SUPPLEMENTARY INFORMATION

Novel strategies for recycling poly(butylene adipate-co-terephthalate)-starch-based plastics: selective solubilization and depolymerization-repolymerization process

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S1. Screening condition for selective solubilization experiment

Table S1. Selective solubilization of PBAT screening: solvents temperature and time

Entry	Conditions					Yield (%)[a]		
	Temp (°C)	n° extr.	time (min)	Solvent	Concentration (wt %)	Starch	PBAT	Total recovery[b]
1	rt	1	30	CH_2Cl_2	4%	30	62	97.5
2	rt	1	120	EtOAc	3%	No separation observed		
3	rt	1	120	γ-VL	3%	No separation observed		
4	rt	1	120	acetone	3%	No separation observed		
5	rt	1	120	2-MeTHF	3%	No separation observed		
6	rt	1	120	Et-lactate	3%	No separation observed		
7	rt	1	120	DMC	3%	No separation observed		
8	50	1	120	DMC	3%	No separation observed		
9	90	1	120	γ-VL	3%	27	50	82.5
10	90	1	120	DMC	3%	33	43	81.8
11	90	2	25	DMC	5%	13.6	80.9	-
12	90	2	25	acetone	5%	52.4	40	-
13	90	2	25	2-MeTHF	5%	18.2	76.5	-
14	90	2	25	Et-lactate	5%	No separation observed		

[a] on total input material

[b] considering sorbitol (≈5.5%)

S2. Screening conditions for depoly-repoly technique

Table S2 Depoly-repoly technique and monomers screening of catalysts, time and temperature

Entry	Depoly Conditions[a]				Yield (%)[b]		
	Temp (°C)	time (h)	Cat.	Cat. loading (mol%)	Starch fraction	PBAT	Total recovery[c]
1	140	5	-	-	91	-	-
2	140	5	H_2SO_4	3	4	-	-
3	140	5	K_2CO_3	3	37	0	37
4	140	7	K_2CO_3	3	30.1	0	30
5	140	5	NaOH	3	45	0	45
6	140	7	NaOH	3	37	0	37
7	140	7	NaOH	4	30	0	30
8	140	7	DBU	3	40	0	40
9	140	7	DBU	5	31	0	31

[a] conc. (wt%) of SBP in MeOH was 20% for all entries

[b] on total input material

[c] considering sorbitol (≈5.5%)

S3. Screening catalysts conditions for monomer recovery

Table S3 Monomers recovery: screening of catalysts, time and temperature

Entry	Methanolysis Conditions[a]			Monomers conversion (%)[b]
	Cat.	Cat. loading (mol%)	Conc. (wt %)	
1	DBU	10	20	95.2
2	NaOH	4	10	91.1
3	K_2CO_3	3	10	93.5
4	$Zn(OAc)_2$	3	10	95.1
5	$(But)_2SnO$	0.5	20	94.2
6	$Sn(But)_2(OAc)_2$	0.5	20	95.8

[a] all reactions have been conducted at 140 °C; 7h

[b] obtained by GC-MS analysis

S4. Reagents and solvents recovery yields for E-factor quantification

Table S4 Materials used in the three processes recovery yield

Process	Recovery yield (%)[a]				
	Overall product	EtOAc	MeOH	Cy	H_2O
Sorbitol removal[b]	-	Not used	99	Not used	Not used
Selective solubilization	98.6	99	Not used	Not used	Not used
Depoly-Repoly process	95	Not used	98	Not used	Not used
Monomer recovery	93.5	Not used	93[c]	98	99

[a] solvents comprise also the fraction used for the washing step and were recovered through distillation under ambient pressure; catalysts used for depoly-repoly and monomer recovery processes accounts for the 0.02-0.03 wt% respect all the materials in input and they were not recovered

[b] sorbitol removal is the pretreatment step, common for all the following processes

[c] 4% is of the not recovered MeOH is actually lost, 3% is incorporated in the final products DMT and DMA

S5. DMTA spectra recorded in the 0 - 70°C range, showing E' for the analyzed polymers: A) E1 (—), B) E2 (—), C) E3 (—), D) E4 (—), E) R3 (—).

