Supplementary Information (SI) for Green Chemistry.
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Supporting Information

Polyamide Depolymerization: Unlocking Polymer Dissolution with Dicarboxylic Acids

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1. State of the art: main advantages of this work compared to previous ones

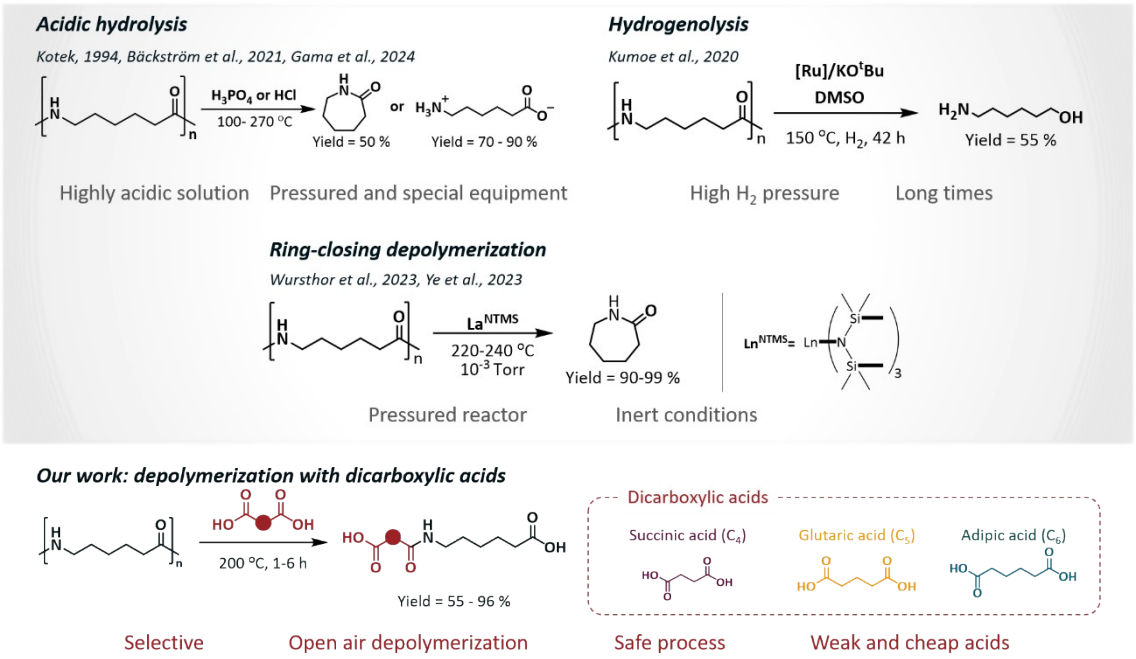

Figure S1. Approaches for chemical recycling of PA6.[1–6]

2. PA6-dicarboxylic acid interactions: analysis of the morphology and thermal structure

2.1. Crystallinity Studies

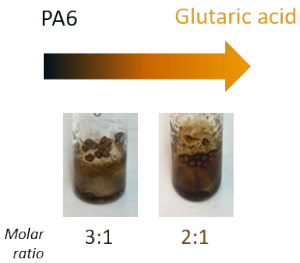

Figure S2. Pictures of the crude products of the dissolution of PA6 with Glutaric acid (1h, 150 °C).

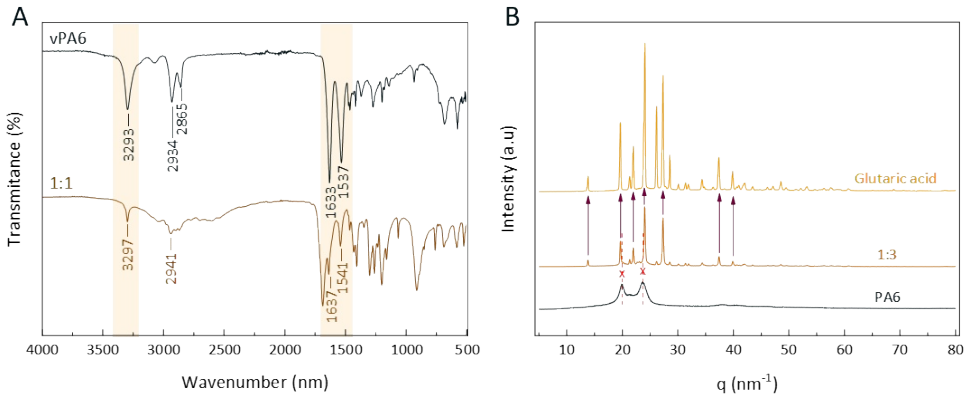

Figure S3. (A) FTIR analysis of the virgin PA6 and the dissolved PA6 in 1 molar equivalent of glutaric acid; (B) WAXS spectra of the mixture of PA6:Glutaric acid (1:3) compared to raw materials (Dissolution conditions: 1h, 150 °C).

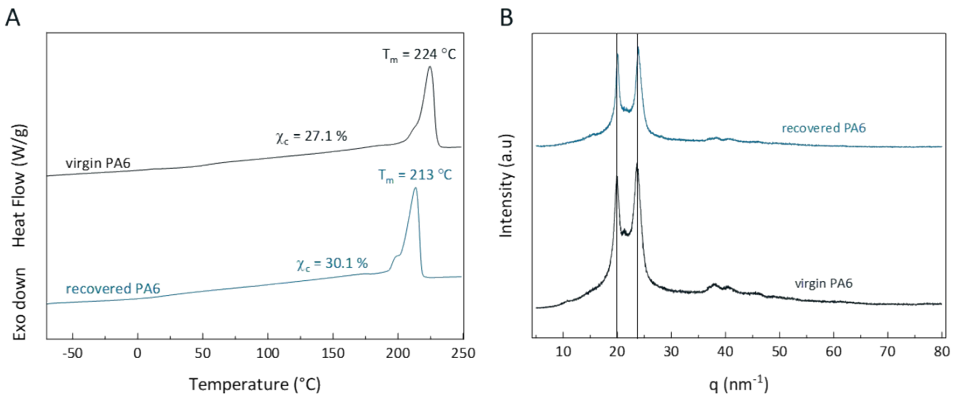

Figure S4. P6 recovered by precipitation in water from the dissolution in glutaric acid (Reaction conditions: PA6 to glutaric acid 1:3 molar eq., 1h, 150 °C). (A) DSC second heating scan of the recovered PA6 compared to virgin PA6 (-90 to 250 °C, 10 °C/min). (B) WAXS spectra of the recovered PA6 compared to the virgin PA6.

3. Depolymerization of PA6 with dicarboxylic acids

3.1. Effect of the acid chain length

Succinic acid (C_4)

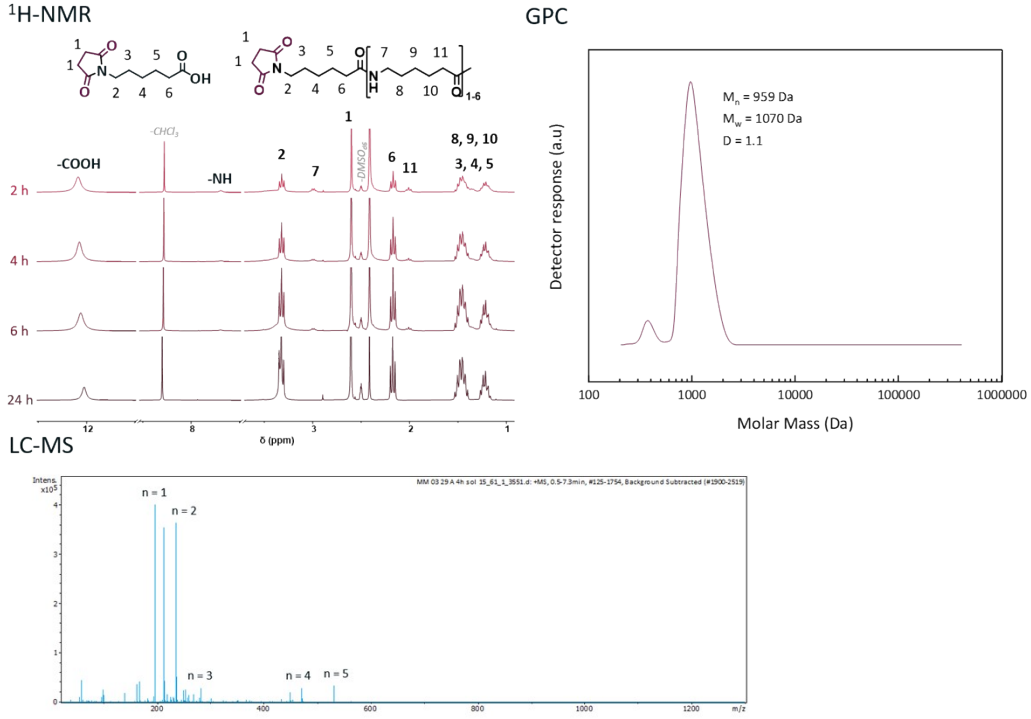

Figure S5. Soluble fraction of the depolymerization of PA6 with glutaric acid analyzed by ^{1}H-NMR over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS) and GPC (in water) and LC-UV-HRMS (in THF) at 4 hours.

