

**Pacific Northwest NATIONAL LABORATORY**

**WASHINGTON STATE UNIVERSITY**

# Envisioning a Circular Bioeconomy: Challenges and Opportunities

**Jonathan L. Male**

December 7, 2022

Advanced BioSystems Workshop –  
Toward a Circular Bioeconomy

**BIOIN** BIOPRODUCTS INSTITUTE

**BATTELLE** U.S. DEPARTMENT OF ENERGY

PNNL-SA-180287

PNNL is operated by Battelle for the U.S. Department of Energy

1

## Challenge: Turn carbon into a renewable feedstock for difficult-to-decarbonize fuels and products

2019 U.S. Green House Gas Emissions

Sector	Percentage
Transportation	33%
Electric Power	24%
Industry	21%
Buildings	13%
Agriculture	9%
Passenger Cars	21%
Light Trucks	30%
Medium and Heavy Vehicles (including Buses)	21%
Off Road	9%
Rail	2%
Water	3%
Aviation	11%
Other (Pipeline/Military/Lubricants)	3%

- Closing the Carbon Cycle is about recycling carbon and hydrogen with efficient and resilient catalysts
- Success would result in sustainable low-carbon, net-zero carbon fuels and products

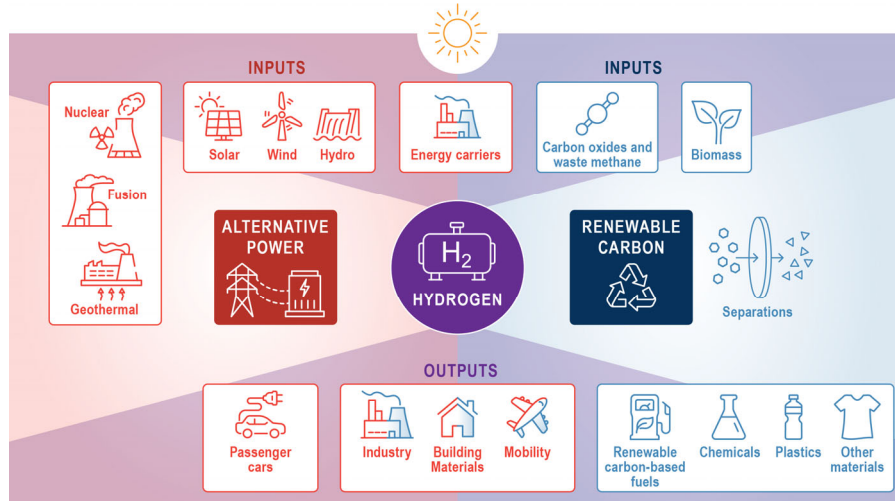
**BIOIN** BIOPRODUCTS INSTITUTE

Credit: Transportation Decarbonization Strategy by Deputy Assistance Secretary for Sustainable Transportation Michael Berube

