SUPPORTING INFORMATION

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Biomass Waste-Derived Recyclable Heterogeneous Catalyst for Aqueous Aldol Reaction and Depolymerization of PET Waste

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 Table S1: XRF Analysis of OPA Catalyst.

Metal oxides	Concentration (%)		
CaO	31.322		
K ₂ O	29.38		
SO_3	15.792		
MgO	7.037		
P_2O_5	5.188		
Fe ₂ O ₃	2.152		
SiO ₂	1.115		
MnO	0.268		
CuO	0.201		
Cr ₂ O ₃	0.129		
Al_2O_3	0.123		
Na ₂ O	0.108		
	Metal oxides CaO K_2O SO_3 MgO P_2O_5 Fe_2O_3 SiO_2 MnO CuO Cr_2O_3 Al_2O_3 Na_2O		

Figure S1: EDX data of fresh OPA





EDAX TEAM

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eZAF Smart Quant Results

Element	Weight %	Atomic %	Net Int.	Error %	Kratio	Z	R	А	F
ОК	53.13	72.16	2,109.72	9.80	0.09	1.06	0.96	0.16	1
MgK	1.85	1.65	349.86	9.34	0.01	0.98	1	0.45	1.01
AIK	0.27	0.22	65.20	25.30	0.00	0.95	1	0.58	1.01
SiK	2.71	2.10	802.50	5.46	0.02	0.97	1.01	0.71	1.02
ΡK	6.18	4.34	1,628.71	3.97	0.05	0.93	1.02	0.79	1.02
SK	1.53	1.04	421.28	7.52	0.01	0.95	1.02	0.82	1.03
кк	0.98	0.54	244.49	8.95	0.01	0.9	1.04	0.96	1.12
CaK	32.51	17.62	6,262.06	1.69	0.29	0.92	1.04	0.97	1
FeK	0.84	0.33	73.68	30.81	0.01	0.82	1.07	0.98	1

Figure S4 (a): SEM image of the recovered OPA



Figure S4 (b): SEM image of the recovered OPA



Figure S4 (c): SEM image of the recovered OPA



Figure S4 (d): SEM image of the recovered OPA



Figure S5 (a): TEM image of the recovered OPA



Figure S5 (b): TEM image of the recovered OPA



Figure S5 (c): TEM image of the recovered OPA



Figure S5 (d): TEM image of the recovered OPA



Figure S6: HPLC data of the crude product

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Figure S7: HPLC data of the recrystallized DMT



Figure S8: HPLC data of commercial ethylene glycol

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Figure S9: HPLC data of HEMT

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1 Peak1

2155448

100.00

214317

100.00

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Figure S10: HPLC data of BHET

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Figure S11: ¹H NMR data of recrystallized DMT





Figure S13: IR data of recrystallized DMT



Scheme S1: Proposed reaction mechanism of aldol reaction with OPA catalyst Step 1: Generation of Carbanion and formation of enolate



Figure S14: HPLC data of the crude product using a reduced methanol loading



Breeze 2 HPLC System



Figure S15: HPLC data of the crude product after 2 hrs reaction time

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Table S2: Methanolysis of Colored PET Bottles using OPA

S1.	PET Color	Catalyst	MeOH	Time (h)	Temp.	DMT
No.		Loading	(mL)		(°C)	Yield %
1.	Clear	30	5	1	200	78
2.	Red	30	5	1	200	69
3.	Light-Yellow	30	5	1	200	61
4.	Green	30	5	1	200	74

Figure S16: UV-Vis Spectra of DMT monomer obtained from clear, red, light-yellow and green PET bottles. The samples were dissolved in chloroform (5 mg/ml).



