DOE Bioenergy Technologies Office (BETO) 2021
Project Peer Review
DE-EE-0008250, WBS 3.5.1.501

Multi-stream Integrated Biorefinery Enabled by Waste Processing

03/22/2021
Systems Development and Integration

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Project Overview

- History: DE-FOA-0001689; Topic Area 2: High value products from waste/or other undervalued streams in an integrated biorefinery. The project has made significant progresses and is ready for BP2 verification.
- Project Goal

The project will leapfrog the technologies to enable multi- stream integrated biorefinery (MIBR) as measured by a set of complex technical targets including:

- Carbon fiber: 100GPa elastic modulus and 2GPa tensile strength, ready for commercialization.
- Asphalt binder modifier: Increasing rutting temp by 10^{o}C (about 1 PG).
- Bioconversion: 60% solubilized biorefinery waste, 25 g/L lipid titer, and 30% conversion rate
- **Integrating 2 out of the 3 aforementioned products to achieve MESP reduction by $0.5.**

- BETO Missions and Broader Energy/Environmental Challenges Addressed:

1. Improve biorefinery economics and sustainability
2. Produce high value bioproducts and manage biorefinery waste
3. Reduce carbon emission by complete biomass usage
4. Light-weighted material to reduce fuel and energy consumption in transportation sector
5. Asphalt binder modifier to improve infrastructure resilience to global climate changes
6. Biorefinery waste for lipid production will alleviate the feedstock limitations for biodiesel industry

Project Overview: Heilmeier Catechism Summary

- What we are trying to do?
  - Transform biorefinery economics and sustainability with the value-added products from waste stream
  - Produce quality carbon fiber for broad applications.
  - Produce asphalt binder modifier to enhance high temperature performance and add value.
  - Produce lipid for biodiesel industry from biorefinery waste.
- What is the state of the art? What is the limit?
  - Part of the lignin waste are burned to power the operation, and the remaining were disposed as waste.
  - Limited value recovery and low overall carbon efficiency – The success of modern biorefinery depends on the value-added products such as DDGS.
  - Lignin-based carbon fiber has low quality, and lignin to improve high temp PG was barely explored.
- Why is the project important?
  - Bring down the biofuel price to competitive range to enable lignocellulosic biorefinery ($259 billion economic potential and 1.1 million jobs).
  - Produce low cost carbon fiber ($4.7 billion market with 11% annual growth) and asphalt binder modifiers with unique features (part of the $10.5 billion market).
- What are the risks?
  - Lack of structure-function relationship understanding makes it difficult to produce quality products
  - Biorefinery integration and scale up. High cost for research on each stream.

