

Supplementary Information for:

Carbon Dioxide-Negative Composite Materials: an Economically Viable Solution for CO₂ Sequestration

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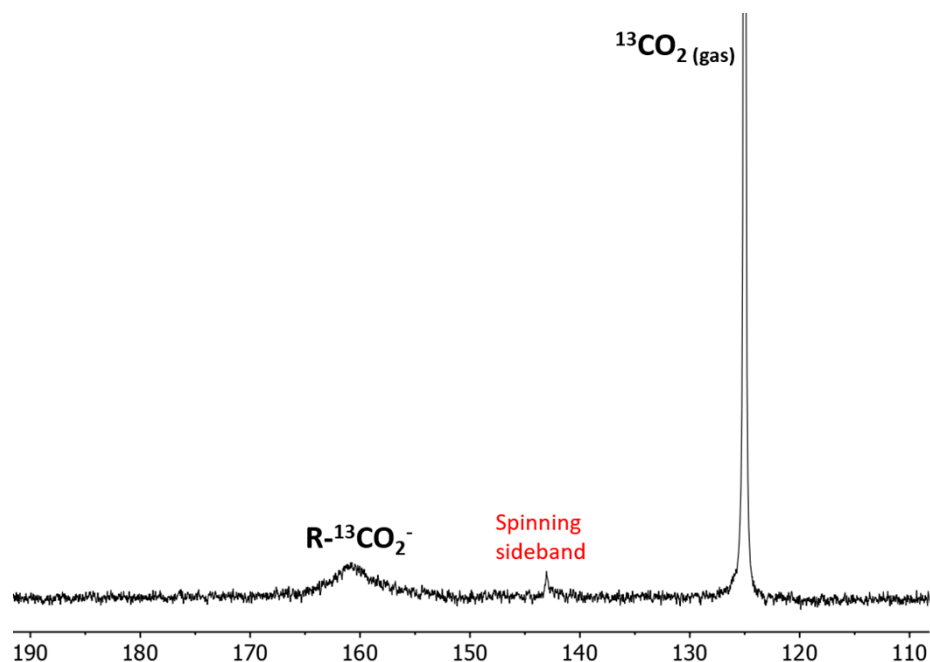


Figure S1. ^{13}C NMR of alkaline lignin, 1 equivalent of NaOH, 99% ^{13}C -enriched CO_2 at 300 psig, at 90 °C.
*Lignin is natural abundance and appears below the baseline.

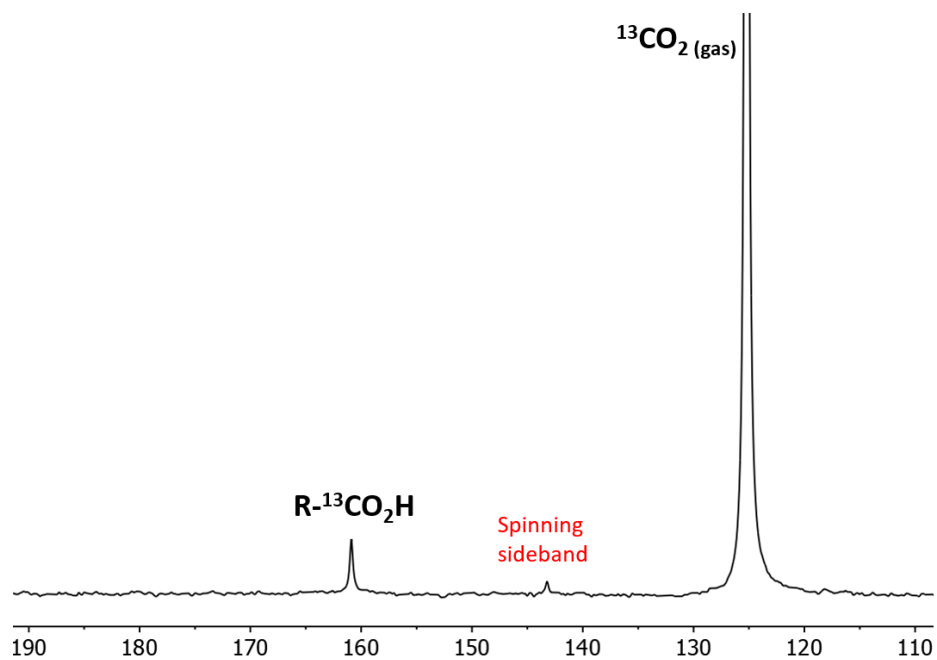


Figure S2. ^{13}C NMR of alkaline lignin, 1 equivalent of NaOH, ^{13}C -enriched CO_2 at 300 psig, at 90 °C followed by *in-situ* 1 M H_2SO_4 addition. *Lignin is natural abundance and appears below the baseline.