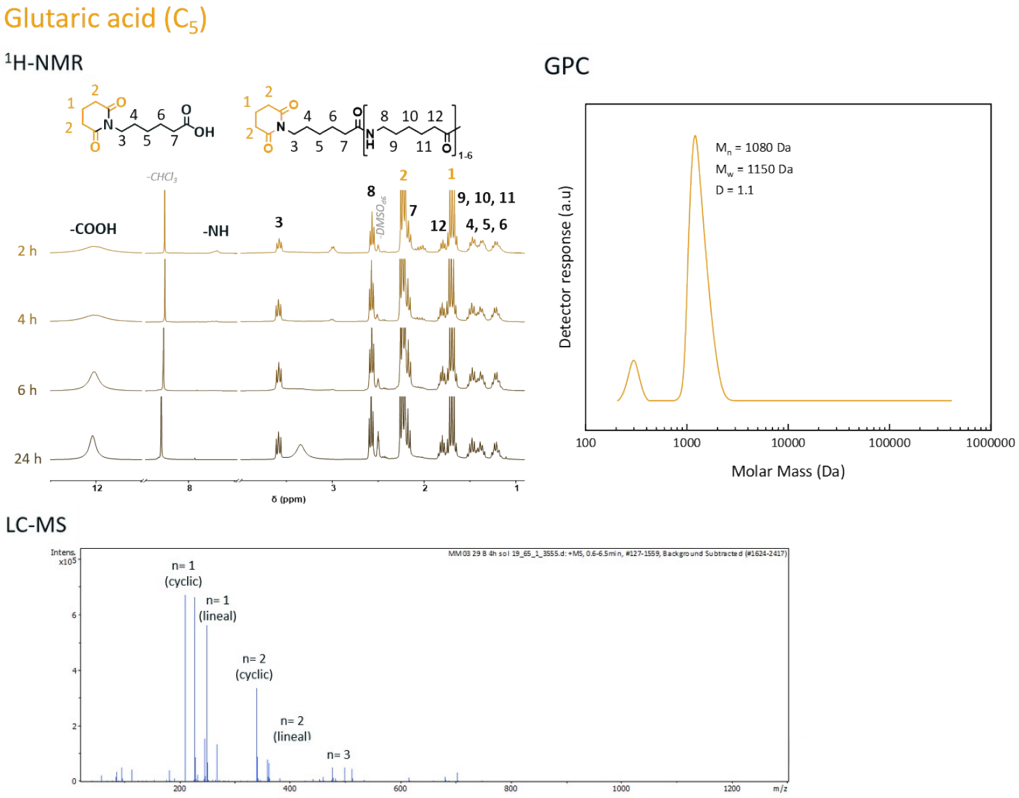

Figure S6. Soluble fraction of the depolymerization of PA6 with glutaric acid analyzed by ^{1}H-NMR over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS) and GPC (in water) and LC-UV-HRMS (in THF) at 4 hours.

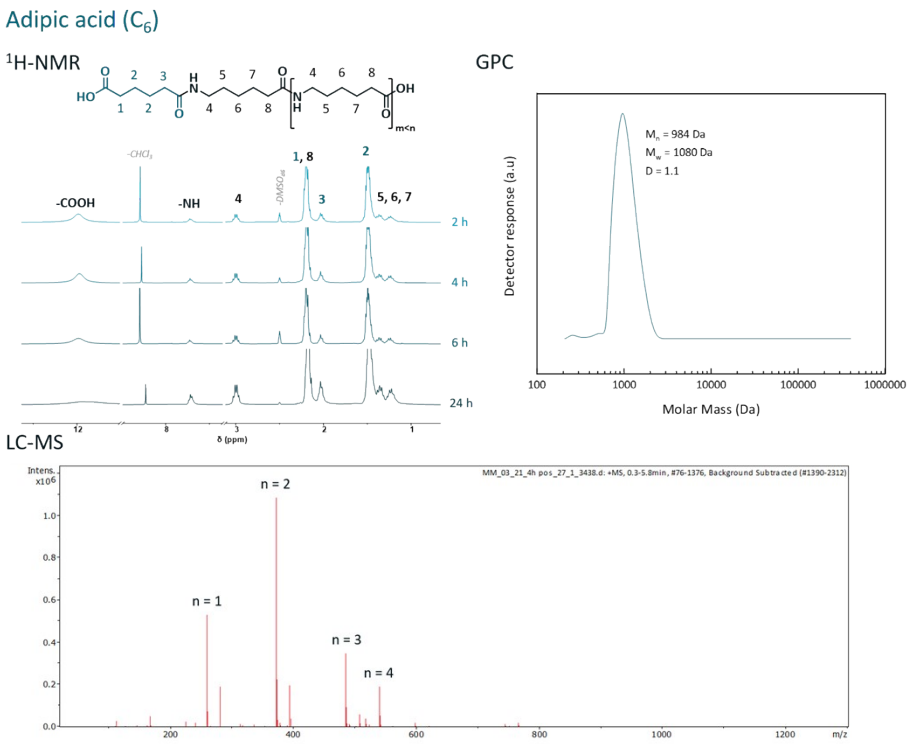

Figure S7. Soluble fraction of the depolymerization of PA6 with adipic acid analyzed by ^{1}H-NMR over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS) and GPC (in water) and LC-UV-HRMS (in THF) at 4 hours.

3.2. Screening of conditions

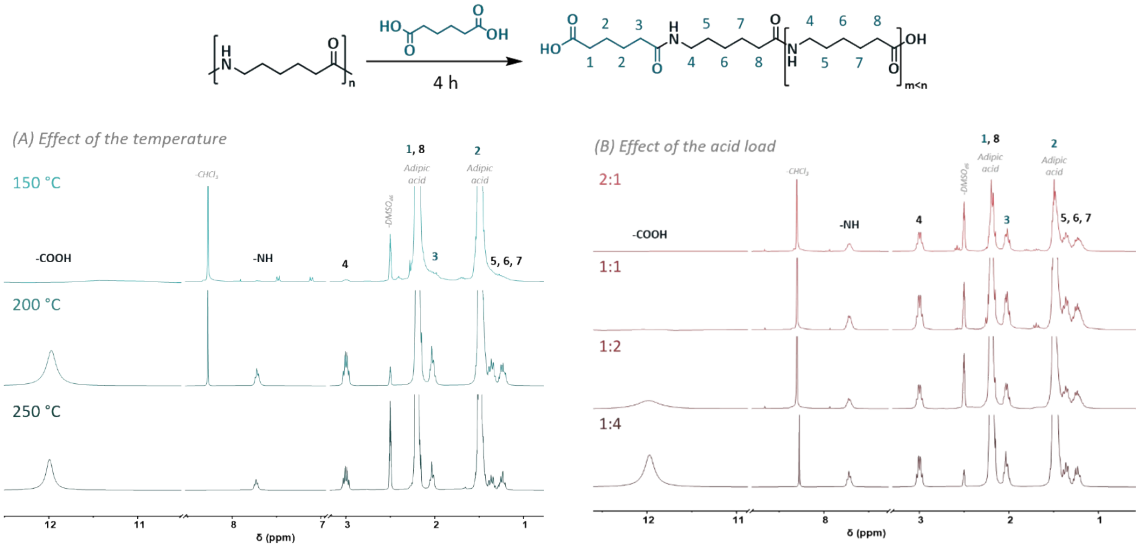

Figure S8. Effect of the temperature and acid load on the depolymerization of PA6 with adipic acid. A) Soluble fraction of the depolymerization of PA6 at different temperatures (1:4 molar eq. PA6:Adipic acid, 4 h) analyzed by ^{1}H-NMR over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS). B) Soluble fraction of the depolymerization of PA6 at different acid loads (200 °C, 4 h) analyzed by ^{1}H-NMR over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS).

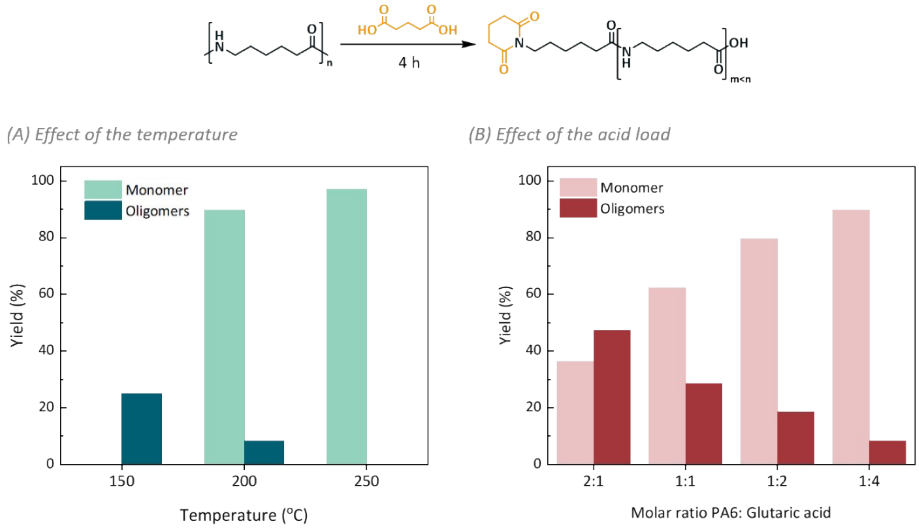

Figure S9. Effect of the temperature and acid load on the depolymerization of PA6 with glutaric acid. (A) Monomer and oligomer yield at different temperatures (ratio PA6 to acid 1:4, 4h); (B) Monomer and oligomer yield at different acid loads (4h at 200 °C). Yield of products was measured by ^{1}H-NMR (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS).

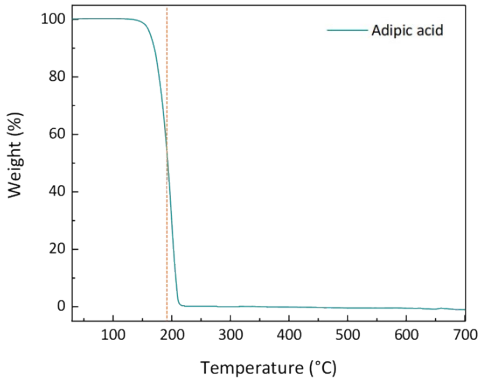

Figure S10. Thermogravimetric analysis of adipic acid.

4. Mechanism of PA6 depolymerization of with dicarboxylic acids

4.1. Model reactions

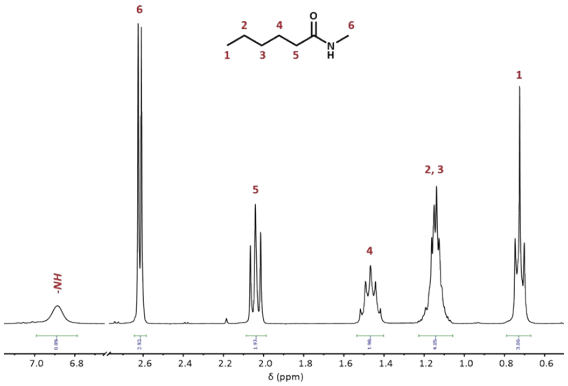

Figure S11. ^{1}H-NMR spectra of the N- methyl hexanamide (in $DMSO_{d6}$)