Management, Team, and Industrial Engagement

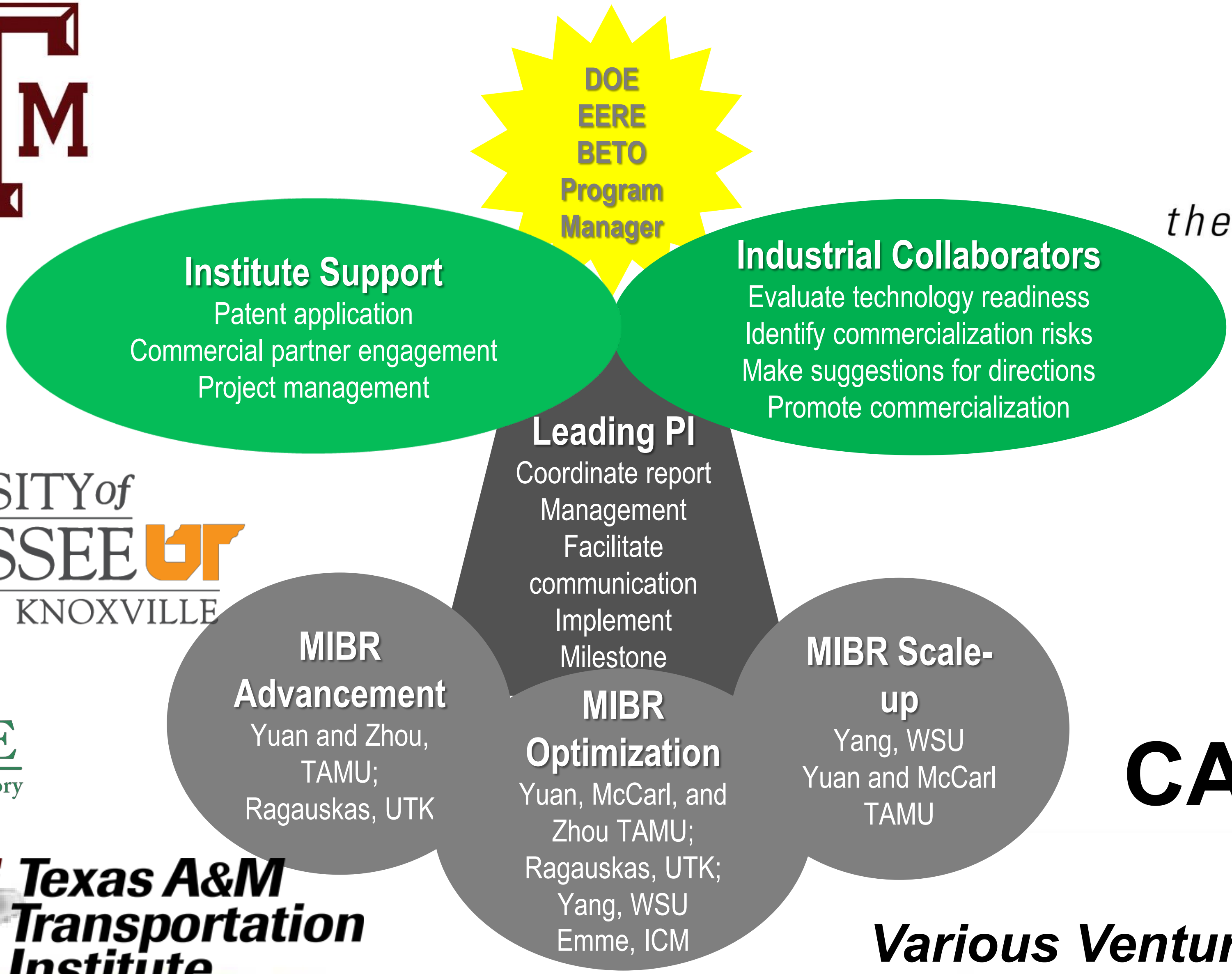
A&M

WSU

THE UNIVERSITY of TENNESSEE UT KNOXVILLE

OAK RIDGE National Laboratory

Texas A&M Transportation Institute



ICM® the energy of innovation™

ADM®

POET™ Energy inspired.™

CARBONFIT™

Various Venture Capital Companies

Management

- Defined and measurable milestones were laid out for technology development and commercialization.
- Go/No-Go milestones were set at the end of each year and each of the three budget periods.
- Monthly group teleconferences and teleconference with program management were implemented to evaluate the progresses against milestones. Risk mitigation strategies were designed and technoeconomic analysis guided the development.
- Monthly teleconferences between the PI and the program management are implemented to evaluate progresses, mitigate risks, and address challenges.
- Engage industrial partners and advisors including ICM inc. and others for deliverables relevant to EERE MYPP.
- Integrate TEA throughout the project to ensure the relevance of the project outcome. TEA and LCA will guide the technology development and identify economic drivers.

Management Approach – Go/No-Go Milestones

Time Point		Benchmark		End of BP2		End of the Project	
Product	**Metrics**	Milestones	Actual	Milestones	Actual	Milestones	Actual
Carbon Fiber	**MOE**	20GPa		50GPa		100GPa	
	Tensile	100MPa		1GPa		2GPa	
Asphalt Binder Modifier	**Rutting Temp Incr.**	7^oC		10^oC		10^oC	
	Low temp	Same		Same		Same	
Lipid for Biodiesel	**Titer**	10g/L		15g/L		25g/L	
	Conversion	30%		30%		40%	
Economic Outcome	**MESP[1]**	N.A.		N.A.		N.A.	
	~$/GGE[2]	N.A.		N.A.		~$3/GGE	