December 7, 2022 2

2

## Bioproducts Institute (Bio-In) focused on net-zero carbon solutions in hard to electrify market segments

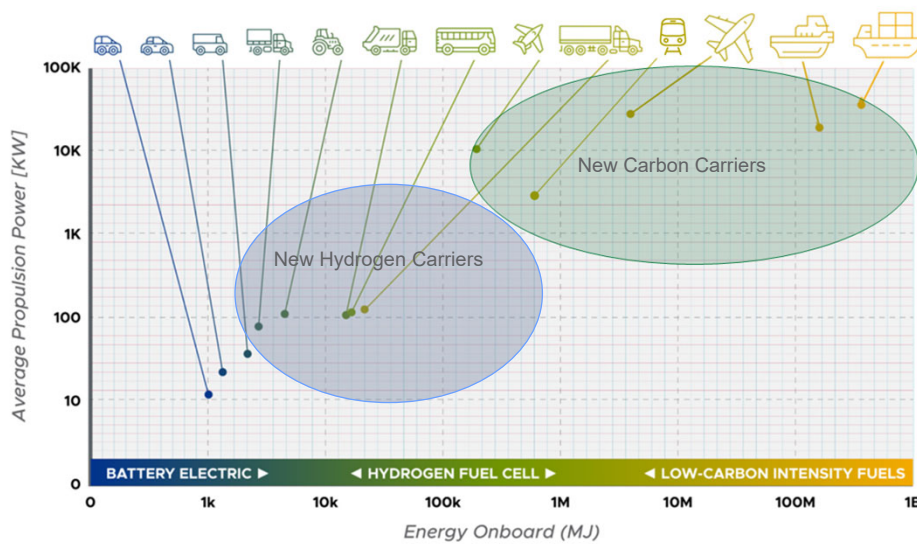


Based on Turning off the Tap for Fossil Carbon - Future Prospects for a Global Chemical and Derived Material Sector Based on Renewable Carbon. *Ferdinand Kähler, Michael Carus, Olaf Porc, and Christopher vom Berg, Nova Institute for Ecology and Innovation April 2021*  
[www.renewable-carbon.eu/publications](http://www.renewable-carbon.eu/publications)

3

3

## Aviation, marine, and rail will require low carbon intensity fuels for decades



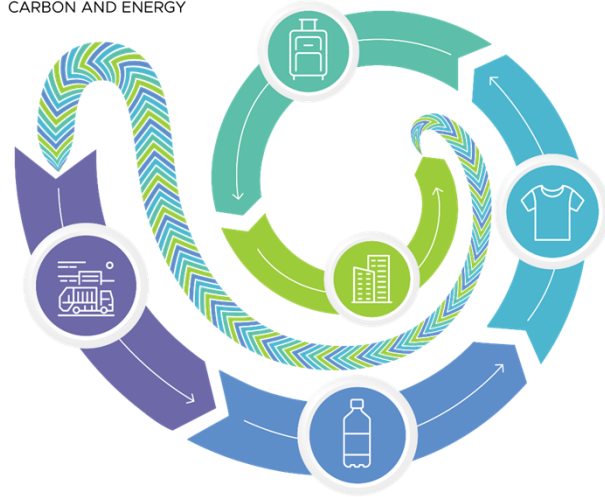
December 7, 2022 4

4

## Keeping carbon in play – Every carbon is reused, ideally multiple times

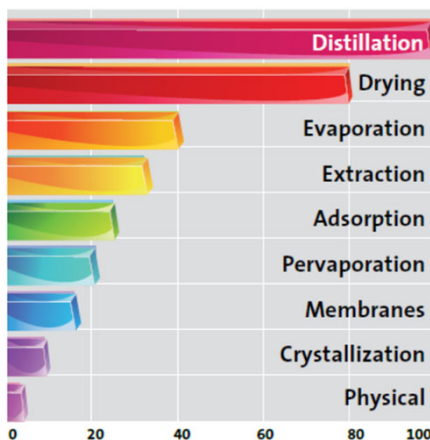
- A spiral economy aims for long life and recyclability of materials and chemicals.
- Once a material or chemical is considered waste, an evaluation determines whether to repurpose it as a similar product or to convert it into a different product.
- At some point, it may be most energetically favorable to break the material or chemical down to a small unit and start over again.
- Opportunity: Develop efficient conversion processes for complex waste streams enabling longer-lived products and reducing the carbon footprint

**INCREASING CARBON RE-USE**  
LEVERAGING EMBODIED CARBON AND ENERGY



5

## Energy Efficient Separations



Challenge: Thermal separations consume 10–15% of the nation's energy.

Opportunity: Increase energy efficiency through advanced and reactive separations to concomitantly reduce CO<sub>2</sub> emissions.

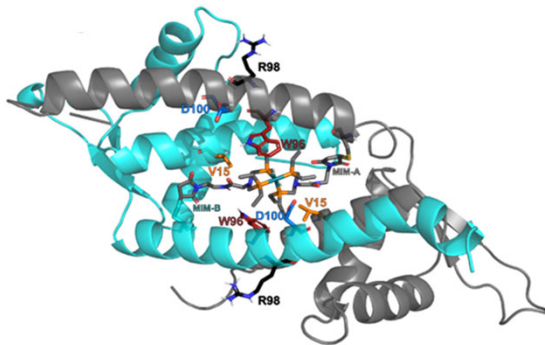
- Create active sites with needed geometry to enable reactive distillations
- Develop membranes for electrochemical separations
- Design cascade processes for the energy-efficient and selective adsorption/release of trace species
- Create separations processes for products from the conversion of complex source streams