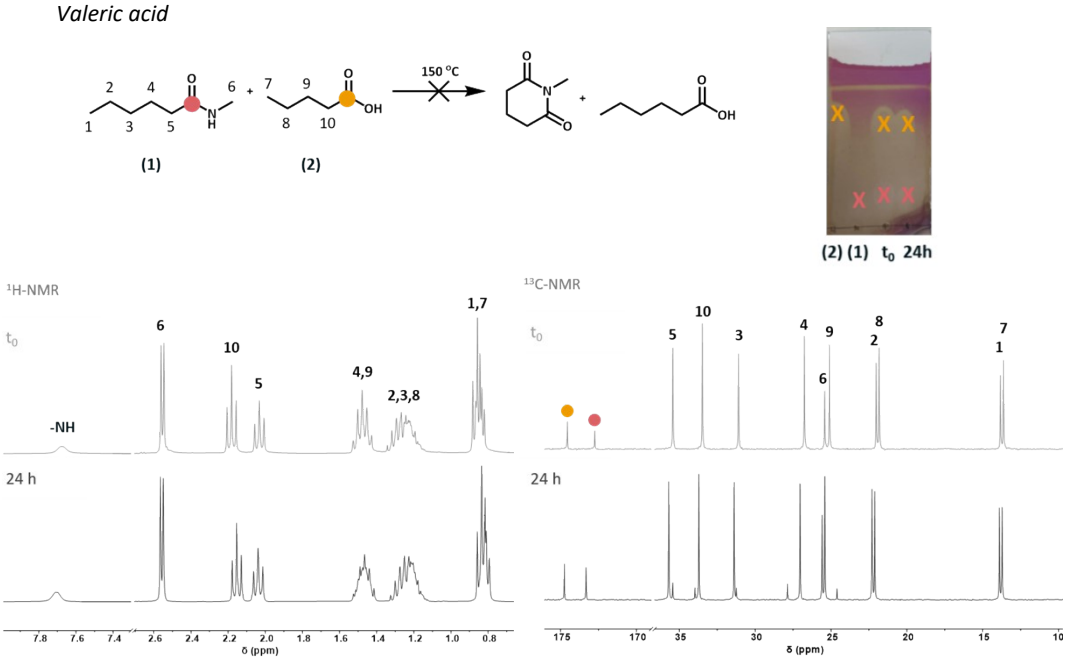

Figure S12. Model reaction using valeric acid as an example of a monoacid. ^{1}H-NMR and ^{13}C-NMR spectra of the reactants and the crude of the reaction at 24 h (dissolved in $DMSO_{d6}$). Picture of the TLC of the starting materials and the crude after 24 h compared with the virgin reactants. (Reaction conditions: 1 to 1 molar eq., 150 °C, 24 h).

Glutaric acid

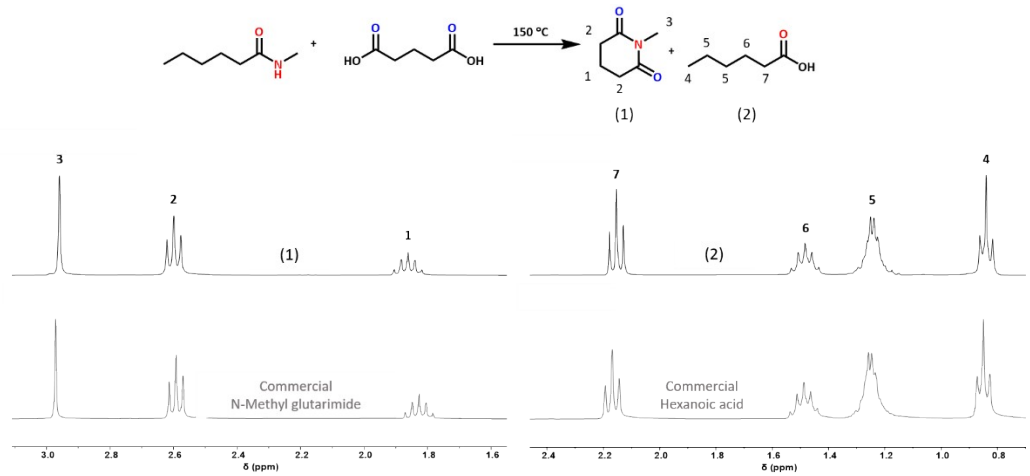

Figure S13. Model reaction using glutaric acid as an example of diacid. ^{1}H-NMR spectra of the products, glutarimide and hexanoic acid, isolated from the crude of the reaction by column chromatography (hexane:ethyl acetate). (Reaction conditions: 1 to 1 molar eq., 150 °C, 24 h).

Glutaric anhydride

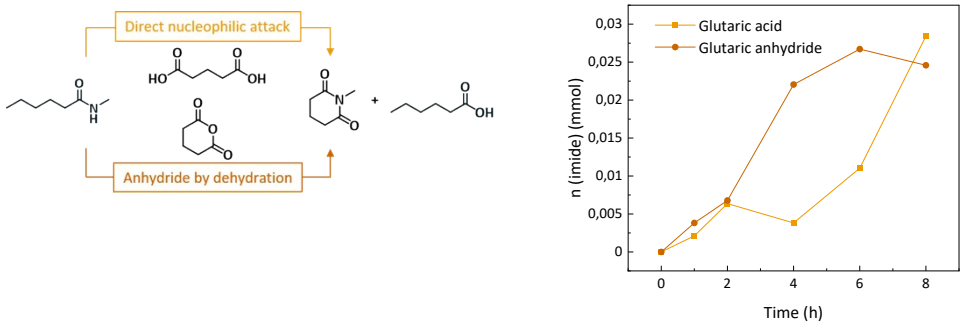

Figure S14. Comparison between the formation of imide product of the reaction with N-methyl hexanamide with glutaric acid or glutaric anhydride measured by ^{1}H-NMR (8h at 150 °C).

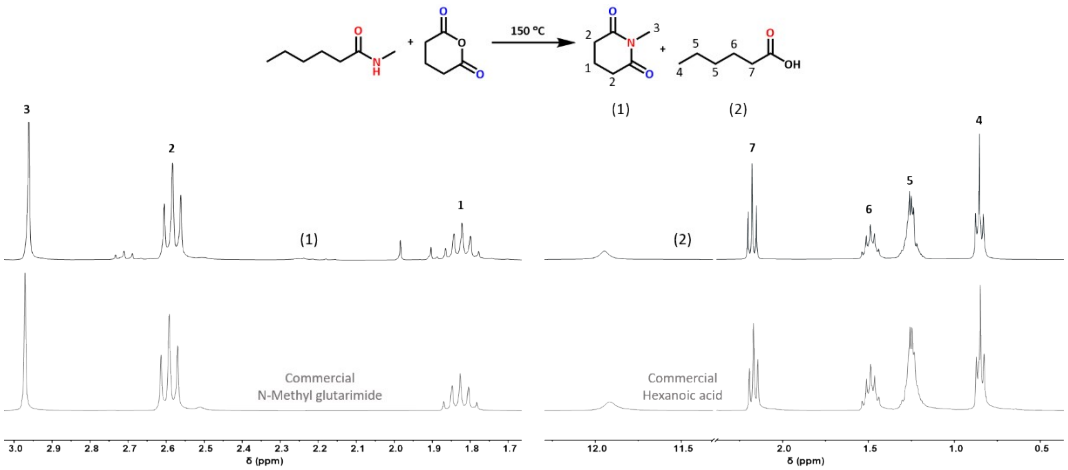

Figure S15. Model reaction using glutaric anhydride. ^{1}H-NMR spectra of the products, glutarimide and hexanoic acid, isolated from the crude of the reaction by column chromatography (hexane:ethyl acetate).(Reaction conditions: 1 to 1 molar eq., 150 °C, 24 h).

4.2. Density Functional Theory

The imaginary frequencies (in cm-1) of all the presented transition states, and the xyz coordinate files of all the presented structures.

IMAGINARY FREQUENCIES OF TRANSITION STATES

TS	**Frequency cm^{-1}**
TS_Dehydration_anhydre	-549.66
TS1_Glutaric	-1317.45
TS2_Glutaric_condesation	-452.59
TS2_Glutaric_hydrolisis	-1262.54
TS2_Succininc_condensation	-470.160
TS2_Adipic_condensation	-433.560
TS1_Adipic	-1309.49
TS2_Adipic_hydrolisis	-933.170

Table S1. Imaginary frequencies (in cm^{-1}) obtained from the frequency calculations of all the presented transition states, calculated at the M06-2X/6-31+G(d,p) level of theory.

Imide formation as a function of the diacid:

The reaction energy profiles of the intramolecular condensation reaction of the intermediates to form the imides have been calculated and are shown inf Figure S14 (a). As it can be seen, the activation barrier for intramolecular condensation increases with chain length; Adipic Acid 53.09 kcal/mol, Glutaric Acid: 62.39 kcal/mol and Adipic Acid 72.92 kcal/mol. As the chain grows longer, the number of internal degrees of freedom rises, reducing the likelihood of the intermediates adopting the optimal geometry required for the reaction to proceed efficiently. Moreover, shorter chains form smaller cycles that are thermodynamically more stable due to reduced flexibility and stronger intramolecular interactions; with the measured product stability, DG_{rxn}, being –22.63, –11.29 and –1.61 kcal/mol for succinimide, glutarimide and adipimide, respectively. Besides, the hydrolysis reaction of the adipic intermediate has also been characterized (Figure S15b). As condensation of the adipic intermediate has been labelled too high to occur at experiment conditions, we have studied an alternative mechanism to rationalize the formation of the amide. In this alternative mechanism, the water molecule produced in the first step of the reaction (TS1) is now able to hydrolyze the intermediate to yield the experimentally observed amide and a carboxylic acid, showing a barrier of 58.45 kcal/mol, making it accessible at reaction conditions. These factors combined rationalize both the experimentally observed products and the measured kinetics.

