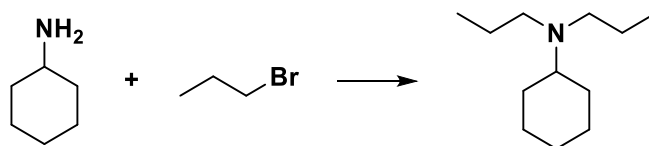


# Recycling of multilayer flexible packaging waste through delamination with recoverable Switchable Hydrophilicity Solvents

Roberta Mastroddi, Chiara Samorì, Martina Vagnoni, Chiara Gualandi, Paola Galletti and Emilio Tagliavini

**Figure S1.** General procedure for the synthesis of *N,N*-dipropylcyclohexylamine and characterization.



In a two-necked round bottom flask under nitrogen atmosphere and flame dried, cyclohexylamine (15 mmol, 1eq), acetonitrile (50 ml) and  $K_2CO_3$  (45 mmol, 3 eq) were added and stirred at room temperature for 15 minutes. Then propylbromide (45 mmol, 3 eq) was added dropwise and the solution was refluxed at 70°C overnight. The reaction was monitored through GC-MS analysis. After the completion, the reaction was quenched with water and extracted three times with a 9:1  $CH_2Cl_2/MeOH$  mixture; the organic layers were combined and then washed with brine, filtered and concentrated under reduced pressure to afford *N,N*-dipropylcyclohexylamine as a light yellow liquid (76% yield, 7.6 mmol, 1.g).

GC-MS\*:  $t_r = 10.70$  min,  $m/z = 183$  (M+); 154; 140; 72.

$^1H$  NMR (400 MHz,  $CDCl_3$ ):  $\delta$  2.42 (tt,  $J = 10.1, 3.9$  Hz, 1H), 2.07 – 1.05 (m, 24H).

\* GC-MS spectra were recorded by EI ionization at 70 eV on a Hewlett-Packard 5971 with GC injection on a Zebron ZB-5MS column (30 m L x 0.25 mm ID x 0.25  $\mu$ m).

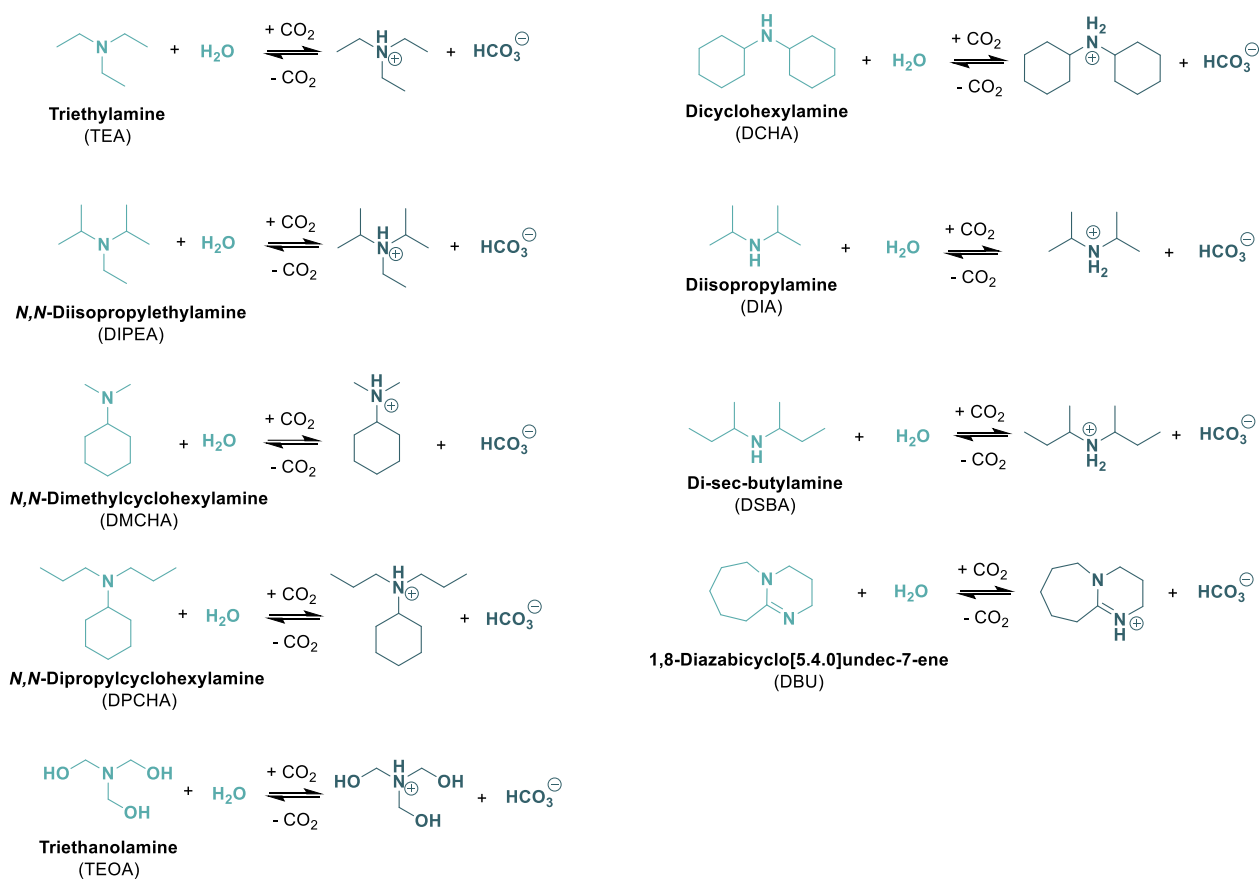
**Table S1.** Classification and features of the three waste categories treated.

