Supplementary Materials for

Marine waste upcycling—recovery of nylon monomer from fishing net waste using seashell waste-derived catalysts in a CO_2-mediated thermocatalytic process

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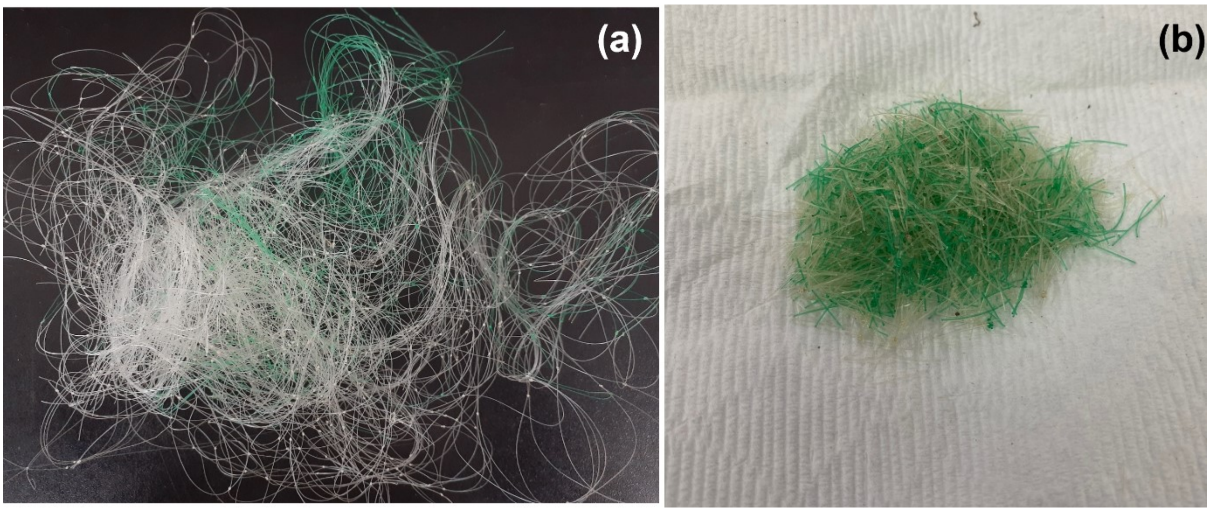


Figure S1. Ghost net used as the feedstock in this study: (a) before cutting and (b) after cutting.