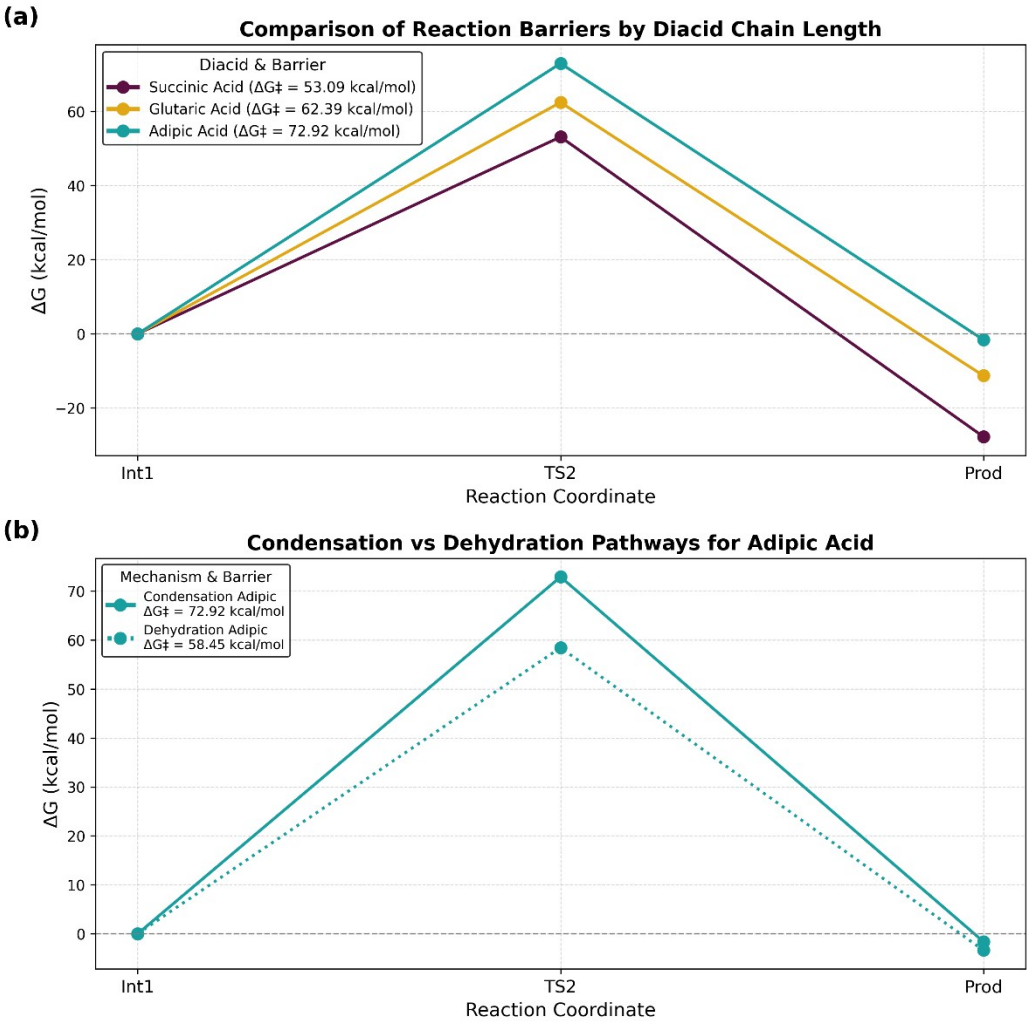

Figure S16. (a) Reaction energy profile of the intermolecular condensation to form the amide for the different diacids, with the energy barriers for the different steps of the reaction and (b) Reaction energy profile of both the condensation and the hydrolysis mechanisms of the adipic acid, both calculated at the M06-2X/6-311++G(2df,2p)//M06-2X/6-31+G(d,p) level of theory.

XYZ coordinate files

Glutaric

Anhydride formation

React

41

C	-4.04500	0.40180	-0.59510
H	-4.43450	-0.61270	-0.45880
H	-3.26950	0.56650	0.15820
H	-4.86340	1.10110	-0.39270
C	-3.50370	0.59080	-2.01050
H	-3.09310	1.60290	-2.11800
H	-2.67010	-0.10030	-2.18750

C	-4.56890	0.36880	-3.08310
H	-5.40180	1.06800	-2.91980
H	-4.98850	-0.64140	-2.96920
C	-4.02160	0.53880	-4.49810
H	-3.18300	-0.15210	-4.64850
H	-3.61020	1.54580	-4.62770
C	-5.08450	0.30600	-5.56650
H	-5.91350	1.01410	-5.43130
H	-5.52460	-0.69710	-5.48190
C	-4.55480	0.51360	-6.97650
N	-5.26030	-0.10120	-7.96100
H	-5.94000	-0.81050	-7.71410
C	-4.92090	0.11110	-9.35360
H	-3.90460	-0.23130	-9.58200
H	-5.63170	-0.43490	-9.97560
H	-4.96900	1.17530	-9.59790
O	-3.57940	1.21460	-7.22630
C	-5.14970	-3.56660	-7.26730
C	-3.85790	-3.10190	-7.88170
H	-3.34940	-3.94450	-8.35800
H	-4.10660	-2.35550	-8.63900
C	-2.96290	-2.47680	-6.79760
H	-2.54990	-3.26040	-6.15540
H	-3.56150	-1.82820	-6.14980
C	-1.84190	-1.65230	-7.41630
H	-2.24350	-0.79220	-7.96740
H	-1.24410	-2.23930	-8.12390
C	-0.91210	-1.09410	-6.37010
O	-5.06820	-4.80630	-6.76040
H	-5.91720	-5.01020	-6.33730
O	0.10270	-0.40420	-6.91940
H	0.63420	-0.04170	-6.19410
O	-6.15950	-2.90050	-7.17600
O	-1.03440	-1.23310	-5.17610

TS_dehydration

41

C	-3.658804	4.485499	-1.397750
C	-3.187360	3.098469	-0.980988
H	-2.986966	2.529455	-1.893105
H	-2.223346	3.215149	-0.468531
C	-4.181593	2.368917	-0.079261
H	-3.831741	1.347793	0.098863
H	-5.157013	2.285330	-0.571735
C	-4.353973	3.041996	1.279262
H	-3.393331	3.147118	1.794501
H	-5.018552	2.458961	1.926748
C	-4.934900	4.419183	1.263790
O	-4.498021	5.067342	-0.550377
H	-5.841442	4.711908	-0.378233
O	-6.418325	4.296781	0.415933
H	-6.992391	4.982987	0.792427
O	-3.283430	5.006328	-2.425553
O	-4.911218	5.318538	2.015017
C	-6.455610	2.724671	-3.059729
H	-7.010804	3.124833	-3.914242
H	-7.177338	2.283959	-2.364644
H	-5.976124	3.578063	-2.564371
C	-5.407182	1.711824	-3.516952
H	-4.907994	1.273039	-2.641697
H	-5.894687	0.875514	-4.033830
C	-4.350878	2.340433	-4.424898
H	-3.958084	3.248168	-3.947330
H	-4.825821	2.670117	-5.359751
C	-3.194026	1.394142	-4.734940
H	-3.552556	0.520626	-5.294658
H	-2.762888	1.002207	-3.806225
C	-2.080782	2.085021	-5.517032
H	-1.748224	2.974019	-4.963809
H	-2.447977	2.436509	-6.489015
C	-0.868855	1.186799	-5.700560

N	-0.147357	1.388292	-6.836665
H	-0.414258	2.129937	-7.464175
C	1.085766	0.659580	-7.066043
H	1.446709	0.880845	-8.070809
H	1.851343	0.936497	-6.334658
H	0.902984	-0.413212	-6.974386
O	-0.541347	0.343066	-4.877487

Anhydride

41

C	-5.434989	-0.034970	0.676254
H	-4.749240	-0.867202	0.863255
H	-5.595721	0.493285	1.620454
H	-6.396458	-0.451882	0.357626
C	-4.865015	0.891425	-0.394786
H	-5.524736	1.756320	-0.541915
H	-3.904307	1.299789	-0.048199
C	-4.651535	0.179864	-1.729532
H	-5.615492	-0.200374	-2.096615
H	-4.012228	-0.698409	-1.567138
C	-4.019423	1.083089	-2.785173
H	-3.107333	1.537981	-2.377846
H	-4.680009	1.927521	-3.013898
C	-3.678564	0.329290	-4.064549
H	-4.594099	-0.015813	-4.562047
H	-3.089219	-0.565295	-3.824926
C	-2.887891	1.169334	-5.051350
N	-2.385927	0.508831	-6.119086
H	-2.355903	-0.498495	-6.060784
C	-1.444577	1.168545	-7.006599
H	-0.501752	1.378075	-6.490035
H	-1.258544	0.524959	-7.866756
H	-1.872826	2.111811	-7.347443
O	-2.719394	2.383018	-4.917873
C	-1.488768	-0.205723	-0.368687
C	-0.686559	0.509925	-2.572947

C	-0.799752	0.979324	0.254717
H	0.237480	0.678743	0.456941
H	-1.285527	1.157753	1.215315
C	-0.833430	2.190234	-0.672256
H	-1.858774	2.537183	-0.831845
H	-0.289305	3.026103	-0.226590
C	-0.205907	1.824277	-2.010523
H	-0.411168	2.583986	-2.769021
H	0.884354	1.727874	-1.921406
O	-1.336706	-0.373548	-1.733464
O	-0.541454	0.164672	-3.712833
O	-2.137842	-1.023442	0.216183
O	-2.388614	3.998352	-2.686454
H	-2.617409	3.502253	-3.495945
H	-2.595438	4.919437	-2.867024

Proposed Mechanism

React
41

C	-4.04500	0.40180	-0.59510
H	-4.43450	-0.61270	-0.45880
H	-3.26950	0.56650	0.15820
H	-4.86340	1.10110	-0.39270
C	-3.50370	0.59080	-2.01050
H	-3.09310	1.60290	-2.11800
H	-2.67010	-0.10030	-2.18750
C	-4.56890	0.36880	-3.08310
H	-5.40180	1.06800	-2.91980
H	-4.98850	-0.64140	-2.96920
C	-4.02160	0.53880	-4.49810
H	-3.18300	-0.15210	-4.64850
H	-3.61020	1.54580	-4.62770
C	-5.08450	0.30600	-5.56650
H	-5.91350	1.01410	-5.43130
H	-5.52460	-0.69710	-5.48190
C	-4.55480	0.51360	-6.97650

N	-5.26030	-0.10120	-7.96100
H	-5.94000	-0.81050	-7.71410
C	-4.92090	0.11110	-9.35360
H	-3.90460	-0.23130	-9.58200
H	-5.63170	-0.43490	-9.97560
H	-4.96900	1.17530	-9.59790
O	-3.57940	1.21460	-7.22630
C	-5.14970	-3.56660	-7.26730
C	-3.85790	-3.10190	-7.88170
H	-3.34940	-3.94450	-8.35800
H	-4.10660	-2.35550	-8.63900
C	-2.96290	-2.47680	-6.79760
H	-2.54990	-3.26040	-6.15540
H	-3.56150	-1.82820	-6.14980
C	-1.84190	-1.65230	-7.41630
H	-2.24350	-0.79220	-7.96740
H	-1.24410	-2.23930	-8.12390
C	-0.91210	-1.09410	-6.37010
O	-5.06820	-4.80630	-6.76040
H	-5.91720	-5.01020	-6.33730
O	0.10270	-0.40420	-6.91940
H	0.63420	-0.04170	-6.19410
O	-6.15950	-2.90050	-7.17600
O	-1.03440	-1.23310	-5.17610

TS1_Glutaric

41

C	0.768845	-0.366044	-1.383344
C	-0.698504	-0.021603	-1.396341
H	-1.006877	0.148602	-0.357528
H	-0.799457	0.921428	-1.938305
C	-1.564156	-1.123026	-2.015419
H	-1.456913	-2.043950	-1.434284
H	-1.231961	-1.337928	-3.036263
C	-3.034978	-0.704961	-2.032021
H	-3.179585	0.186474	-2.655315