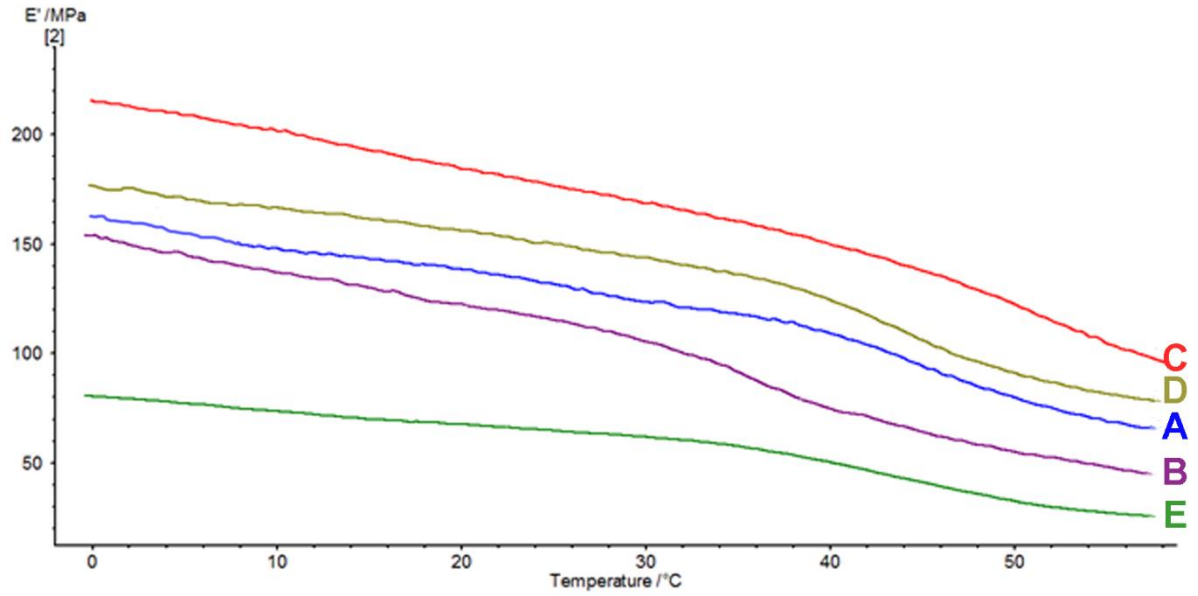


Figure S5 DMTA spectra recorded in the 0 - 70°C range, showing E' for the analyzed polymers: A) E1 (—), B) E2 (—), C) E3 (—), D) E4 (—), E) R3 (—).

S6. GC-MS analysis of recovered sorbitol

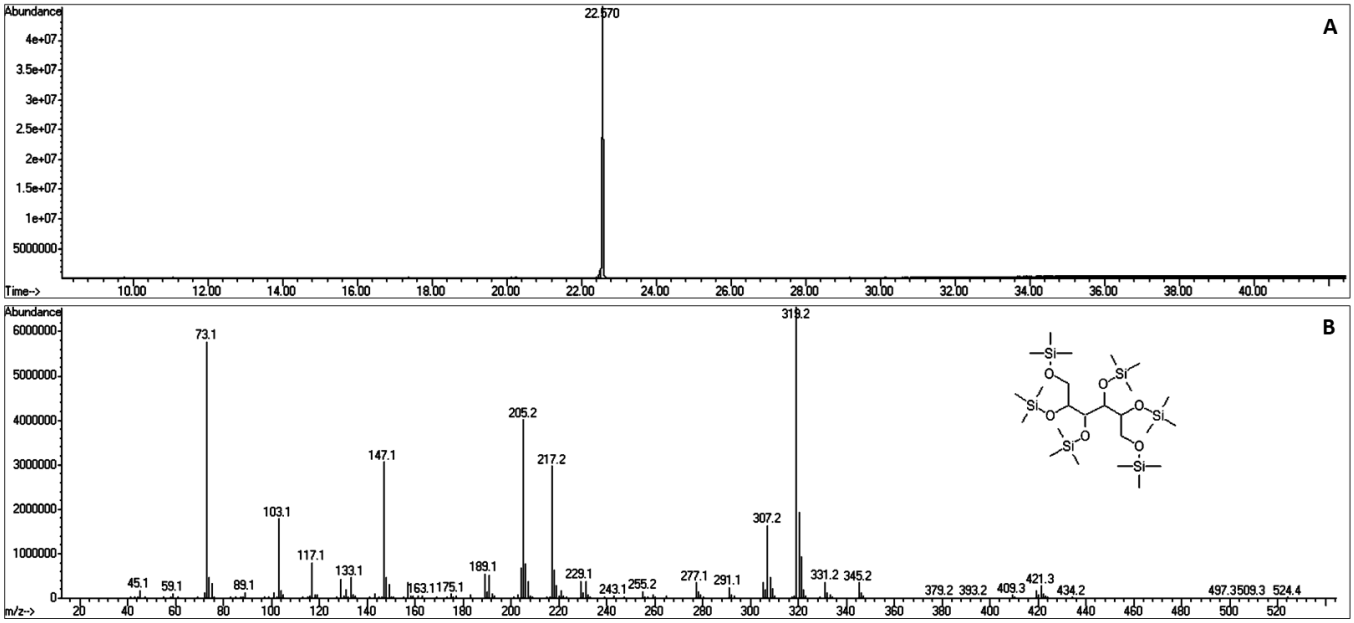
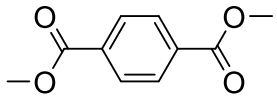
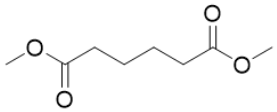