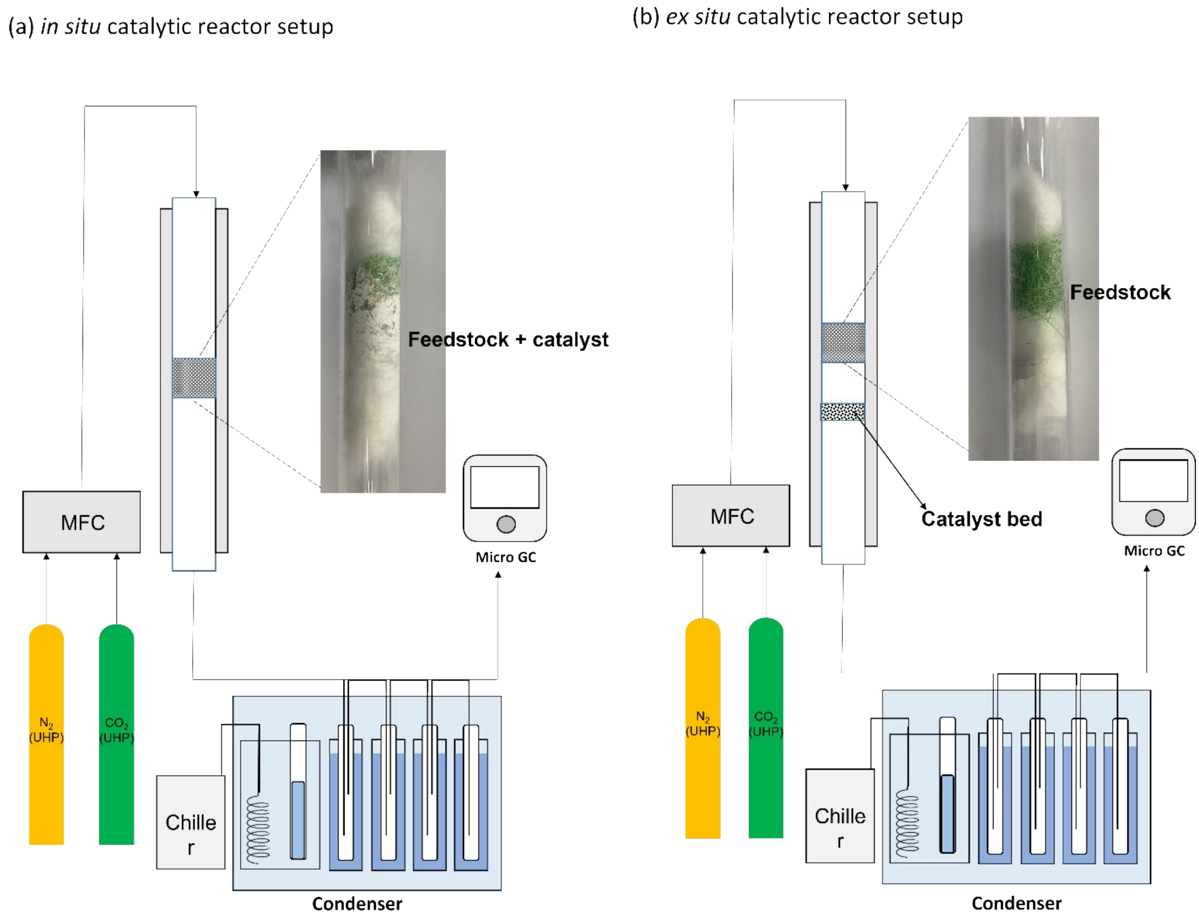


Figure S2. Schematic diagram of (a) *in situ* catalytic reactor setup and (b) *ex situ* catalytic reactor setup used for thermocatalytic conversion of the ghost net feedstock.

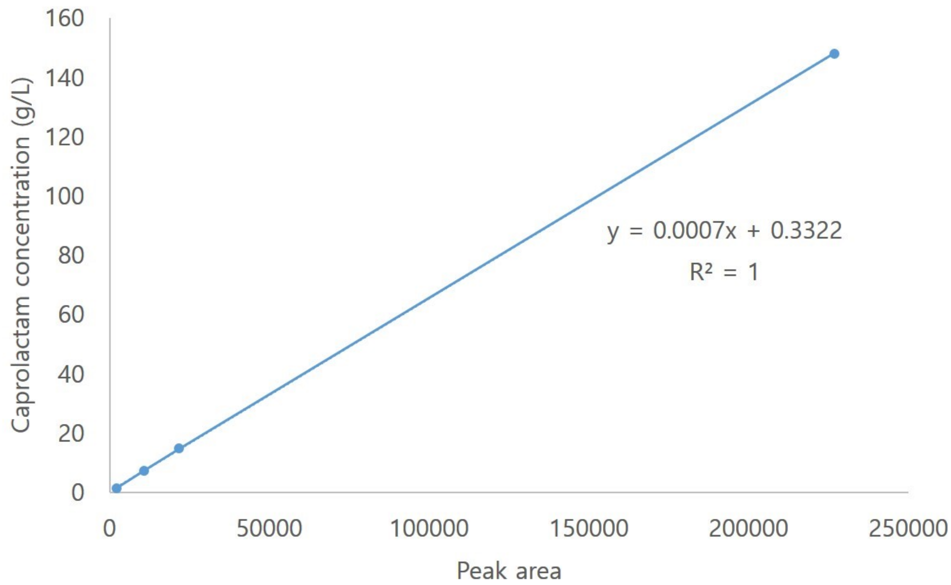


Figure S3. Four-point calibration curve used to quantify caprolactam in the pyrolytic product using a GC-FID.