H	-3.398510	-0.486604	-1.028736
C	-3.848675	-1.804793	-2.654455
O	1.001474	-1.578336	-0.836631
H	1.960107	-1.722146	-0.855036
O	-4.100392	-2.914237	-1.360777
H	-3.988767	-3.795204	-1.748443
O	1.660771	0.334667	-1.794718
O	-3.656730	-2.351480	-3.705123
C	-9.791635	4.825515	-2.178575
H	-10.729843	4.277906	-2.316565
H	-10.011252	5.725117	-1.597267
H	-9.443184	5.141718	-3.167396
C	-8.747642	3.951278	-1.487268
H	-7.825669	4.526183	-1.333882
H	-9.103737	3.667807	-0.488817
C	-8.422357	2.686353	-2.279949
H	-8.065914	2.969776	-3.280697
H	-9.346122	2.110589	-2.434875
C	-7.379475	1.806854	-1.593681
H	-7.728283	1.516131	-0.596397
H	-6.455261	2.373934	-1.434312
C	-7.065143	0.551807	-2.398009
H	-6.692892	0.814076	-3.398109
H	-7.978502	-0.032891	-2.568532
C	-6.032483	-0.332573	-1.720875
N	-5.629028	-1.505985	-2.378072
H	-5.156360	-2.445867	-1.584327
C	-6.361856	-1.944328	-3.564039
H	-5.910005	-2.870582	-3.919832
H	-7.410375	-2.134239	-3.321413
H	-6.302658	-1.216068	-4.378784
O	-5.552313	-0.040474	-0.641391

Int1

41

C	1.320787	4.519585	-2.744509

C	0.046564	0.392902	-3.542298
C	0.654089	3.723502	-1.653657
H	-0.385111	4.055369	-1.597314
H	1.152337	3.932557	-0.703382
C	0.724302	2.217771	-1.956597
H	1.767221	1.885946	-1.957584
H	0.217908	1.670632	-1.158469
C	0.062402	1.889473	-3.293924
H	-0.969955	2.252266	-3.280503
H	0.576573	2.393757	-4.117482
O	2.666254	4.447017	-2.674847
O	0.210057	-0.385426	-2.614313
O	0.754199	5.121404	-3.625737
C	-4.984329	4.818616	-7.864825
H	-4.344588	5.692211	-7.702007
H	-5.605230	5.016234	-8.743003
H	-5.647695	4.727336	-6.998288
C	-4.144731	3.555386	-8.042790
H	-4.801970	2.696901	-8.231099
H	-3.507655	3.656214	-8.930560
C	-3.266852	3.257609	-6.828366
H	-3.904242	3.156332	-5.938366
H	-2.609140	4.117511	-6.638056
C	-2.423798	1.995982	-7.001596
H	-1.782427	2.089378	-7.884428
H	-3.072882	1.132879	-7.188540
C	-1.559350	1.724100	-5.773485
H	-2.183122	1.603005	-4.879544
H	-0.904916	2.583852	-5.595585
C	-0.729221	0.465716	-5.945019
N	-0.061162	-0.107646	-4.840411
H	3.023729	4.948506	-3.423975
C	0.356720	-1.503953	-5.029375
H	1.077025	-1.756969	-4.255958
H	-0.504058	-2.174565	-4.953195

H	0.792909	-1.599616	-6.020832
O	-0.695664	-0.121896	-7.007177
O	-2.609997	0.277539	-2.752972
H	-1.983586	-0.315790	-2.316432
H	-3.491695	-0.016030	-2.504302

TS2_Glutaric_condensation

41

C	-3.006123	1.890084	-3.263314
C	-2.083468	1.570782	-2.099816
H	-1.369806	2.397967	-2.062389
H	-2.628004	1.507245	-1.156382
C	-1.373161	0.242947	-2.362294
H	-0.587062	0.095989	-1.617515
H	-0.888013	0.266671	-3.344677
C	-2.360189	-0.924997	-2.271283
H	-2.832898	-0.926669	-1.281459
H	-1.877754	-1.891256	-2.431971
C	-3.479040	-0.834532	-3.269055
O	-4.446594	2.504983	-2.623822
H	-4.323865	2.890153	-1.741904
O	-6.244695	-2.026986	-1.574034
H	-5.649162	-2.461948	-2.194532
O	-2.659830	2.432683	-4.278399
O	-3.937168	-1.777078	-3.861313
C	-11.244925	2.138665	-1.504502
H	-11.600166	1.142134	-1.786389
H	-11.887683	2.509071	-0.701209
H	-11.377005	2.795983	-2.370384
C	-9.780198	2.091478	-1.074917
H	-9.451925	3.093326	-0.769144
H	-9.673406	1.447541	-0.193074
C	-8.861064	1.577861	-2.181866
H	-8.965328	2.221798	-3.066594
H	-9.186417	0.573654	-2.486240
C	-7.395749	1.521036	-1.756234

H -7.285781 0.874275 -0.879490

H -7.059334 2.523764 -1.458732

C -6.497848 0.997678 -2.875403

H -6.586697 1.628920 -3.761323

H -6.800727 -0.025661 -3.135430

C -5.060478 0.913920 -2.403427

N -4.029940 0.504199 -3.535669

H -5.745339 -1.280358 -1.208906

C -4.528917 0.607331 -4.924666

H -5.331276 -0.110063 -5.086476

H -4.859051 1.629979 -5.092281

H -3.697323 0.399288 -5.598780

O -4.773491 0.475442 -1.282906

TS2_Glutaric_hydrolysis

41

C 0.454275 4.734307 -2.368550

C 0.757401 0.641992 -3.925230

C 0.698759 3.601910 -1.406164

H -0.254209 3.375985 -0.921643

H 1.423269 3.915768 -0.651097

C 1.226364 2.365417 -2.154493

H 2.208382 2.570599 -2.588137

H 1.361893 1.555488 -1.430915

C 0.268573 1.917894 -3.260667

H -0.743290 1.785073 -2.860769

H 0.207823 2.699717 -4.026270

O 1.579150 5.420450 -2.646501

O 1.943520 0.445160 -4.103107

O -0.604851 4.989035 -2.893465

C -4.435912 5.029374 -8.125050

H -3.567316 5.395725 -8.682001

H -5.326582 5.220200 -8.729663

H -4.517797 5.623049 -7.208747

C -4.287168 3.544613 -7.800437

H -5.179669 3.190281 -7.269317

H	-4.230720	2.965287	-8.730497
C	-3.050326	3.252894	-6.952010
H	-3.106009	3.829009	-6.017753
H	-2.156040	3.610565	-7.481338
C	-2.889827	1.768143	-6.632415
H	-2.810881	1.184587	-7.555601
H	-3.780916	1.400039	-6.107309
C	-1.654639	1.505301	-5.768114
H	-1.707588	2.081855	-4.844645
H	-0.743574	1.804579	-6.302483
C	-1.525584	0.031940	-5.489185
N	-0.212206	-0.315692	-4.261554
H	1.354997	6.099098	-3.302202
C	0.342923	-1.628965	-4.604676
H	1.030147	-1.946513	-3.819061
H	-0.481493	-2.334703	-4.695577
H	0.885183	-1.586090	-5.551957
O	-1.637994	-0.860140	-6.261883
O	-2.509033	-0.266460	-3.988563
H	-1.329965	-0.319789	-3.680574
H	-3.028568	0.482523	-3.669081

Product (imide)

41

C	-0.80320	1.24800	-3.29010
C	0.91790	0.02490	-4.55480
C	-0.41930	2.52000	-4.01020
H	0.38120	3.00030	-3.43160
H	-1.29010	3.17800	-3.97170
C	0.05300	2.23140	-5.43080
H	-0.75450	1.77340	-6.01320
H	0.34650	3.15800	-5.93100
C	1.22350	1.26170	-5.36680
H	1.52910	0.90760	-6.35260
H	2.09590	1.74020	-4.90120
O	-0.93730	-0.75050	-6.42060

O	1.47950	-1.03320	-4.75070
O	-1.75530	1.21240	-2.51870
C	-3.29000	4.80240	-8.72250
H	-2.49280	5.28400	-8.14610
H	-3.13090	5.04060	-9.77760
H	-4.23960	5.25340	-8.41620
C	-3.30040	3.29440	-8.48280
H	-4.08760	2.82770	-9.08800
H	-2.35200	2.85760	-8.82140
C	-3.51770	2.93330	-7.01470
H	-4.46480	3.37000	-6.66720
H	-2.72560	3.40090	-6.40930
C	-3.52210	1.42630	-6.76830
H	-2.58390	0.98890	-7.13170
H	-4.32490	0.94900	-7.34040
C	-3.68830	1.09270	-5.29000
H	-4.66910	1.42610	-4.92300
H	-2.95050	1.62850	-4.68250
C	-3.59300	-0.39150	-5.00050
N	-0.04150	0.12050	-3.53270
H	-1.78880	-1.20030	-6.52590
C	-0.35690	-1.11930	-2.81580
H	-0.87330	-0.85840	-1.89490
H	-1.00300	-1.75090	-3.43060
H	0.57470	-1.64200	-2.60770
O	-3.71780	-1.24500	-5.84990
O	-3.38650	-0.74510	-3.72580
H	-0.27060	-1.44120	-6.32020
H	-3.12470	0.01140	-3.16830

Succinic
Reac
38

C	-1.198056	-1.035658	0.082483
C	-2.055659	-1.443808	-1.094622
H	-1.682842	-2.394570	-1.492655
H	-3.088359	-1.619920	-0.771157

C	-2.048348	-0.380255	-2.180573
H	-1.014489	-0.161143	-2.472194
H	-2.476243	0.551720	-1.800202
C	-2.844939	-0.862405	-3.377901
O	-1.247527	-1.867217	1.148111
H	-1.863900	-2.593347	0.987759
O	-5.124173	-0.974649	-1.780174
H	-5.250058	-1.329909	-2.676615
O	-0.496422	-0.059583	0.114697
O	-3.069854	-2.032334	-3.574080
C	-8.088827	2.238110	-9.733423
H	-7.291206	2.241319	-10.483645
H	-9.025013	1.990759	-10.241059
H	-8.179313	3.256054	-9.340062
C	-7.779246	1.246914	-8.613748
H	-8.602062	1.238681	-7.887809
H	-7.719710	0.230990	-9.023888
C	-6.474462	1.570097	-7.887545
H	-6.532922	2.588629	-7.477408
H	-5.650099	1.578402	-8.614891
C	-6.159788	0.582625	-6.765974
H	-6.098706	-0.435684	-7.165612
H	-6.975280	0.571070	-6.034461
C	-4.854404	0.921019	-6.055929
H	-4.903414	1.935457	-5.638128
H	-4.018903	0.925281	-6.768374
C	-4.536499	-0.060464	-4.940707
N	-3.326197	0.137053	-4.262532
H	-5.974128	-0.604352	-1.524192
C	-2.658750	1.438610	-4.322110
H	-2.554062	1.760737	-5.358337
H	-3.205393	2.203162	-3.760028
H	-1.656508	1.351727	-3.908622
O	-5.308835	-0.951216	-4.645293