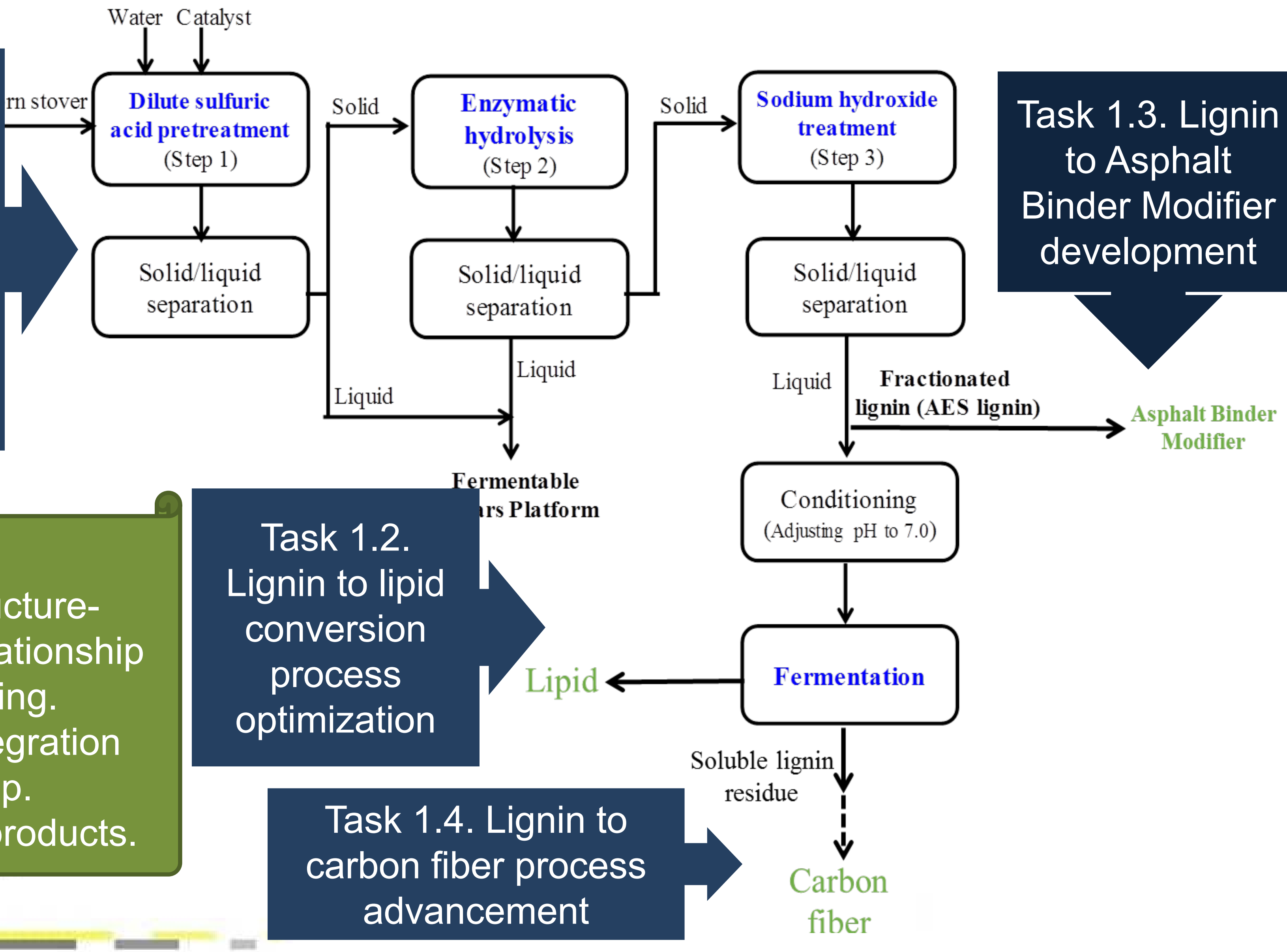
Technical Advancements Derive Economic Output and TEA guide Tech Development

1. Minimal Ethanol Selling Price
2. Gasoline Gallon Equivalent

- **Defined S.M.A.R.T. Go/No-Go milestones were set and implemented to ensure project progresses.**
- **The technical milestones were designed in a way to ensure that the economic targets can be achieved. Full ASPEN model was built.**
- **Down-selection to two product streams based on TEA and performance.**

Technical Approach – Process View

Task 1.1. Advance Pretreatment and fractionation technologies to enable three streams