Sample	Use	Composition
Group 1	snacks, biscuits, chips, dried fruits	Al < 4% (powder) PP ~ 98%
Group 2	food, animal feed, coffee bags	13 < Al < 20% (foil) PE (internal layer) PET (external layer) Polyurethane and polyacrylate (tie layer)
Group 3	food, animal feed	1 < Al < 20% (powder or foil) PE (internal layer) PET (external layer) Polyurethane and polyacrylate (tie layer)

**Table S2.** Delamination of sample **B** with DMCHA varying the sample loading.

Sample loading (wt%)	Delamination	Total time (h)	Additive solubilisation
1	Yes	2.5	Yes
1.5	Yes	2.5	Yes
2	Yes	2.5	Yes
3	Yes	2.5	Yes
4	Yes	3	No
5	Yes	3	No

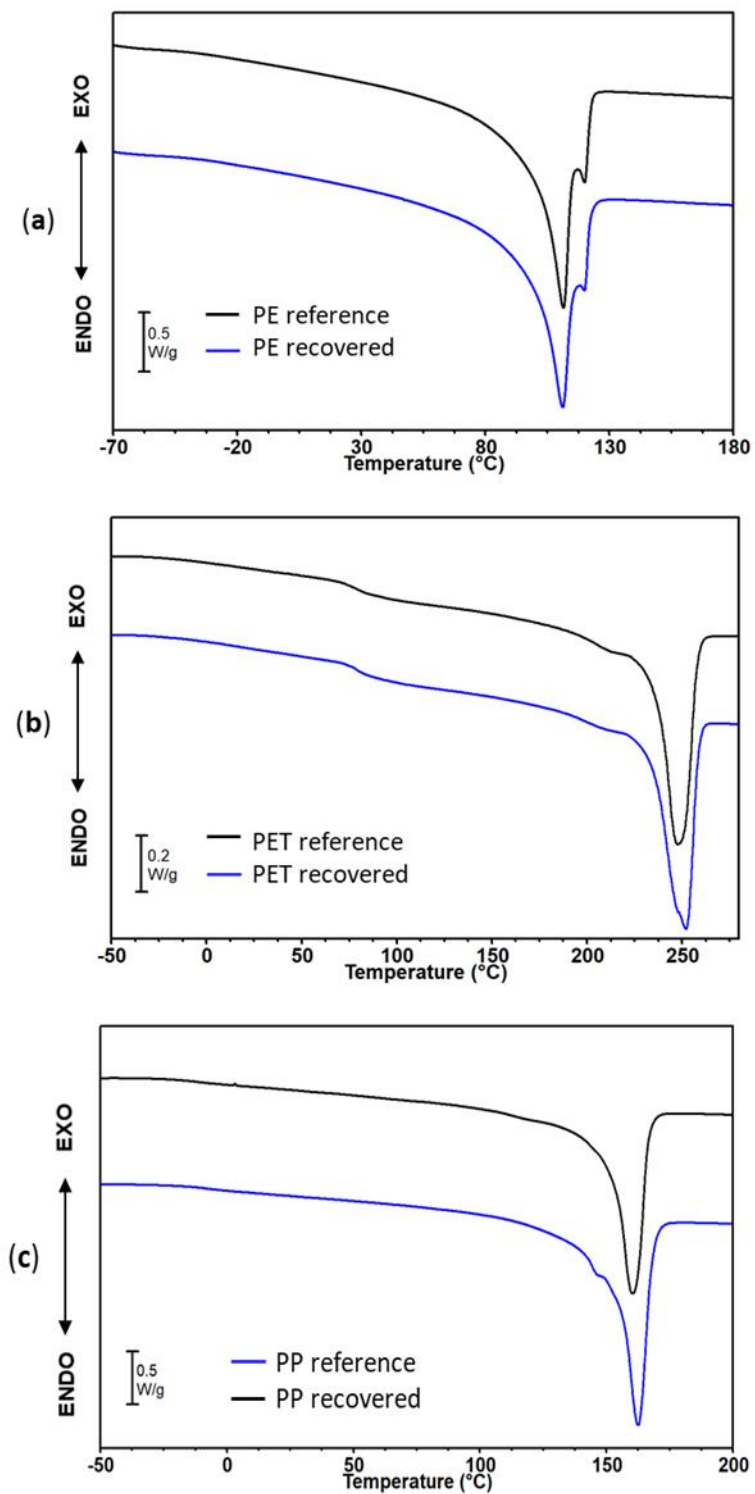
**Figure S1.** Chemical structures of the Switchable Solvents before and after CO<sub>2</sub> addition.