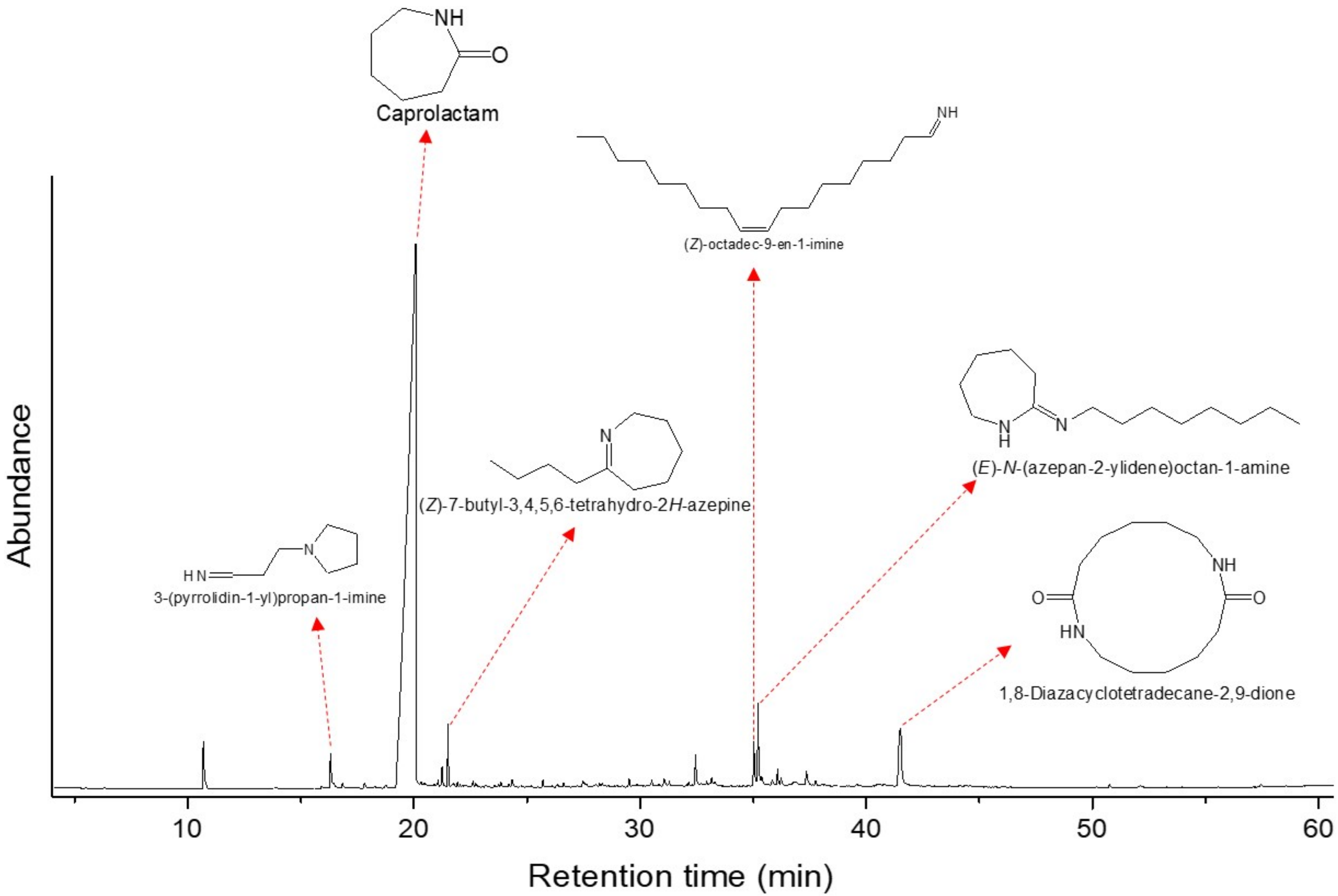


Figure S4. A representative GC-MS chromatogram of the liquid product produced via thermocatalytic conversion of the fishing net waste.

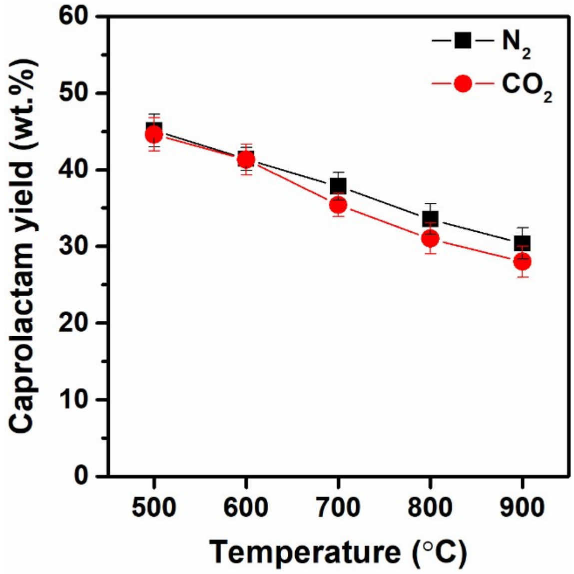


Figure S5. Yield of caprolactam (on the basis of weight of the feedstock) produced from non-catalytic thermal conversion of the fishing net waste under N_2 and CO_2 environments as a function of temperature.