Table S3: Reusability of the Catalyst

Sl.	No.	of	Catalyst	MeOH	Time	Temp.	DMT
No.	Cycle		Loading		(h)	(°C)	Yield %
1	First		30	5	1	200	78
2	Second		30	5	1.5	200	73
3	Third		30	5	2.5	200	71
4	Fourth		30	5	3.5	200	68

NMR SPECTRAL DATA OF THE ALDOL PRODUCT:

2-(hydroxy(4-nitrophenyl)methyl)cyclohexan-1-one (7a, white solid):



¹H NMR (400 MHz, CDCl₃): δ 8.22 - 7.47 (m,4H), 5.48 (d, *J* = 2.4 Hz, 1H for *syn*), 4.90 (d, *J* = 8.4hz, 1H for *anti*), 2.48 - 1.54 (m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 214.95, 214.26, 149.30, 148.58, 128.11, 126.85, 123.81, 123.70, 74.28, 70.38, 57.43, 57.04, 42.91, 42.85, 31.00, 28.08, 27.87, 26.16, 25.02, 24.94.

2-(hydroxy(3-nitrophenyl)methyl)cyclohexan-1-one (7b, oily liquid):



¹H NMR (400 MHz, CDCl₃): δ 8.21 - 7.51 (m, 4H), 5.48 (d, *J* = 2.0 Hz, 1H for *syn*), 4.89 (d, *J* = 8.4 Hz, 1H for *anti*), 2.62 - 1.55 (m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 215.06, 214.33, 148.55, 144.06, 143.51, 133.42, 132.18, 129.53, 129.38, 123.10, 122.32, 122.26, 121.11, 74.28, 70.15, 57.37, 57.00, 42.89, 42.84, 30.98, 28.09, 27.86, 26.15, 24.99, 24.90.

2-(hydroxy(2-nitrophenyl)methyl)cyclohexan-1-one (7c, oily liquid):



¹H NMR (400 MHz, CDCl₃): δ 8.11 - 7.42 (m, 4H), 5.96 (d, *J* = 2.0 Hz, 1H for *syn*), 5.47 (d, *J* = 8.4 Hz, 1H for *anti*), 2.46 - 1.55 (m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 214.28, 188.34, 137.22, 134.28, 133.92, 133.37, 129.88, 129.86, 128.17, 124.89, 124.74, 66.93, 55.08, 42.80, 28.23, 26.76, 25.10.

2-((4-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7d, white solid):



¹H NMR (400 MHz, CDCl₃): δ 7.41 - 7.10 (m, 4H), 5.27 (s, 1H for *syn*), 4.69-4.66 (dd, *J*₁ = 2.8 Hz, *J*₂ = 2.4 Hz, 1H for *anti*), 3.91 (d, *J* =2.8 Hz, 1H for OH), 2.50-1.03 (m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 213.96, 213.32, 142.97, 141.25, 131.43, 131.36, 127.98, 127.54, 121.72, 121.12, 74.13, 70.32, 57.43, 57.12, 42.67, 30.85, 28.12, 27.86, 26.14, 25.24, 24.97.

2-((3-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7e, white solid):



¹H NMR (400 MHz, CDCl₃): δ 7.25 - 7.08 (m, 4H), 5.28 (s, 1H for *syn*), 4.68 – 4.66 (d, *J* = 8.8 Hz, 1H for *anti*), 3.96 (s, 1H for OH), 2.52 - 1.46 (m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 215.41, 143.31, 134.58, 129.83, 129.67, 128.28, 127.38, 126.25, 125.53, 124.10, 74.50, 70.31, 57.50, 57.20, 42.89, 31.01, 28.13, 27.95, 26.16, 25.05, 24.93.

2-((2-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7f, white solid):



¹H NMR (400 MHz, CDCl₃): δ 7.55 - 7.18 (m, 4H), 5.71 (2, 1H for *syn*), 5.36 – 5.33 (dd, *J*₁ = 3.6 Hz, *J*₂ = 4.0 Hz, 1H for *anti*), 4.05 (d, *J* = 3.6, 1H for OH), 2.69 - 1.53 (m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 215.27, 214.79, 139.10, 138.58, 132.97, 130.86, 129.22, 128.75, 128.54, 128.26, 128.20, 127.26, 126.61, 70.45, 67.72, 57.60, 53.57, 42.73, 42.67, 30.40, 27.95, 27.82, 25.97, 24.92, 24.86.

