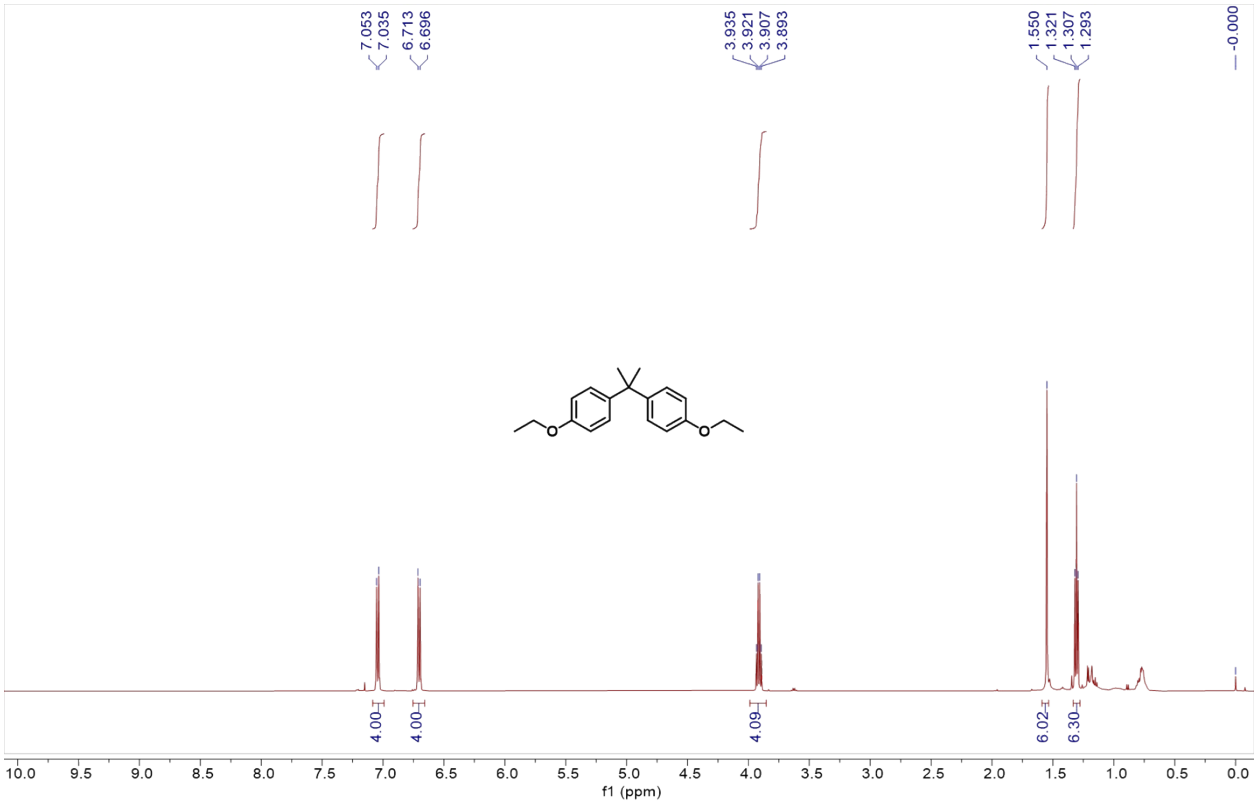
S7. ^{1}H NMR and ^{13}C NMR spectra of all compounds