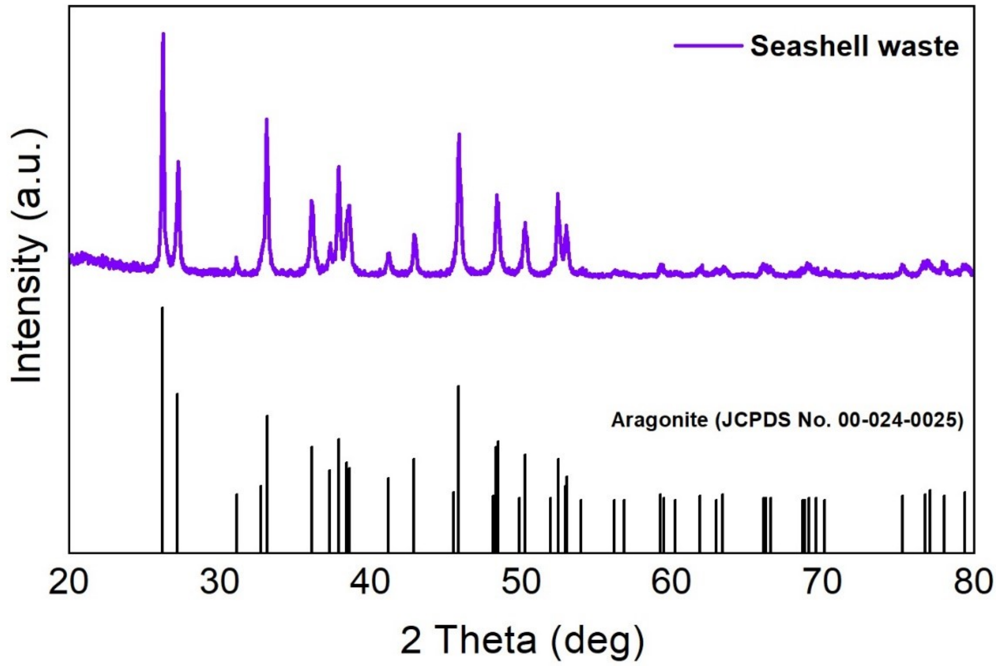


Figure S6. XRD pattern of seashell waste.

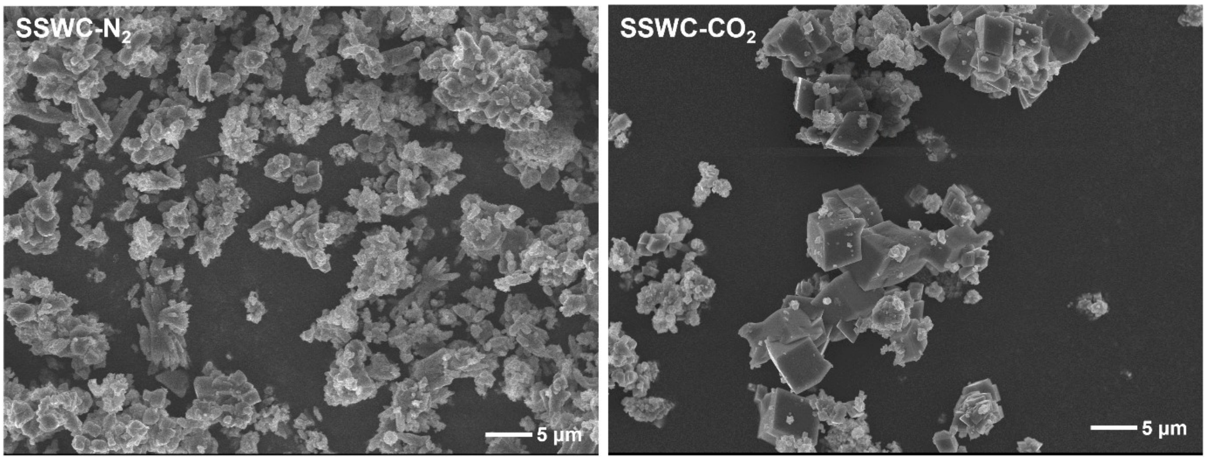


Figure S7. Scanning electron microscopy (SEM) images of SSWC-N_2 and SSWC-CO_2 with a magnification of 5 μm.