2-((4-bromophenyl)(hydroxy)methyl)cyclohexan-1-one (7g, white solid):



¹H NMR (400 MHz, CDCl₃): δ 7.48 - 7.17 (m, 4H), 5.33 (d, *J* = 2.4 Hz, 1H for *syn*), 4.75 (d, *J* = 8.8 Hz, 1H for *anti*), 2.55 - 1.52(m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 215.27, 214.57, 140.53, 140.02, 131.49, 131.26, 128.75, 127.55, 121.72, 120.77, 74.19, 70.15, 57.33, 56.99, 42.67, 30.76, 27.93, 27.73, 25.96, 24.85, 24.71.

2-(hydroxy(phenyl)methyl)cyclohexan-1-one (7h, white solid):



¹H NMR (400 MHz, CDCl₃): δ 7.34 - 7.26 (m, 5H), 5.39 (d, *J* = 2.4 Hz, 1H for *syn*), 4.78 (d, *J* = 8.8 Hz, 1H for *anti*), 2.47 - 1.52(m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 215.78, 215.04, 141.72, 141.17, 128.63, 128.40, 128.15, 127.28, 127.23, 126.01, 75.38, 70.89, 57.67, 57.44, 42.93, 31.10, 28.19, 28.05, 26.26, 25.13, 24.98.

2-(hydroxy(p-tolyl)methyl)cyclohexan-1-one (7i, white solid):



¹H NMR (400 MHz, CDCl₃): δ 7.25-7.13 (m, 4H), 5.34 (d, *J* = 1.6 Hz, 1H for syn), 4.75 (d, *J* = 8.8 Hz, 1H for anti), 2.60 - 1.52 (m, 12H); ¹³CNMR (100 MHz, CDCl₃): δ 215.85, 215.07, 138.74, 138.22, 137.80, 136.79, 129.29, 129.08, 127.17, 125.94, 74.80, 70.82, 57.68, 57.49, 42.91, 31.12, 28.19, 28.06, 26.35, 25.13, 24.90, 21.38, 21.30.

2-(hydroxy(4-methoxyphenyl)methyl)cyclohexan-1-one (7j, light yellow solid):



¹H NMR (400 MHz, CDCl₃): δ 7.86 - 7.82 (m, 4H), 5.32 (d, *J* = 2.4 Hz, 1H for syn), 4.74 (d, *J* = 8.8 Hz, 1H for *anti*), 3.89 (s, 3H, for *syn*-isomer), 3.80 (s, 3H, for *anti*-isomer), 2.59 - 1.25 (m, 9H); ¹³CNMR (100 MHz, CDCl₃): δ 215.87, 215.11, 191.05, 159.52, 158.86, 133.86, 133.40, 132.23, 128.39, 127.16, 114.56, 114.03, 113.82, 74.51, 70.65, 57.74, 57.51, 55.50, 42.91, 42.89, 31.08, 28.20, 28.03, 26.43, 25.11, 24.96.

4-hydroxy-4-(4-nitrophenyl)butan-2-one (7k, light yellow solid):



¹H NMR (400 MHz, CDCl₃): δ 8.27 - 7.09 (m, 4H), 5.17 - 5.07 (dd, *J* = 2.4, 2.4 Hz, 1H), 3.02 - 1.73 (m, 6H); ¹³CNMR (100 MHz, CDCl₃): δ 197.74, 148.88, 140.91, 140.31, 130.62, 129.04, 128.72, 124.45, 123.99, 50.80, 44.64, 28.27.