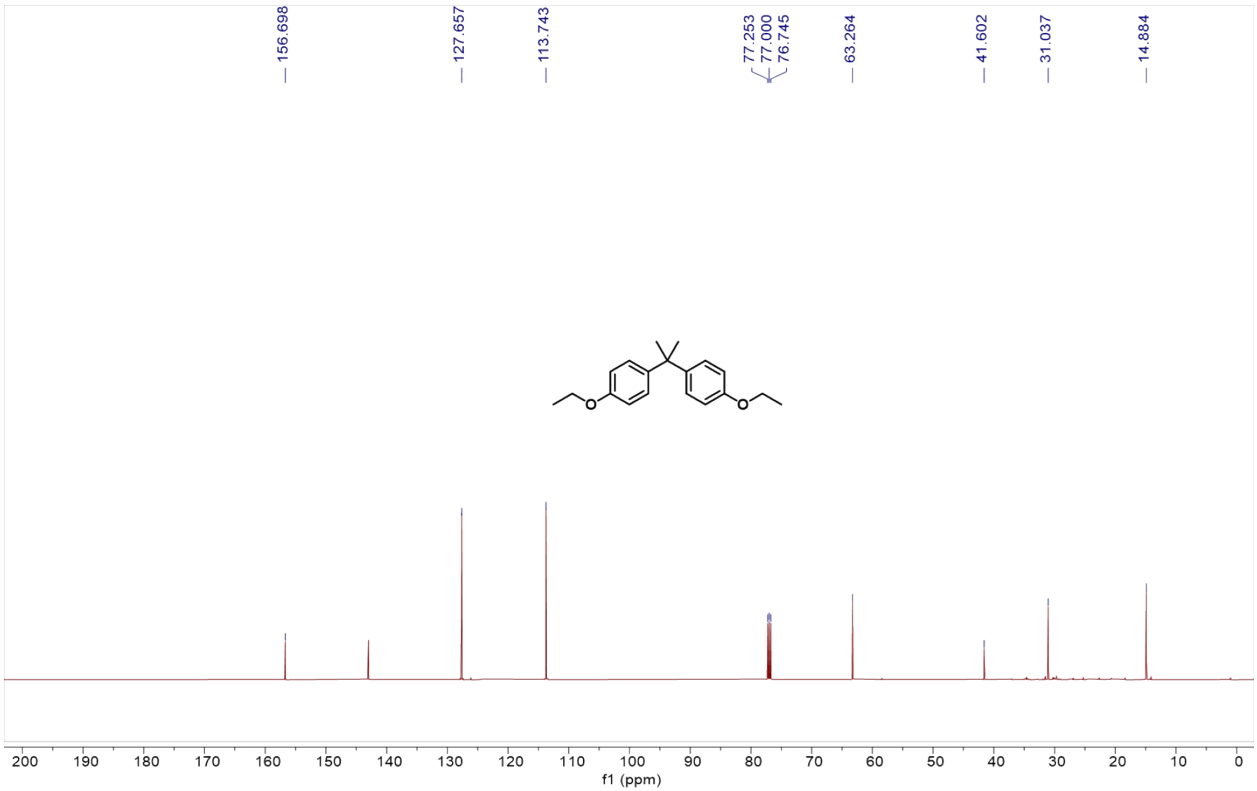
^{1}H NMR of **3a** (600 MHz, $CDCl_3$) and ^{13}C NMR of **3a** (151 MHz, $CDCl_3$)