Sustainable Separation Processes: A Road Map to Accelerate Industrial Application of Less Energy-Intensive Alternative Separations (ALTSEP)

6

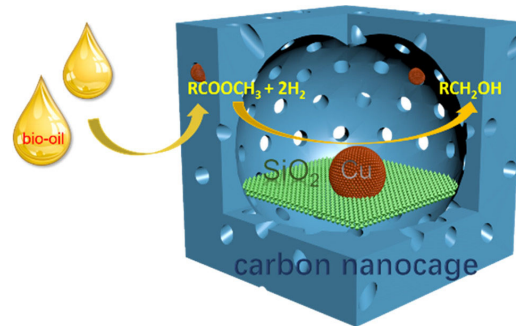
## Dramatically Improve Catalyst Efficiency and Resilience

Using nature as inspiration . . .



**BIOIN**  
BIOPRODUCTS INSTITUTE

. . . for creating synthetic catalysts



December 7, 2022 7

7

## Analysis that supports strategy and research

TEA and LCA

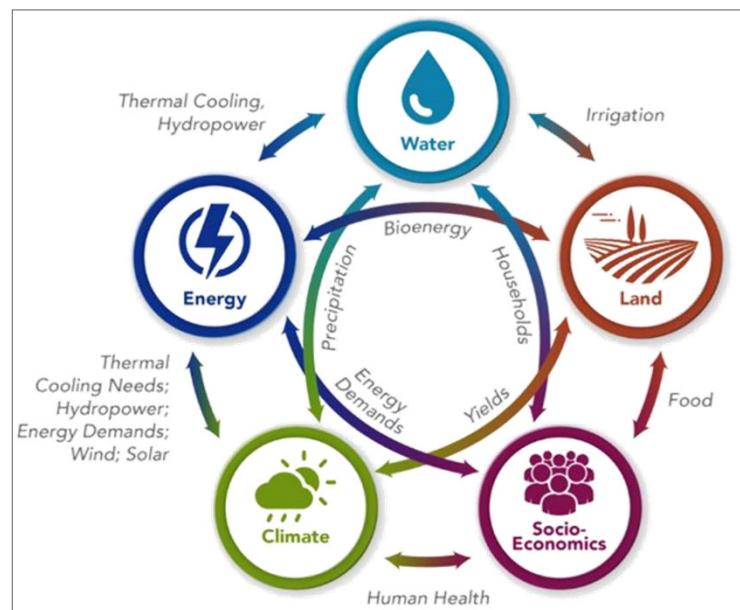
- Flowsheet simulations in ChemCAD or AspenPlus
- Discounted cash flow analysis, lifecycle inventory, and multivariate sensitivity analysis

Assessment tools

- High resolution physical-GIS models
- Full supply chain assessment

Global

- Global modeling of energy, land, water and human activity (GCAM)



**BIOIN**  
BIOPRODUCTS INSTITUTE

December 7, 2022 8

8

Advancing science that reduces the environmental impact of fuels and products while retaining carbon that must stay in the economy

Continued leadership in Sustainable Aviation Fuel

Wet wastes      Biocrude      Sustainable aviation fuel

Hydrothermal liquefaction      Upgrading

\$31 million in new projects

**Turning today's waste into tomorrow's carbon resource**

15 Distinguished Research Graduate Program Fellows since 2018, Tutored 11 Post-Docs and Students

Carbon fabric mat      Commercial epoxy resin      Polymer solution      Recycled fabric mat

Upcycling

Recyclable High-Performance CFRP

High  $T_g$  (>200 °C)  
Hydrothermal recyclability  
Robust properties

62 research publications

December 7, 2022 9

9

**Accelerate technological marketability by solving more than one problem at once**

Image from Gas Cleaning Technologies LLC

**Steel mill off-gas**

Image from Environmental Science & Engineering Magazine

**Sewage sludge**

Image by Picasa

**Manures**

Image from: cementis.com

**Municipal solid waste**

cost-advantaged feedstocks

Each of these represent:

- Stranded carbon resources
- GHG emissions
- Ecological liability
- Economic liability
- Social liability

10

## Bio-In is focused on management of carbon fluxes – Polymers challenge

**Current situation**

**Target situation**

**Technology challenges and opportunities:**

- Plastic wastes result in environmental problems
- Contaminants are a challenge for recycling
- Opportunities to store carbon in buildings and infrastructure