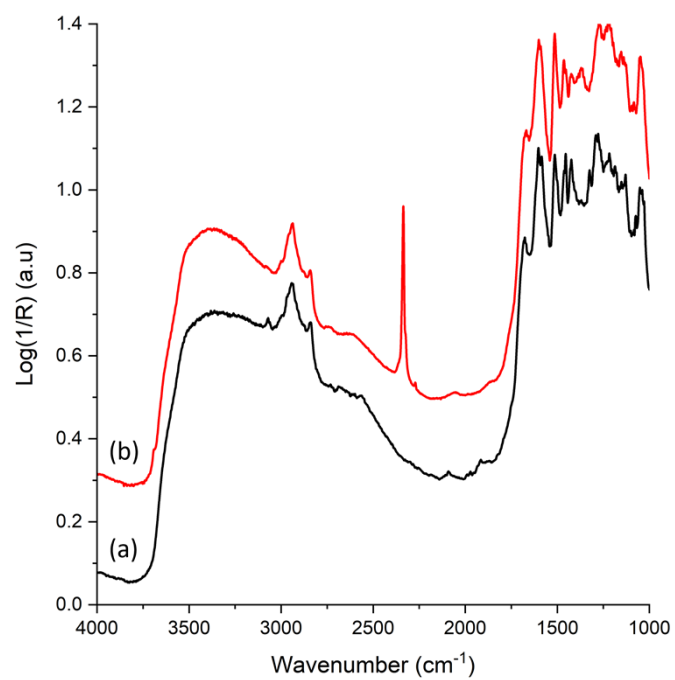
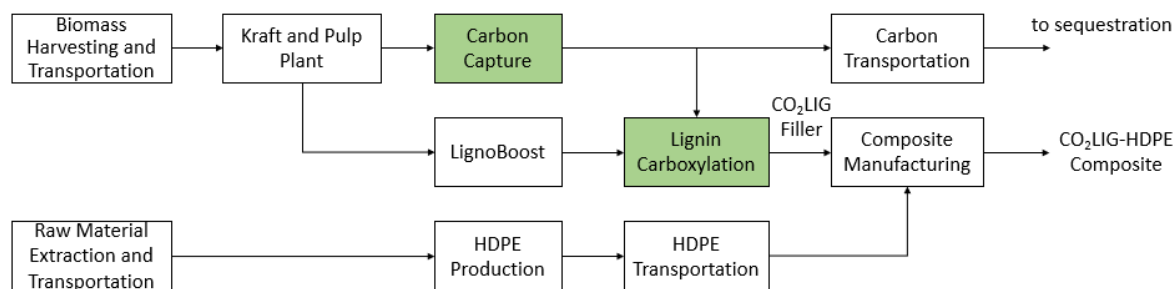
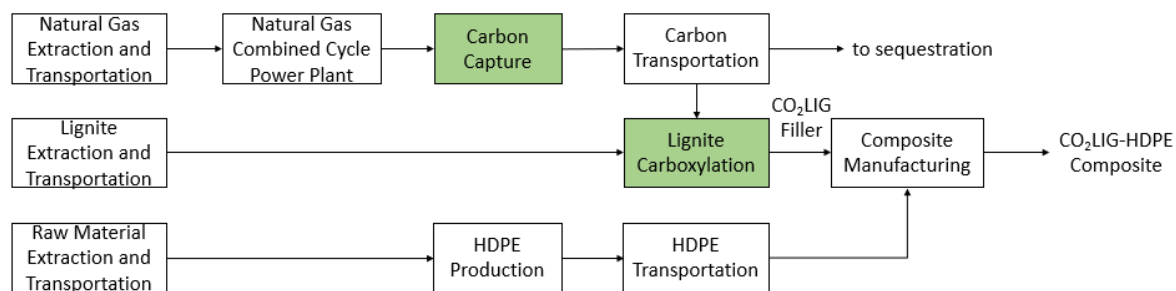


Figure S3 (a) DRIFT spectrum of alkaline lignin mixed with benzoic acid and KBr and (b) in situ DRIFT spectrum of alkaline lignin under nitrogen flow after cooling.

(a) Lignin case



(b) Lignite case



■ Included in TEA with rigorous process models developed in Aspen Plus

□ Included in both TEA and LCA with mass and energy balance or life cycle inventory data from literature or databases

Figure S4. System boundary for TEA and LCA: (a) lignin case and (b) lignite case.