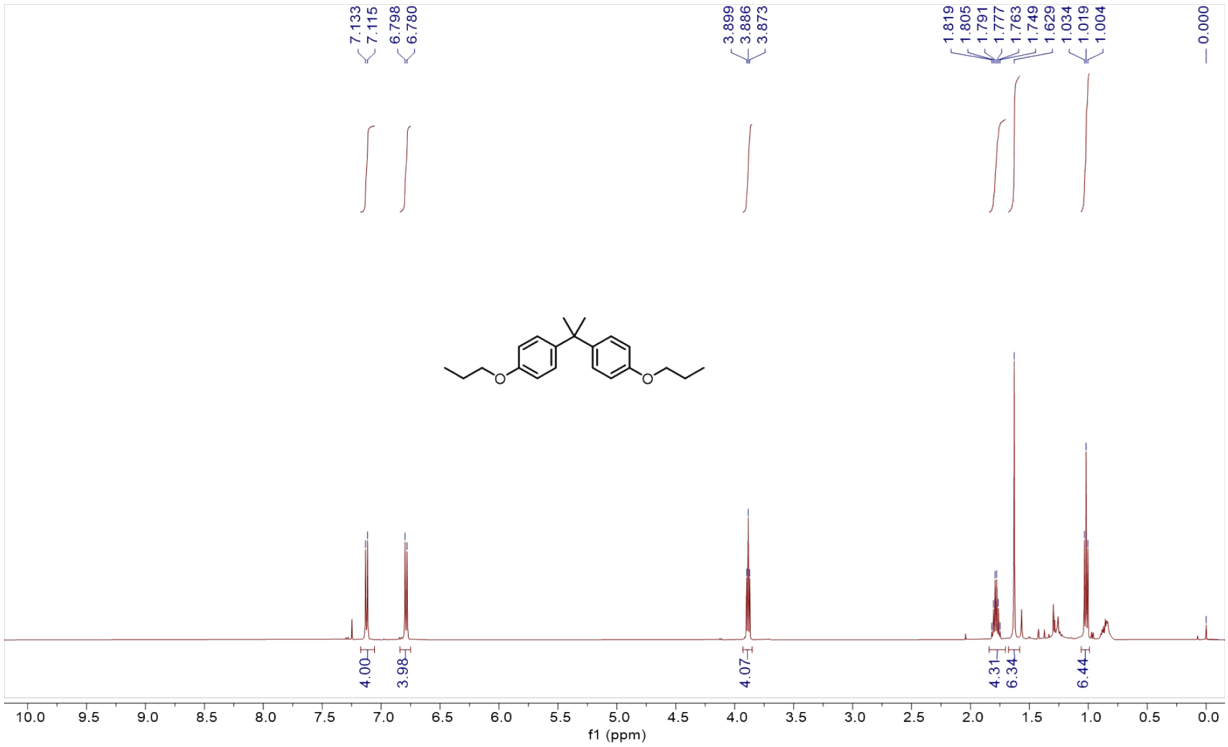


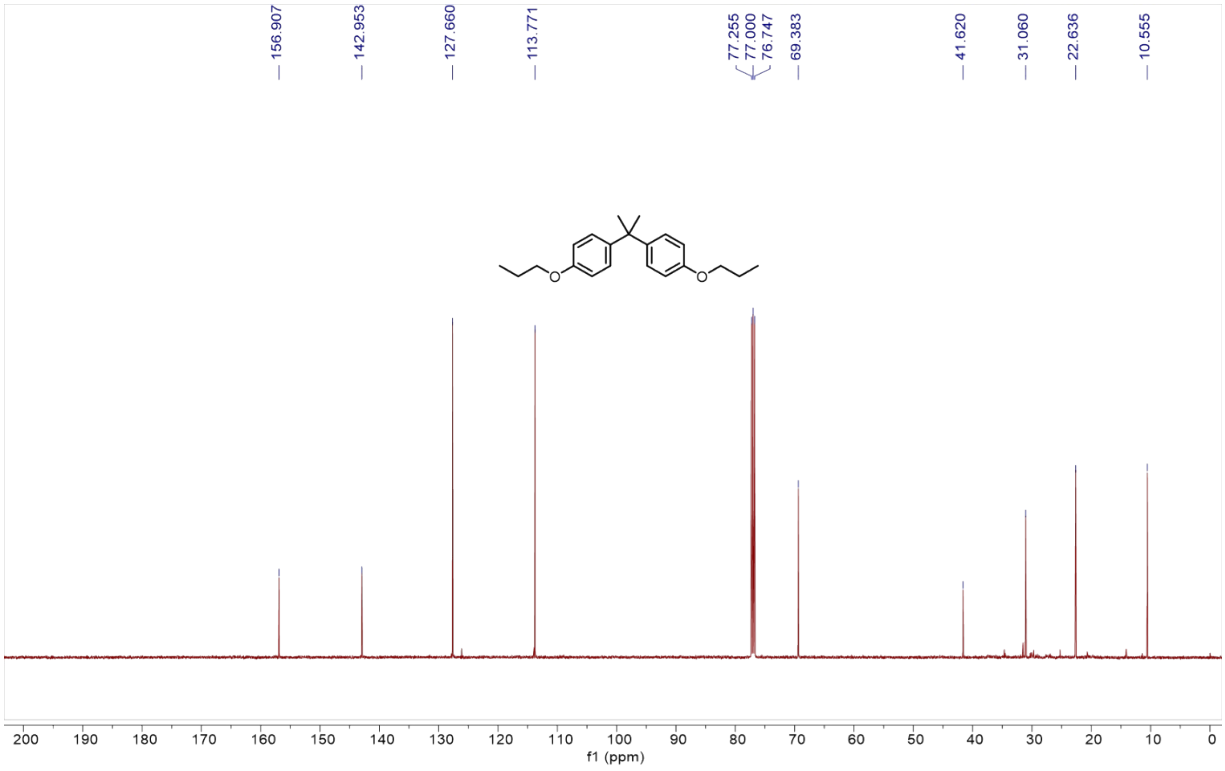
^{1}H NMR of **3b** (600 MHz, $CDCl_3$) and ^{13}C NMR of **3b** (151 MHz, $CDCl_3$)