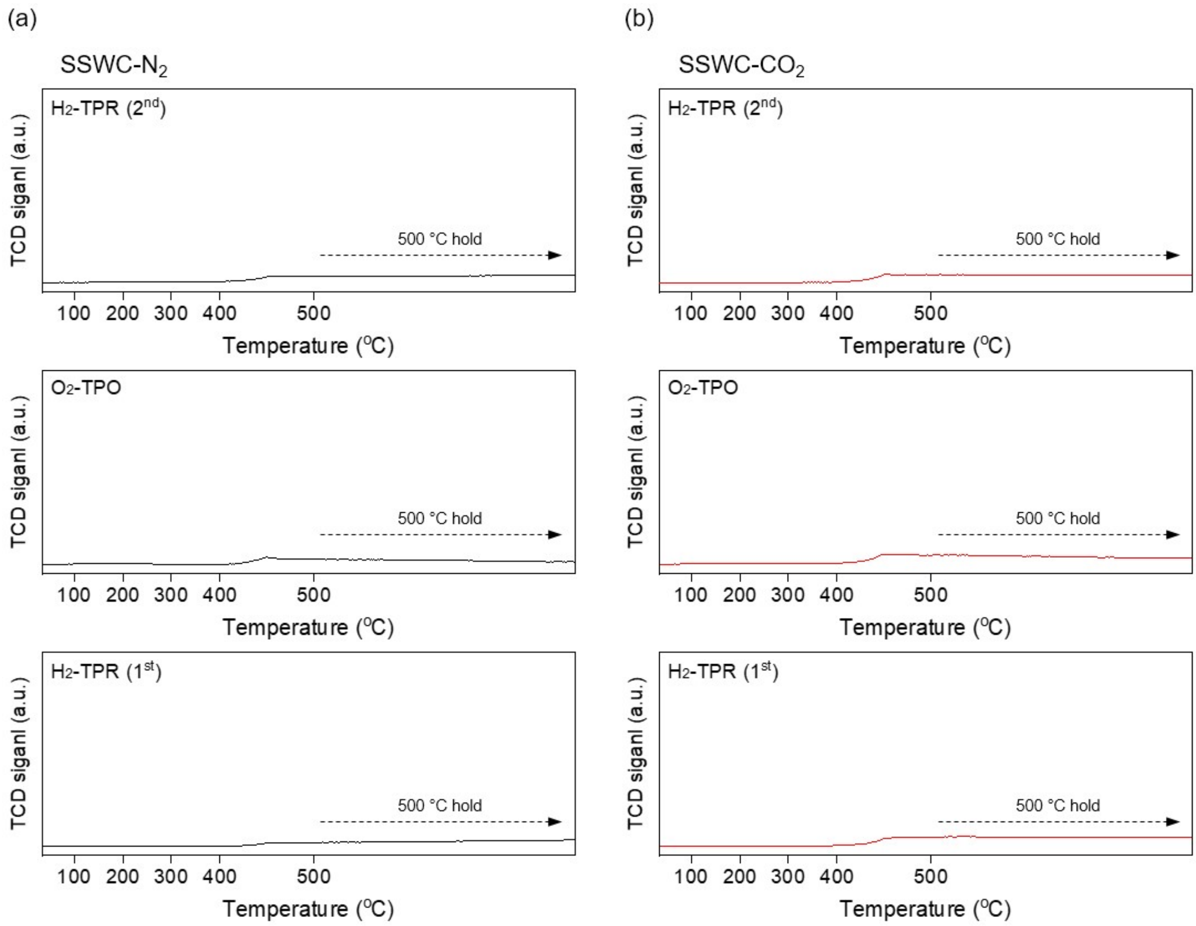


Figure S8. The sequence analysis of H_2-TPR, O_2-TPO, and H_2-TPR for (a) SSWC-N_2 and (b) SSWC-CO_2 catalysts.

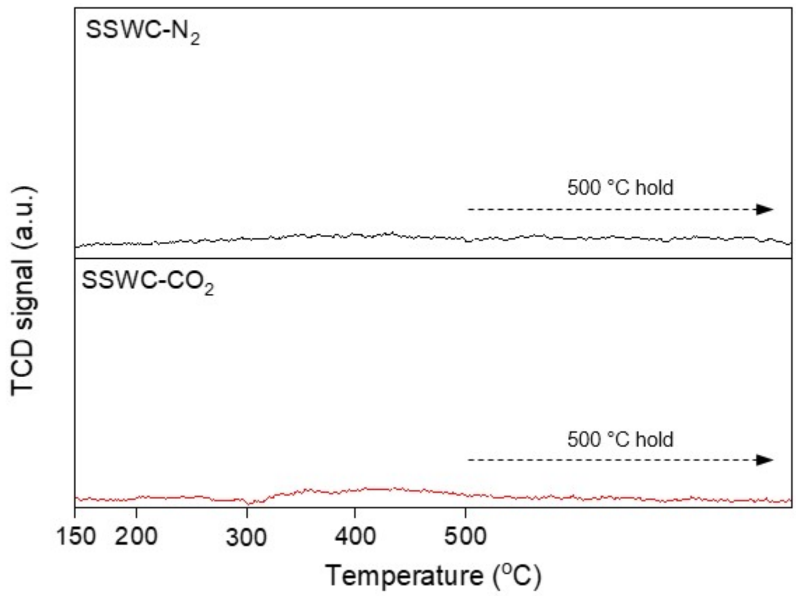


Figure S9. NH_3-TPD profiles of the SSWC-N_2 and SSWC-CO_2 catalysts.