Task 1.3. Lignin to Asphalt Binder Modifier development

Task 1.2. Lignin to lipid conversion process optimization

Task 1.4. Lignin to carbon fiber process advancement

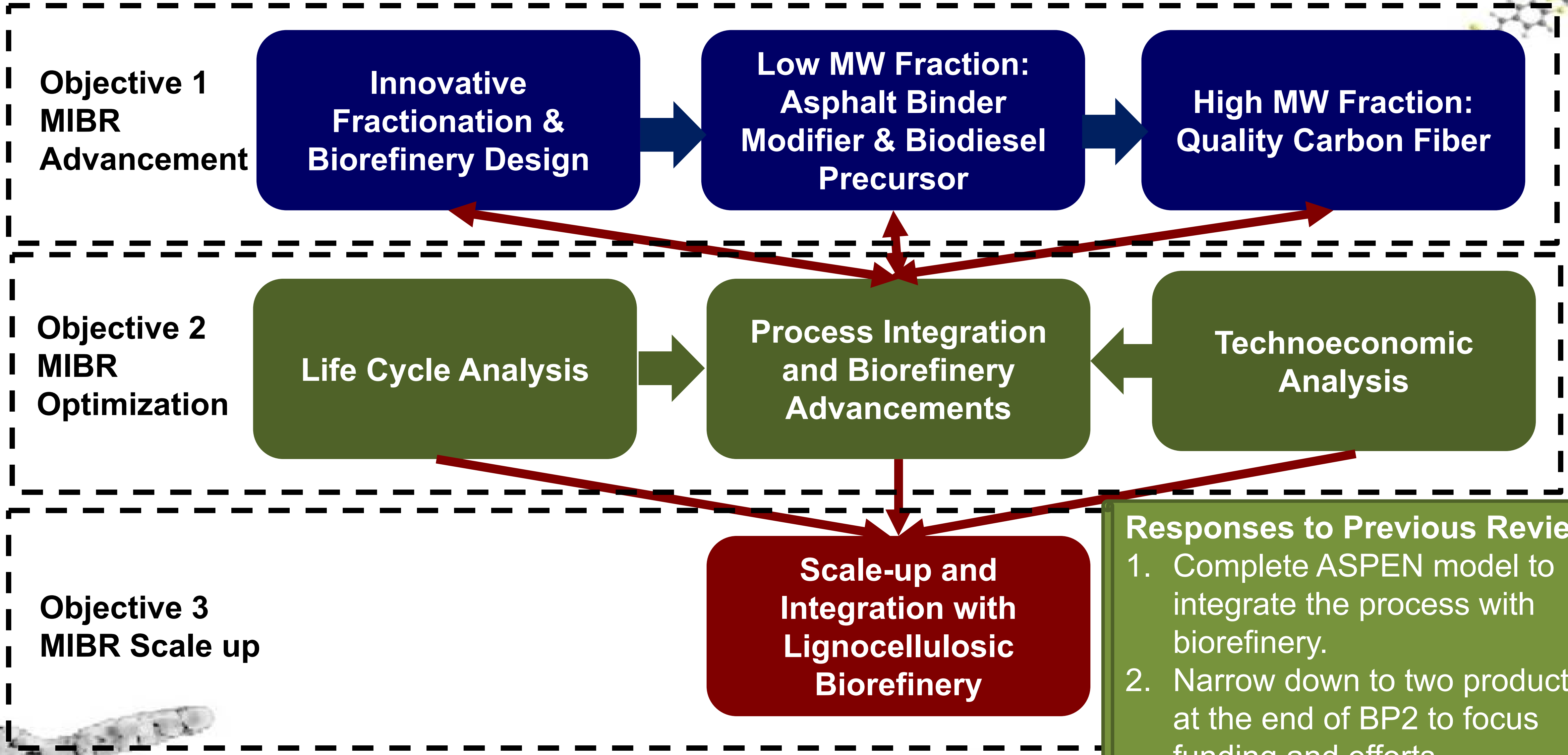
Challenges:

1. Lack of structure-function relationship understanding.
2. System integration and scale up.
3. Too many products.

Process Integration and Biorefinery Advancements, TEA and LCA

Scale-up and Integration with Lignocellulosic Biorefinery

Technical Approach – Integration of Tasks



Responses to Previous Review:

1. Complete ASPEN model to integrate the process with biorefinery.
2. Narrow down to two products at the end of BP2 to focus funding and efforts.

Broad Scientific Impact and Transformative Industrial Impact

- Transformative Industrial Impact
  1. Develop two out of three product streams to bring down the MESP to below $3/GGE range.
  2. Constantly engage biorefinery companies like ICM, ADM, and POET.
  3. Constantly engage carbon fiber industry, national biodiesel association, and investors.
  4. TEA has shown significant potential of the platform to transform biorefinery economics.
- Broad Energy and Environmental Impacts– Well Addressing BETO Missions
  1. Improve biorefinery sustainability and cost-effectiveness with value-added products from waste.
  2. Provide low-cost carbon fiber to improve energy efficiency for US energy sector, with applications on wind turbine, automobile and others.
  3. Alleviate the feedstock shortage at biodiesel industry.
  4. Enhance asphalt high temperature performance to improve infrastructure resilience to climate changes.
- Broad Scientific Impacts
  1. 28 publications with total impact factors at 190.
  2. Two PCT patent applications.
  3. Numerous scientific presentations and special events to engage companies.

Impact: Lignin Utilization to Enable Economic and Sustainable Multi-stream Biorefinery

BETO Missions:

- Improve biorefinery economics and sustainability
- Produce high value bioproducts and manage biorefinery waste
- Reduce carbon emission by complete biomass usage