TS2_Succininc_condensation

38

C	-3.485846	1.924232	-3.148361
C	-2.104848	1.684394	-2.539152
H	-1.391060	1.754633	-3.364677
H	-1.870296	2.477755	-1.825992
C	-2.110047	0.278835	-1.930841
H	-2.530228	0.261038	-0.917770
H	-1.136582	-0.211159	-1.909693
C	-3.057587	-0.510208	-2.796273
O	-4.525276	2.089235	-1.842132
H	-4.106473	2.027050	-0.969002
O	-5.898204	-2.399060	-2.919974
H	-5.020080	-2.737565	-3.133364
O	-3.751264	2.686672	-4.046318
O	-2.985613	-1.661785	-3.118409
C	-11.227414	2.605949	-1.627688
H	-11.691682	1.757931	-2.141761
H	-11.905655	2.928479	-0.832956
H	-11.141460	3.425262	-2.349035
C	-9.856626	2.222240	-1.074478
H	-9.420428	3.076372	-0.540963
H	-9.967375	1.419072	-0.334963
C	-8.889943	1.767524	-2.166689
H	-8.776806	2.571273	-2.907420
H	-9.324429	0.911309	-2.701211
C	-7.517498	1.381975	-1.618599
H	-7.618990	0.574818	-0.884686
H	-7.073734	2.238395	-1.096144
C	-6.567108	0.931357	-2.727007
H	-6.425667	1.729221	-3.462331
H	-6.969855	0.045495	-3.233453
C	-5.234002	0.533056	-2.138254
N	-4.106628	0.378617	-3.292796
H	-5.785094	-1.966605	-2.060710
C	-4.585259	0.042003	-4.647309

H -5.296221 -0.782177 -4.584418

H -5.034798 0.941840 -5.067044

H -3.729866 -0.249757 -5.258267

O -5.094058 -0.213416 -1.176163

Prod

38

C -2.659797 1.215220 -3.391302

C -2.071515 0.425289 -2.243683

H -0.982815 0.422579 -2.342168

H -2.332202 0.939947 -1.317558

C -2.700342 -0.964932 -2.370358

H -3.389650 -1.189809 -1.552434

H -1.980221 -1.782777 -2.437986

C -3.514435 -0.907981 -3.649030

O -5.092602 3.423728 -3.002325

H -4.143225 3.236236 -3.162607

O -6.688369 -0.291010 -3.522751

H -6.268456 -1.058316 -3.933014

O -2.529300 2.407755 -3.605041

O -4.188836 -1.790556 -4.134767

C -11.351639 1.220995 0.291814

H -11.818181 0.857783 -0.629700

H -11.764074 0.646189 1.125592

H -11.648957 2.266492 0.425141

C -9.831725 1.092013 0.218086

H -9.385101 1.434374 1.160499

H -9.554651 0.035951 0.109175

C -9.231299 1.886529 -0.940173

H -9.506367 2.945563 -0.834125

H -9.675078 1.542228 -1.884429

C -7.712477 1.754902 -1.025128

H -7.447462 0.699834 -1.150105

H -7.255871 2.086861 -0.084062

C -7.137545 2.562055 -2.184607

H -7.371250 3.627670 -2.098097

H -7.570866 2.214355 -3.132863

C -5.648701 2.412078 -2.341882

N -3.398206 0.365285 -4.186927

H -6.104734 -0.049143 -2.789159

C -4.152621 0.808362 -5.346651

H -5.173129 1.069767 -5.050595

H -3.645235 1.674708 -5.770699

H -4.188813 -0.010075 -6.066254

O -5.006253 1.450650 -1.946127

Adipic
Reac

44

C -4.381310 -0.315838 -1.004559

H -5.009985 -1.212171 -0.976101

H -3.568705 -0.453336 -0.286141

H -4.990840 0.527955 -0.664724

C -3.848528 -0.068521 -2.414085

H -3.201522 0.817404 -2.417053

H -3.214731 -0.910556 -2.723735

C -4.963097 0.122145 -3.441408

H -5.602921 0.961882 -3.135598

H -5.610329 -0.768627 -3.440815

C -4.434068 0.368079 -4.851686

H -3.771432 -0.459425 -5.140008

H -3.812803 1.269258 -4.878478

C -5.550693 0.501521 -5.885861

H -6.135667 1.408752 -5.688786

H -6.244118 -0.345610 -5.821578

C -4.994320 0.608744 -7.295250

N -5.704011 -0.035725 -8.259005

H -6.351148 -0.754594 -7.959083

C -5.175359 -0.107905 -9.607476

H -4.265903 -0.721276 -9.650846

H -5.932689 -0.536973 -10.263987

H -4.919830 0.895214 -9.951715

O -3.970744 1.235560 -7.547166

C -6.035759 -3.334805 -6.023137

C	-2.282805	-2.476231	-6.518149
H	-1.774646	-3.445573	-6.458617
H	-2.182701	-2.022028	-5.524967
C	-1.585707	-1.583431	-7.537925
H	-2.059886	-0.591987	-7.580558
H	-1.654061	-1.995481	-8.552004
C	-0.128570	-1.378616	-7.218813
O	-6.691731	-4.184481	-5.216468
H	-7.637115	-4.119472	-5.424295
O	0.509753	-0.696718	-8.190771
H	1.429823	-0.583258	-7.907144
O	-6.614902	-2.658469	-6.845942
O	0.435652	-1.759694	-6.221232
C	-3.756637	-2.659524	-6.870093
H	-4.201640	-1.685958	-7.092479
H	-3.856464	-3.251610	-7.788676
C	-4.554356	-3.315134	-5.749693
H	-4.424731	-2.758361	-4.809550
H	-4.223563	-4.338025	-5.542842

TS1_Adipic

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C	1.696354	0.542797	-0.929573
C	-1.581727	-1.197627	-2.042621
H	-1.500609	-2.096723	-1.418692
H	-1.263721	-1.477850	-3.053540
C	-3.046673	-0.758735	-2.074862
H	-3.172587	0.119320	-2.720968
H	-3.408058	-0.508406	-1.078527
C	-3.878898	-1.855861	-2.674392
O	2.972349	0.112747	-0.834251
H	3.504027	0.857049	-0.512687
O	-4.136972	-2.948923	-1.359743
H	-4.042770	-3.834728	-1.740647
O	1.372905	1.668865	-0.641641
O	-3.707324	-2.422657	-3.717371
C	-9.732262	4.855945	-2.171699

H	-10.676710	4.321021	-2.317027
H	-9.943029	5.754633	-1.585641
H	-9.375833	5.174049	-3.157073
C	-8.702053	3.964454	-1.481823
H	-7.773610	4.526736	-1.321024
H	-9.065674	3.679215	-0.486604
C	-8.389212	2.700418	-2.280987
H	-8.025135	2.985526	-3.278505
H	-9.319541	2.137271	-2.443273
C	-7.360244	1.803926	-1.595898
H	-7.716602	1.512338	-0.601514
H	-6.429317	2.357790	-1.429705
C	-7.059141	0.548959	-2.405438
H	-6.677355	0.811479	-3.402004
H	-7.979481	-0.022104	-2.584302
C	-6.041565	-0.352555	-1.727782
N	-5.658294	-1.532828	-2.383904
H	-5.187057	-2.469460	-1.585378
C	-6.406952	-1.966516	-3.560992
H	-5.961550	-2.893072	-3.924384
H	-7.453110	-2.154527	-3.306099
H	-6.356944	-1.236518	-4.374935
O	-5.556273	-0.067556	-0.648755
C	-0.679174	-0.096916	-1.490229
H	-0.756927	0.802472	-2.111604
H	-1.010115	0.195243	-0.487366
C	0.776897	-0.540520	-1.431678
H	1.141661	-0.846470	-2.420126
H	0.905504	-1.414100	-0.781638

Int1

44

C	-1.917261	0.262197	0.075410
C	-1.288002	-1.001155	-0.495923
H	-0.654646	-1.412246	0.294456
H	-2.050008	-1.745895	-0.747364
C	-1.261648	-0.317462	-2.959906

H	-0.638658	0.200862	-3.696483
H	-2.061949	0.379112	-2.681786
C	-1.842694	-1.582185	-3.614884
H	-2.302702	-2.225626	-2.864306
H	-1.044267	-2.137605	-4.112772
C	-2.841781	-1.279344	-4.708387
O	-3.241225	0.420099	-0.088298
H	-3.619323	-0.274125	-0.661342
O	-6.170790	-2.635184	-3.592689
H	-6.534536	-3.474292	-3.892077
O	-1.265036	1.103934	0.639678
O	-2.621081	-1.574461	-5.862759
C	-9.535202	2.016270	0.047683
H	-10.307922	1.646747	-0.634726
H	-9.945180	1.996128	1.061019
H	-9.334324	3.060928	-0.211747
C	-8.265083	1.175338	-0.059916
H	-7.513593	1.545379	0.648393
H	-8.481067	0.139389	0.230879
C	-7.672892	1.185894	-1.468173
H	-7.459231	2.223615	-1.761084
H	-8.422180	0.810771	-2.179638
C	-6.398644	0.350796	-1.579457
H	-6.611122	-0.687582	-1.299081
H	-5.652844	0.727177	-0.870177
C	-5.830868	0.376506	-2.993302
H	-5.577902	1.403955	-3.287911
H	-6.582311	0.017184	-3.705527
C	-4.595619	-0.484163	-3.149280
N	-4.027103	-0.545058	-4.405942
H	-5.598515	-2.832948	-2.840503
C	-4.801388	-0.078901	-5.562883
H	-5.653942	-0.742320	-5.734491
H	-5.149257	0.939864	-5.396063
H	-4.151923	-0.103230	-6.433116

O	-4.132062	-1.105253	-2.191833
C	-0.426175	-0.657659	-1.721194
H	0.218092	0.186378	-1.455948
H	0.231864	-1.504531	-1.948553