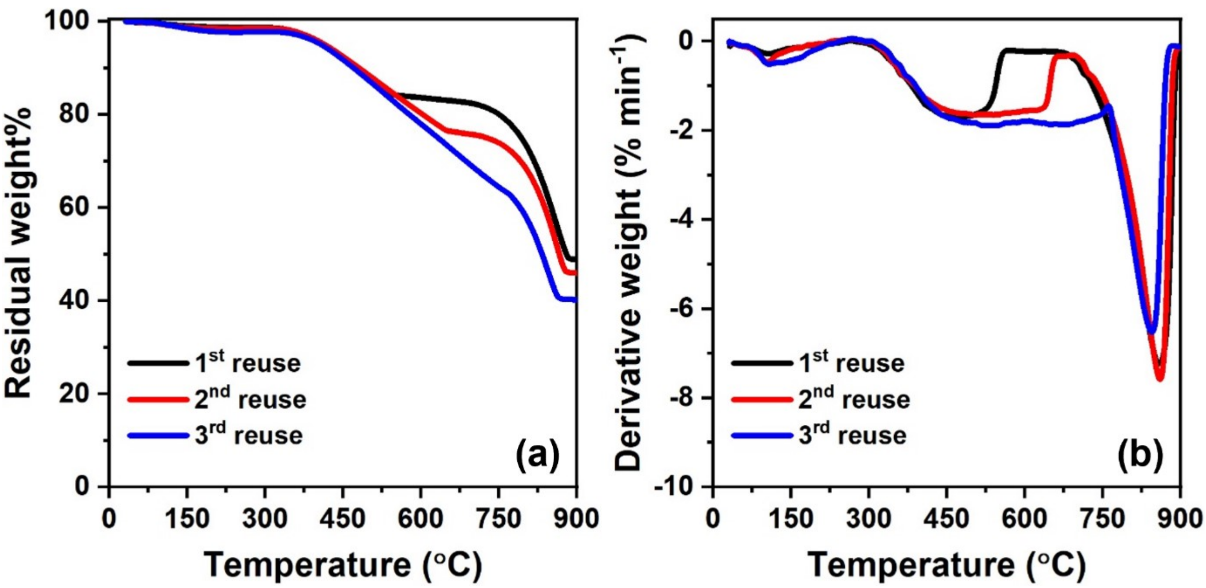


Figure S10. Thermogravimetric analysis of spent SSWC-CO_2 catalyst in air: (a) residual mass change in terms of temperature; and (b) derivative weight variation in terms of temperature.

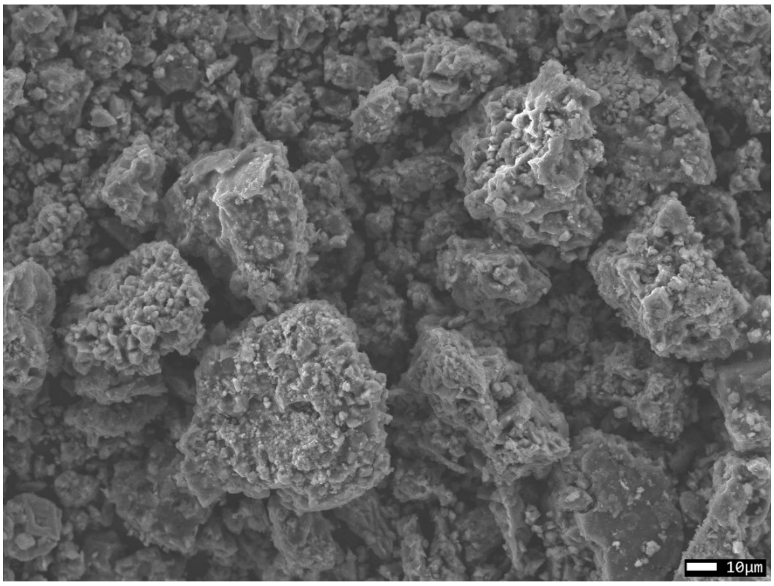


Figure S11. SEM image of the spent SSWC-CO_2 after 3rd reuse.