Figure S6 Analysis of the extracted fraction of PBAT with MeOH: A) chromatograph with retention time; B) mass spectrum of the peak at retention time of 22.57, corresponding to silylated sorbitol.

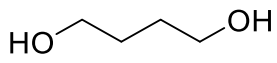
S7. ^{1}H NMR and ^{13}C NMR of monomers recovered through selective depolymerization



DMT: White solid (f.p.:140-142 °C). **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.08 (s, 4H), 3.93 (s, 6H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 166.24, 133.87, 129.51, 52.39.



DMA: Colorless liquid. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 3.65 (s, 6H), 2.37 – 2.25 (m, 4H), 1.69 – 1.60 (m, 4H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 173.71, 51.50, 33.64, 24.34.



1,4-BD: Colorless liquid. **^{1}H NMR** (400 MHz, D_2O) δ 3.53 – 3.40 (m, 4H), 1.49 – 1.37 (m, 4H). **^{13}C NMR** (100 MHz, D_2O) δ 61.41, 27.74.

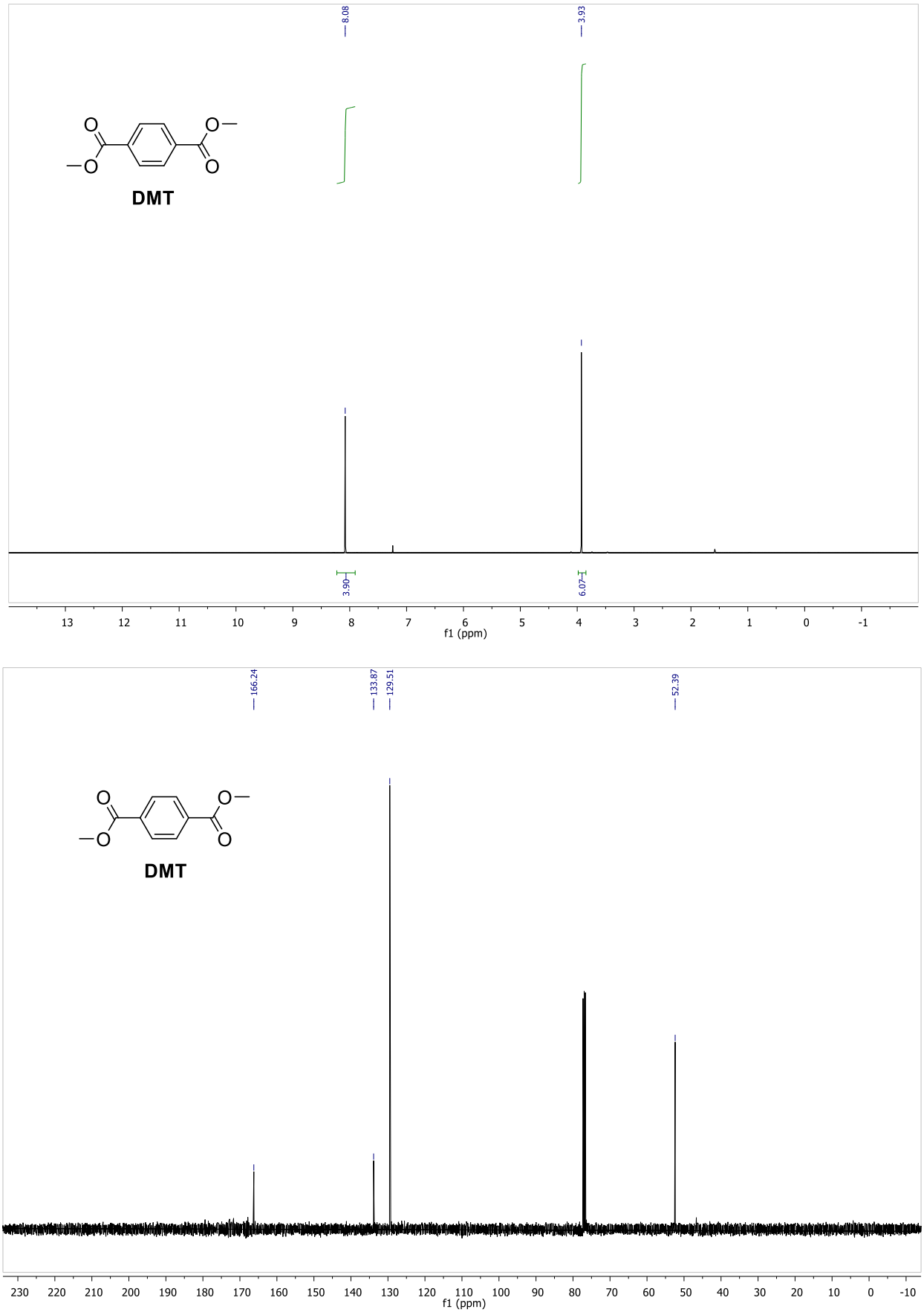
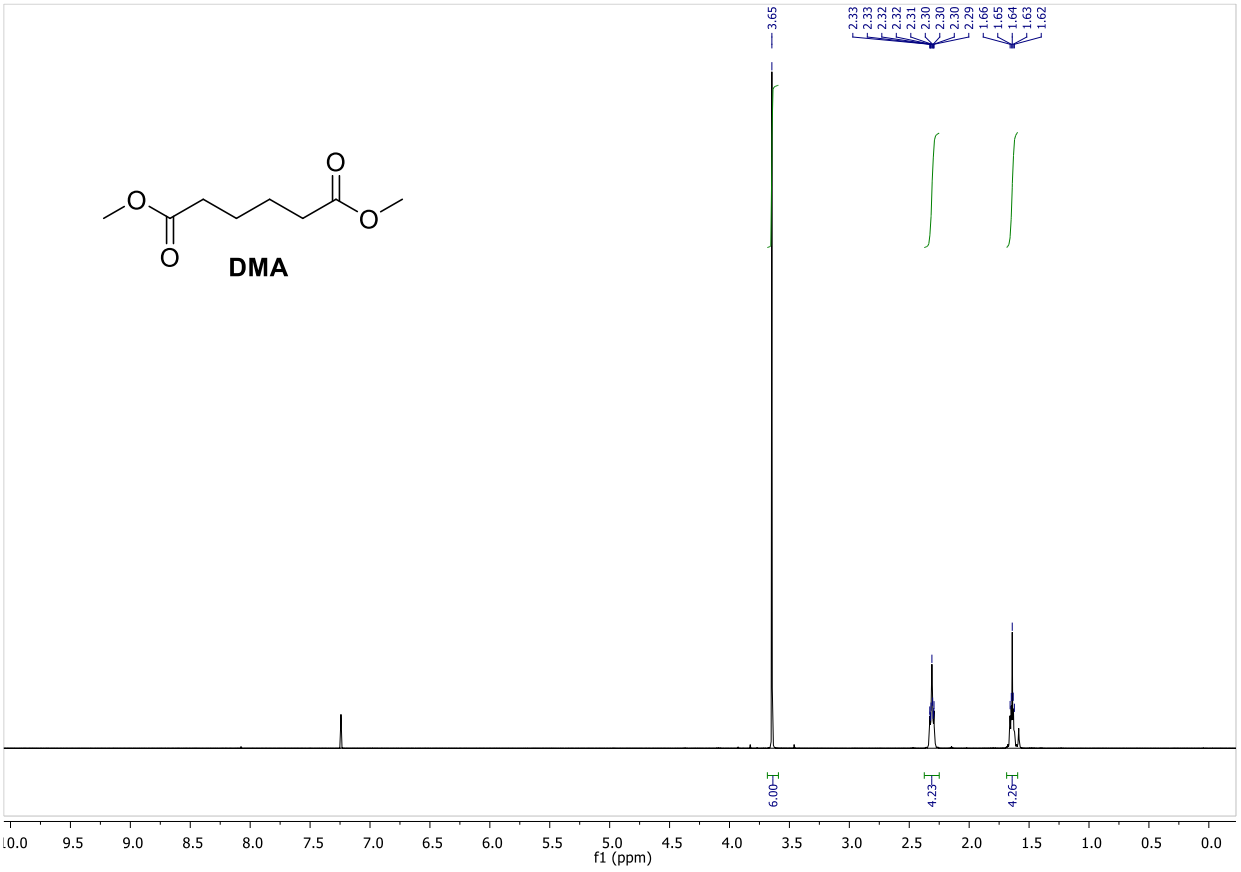