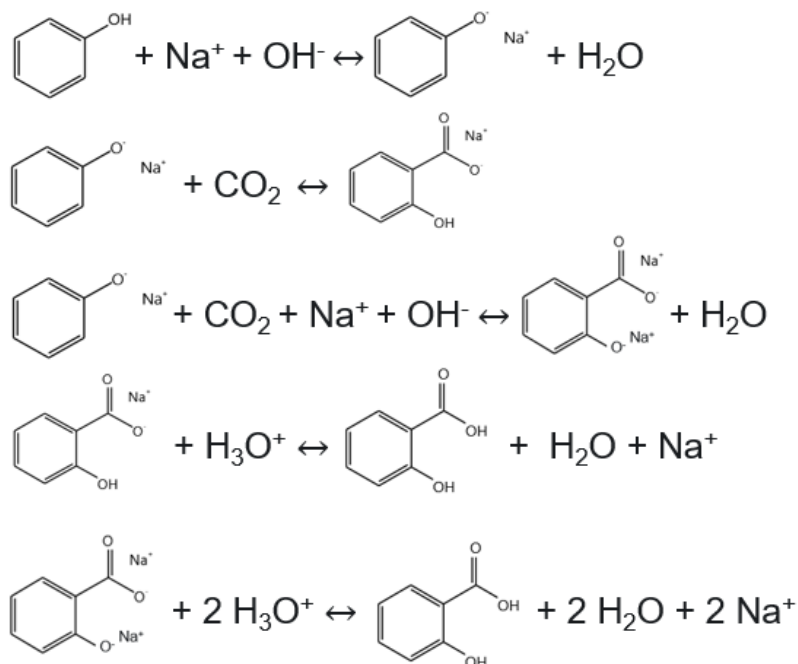


Figure S6. Representative reactions in the carboxylation unit.

Supplementary Tables 1 – 3

Table S1. Data sources for other process units.

Unit	Reference
Kraft and pulp plant with LignoBoost	5
HDPE supply chain	DATASMART LCI Package v.2021.1
Natural gas combined cycle power plant	6
Composite manufacturing	(Sommerhuber et al., 2017)
Biomass supply chain	DATASMART LCI Package v.2021.1
Lignite supply chain	U.S. Life Cycle Inventory (LCI) Database

Table S2. Key economic and price assumptions.

Assumption	Value
Pricing basis	2018 Q4

Scale, lignin	6.1 metric ton/h
Scale, lignite	122 metric ton/h
Lignin	\$300/metric ton
Lignite	\$20.1/metric ton
NaOH	\$1.02/kg
CaO	\$0.12/kg
NaCl	\$0.59/kg
HCl	\$0.44/kg
H ₂ SO ₄	\$0.1/kg
Acetone	\$0.99/kg
Cooling water	\$0.15/MGal
Electricity	\$50.4/MWh

Table S3. Mass and energy balances of the carboxylation unit.

Product	Lignin-HCl	Lignin-H ₂ SO ₄	Lignite-H ₂ SO ₄
CO ₂ LIG filler	4,644 kg/h	3,871 kg/h	46,768 kg/h
Co-product			
NaCl	339 kg/h		
Raw Materials			
Lignin (5wt% H ₂ O)	6,110 kg/h	6,110 kg/h	
Lignite (35wt% H ₂ O)			122,200 kg/h
NaOH	232 kg/h	232 kg/h	9,360 kg/h
CO ₂	256 kg/h	256 kg/h	3,100 kg/h
HCl	212 kg/h		
Acetone	21 kg/h		
H ₂ SO ₄		1138 kg/h	22,740 kg/h
CaO		490 kg/h	6,440 kg/h
Utilities			
Electricity	1865 kW	475 kW	10,377 kW
Emissions			
Wastewater		161 Mgal/h	3,174 Mgal/h

Supplementary References

1. Jiang, Y.; Mathias, P. M.; Zheng, R. F.; Freeman, C. J.; Barpaga, D.; Malhotra, D.; Koech, P. K.; Zwoster, A.; Heldebrant, D.J. Energy-effective and Low-cost Carbon Capture from Point-sources Enabled by Water-lean Solvents. *J. Clean. Prod.* **2023**, *388*, 135696.
2. Wooley R. J.; Putsche, V.; Development of an Aspen Plus Physical Property Database for Biofuel Components. NREL/TP-425-20685, 1996.
3. Frischknecht, R. *et al.* The ecoinvent database: Overview and methodological framework. *Int J Life Cycle Ass* **10**, 3-9 (2005). <https://doi.org/10.1065/lca2004.10.181.1>
4. Sommerhuber, P. F., Wenker, J. L., Rüter, S. & Krause, A. Life cycle assessment of wood-plastic composites: Analysing alternative materials and identifying an environmental sound end-of-life option. *Resour Conserv Recy* **117**, 235-248 (2017). <https://doi.org/10.1016/j.resconrec.2016.10.012>
5. Culbertson, C., Treasure, T., Venditti, R., Jameel, H. & Gonzalez, R. Life Cycle Assessment of lignin extraction in a softwood kraft pulp mill. *Nord Pulp Pap Res J* **31**, 30-U247 (2016).
6. Skone, T., Schivley, G., Jamieson, M., Marriott, J., Cooney, G., Littlefield, J., Mutchek, M., Krynock, M., Shih, C., 2018. Life Cycle Analysis: Natural Gas Combined Cycle (NGCC) Power Plants (No. DOE/NETL--2018/1890, 1562914). <https://doi.org/10.2172/1562914>.