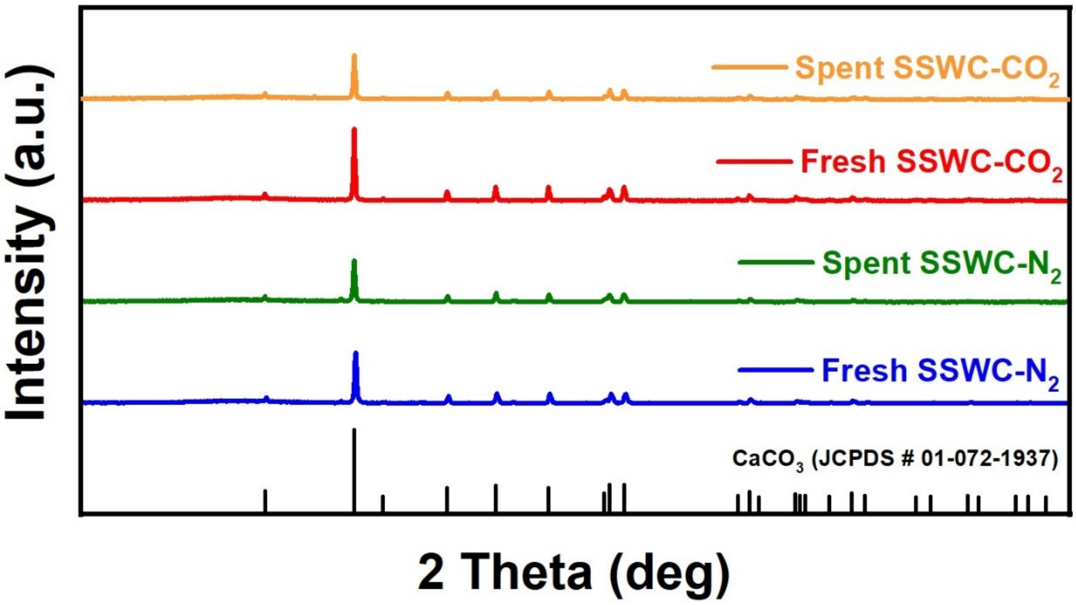


Figure S12. XRD patterns of fresh and spent SSWC-N_2 and fresh and spent SSWC-CO_2 catalysts.

Table S1. Specification, column information, and analytical conditions for the micro-GC.

Conditions		Module A	Module B
Column		Rt-Molsieve 5A	Rt-Q-Bond
Sample Pump setting	Sample pump mode	Continuous	Continuous
	Sample pump time	20 s	20 s
Column setting	Carrier gas	Argon (≥99.999%)	Helium (≥99.999%)
	Column pressure	20 psi	17 psi
	Initial temperature	50 °C (40 s)	50 °C (30 s)
	Ramping time	50 s	60 s
	Final temperature	100 °C (40 s)	110 °C (40 s)
	Total analysis time	130 s	130 s
Injector setting	Inject time	30 ms	30 ms
	Injector temperature	90 °C	90 °C
TCD setting	TCD temperature	70 °C	70 °C
	Data rate	50 Hz	50 Hz

Table S2. Specification, column information, and analytical conditions for the GC-MS.

Column	HP-5MS Ultra Inlet column (0.25 mm × 0.25 um × 30 m)	
Oven setting	Initial temperature	40 °C (1 min)
	Ramping	3 °C min^{-1}
	Final temperature	280 °C (19 min)
	Total analysis time	100 min
Column setting	Carrier gas	Helium (≥99.999%)
	Carrier gas flow	1.5 mL min^{-1}
	Column flow	1 mL min^{-1}
Injector setting	Injection mode	Splitless
	Injection volume	1 μL
	Injection temperature	275 °C
MS setting	Aux temperature	300 °C
	m/z range	50~500 amu

Table S3. Information of byproducts identified in the liquid product produced via thermocatalytic conversion of the fishing net waste.