4-hydroxy-4-(3-nitrophenyl)butan-2-one (7l, light pinkish white solid):



¹H NMR (400 MHz, CDCl₃): δ 8.41 - 7.53 (m, 4H), 5.19 - 5.07 (dd, *J* = 2.4, 2.4 Hz, 1H), 3.00 - 1.69 (m, 6H); ¹³CNMR (100 MHz, CDCl₃): δ 197.76, 151.23, 140.41, 136.54, 133.97, 130.28, 129.64, 124.94, 122.84, 51.98, 28.31.

4-(4-chlorophenyl)-4-hydroxybutan-2-one (7m, white solid):



¹H NMR (400 MHz, CDCl₃): δ 7.32 - 7.23 (m, 4H), 5.14 - 5.10 (dd, *J* =3.6, 3.6 Hz, 1H), 2.98 - 2.14 (m, 6H); ¹³CNMR (100 MHz, CDCl₃): δ 198.08, 141.88, 136.47, 132.96, 129.40, 129.28, 127.50, 51.28, 27.67.

4-hydroxy-4-phenylbutan-2-one (7n, colourless liquid):



¹H NMR (400 MHz, CDCl₃): δ 7.33 - 7.23 (m, 5H), 5.13 - 5.10 (dd, *J* =3.6, 3.6 Hz, 1H), 2.99 - 2.15 (m, 6H); ¹³CNMR (100 MHz, CDCl₃): δ 209.17, 143.03, 128.66, 127.79, 125.78, 69.99, 52.16, 30.88.

4-hydroxy-4-(p-tolyl)butan-2-one (70, light yellow liquid):



¹H NMR (400 MHz, CDCl₃): δ 7.62 - 7.18 (m, 4H), 5.51 - 5.48 (dd, *J* = 2.4, 2.4 Hz, 1H), 3.00 - 2.64 (m, 6H), 2.20 (s, 3H); ¹³CNMR (100 MHz, CDCl₃): δ 209.38, 140.35, 131.34, 129.54, 128.79, 127.45, 127.29, 50.23.

4-hydroxy-4-(4-methoxyphenyl)butan-2-one (7p, white solid):



¹H NMR (400 MHz, CDCl₃): δ 7.51 - 7.6.91 (m, 4H), 5.53 - 5.46 (dd, *J* = 2.4, 2.4 Hz,1H), 3.84 (s, 3H), 3.01 - 2.35 (m, 6H); ¹³CNMR (100 MHz, CDCl₃): δ 198.63, 161.87, 143.49, 130.19, 127.33, 125.28, 114.69, 55.63, 27.61.

3-hydroxy-3-(4-nitrophenyl)-1-phenylpropan-1-one (7q, white solid):



¹H NMR (400 MHz, CDCl₃): δ 8.24 - 7.26 (m, 9H), 5.47 - 5.44 (dd, *J* = 3.7, 3.2 Hz, 1H), 3.44 - 3.31 (m, 3H); ¹³CNMR (100 MHz, CDCl₃): δ 199.73, 150.45, 147.62, 136.43, 134.25, 129.07, 128.39, 126.80, 124.04, 69.47, 47.20.

3-hydroxy-3-(3-nitrophenyl)-1-phenylpropan-1-one (7r, light yellow solid):



¹H NMR (400 MHz, CDCl₃): δ 8.32 - 7.46 (m, 9H), 5.46 - 5.44 (dd, *J* = 3.2, 3.6 Hz, 1H), 3.46 - 3.33 (m, 3H); ¹³CNMR (100 MHz, CDCl₃): δ 199.80, 148.68, 145.34, 136.45, 134.20, 132.17, 129.74, 129.04, 128.39, 122.81, 121.11.