Figure S7a ^{1}H NMR (up) and ^{13}C NMR (down) of DMT.



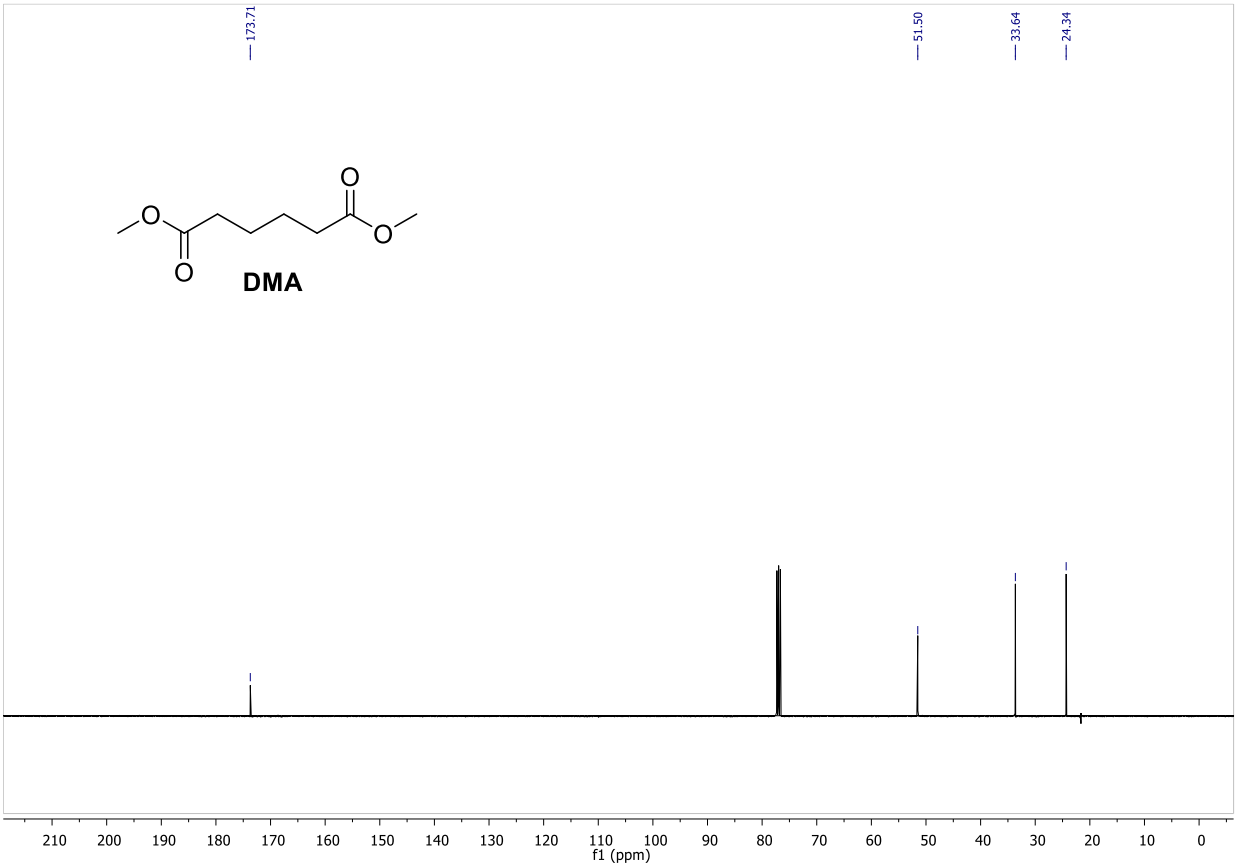


Figure S7b ^{1}H NMR (up) and ^{13}C NMR (down) of DMA.