Cellulosic Biomass

Lignin Waste

Lignin Processing

Additional Bioproducts and Biofuel Streams

Carbohydrate Conversion

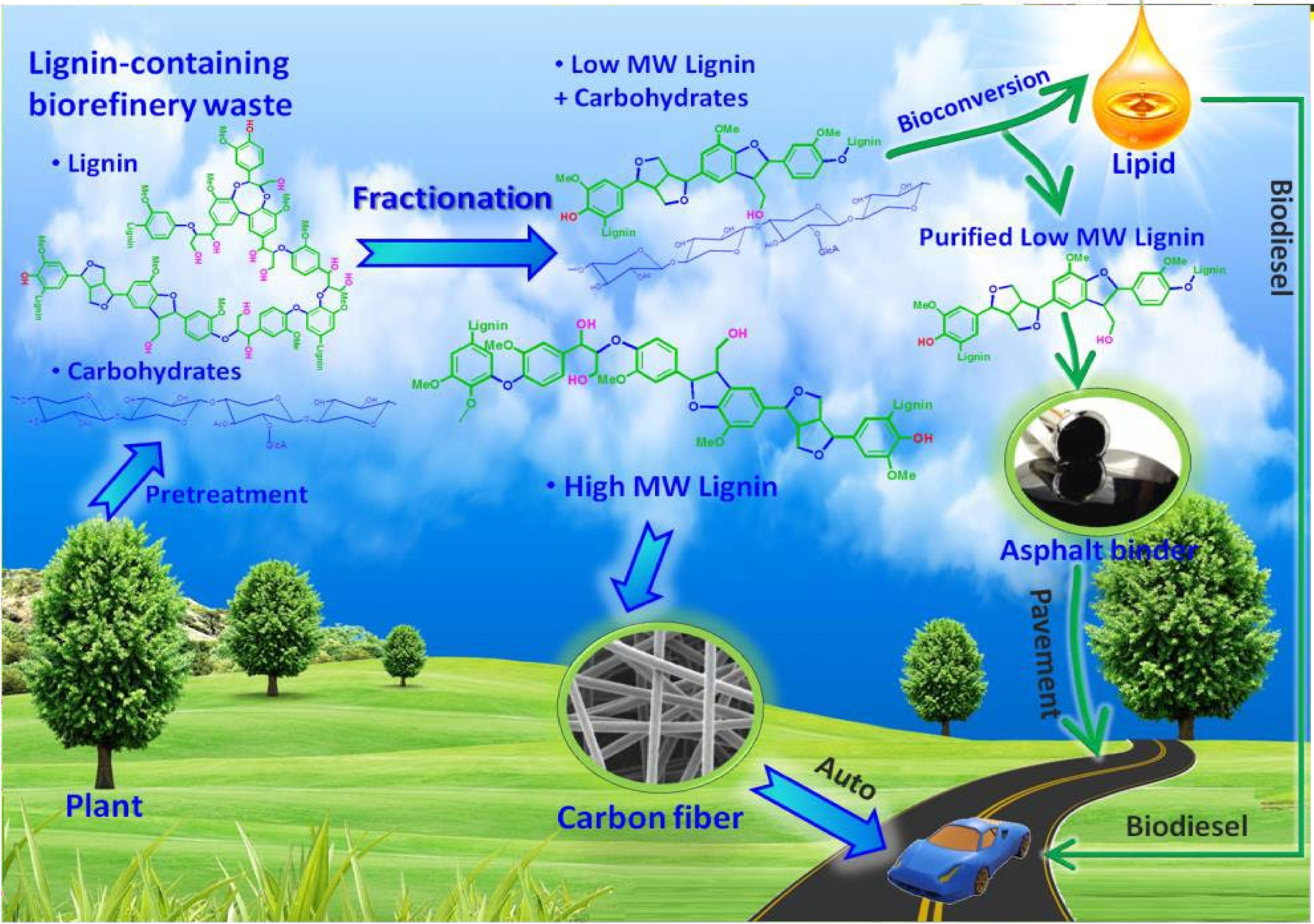
Pretreatment

Hydrolysis

Fermentation

Fuel finishing and blending

Xie, et al. Industrial Biotechnology 12 (3), 161-167



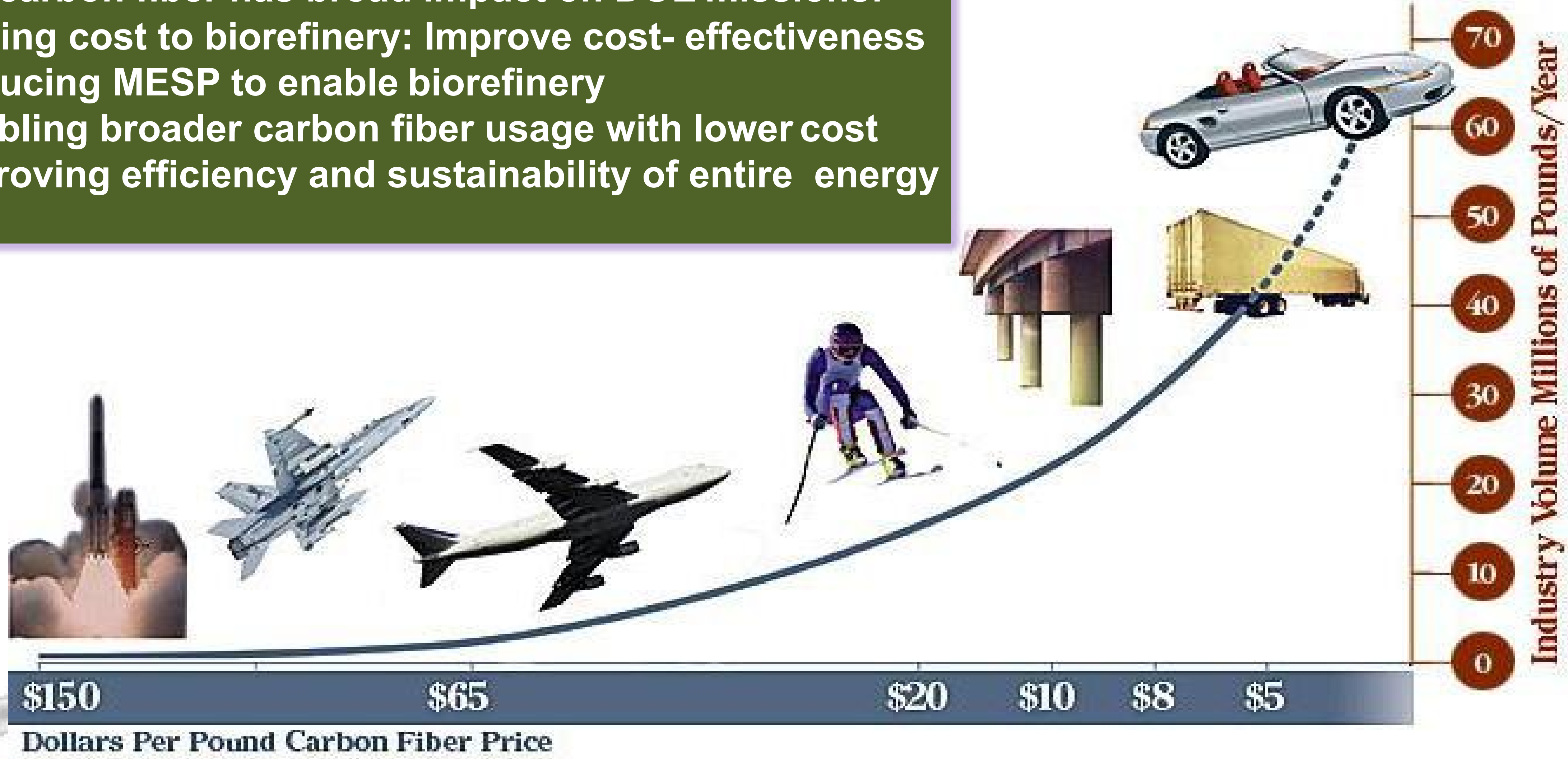
MIBR for Cost-effective and Sustainable Biorefinery and Bioeconomy

Q. Li et al. *Green Chem.*, 19, 1628–1634

Impact: Lignin as a Promising Substitute for Carbon Fiber Precursor