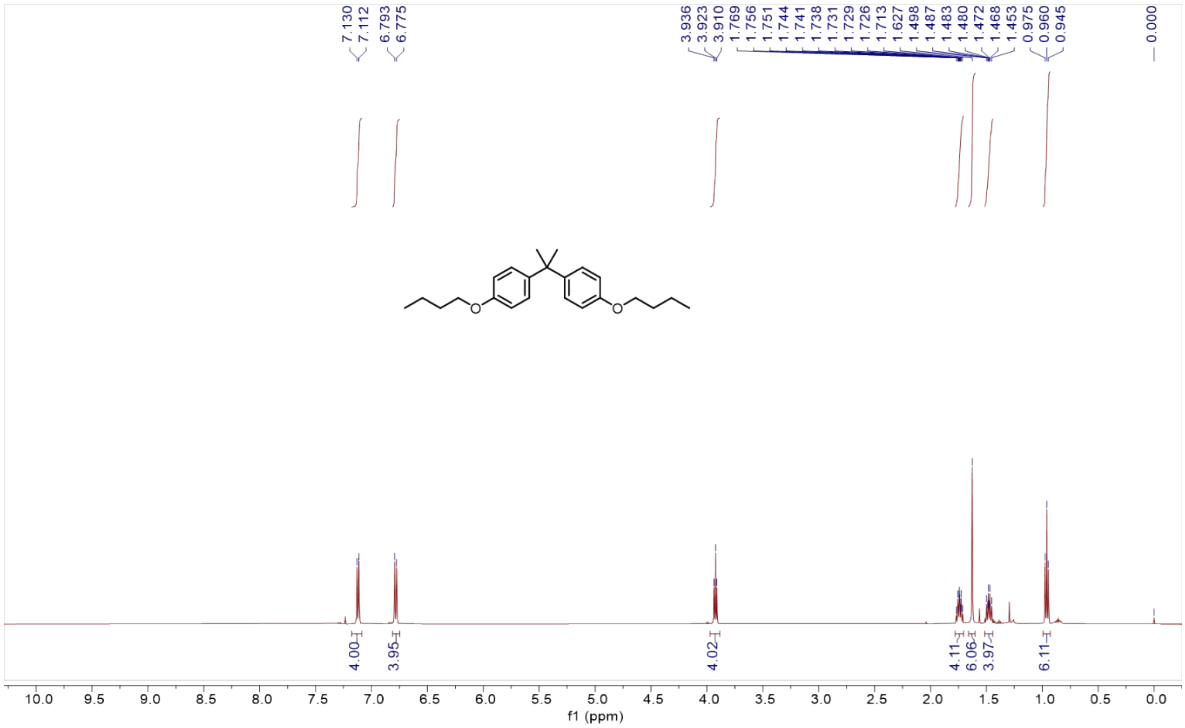


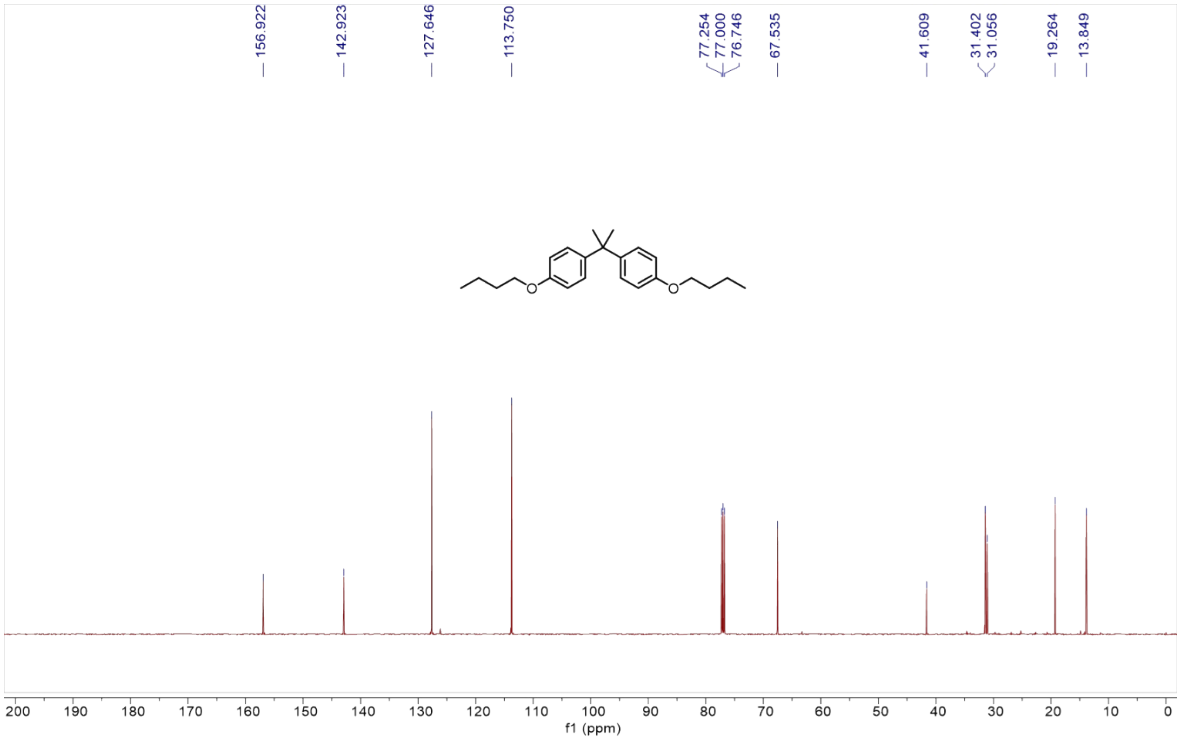
^{1}H NMR of **3c** (600 MHz, $CDCl_3$) and ^{13}C NMR of **3c** (151 MHz, $CDCl_3$)