**Product lifetime distribution**

Years

- Packaging
- Other and textiles
- Transportation
- Building & construction
- Consumer & Institutional products
- Electrical & electronic
- Industrial machinery

**Product lifetime distribution**

Years

- Packaging
- Other and textiles
- Transportation
- Building & construction
- Consumer & Institutional products
- Electrical & electronic
- Industrial machinery

Production, use, and fate of all plastics ever made. Geyer, R.; Jambeck, J. R.; Law, K. L. Sci. Adv. 2017, 3, 7, e1700782 DOI: [10.1126/sciadv.1700782](https://doi.org/10.1126/sciadv.1700782)

December 7, 2022 11

11

## Bioproducts Institute (Bio-In) science themes

### Closing the carbon Loop

- > 60% carbon efficiency conversion of waste, biomass, CO<sub>x</sub> to diesel and jet
- Reusing each carbon atom more than once — in the future we will target > two uses

### Distributed processing

- Access more blended feedstocks
- Electrocatalysis and reimagining gasification
- Supply chain analyses and ecosystem services

### Decarbonization of hard to electrify market segments

- System level analyses
- Support Institute for Integrated Catalysis and Laboratory Objectives
- Seek most impactful use of renewable carbon

\*This analysis based on carbon basis

December 7, 2022 12

12

## Aviation offers a unique market for Sustainable Aviation Fuels (SAFs)

- Market size (U.S., 2019, transportation)<sup>1</sup>
  - Jet fuel 26.7 billion gallons (market projected ↑)<sup>2,3</sup>
  - Gasoline 137 billion gallons (market projected ↓)<sup>3</sup>
  - Diesel 47.2 billion gallons
- Consumer set is small
  - Airlines, military
  - Top ten airports: 50% of U.S. commercial aviation<sup>4</sup>
- Fuel and delivery
  - Few fuel variants
  - Delivered to plane via hydrant system at major airports



<sup>1</sup> Data from EIA, Monthly Energy Review (Table 3.7c); total diesel use 62.6 B gal, total gasoline 142 B gal)

<sup>2</sup> 26 billion gallons contains 3.6 quadrillion Btu of energy

<sup>3</sup> Global consumption 106 billion gallons (current) projected to double over next 30 years to 230 billion gallons in 2050, IEA Annual Energy Outlook

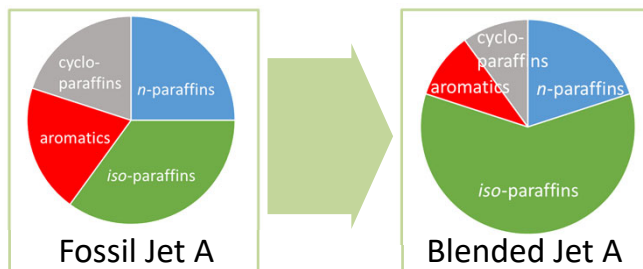
<sup>4</sup> In 2017 the ten largest U.S. airports used 10.5 billion gallons, half of commercial jet fuel use (21 billion), source ASCENT



December 7, 2022 13

13

## Understanding the critical fuel properties to lead to more Sustainable Aviation Fuels (SAFs)



- Reduce soot
- Increase energy content
- To maintain low temperature fluidity
- To achieve thermal stability
- To maintain seal swelling in older planes

Make it better by only producing hydrocarbons that contribute to key fuel properties

PNNL and WSU are developing an understanding of Sustainable Aviation Fuels at molecular levels to move from 50% blends to 100% blends by 2050



December 7, 2022 14

14

## SAF Grand Challenge: A focus of WSU and PNNL moving forward

**By 2030, 3 billion gallons of domestic production**

**Major national needs:**

- Feasibility analysis on the path towards achieving the 2030 goal
- Leverage existing ASTM-approved fuel pathways
- Synthesis gas as a platform for scaling SAF
  - Feedstock expansion
  - Develop more carbon efficient routes for synthesis gas production
- Support ASTM for augmentation of annexes for broader feedstock and process definitions

**By 2050, 35 billion gallons of production**

**Major national needs:**

- New pathways and continued support of broader feedstock and process definitions for existing pathways
- A 100% SAF specification(s) for drop-in and non-drop-in (potentially)