Lignin carbon fiber has broad impact on DOE missions:

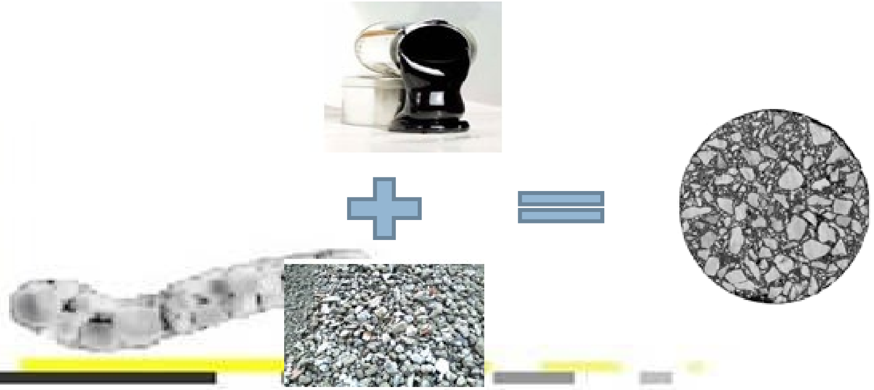
1) Adding cost to biorefinery: Improve cost- effectiveness
2) Reducing MESP to enable biorefinery
3) Enabling broader carbon fiber usage with lower cost
4) Improving efficiency and sustainability of entire energy sector

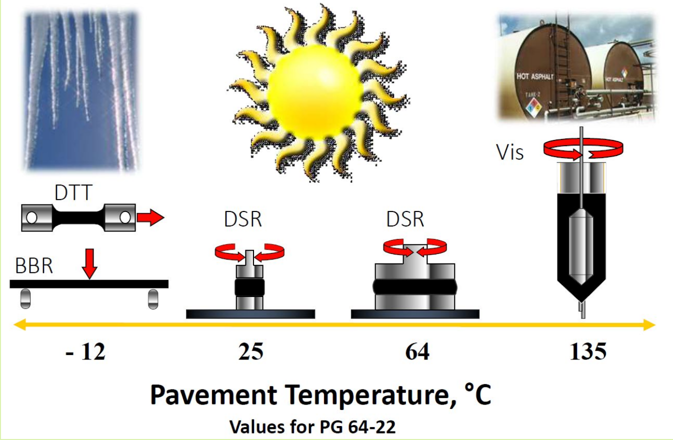


Impact: Asphalt Binder Modifier to Enhance Infrastructure Resilence to Climate Change