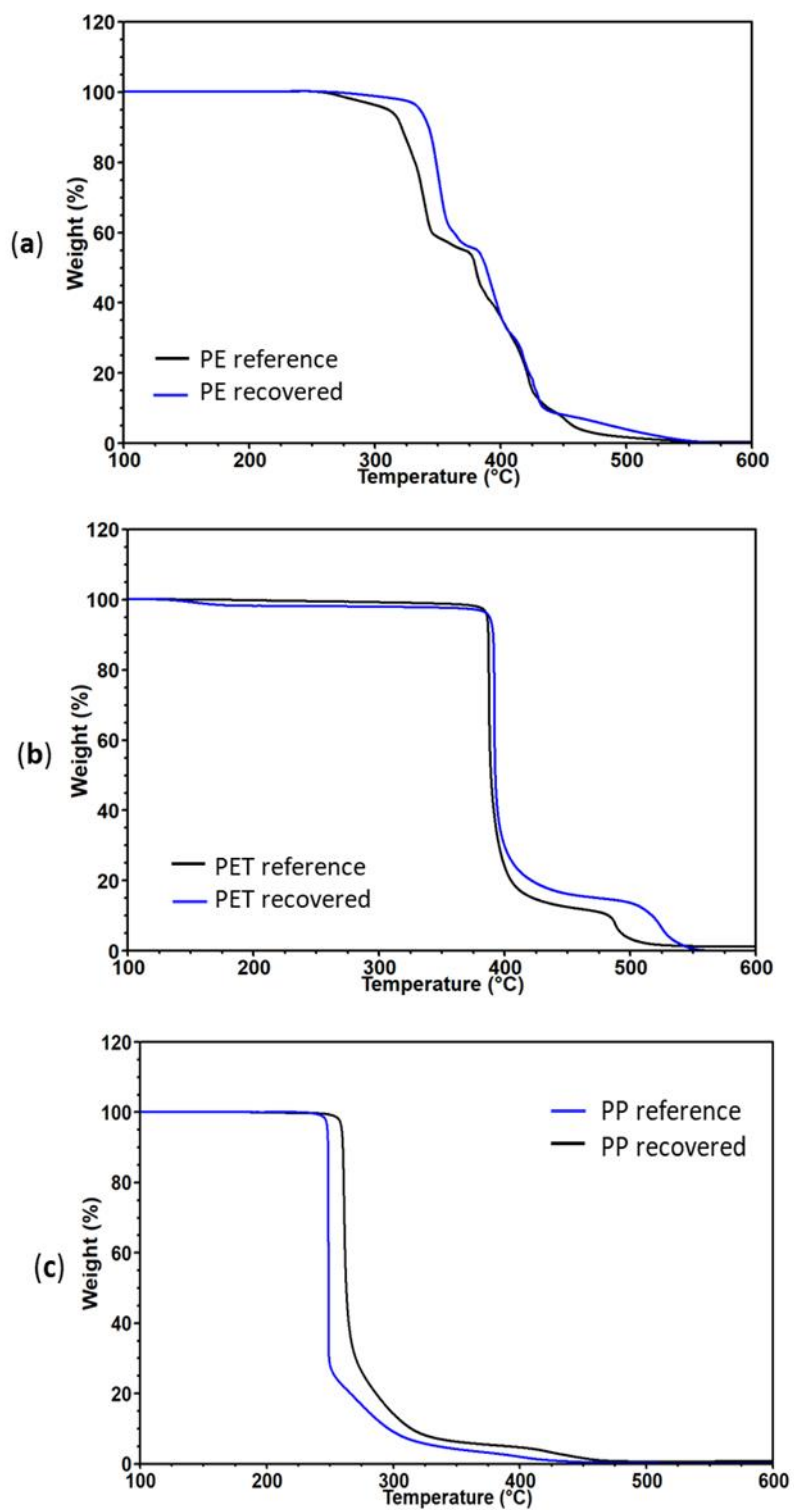
**Table S3.** Delamination of sample **B** with recycled DMCHA.

Cycle	Delamination	Total time (h)	Additive solubilisation
0	Yes	2.5	Yes
1	Yes	2.5	Yes
2	Yes	2.5	Yes
3	Yes	2.5	Yes
4	Yes	2.5	Yes
5	Yes	2.5	Yes
6	Yes	2.5	Yes
7	Yes	2.5	Yes

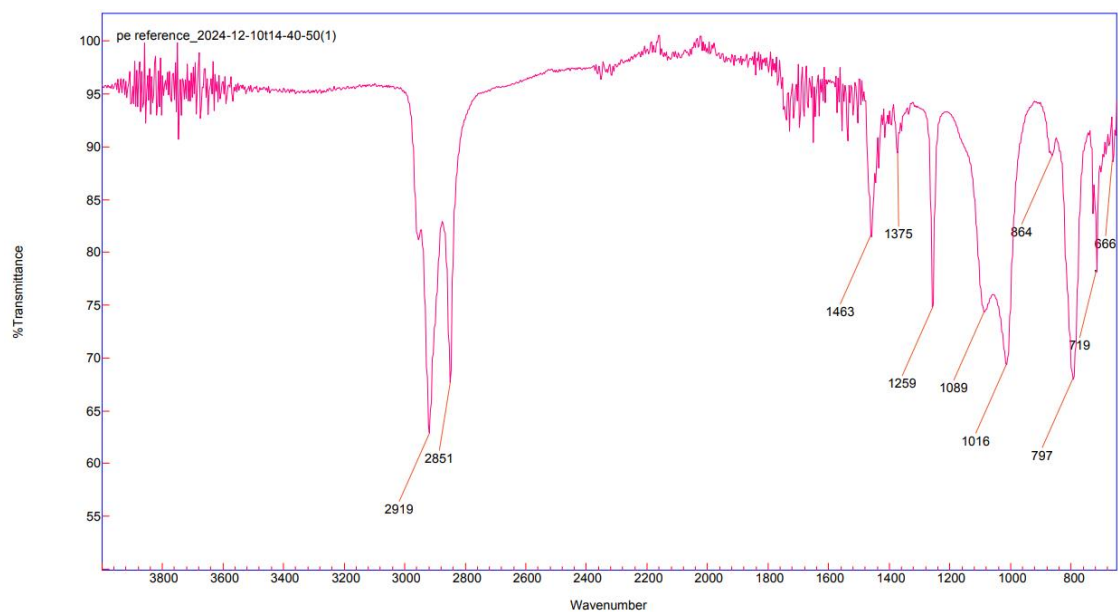
**Figure S3.** DSC curves of the recovered polymers and the corresponding references. (a) PE; (b) PET; (c) PP.