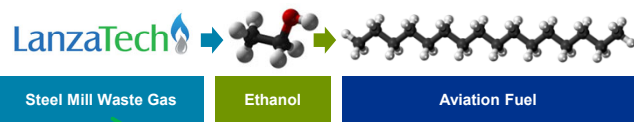
December 7, 2022 15

15

## One example of SAF at PNNL is wastes to alcohol to high quality fuels

**Developed a robust and efficient catalyst that**

- Converted ethanol to aviation fuel molecules
- Alcohol to jet (ATJ) ASTM D7566 Annex A5 approved April 2018 for up to 50% blend
- LanzaTech licensed for scale-up



Bioenergy Technologies Office co-funding



R&D (started in 2010, internally funded)

Build 10M gpy



December 7, 2022 16

16



## Reformers will be important for H<sub>2</sub> and SAF

### Steam-methane reforming (SMR)

- $\text{CH}_3\text{OH} \rightarrow \text{CO} + 2\text{H}_2$
- $\text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2$
- $\text{H}_2\text{O} + \text{CO} \rightarrow \text{CO}_2 + \text{H}_2$

### Dry methane reforming (DMR)

- $\text{CH}_4 + \text{CO}_2 \rightarrow 2\text{CO} + 2\text{H}_2$

### Partial oxidation (POX)

- $\text{CH}_4 + 1/2\text{O}_2 \rightarrow \text{CO} + 2\text{H}_2$

### Methane tri-reforming

- Combines SMR, DMR, and POX of methane

### Autothermal reforming (ATR)

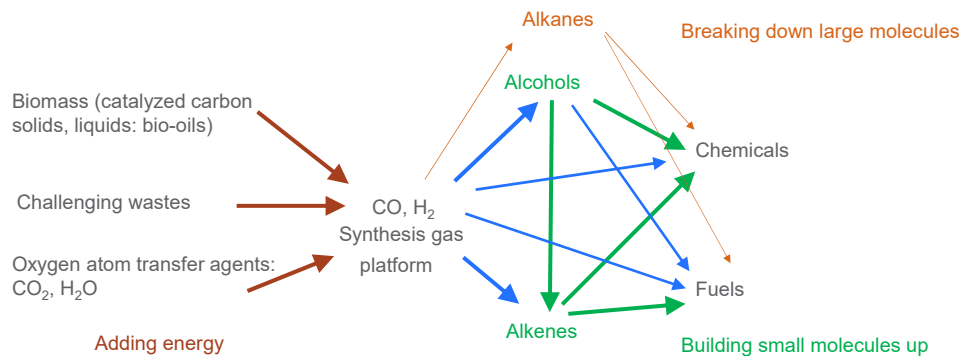
WSU and PNNL opportunity for optimization of H<sub>2</sub>:CO ratios, heat integration and utilization of the CO<sub>2</sub> generated in making a SAF

### *In general...*

Increasing efficiency, yields and conversion, H<sub>2</sub>/CO ratios, CO<sub>2</sub> utilization, and catalyst stability due to presence of H<sub>2</sub>O and O<sub>2</sub>

17

## PNNL and WSU will expand SAF through expanding impact of synthesis gas platform



- Develop SAF from more diverse biomass and waste streams
- Minimize and utilize any CO<sub>2</sub> formed along the way
- Drive carbon efficiency to desired products > 60%
- Leverage PNNL's leadership in conversion of alkenes and alcohols to fuels and chemicals

18

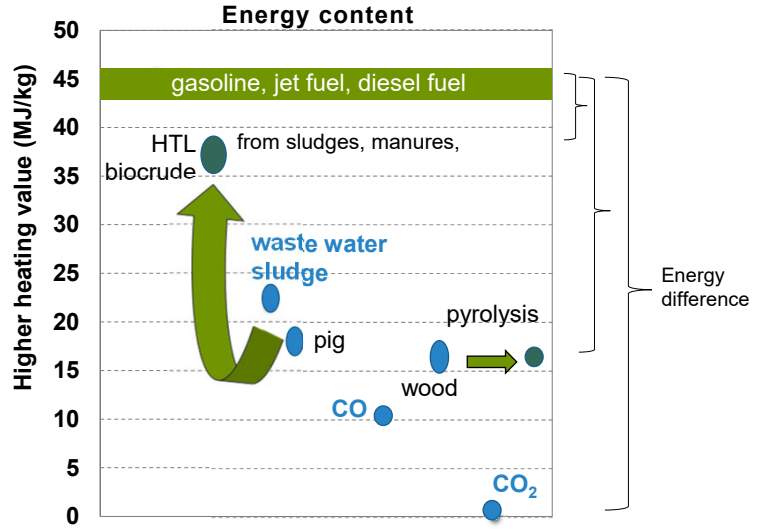
## There are other carbon sources, that like CO<sub>2</sub> are a problem, but which contain energy