TS2_Adipic_condensation

44

C	-3.41170	2.13110	-3.41670
C	-2.43400	2.17800	-2.25530
H	-1.96640	3.16450	-2.31180
H	-2.93910	2.06080	-1.29530
C	-1.19880	-0.47330	-2.31140
H	-0.47880	-1.01780	-1.69550
H	-0.73860	-0.32810	-3.29540
C	-2.42110	-1.48340	-2.42340
H	-2.86290	-1.61350	-1.42790
H	-2.23690	-2.46830	-2.85750
C	-3.47900	-0.83450	-3.26910
O	-4.44660	2.50500	-2.62380
H	-4.32390	2.89020	-1.74190
O	-6.24470	-2.02700	-1.57400
H	-5.64920	-2.46190	-2.19450
O	-3.22540	2.64870	-4.48540
O	-3.93720	-1.77710	-3.86130
C	-11.24490	2.13870	-1.50450
H	-11.60020	1.14210	-1.78640
H	-11.88770	2.50910	-0.70120
H	-11.37700	2.79600	-2.37040
C	-9.78020	2.09150	-1.07490
H	-9.45190	3.09330	-0.76910
H	-9.67340	1.44750	-0.19310
C	-8.86110	1.57790	-2.18190
H	-8.96530	2.22180	-3.06660
H	-9.18640	0.57370	-2.48620
C	-7.39570	1.52100	-1.75620
H	-7.28580	0.87430	-0.87950

H	-7.05930	2.52380	-1.45870
C	-6.49780	0.99770	-2.87540
H	-6.58670	1.62890	-3.76130
H	-6.80070	-0.02570	-3.13540
C	-5.06050	0.91390	-2.40340
N	-4.02990	0.50420	-3.53570
H	-5.74530	-1.28040	-1.20890
C	-4.52890	0.60730	-4.92470
H	-5.33130	-0.11010	-5.08650
H	-4.85910	1.63000	-5.09230
H	-3.69730	0.39930	-5.59880
O	-4.77350	0.47540	-1.28290
C	-1.50460	1.01710	-2.09300
H	-0.58700	1.52370	-2.30790
H	-1.99390	0.99680	-1.14160

TS2_Adipic_hydrolysis

44

C	-1.736509	4.252246	-3.219303
C	0.821859	0.523994	-3.781814
C	0.984103	3.068391	-3.460521
H	0.535232	3.276099	-4.441752
H	2.060896	2.989916	-3.634923
C	0.469280	1.734297	-2.922542
H	0.933176	1.536874	-1.945610
H	-0.606618	1.775040	-2.746459
O	-1.768112	5.383435	-3.934796
O	1.959158	0.381750	-4.204983
O	-2.404878	3.284373	-3.530792
C	-3.122517	5.542007	-7.908620
H	-2.531526	5.452066	-8.825365
H	-4.023167	6.114928	-8.143892
H	-2.529371	6.128997	-7.197081
C	-3.462868	4.164095	-7.343751
H	-4.094452	4.267548	-6.449967
H	-4.062766	3.603425	-8.070858

C -2.220604 3.351875 -6.982934

H -1.624718 3.906132 -6.240863

H -1.574460 3.253772 -7.865527

C -2.547030 1.968378 -6.422569

H -2.937549 1.314976 -7.209712

H -3.329766 2.061263 -5.660125

C -1.307812 1.338835 -5.785575

H -0.908498 2.012903 -5.030654

H -0.522847 1.151804 -6.527706

C -1.623029 0.009988 -5.166920

N -0.190339 -0.410972 -3.941737

H -2.383425 5.255027 -4.676045

C 0.280498 -1.733890 -4.343350

H 0.975240 -2.124385 -3.594836

H -0.579982 -2.397493 -4.426949

H 0.801272 -1.696777 -5.305543

O -2.020386 -0.977180 -5.688478

O -2.422734 0.299395 -3.667327

H -1.432596 -0.090757 -3.339708

H -2.527678 1.251076 -3.477306

C 0.690516 4.241023 -2.518594

H 1.306955 4.159194 -1.616604

H 0.964279 5.180021 -3.009413

C -0.781789 4.324290 -2.058310

H -1.028464 3.502357 -1.384160

H -0.947276 5.268666 -1.532621

Prod_Adipic_Imide

44

C -2.693986 0.994468 -3.696891

C -1.989118 0.990363 -2.363229

H -1.831423 2.039534 -2.110776

H -2.677914 0.578196 -1.619687

C -0.808563 -1.173030 -2.980773

H 0.008342 -1.815472 -2.640481

H -0.724392 -1.118373 -4.072204

C -2.144354 -1.823811 -2.619008

H	-2.392552	-1.660310	-1.562935
H	-2.110043	-2.906932	-2.756643
C	-3.334272	-1.398103	-3.465268
O	-5.263191	3.207655	-3.971522
H	-4.318416	2.989622	-4.118638
O	-6.692840	-0.611285	-3.027290
H	-6.194673	-1.377510	-3.342209
O	-2.759080	2.009908	-4.377379
O	-4.253555	-2.174386	-3.653249
C	-11.267820	1.936239	0.174145
H	-11.745045	1.265704	-0.547971
H	-11.615458	1.656276	1.172439
H	-11.621136	2.952174	-0.031890
C	-9.746570	1.856306	0.065680
H	-9.287196	2.512909	0.815908
H	-9.411633	0.837662	0.297895
C	-9.236370	2.243880	-1.320977
H	-9.569865	3.264492	-1.557475
H	-9.692557	1.584470	-2.072073
C	-7.716676	2.157271	-1.439228
H	-7.394915	1.132753	-1.226598
H	-7.246982	2.802653	-0.685876
C	-7.233061	2.559381	-2.829129
H	-7.523723	3.582784	-3.085473
H	-7.681463	1.897621	-3.583454
C	-5.745706	2.424768	-3.009672
N	-3.331524	-0.164082	-4.131231
H	-6.046707	-0.081166	-2.538297
C	-4.258218	-0.021804	-5.262583
H	-5.228815	0.345372	-4.916052
H	-3.823688	0.680405	-5.970588
H	-4.395896	-1.000327	-5.718056
O	-5.040598	1.652739	-2.378629
C	-0.660698	0.223127	-2.369874
H	0.096545	0.795104	-2.914034

H -0.317496 0.142657 -1.332928

Prod_Adipic_Amide
44

C -1.441588 4.602385 -2.929974
C 0.900723 0.820461 -4.187991
C 1.048465 2.974266 -2.888750
H 0.697766 3.440615 -3.819012
H 2.139540 3.013032 -2.943235
C 0.604903 1.512875 -2.863736
H 1.136210 0.973399 -2.068951
H -0.465142 1.416735 -2.646048
O -0.998362 5.853475 -3.075788
O 1.734175 1.249627 -4.976624
O -2.123688 4.070673 -3.788792
C -3.919749 4.883079 -9.262427
H -3.657505 4.219656 -10.092988
H -4.747072 5.518789 -9.589488
H -3.056252 5.528080 -9.068627
C -4.288281 4.077596 -8.018295
H -4.574954 4.758840 -7.206825
H -5.169554 3.457844 -8.226672
C -3.146952 3.182181 -7.539701
H -2.262775 3.799462 -7.326957
H -2.858892 2.496210 -8.348153
C -3.506444 2.378532 -6.292355
H -4.390448 1.759936 -6.491538
H -3.775617 3.071923 -5.486680
C -2.351731 1.472759 -5.841977
H -1.458005 2.081925 -5.644390
H -2.102851 0.748883 -6.621751
C -2.688197 0.689684 -4.591817
N 0.166579 -0.292003 -4.432738
H -1.307762 6.186845 -3.933148
C 0.279064 -1.025900 -5.678691
H 1.078087 -1.772883 -5.636964

H	-0.674739	-1.518263	-5.876328
H	0.509975	-0.323606	-6.482836
O	-2.833478	-0.509633	-4.555232
O	-2.813349	1.416402	-3.467070
H	-0.551903	-0.571304	-3.779418
H	-2.651103	2.368483	-3.634974
C	0.558004	3.773968	-1.680335
H	0.852673	3.275224	-0.750190
H	1.027796	4.762316	-1.678098
C	-0.972505	3.961006	-1.655052
H	-1.497728	3.009455	-1.547299
H	-1.253733	4.610373	-0.820486

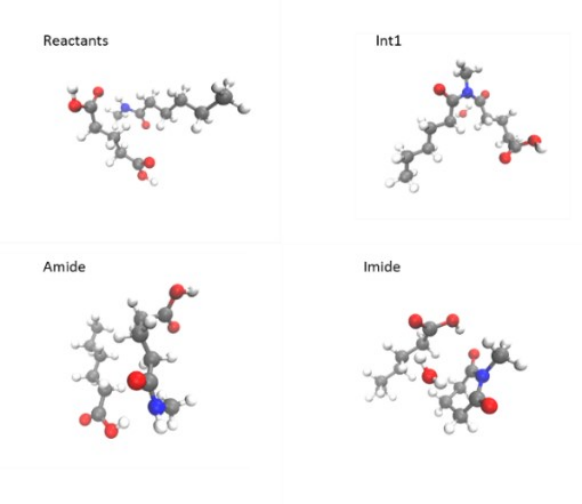


Figure S17. DFT-optimized geometries of the reactants, Int1, amide and imide structures of the proposed mechanism.