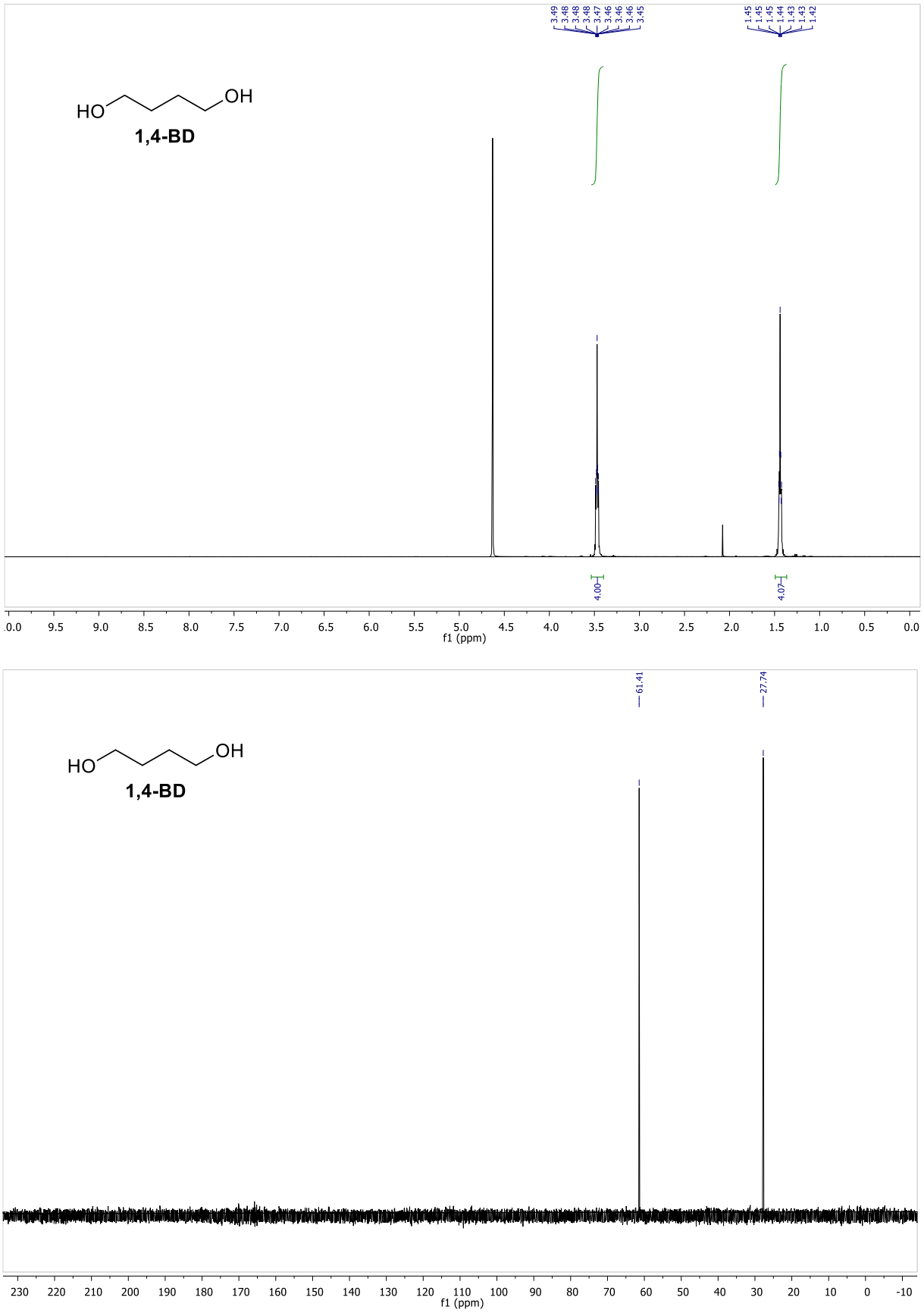


Figure S7c ^{1}H NMR (up) and ^{13}C NMR (down) of 1,4-BD.