Many waste carbon sources in the environment still contain energy

Intermediate oils can be produced using low energy

- 6 MJ/kg to convert wastewater sludge to biocrude oil hydrothermal processing<sup>1</sup>
- 0.5 MJ/kg to convert wood to pyrolysis oil<sup>2</sup>

$\Delta G^\circ$  for CO<sub>2</sub> to CO is 9.2 MJ/kg (0.35 MJ/mol)



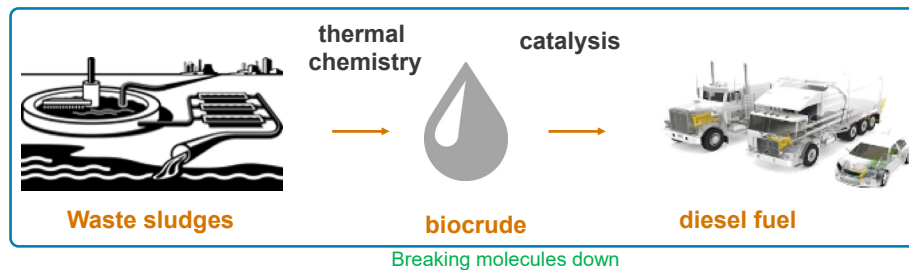
<sup>1</sup> 4.7 MJ/kg of electricity and 1.6 MJ/kg of natural gas  
<sup>2</sup> 0.47 – 0.64 MJ/kg dependent on biomass ash content

December 7, 2022 19

19

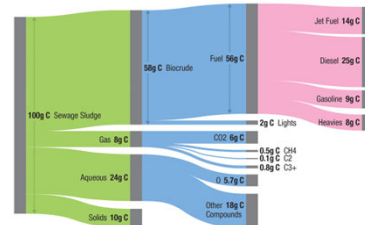
## Recycling carbon – Produce biofuels while solving another problem

Hydrothermal Liquefaction (HTL) of wet waste streams and hydrotreating to fuels



Carbon yield to fuels is typically 60%

An example of industrial symbiosis

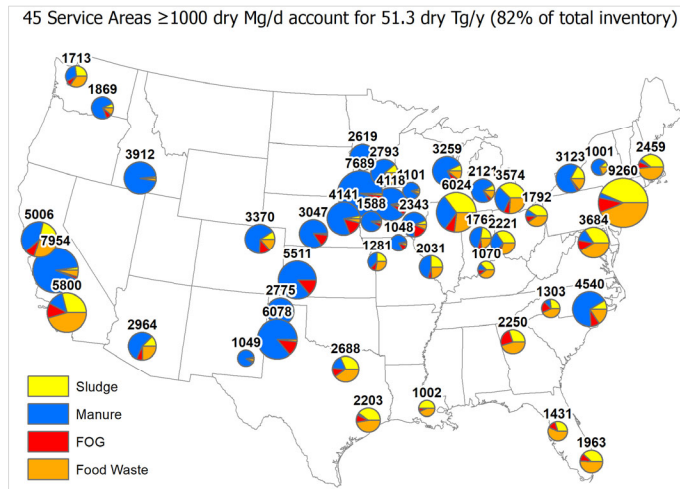


\*This analysis based on carbon basis 2022 20



20

## Wet wastes: Fats, oils, & greases are limited – What else is out there?



### Challenges:

Small scale, distributed, unstable materials

### Opportunities:

Wet wastes are highly collocated

- 80% of non-sludge waste occurs within 25 miles of water resource recovery facilities  $\geq 1$  Mg/d