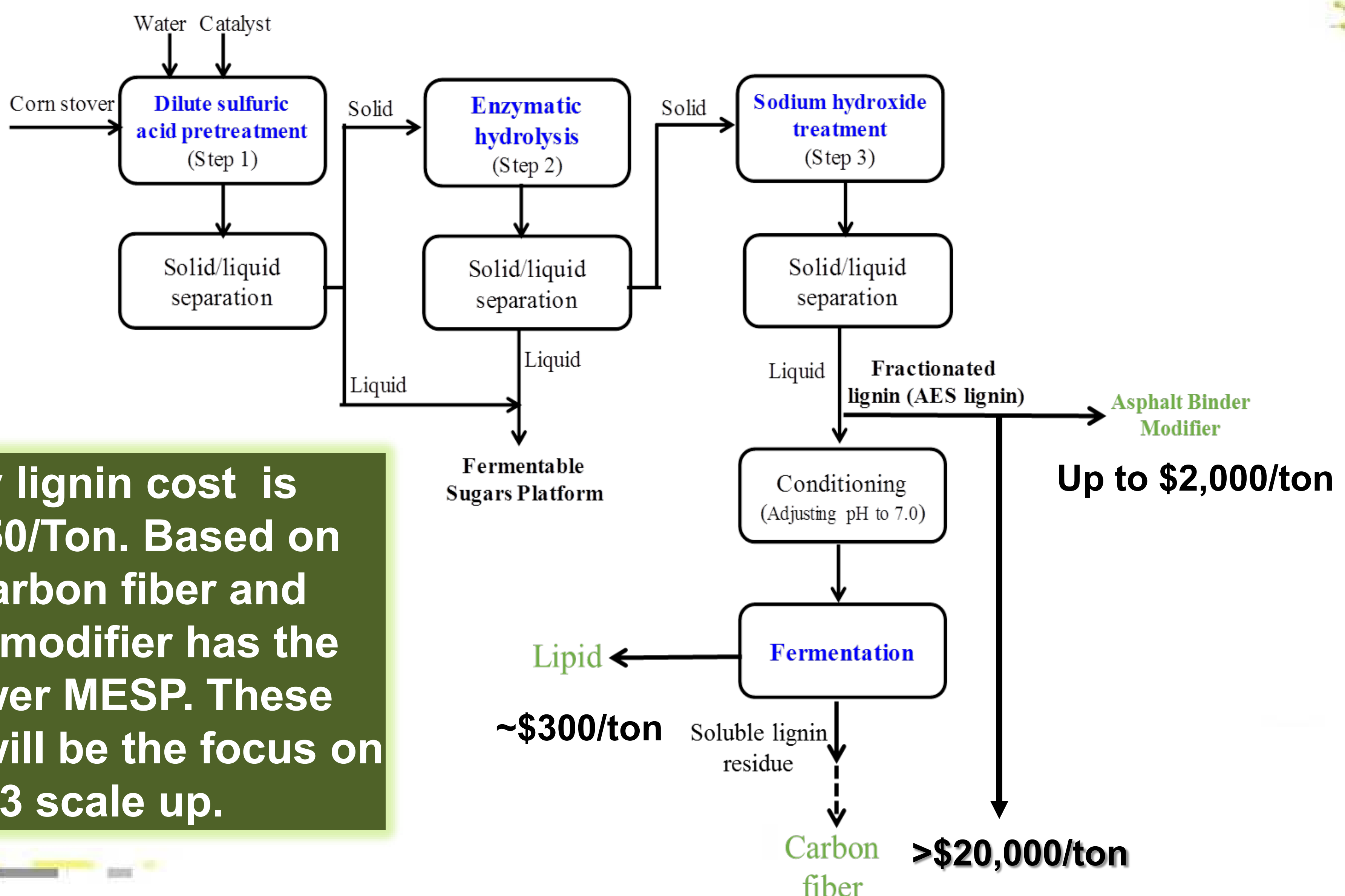
- Asphalt pavement facts:
  - 2.5 million miles of asphalt paved road in US.
  - 3,500 asphalt mix plants in US, producing about 350 million tons of asphalt mixes per year: $21 billion.
  - 17.5 million tons of asphalt binders
    - Binder: $600/ton
    - Market: $10.5 billion

- Asphalt binder functions as a <u>glue</u> in asphalt mixtures. Its quality have big influence on pavement perform.