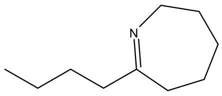
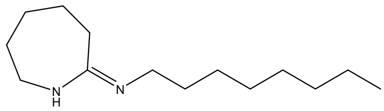
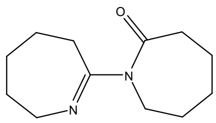
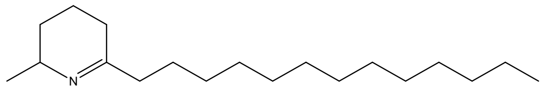
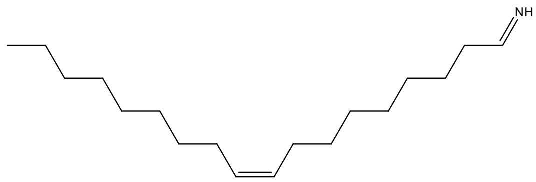
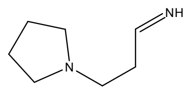
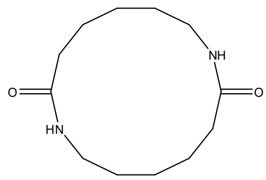
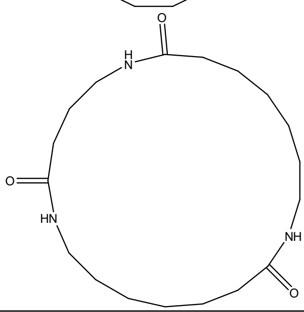
Chemical species	Chemical formula	MW	Chemical structure
Azepines			
7-butyl-3,4,5,6(2*H*)-tetrahydroazepine	$C_{10}H_{19}N$	153.26	
(*E*)-*N*-(azepan-2-ylidene)octan-1-amine	$C_{14}H_{28}N_2$	224.39	
1-(3,4,5,6-tetrahydro-2H-azepin-7-yl)-2-azepanone	$C_{12}H_{20}N_2O$	208.30	
Imines			
2-methyl-6-tridecyl-6-piperidene	$C_{19}H_{37}N$	279.50	
(*Z*)-octadec-9-en-1-imine	$C_{18}H_{35}N$	265.48	
3-(pyrrolidin-1-yl)propan-1-imine	$C_7H_{14}N_2$	126.20	
Cyclic dimers			
1,8-diazacyclotetradecane-2,9-dione	$C_{12}H_{22}N_2O_2$	226.32	
1,6,14-triazacyclohenicosane-2,7,15-trione	$C_{18}H_{33}N_3O_3$	339.47	

Table S4. Composition of the SSWC-N_2 and SSWC-CO_2 catalysts (unit: wt.%).

Constituent	SSWC-N_2	SSWC-CO_2
$CaCO_3$	89.9	96.3
Ba	0.0017	0.0018
Cr	0.0014	0.0152
Cu	0.0002	0.0004
Fe	0.0084	0.0474
K	8.96	2.0
Li	0.00009	0.00017
Mg	0.0113	0.0106
Mn	0.00027	0.0006
Na	0.14	0.11
Ni	0.00098	0.0138
P	0.0506	0.0233
Sr	0.0968	0.10
Si	0.0434	0.0506
Zn	0.00018	0.00026

Table S5. Comparison of caprolactam yields obtained from fishing nets via thermocatalytic processes with different catalysts.

Feedstock	Catalyst	Reaction conditions	Caprolactam yield (wt.%)	Ref.
Fishing net	ZSM-5	N_2 environment; T = 342–476 °C; catalyst/feed = 0.2 (w/w)	83 (GC-MS area%)	1
Fishing net	Scallop shell calcined under air	He environment; T = 410 °C; catalyst/feed = 5 (w/w); residence time = 2 min	66	2
Commercial nylon 6	Scallop shell calcined under air	He environment; T = 410 °C; catalyst/feed = 5 (w/w); residence time = 2 min	70	2
Fishing net waste	Seashell waste-derived catalysts under CO_2 (SSWC-CO_2)	CO_2 environment; T = 500 °C; catalyst/feed = 0.05 (w/w); residence time = 1 min	80	This work

Table S6. Data used for estimating microplastic release from ghost net and the value of caprolactam made via the thermocatalytic upcycling process of ghost net.

Data	Value	Ref.
The amount of ghost nets	640,000 metric tons per year	[3]
Release of microplastic from fishing gear abandoned at sea	1277 microplastic pieces m^{-1}	[4]
Contribution of ghost net to microplastic release	49%	[4]
Density of ghost net	0.00057 g cm^{-1}	Measured ourselves
Caprolactam market price (as of January 16, 2022)	CFR 2,020 USD per ton	[5]

References

1. J. Eimontas, S. Yousef, N. Striūgas and M. A. Abdelnaby, *Renew. Energ.*, 2021, **179**, 1385-1403.

2. N. Chaihad, I. Kurnia, A. Yoshida, C. Watanabe, K. Tei, P. Reubroycharoen, Y. Kasai, A. Abudula and G. Guan, *J. Anal. Appl. Pyrolysis*, 2020, **146**, 104750.

3. M. Charter, R. Carruthers and S. F. Jensen, *Products from waste fishing nets*, Circular Ocean led by the Environmental Research Institute, Scotland, 2018.

4. L. S. Wright, I. E. Napper and R. C. Thompson, *Mar. Pollut. Bull.*, 2021, **173**, 113115.

5. Caprolactam (CPL) price, https://www.echemi.com/productsInformation/pid_Seven1952-caprolactam-cpl.html, (accessed January 16, 2022).