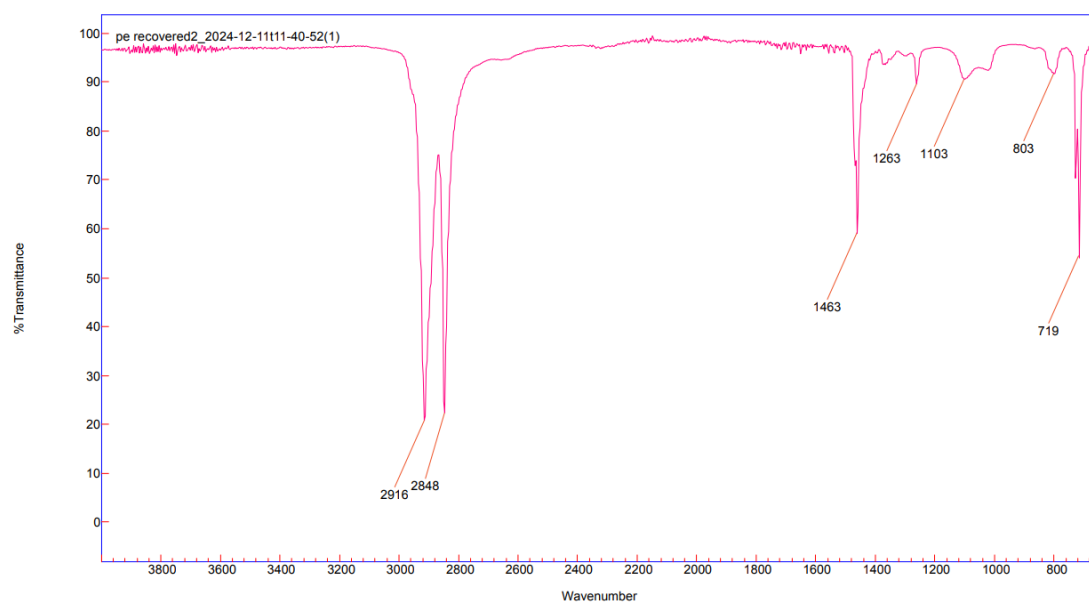
**Figure S4.** TGA curves of the recovered polymers and the corresponding references. (a) PE; (b) PET; (c) PP.