5. Selective depolymerization of PA6 from polymer mixtures and waste

5.1. Fishing nets

A) Insoluble fraction

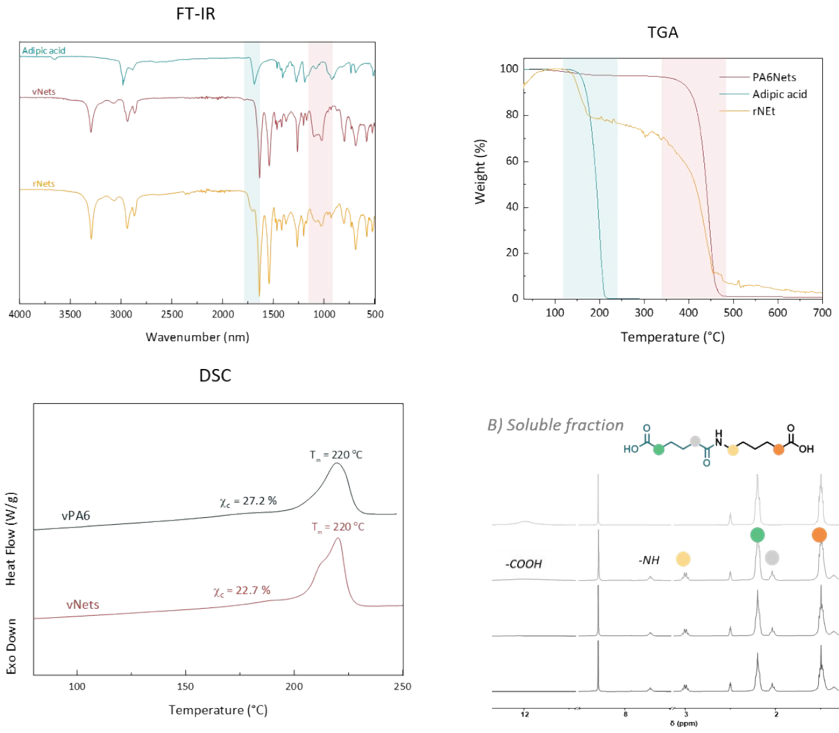
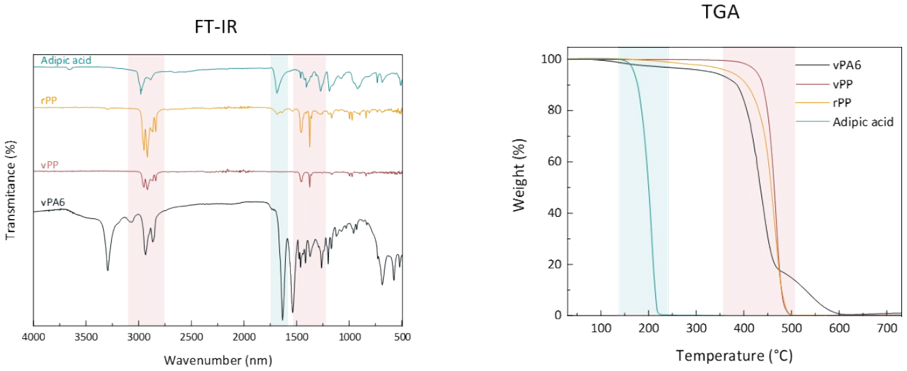


Figure S18. Depolymerization of PA6 from fishing nets with adipic acid. A) Characterization of the insoluble fraction by FT-IR, TGA and DSC compared with virgin reagents. B) Stacked ^{1}H-NMR spectra of the water-soluble products over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS). Reaction conditions: PA6 (0.5 g, 4.4 mmol, 1 eq.) with adipic acid (1.29 g, 8.8 mmol, 2 eq.) for 6 hours at 200 °C.

5.2. PA6-PP

A) Insoluble fraction



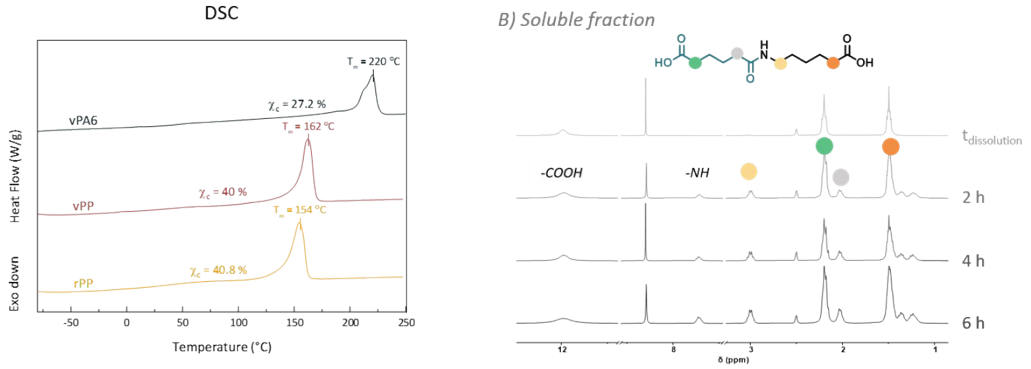


Figure S19. Depolymerization of PA6 from a mixture with PP in adipic acid. A) Characterization of the insoluble fraction by FT-IR, TGA and DSC[7] compared with virgin reagents. B) Stacked ^{1}H-NMR spectra of the water-soluble products over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS). Reaction conditions: PA6 (0.5 g, 4.4 mmol, 1 eq.) mixed with PP (0.19, 4.4 mmol, 1 eq.), in adipic acid (1.29 g, 8.8 mmol, 2 eq.) for 6 hours at 200 °C.

5.3. PA6-PET

A) Insoluble fraction

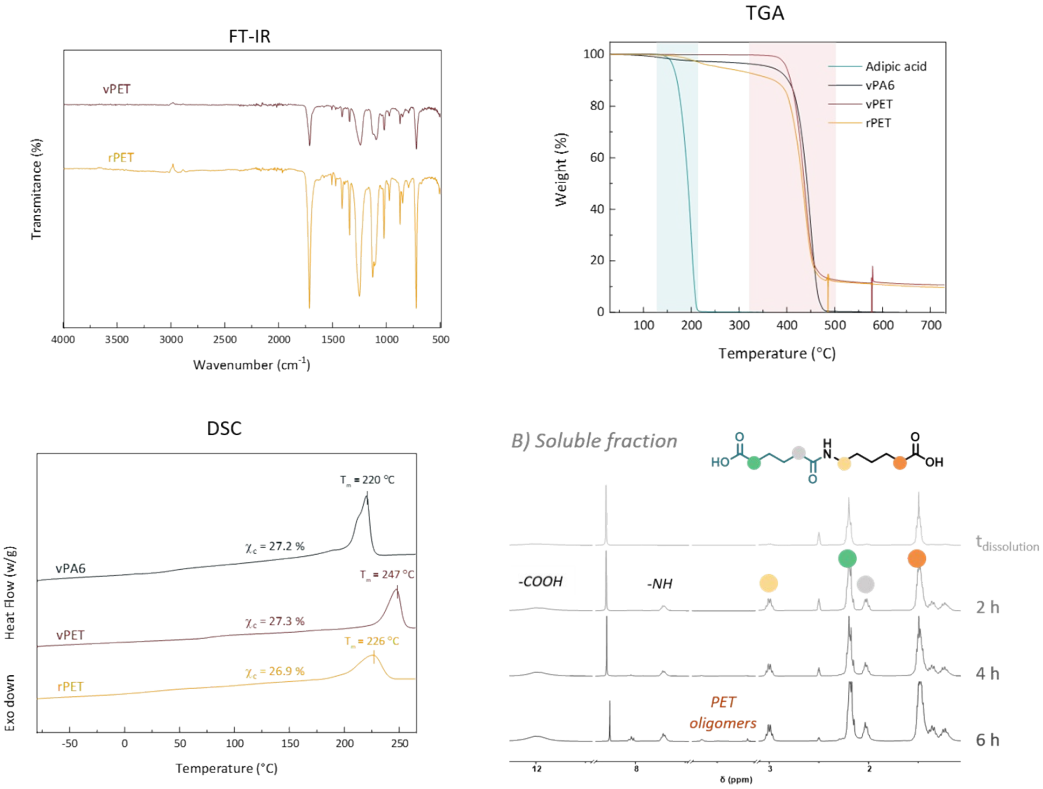


Figure S20. Depolymerization of PA6 from a mixture with PET in adipic acid. A) Characterization of the insoluble fraction by FT-IR, TGA and DSC[8] compared with virgin reagents. B) Stacked ^{1}H-NMR spectra of the water-soluble products over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS). Reaction conditions: PA6 (0.5 g, 4.4 mmol, 1 eq.) mixed with PET (0.84 g, 4.4 mmol, 1 eq.) in adipic acid (1.29 g, 8.8 mmol, 2 eq.) for 6 hours at 200 °C.

5.4. Facemasks (PA6-Cotton)

A) Insoluble fraction

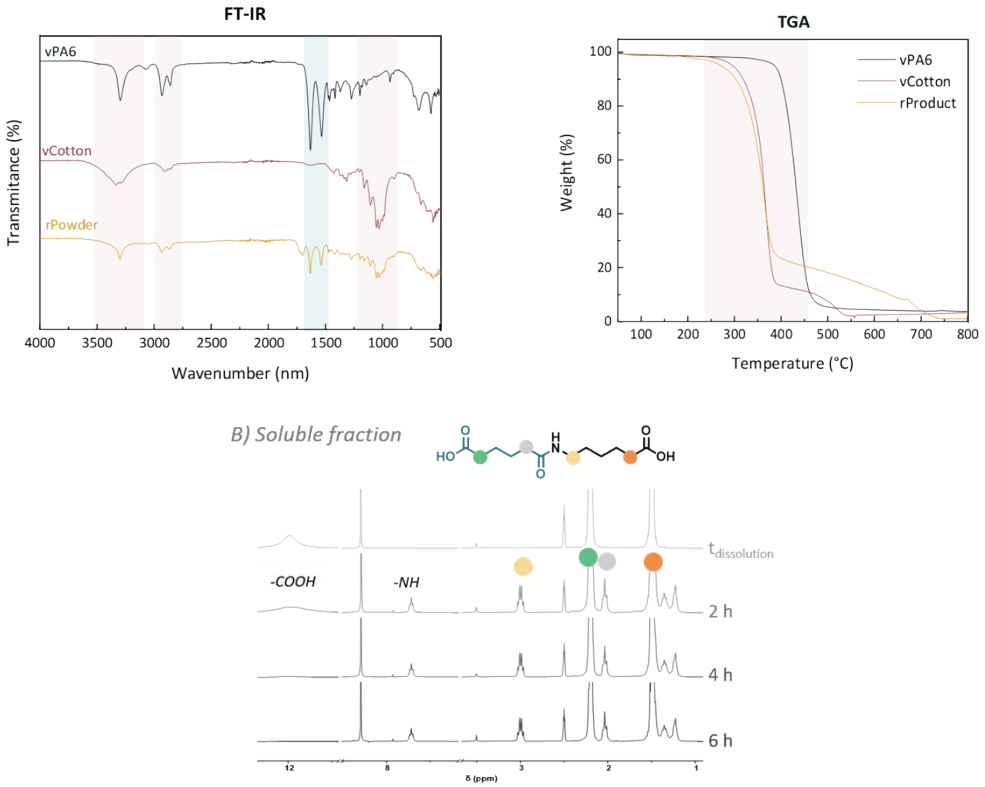


Figure S21. Depolymerization of PA6 from facemask (PA6:cotton 1:1) in adipic acid. A) Characterization of the insoluble fraction by FT-IR, TGA and DSC compared with virgin reagents. B) Stacked ^{1}H-NMR spectra of the water-soluble products over time (dissolved in $DMSO_{d6}$, 10 µL of $CHCl_3$ added as IS). Reaction conditions: PA6 (0.5 g, 4.4 mmol, 1 eq.) mixed with cotton (1.5 g, 4.4 mmol, 1 eq.) in adipic acid (2.6 g, 17.7 mmol, 4 eq.) for 6 hours at 200 °C.

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