Figure S17: ¹H NMR data of 2-(hydroxy(4-nitrophenyl)methyl)cyclohexan-1-one (7a)

Figure S18: ¹³C NMR data of 2-(hydroxy(4-nitrophenyl)methyl)cyclohexan-1-one (7a)





Figure S19: ¹H NMR data of 2-(hydroxy(3-nitrophenyl)methyl)cyclohexan-1-one (7b)

Figure S20: ¹³C NMR data of 2-(hydroxy(3-nitrophenyl)methyl)cyclohexan-1-one (7b)





Figure S21: ¹H NMR data of 2-(hydroxy(2-nitrophenyl)methyl)cyclohexan-1-one (7c)

Figure S22: ¹³C NMR data of 2-(hydroxy(2-nitrophenyl)methyl)cyclohexan-1-one (7c)





Figure S23: ¹H NMR data of 2-((4-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7d)

Figure S24: ¹³C NMR data of 2-((4-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7d)





Figure S25: ¹H NMR data of 2-((3-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7e)

Figure S26: ¹³C NMR data of 2-((3-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7e)





Figure S27: ¹H NMR data of 2-((2-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7f)

Figure S28: ¹³C NMR data of 2-((2-chlorophenyl)(hydroxy)methyl)cyclohexan-1-one (7f)





Figure S29: ¹H NMR data of 2-((4-bromophenyl)(hydroxy)methyl)cyclohexan-1-one (7g)

Figure S30: ¹³C NMR data of 2-((4-bromophenyl)(hydroxy)methyl)cyclohexan-1-one (7g)





Figure S31: ¹H NMR data of 2-(hydroxy(phenyl)methyl)cyclohexan-1-one (7h)

Figure S32: ¹³C NMR data of 2-(hydroxy(phenyl)methyl)cyclohexan-1-one (7h)





Figure S33: ¹H NMR data of 2-(hydroxy(p-tolyl)methyl)cyclohexan-1-one (7i)

Figure S34: ¹³C NMR data of 2-(hydroxy(p-tolyl)methyl)cyclohexan-1-one (7i)





Figure S35: ¹H NMR data of 2-(hydroxy(4-methoxyphenyl)methyl)cyclohexan-1-one (7j)

Figure S36: ¹³C NMR data of 2-(hydroxy(4-methoxyphenyl)methyl)cyclohexan-1-one (7j)





Figure S37: ¹H NMR data of 4-hydroxy-4-(4-nitrophenyl)butan-2-one (7k)

Figure S38: ¹³C NMR data of 4-hydroxy-4-(4-nitrophenyl)butan-2-one (7k)





Figure S39: ¹H NMR data of 4-hydroxy-4-(3-nitrophenyl)butan-2-one (7l)

Figure S40: ¹³C NMR data of 4-hydroxy-4-(3-nitrophenyl)butan-2-one (7l)





Figure S41: ¹H NMR data of 4-(4-chlorophenyl)-4-hydroxybutan-2-one (7m)

Figure S42: ¹³C NMR data of 4-(4-chlorophenyl)-4-hydroxybutan-2-one (7m)





Figure S43: ¹H NMR data of 4-hydroxy-4-phenylbutan-2-one (7n)

Figure S44: ¹³C NMR data of 4-hydroxy-4-phenylbutan-2-one (7n)





Figure S45: ¹H NMR data of 4-hydroxy-4-(p-tolyl)butan-2-one (70)

Figure S46: ¹³C NMR data of 4-hydroxy-4-(p-tolyl)butan-2-one (70)





Figure S47: ¹H NMR data of 4-hydroxy-4-(4-methoxyphenyl)butan-2-one (7p)

Figure S48: ¹³C NMR data of 4-hydroxy-4-(4-methoxyphenyl)butan-2-one (7p)





Figure S49: ¹H NMR data of 3-hydroxy-3-(4-nitrophenyl)-1-phenylpropan-1-one (7q)

Figure S50: ¹³C NMR data of 3-hydroxy-3-(4-nitrophenyl)-1-phenylpropan-1-one (7q)





Figure S51: ¹H NMR data of 3-hydroxy-3-(3-nitrophenyl)-1-phenylpropan-1-one (7r)

Figure S52: ¹³C NMR data of 3-hydroxy-3-(3-nitrophenyl)-1-phenylpropan-1-one (7r)