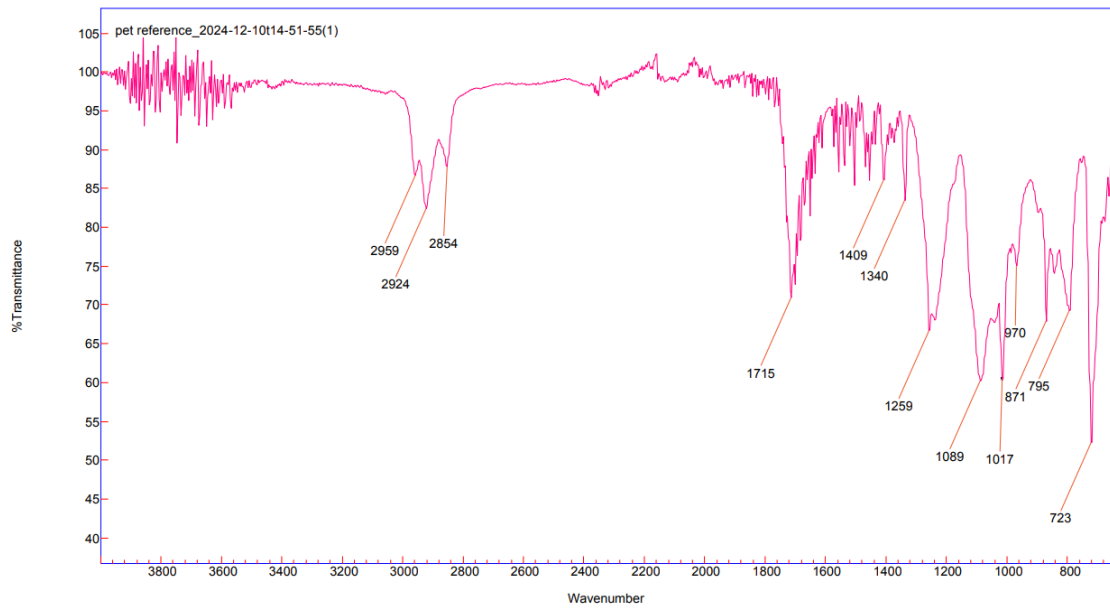
**Figure S5.** FTIR spectra of the recovered polymeric layers and the corresponding references. **(a)** PE reference; **(b)** PE recovered; **(c)** PET reference; **(d)** PET recovered; **(e)** PP reference; **(f)** PP recovered.