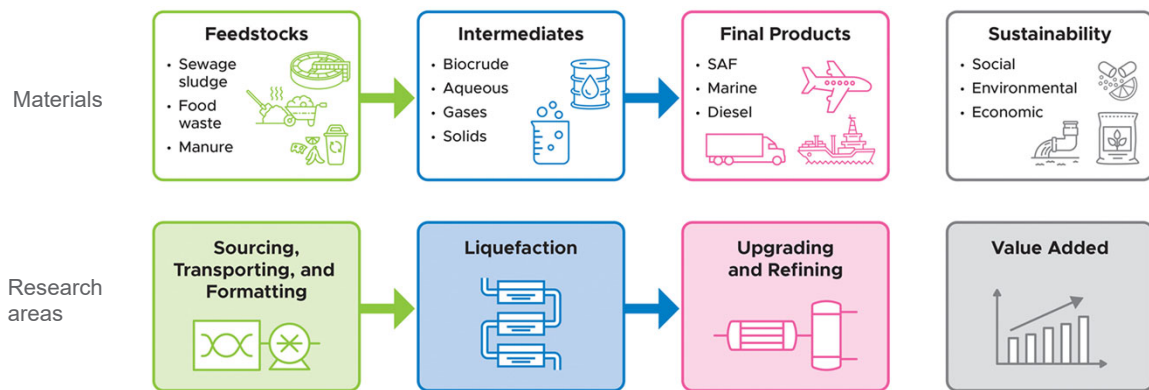
Amount of stranded, energy-rich carbon sources is not trivial



December 7, 2022 21

21

## Hydrothermal liquefaction (HTL) provides a means to convert wet carbon-rich waste into fuels



Research and Development needs:

- Optimize value from other HTL streams
- Co-process in petroleum refineries
- Greater control of jet- and diesel-fractions

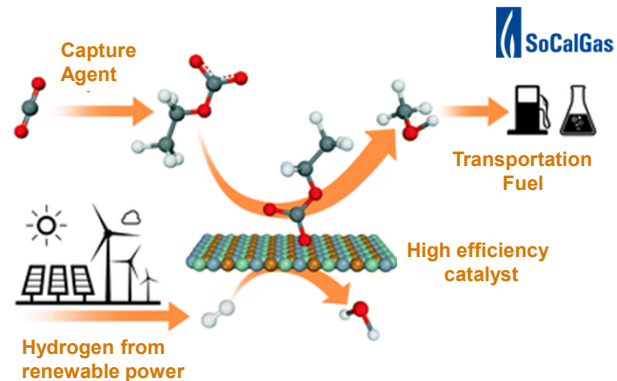


December 7, 2022 22

22

## Capture and convert carbon dioxide to methanol

- Invented the capture agent for more effective CO<sub>2</sub> separations
  - Converted the bound CO<sub>2</sub> directly to methanol
- Future:
  - Implement pilot-scale demonstration
  - Create new separation materials for other waste carbon
  - Expand catalytic reactions of bound CO<sub>2</sub> to chemical building blocks



U.S. Patent 10,961,173 Integrated capture and conversion of CO<sub>2</sub> to methanol process technology

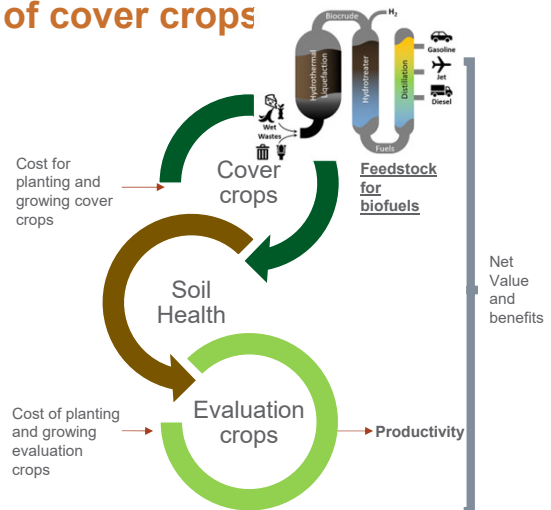


December 7, 2022 23

23

## An example of lowering the carbon intensity of feedstocks: Maximizing the value of cover crops

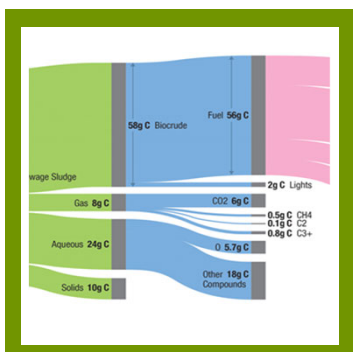
- Goal: Derive greater value from cover crops compared to off-season oil crop.
- Challenge: Develop an understanding of the trade-off impacts of cover crops growth for soil benefits or growth for fuel.
- Approach: Grow cover crops in at least two growing seasons to be able to characterize critical materials attribute (CMA) for fuels production
- Outcome: Leveraging of an underutilized feedstock source for fuel source, and improve agricultural and agronomic practices