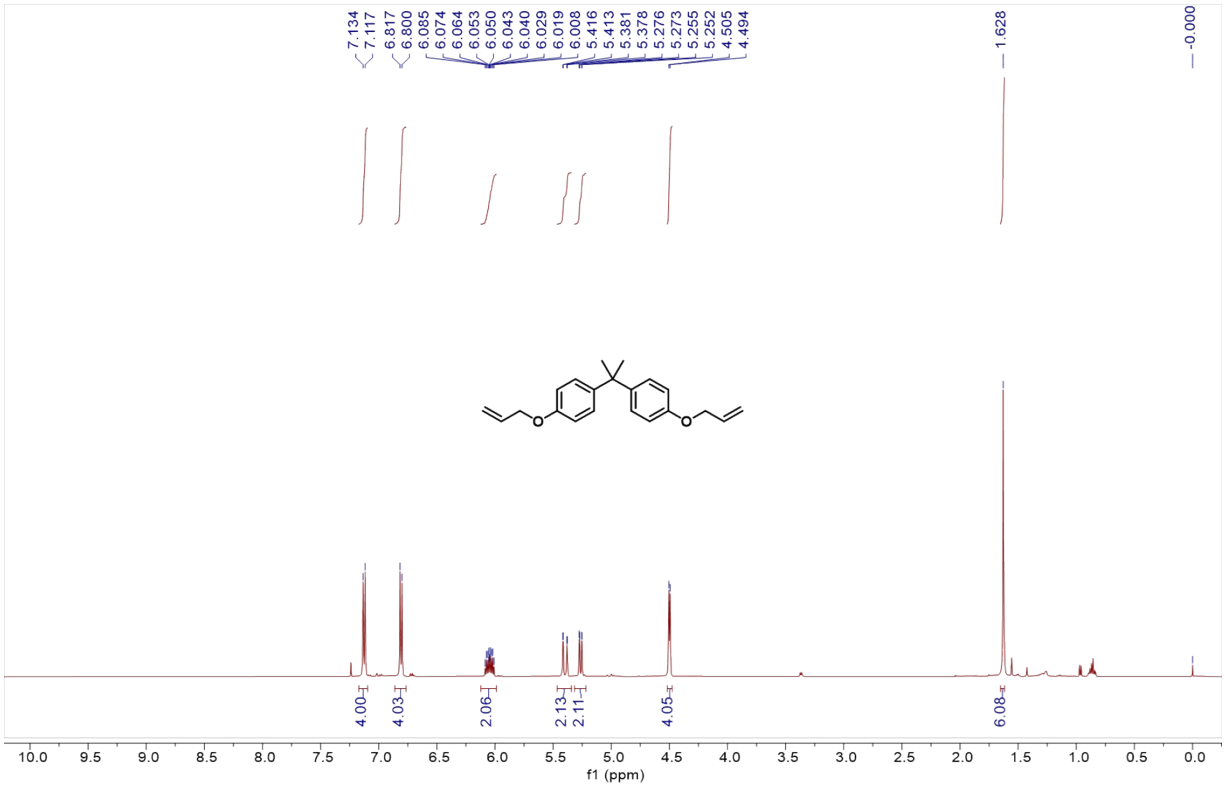


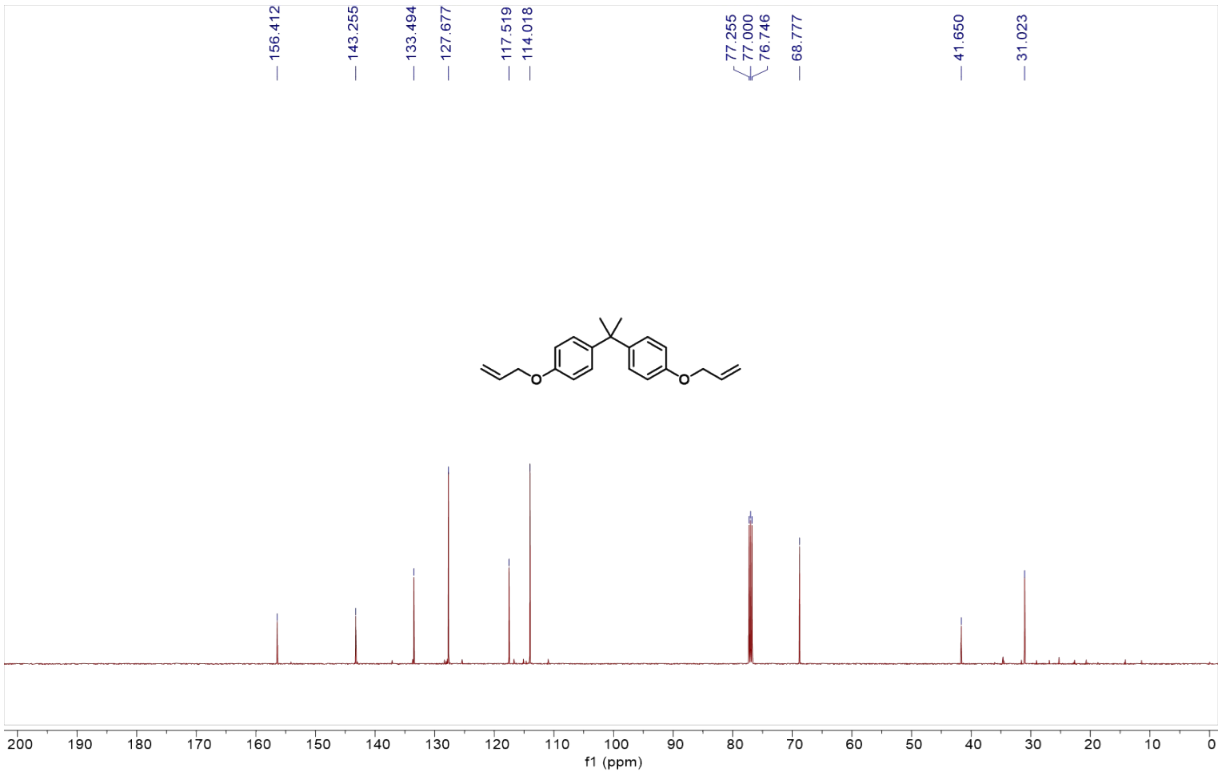
^{1}H NMR of **3d** (600 MHz, $CDCl_3$) and ^{13}C NMR of **3f** (151 MHz, $CDCl_3$)