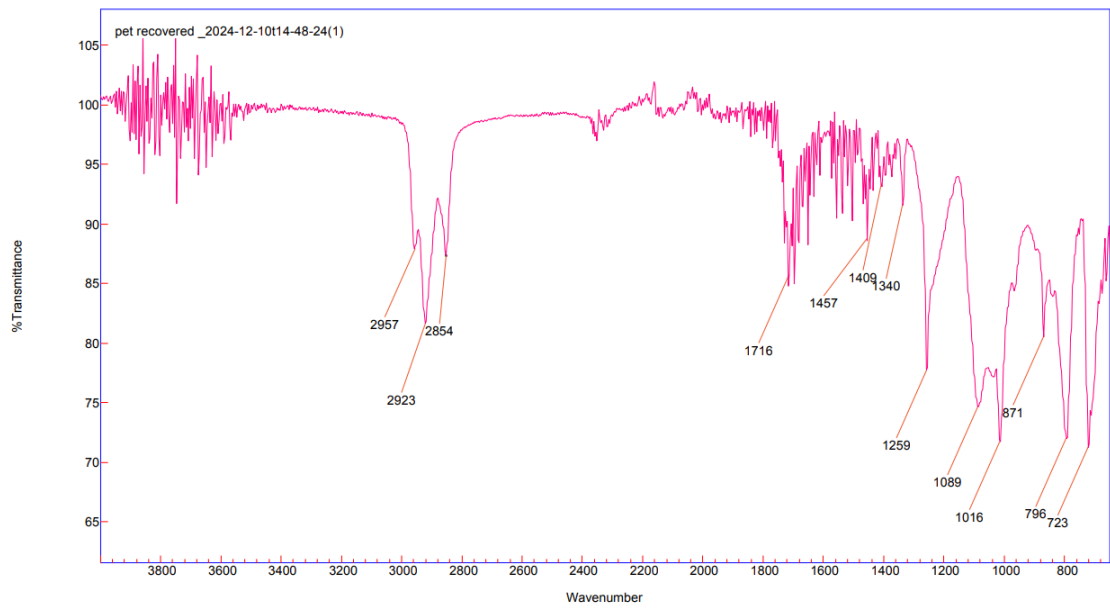
**(a)**



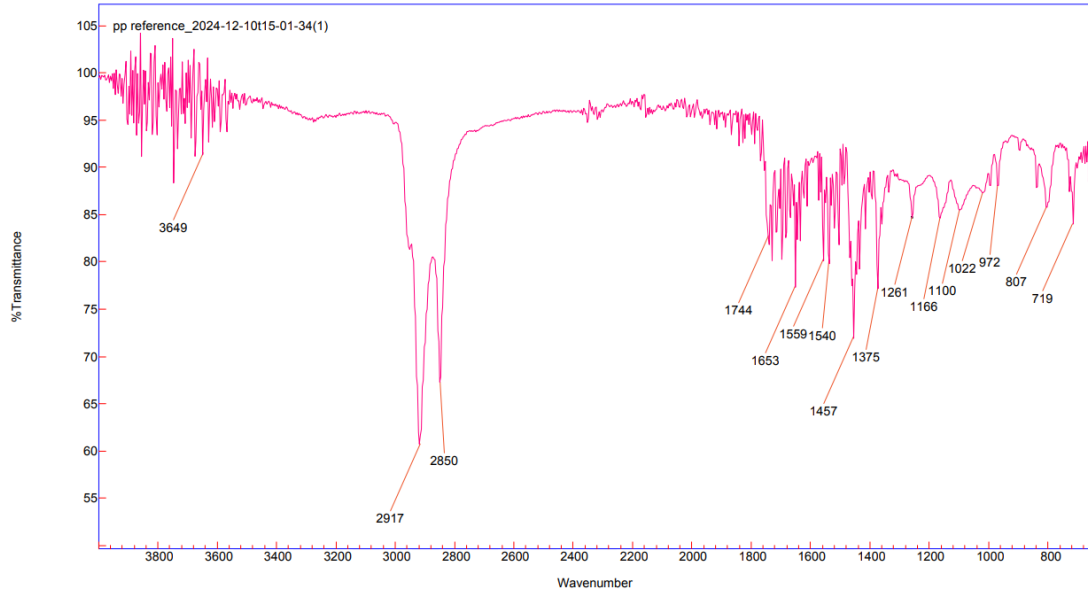
**(b)**



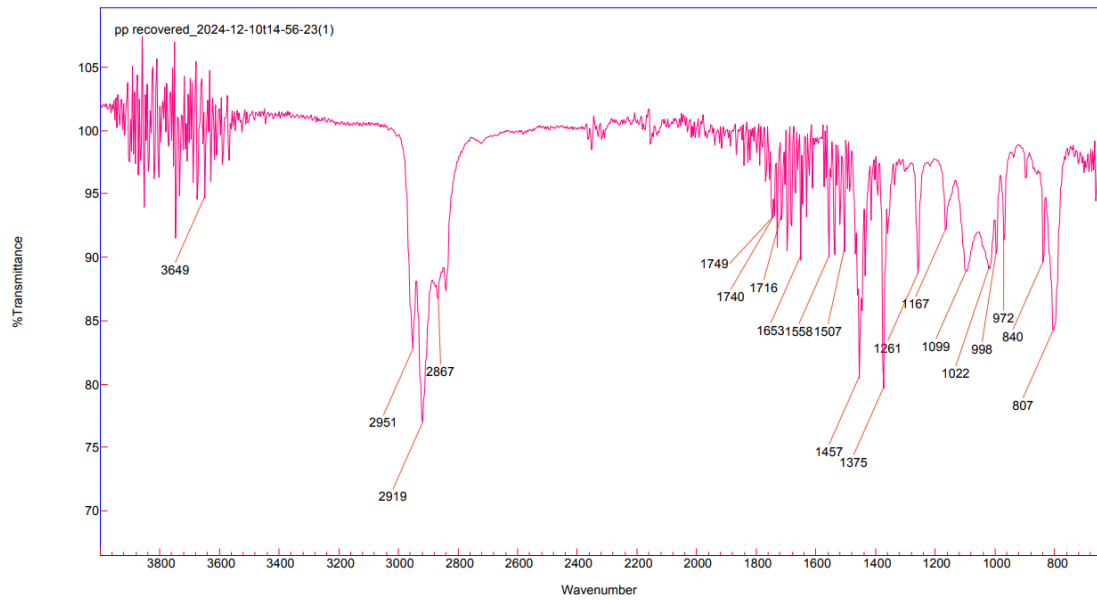
(c)



(d)



(e)



(f)

**Figure S6.**  $^1\text{H}$  NMR spectrum of recovered DMCHA after the switch and back-switch procedure.