December 7, 2022 24

24

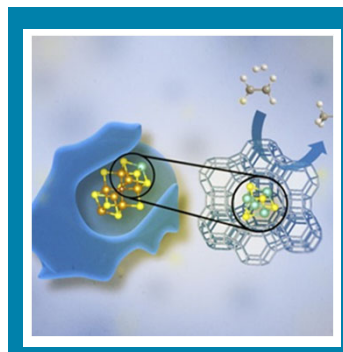
## PNNL and WSU focused on closing the carbon cycle



**Carbon Recycling & Separations**



**Hydrogen Production & Energy Carriers**

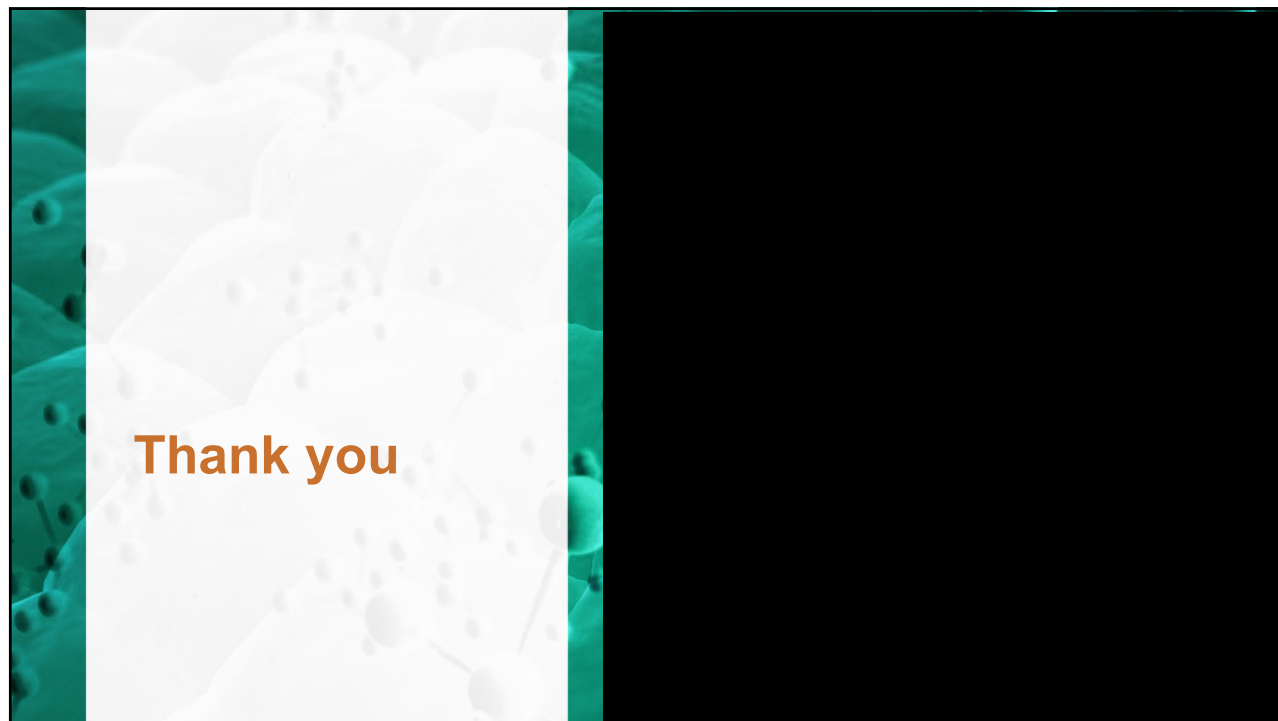


**Catalyst & Conversion Science**



December 7, 2022 25

25



26