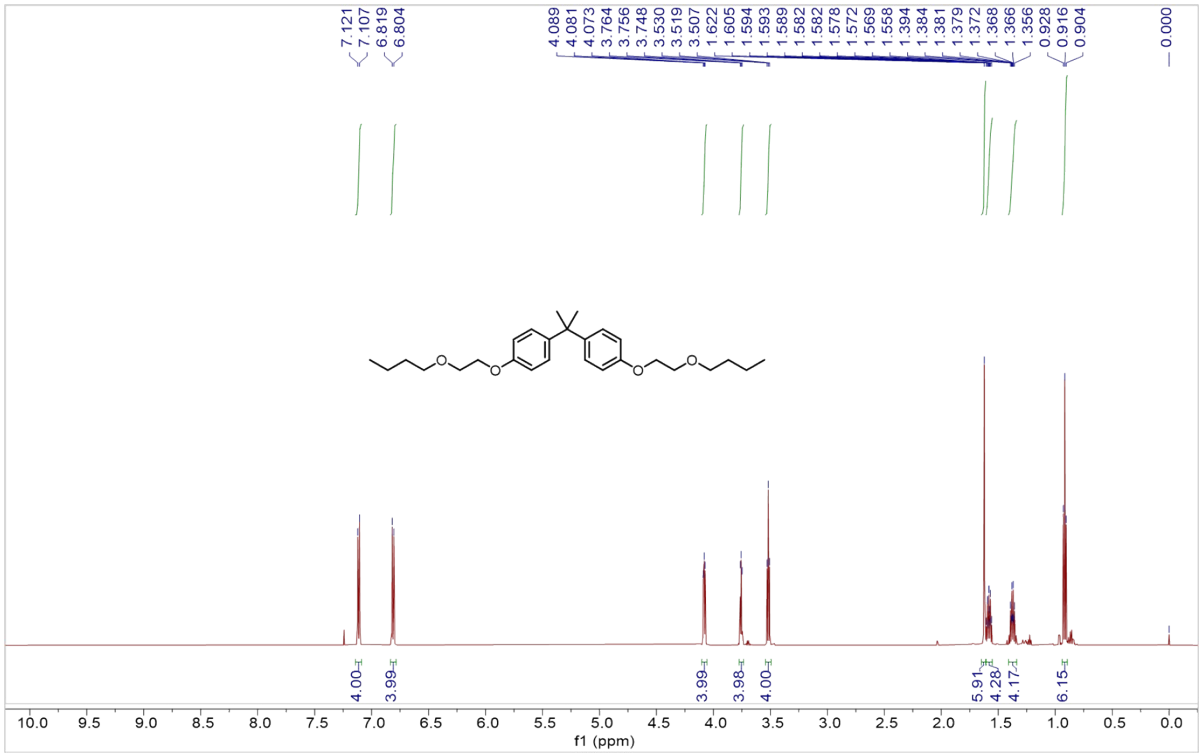


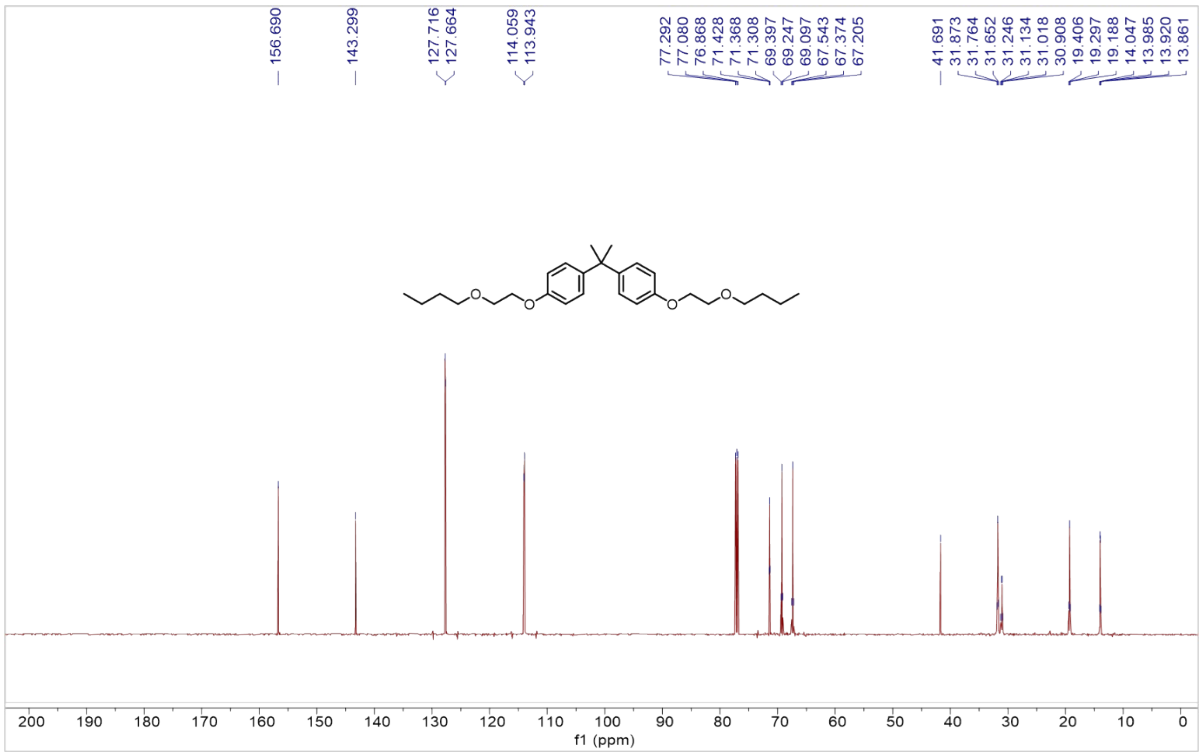
^{1}H NMR of **3e** (600 MHz, $CDCl_3$) and ^{13}C NMR of **3g** (151 MHz, $CDCl_3$)



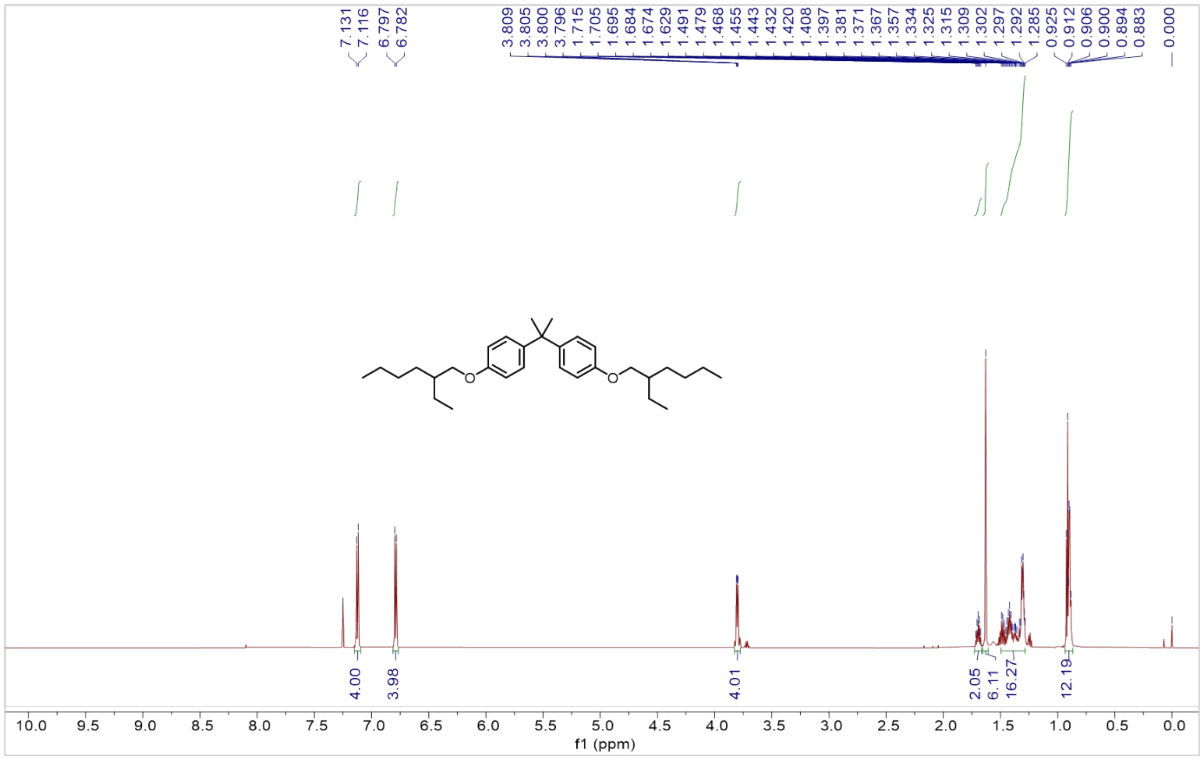


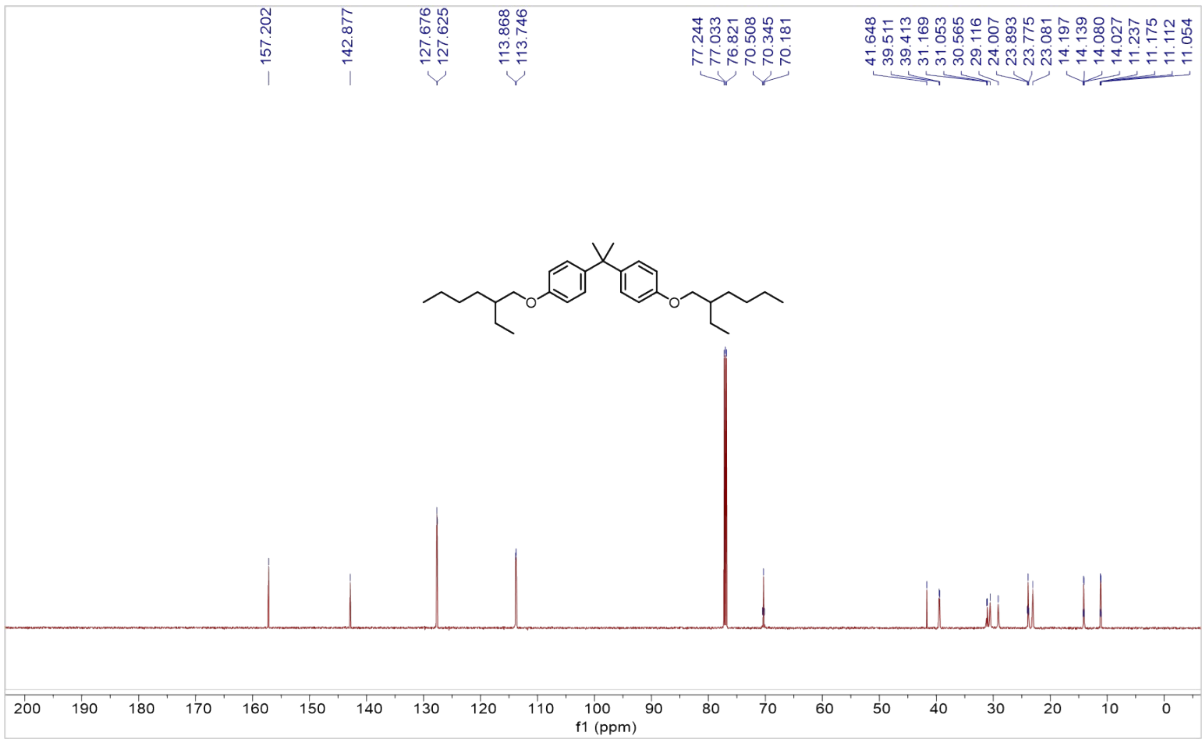
^{1}H NMR of **3f** (600 MHz, $CDCl_3$) and ^{13}C NMR of **3d** (151 MHz, $CDCl_3$)





^{1}H NMR of **3g** (600 MHz, $CDCl_3$) and ^{13}C NMR of **3e** (151 MHz, $CDCl_3$)





S8. Cartesian coordinates for optimized structure.

TS 1

C	-0.65385	-4.9947	6.28901
C	-1.88299	-4.47713	5.85555
C	-1.9891	-3.74659	4.67747
C	-0.84253	-3.44783	3.85964
C	0.38732	-4.04746	4.30451
C	0.47282	-4.77638	5.48448
H	-0.58041	-5.55953	7.21143
H	-2.77512	-4.65009	6.45311
H	-2.94716	-3.34716	4.35984
H	1.27589	-3.87615	3.70228
H	1.43436	-5.18341	5.78841
O	-0.90661	-2.70099	2.82562
P	-1.99169	0.47202	-0.32052
O	-2.2562	1.1045	0.99636
O	-3.28863	0.37523	-1.28224
O	-1.53699	-1.06525	-0.27565
O	-1.02181	1.21429	-1.33612
C	-4.40485	-0.40336	-0.80444
H	-4.75984	-0.02429	0.15669
H	-5.19349	-0.30113	-1.54833
H	-4.12621	-1.4556	-0.71005
C	-1.19329	-1.86915	1.26449
H	-0.90526	-2.88804	1.0077
H	-0.37403	-1.35099	1.76202
H	-2.08866	-1.88918	1.88899
C	-0.2382	2.38081	-0.96462
H	-0.74414	3.26462	-1.35831
H	-0.11194	2.43444	0.11121
H	0.73636	2.2656	-1.43607

TS 2

C	-0.65385	-4.9947	6.28901
C	-1.88299	-4.47713	5.85555
C	-1.9891	-3.74659	4.67747
C	-0.84253	-3.44783	3.85964
C	0.38732	-4.04746	4.30451
C	0.47282	-4.77638	5.48448
H	-0.58041	-5.55953	7.21143
H	-2.77512	-4.65009	6.45311
H	-2.94716	-3.34716	4.35984
H	1.27589	-3.87615	3.70228
H	1.43436	-5.18341	5.78841
O	-0.90661	-2.70099	2.82562
P	-1.99169	0.47202	-0.32052
O	-2.2562	1.1045	0.99636
O	-3.28863	0.37523	-1.28224

O	-1.53699	-1.06525	-0.27565
O	-1.02181	1.21429	-1.33612
C	-4.40485	-0.40336	-0.80444
H	-4.75984	-0.02429	0.15669
H	-5.19349	-0.30113	-1.54833
H	-4.12621	-1.4556	-0.71005
C	-1.19329	-1.86915	1.26449
H	-0.90526	-2.88804	1.0077
H	-0.37403	-1.35099	1.76202
H	-2.08866	-1.88918	1.88899

Int 1

C	-5.34279	1.04634	0.46337
C	-4.07112	1.56613	0.23934
C	-2.99238	0.72941	-0.05832
C	-3.19278	-0.65387	-0.13135
C	-4.47246	-1.18243	0.09416
C	-5.53466	-0.33673	0.38764
H	-6.17279	1.70378	0.69368
H	-3.90452	2.63644	0.29434
H	-2.01442	1.15816	-0.22906
H	-4.61355	-2.25535	0.03285
H	-6.51881	-0.75924	0.55811
O	-2.2139	-1.55321	-0.41149
P	2.60076	0.17034	-0.14149
O	1.92748	0.36101	1.19894
O	3.99487	1.08215	-0.17376
O	1.83255	0.34837	-1.42152
O	3.31635	-1.30937	-0.22452
C	3.91492	2.44775	0.22202
H	3.56708	2.53542	1.2584
H	4.92603	2.86537	0.14495
H	3.24255	3.02007	-0.42937
C	-0.8923	-1.05425	-0.66662
H	-0.87598	-0.41422	-1.5509
H	-0.27717	-1.93445	-0.847
H	-0.49553	-0.50547	0.18978
C	4.08701	-1.7709	0.88754
H	4.93389	-1.1055	1.08106
H	3.47273	-1.8431	1.79006
H	4.46342	-2.76196	0.6263

Int 2

C	-4.37673	1.37017	0.27158
C	-3.03871	1.61759	-0.02833
C	-2.13248	0.56911	-0.20113
C	-2.57318	-0.75401	-0.07134
C	-3.92062	-1.00772	0.23161
C	-4.80982	0.04666	0.39874
H	-5.07205	2.19089	0.40287

H	-2.6829	2.63823	-0.13249
H	-1.09733	0.78768	-0.44045
H	-4.24945	-2.03318	0.31686
H	-5.84697	-0.16534	0.62335
O	-1.77322	-1.83675	-0.22541
P	2.9563	-0.1199	0.21003
O	1.90207	-0.22731	1.33576
O	2.84028	1.51701	-0.33254
O	2.61086	-0.96679	-1.0329
O	4.42264	-0.19066	0.66999
C	1.57564	1.95281	-0.77243
H	0.84996	2.01289	0.05547
H	1.68526	2.95674	-1.2094
H	1.16169	1.28307	-1.54458
C	-0.39443	-1.62265	-0.58841
H	-0.33641	-1.14325	-1.57192
H	0.04284	-2.61908	-0.64673
H	0.15081	-1.03308	0.15313

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