Impact: Multiple Value-adding Products

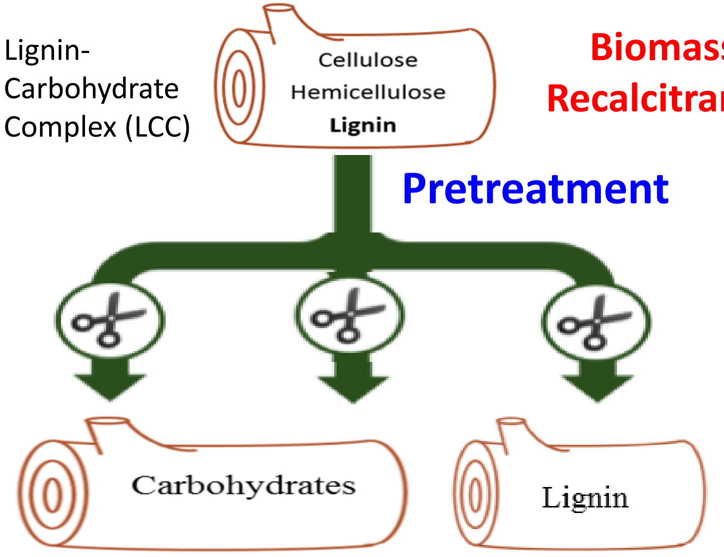


The biorefinery lignin cost is estimated at $50/Ton. Based on current TEA, carbon fiber and asphalt binder modifier has the potential to lower MESP. These two products will be the focus on Budget Period 3 scale up.

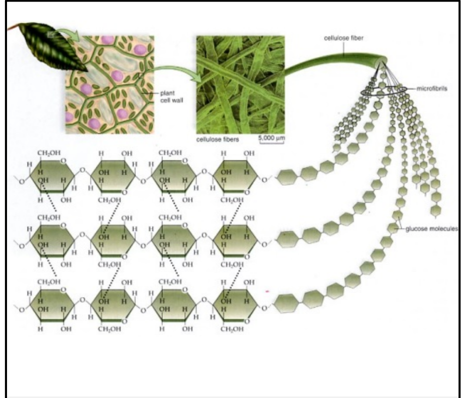
Fundamental Challenges for Lignin Bioproducts

Processing: Carbohydrate vs. Lignin

Lignin-Carbohydrate Complex (LCC)



Biomass Recalcitrance



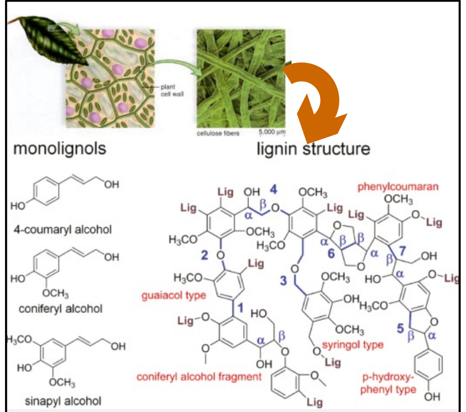
- ❑ Uniform
- ❑ Linear polymer
- ❑ Macromolecules

Enzymatic hydrolysis

Fermentable sugar platform-monosaccharide:
Glucose, xylose, arabinose, mannose, galactose……

???

Monomer Conversion?
Polymer Utilization?
Other Options?



- ❑ Heterogeneity
- ❑ 3D complexity
- ❑ Macromolecules

Hydrophobicity
Interunitery linkages
Functional groups
Molecular weight
Diverse Monomers
Aromatic Compounds

Not All Lignin Are Created Equal!

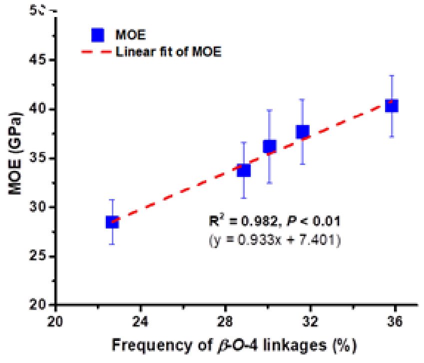
Renders et al, Energ. Environ. Sci. 2017;10(7):1551-7; Renders et al., ACS Sustain. Chem. Eng. 2016, 4(12):6894-904.

Structure-Function Relationship-guided Process Design

DESIGN

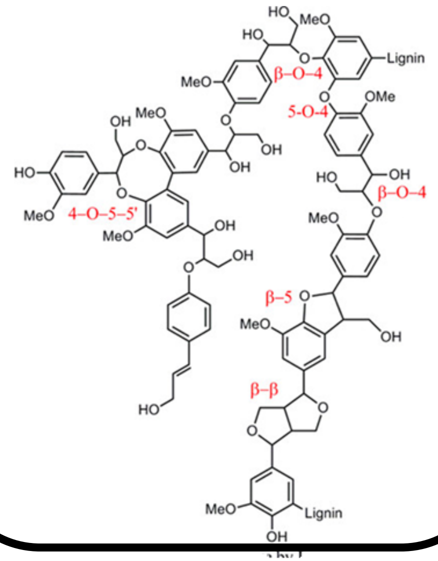
Process Design
Microbial & Chemical Engineering

Feedstock Design



SCIENCE

Lignin Chemistry Structure-Property Relationship



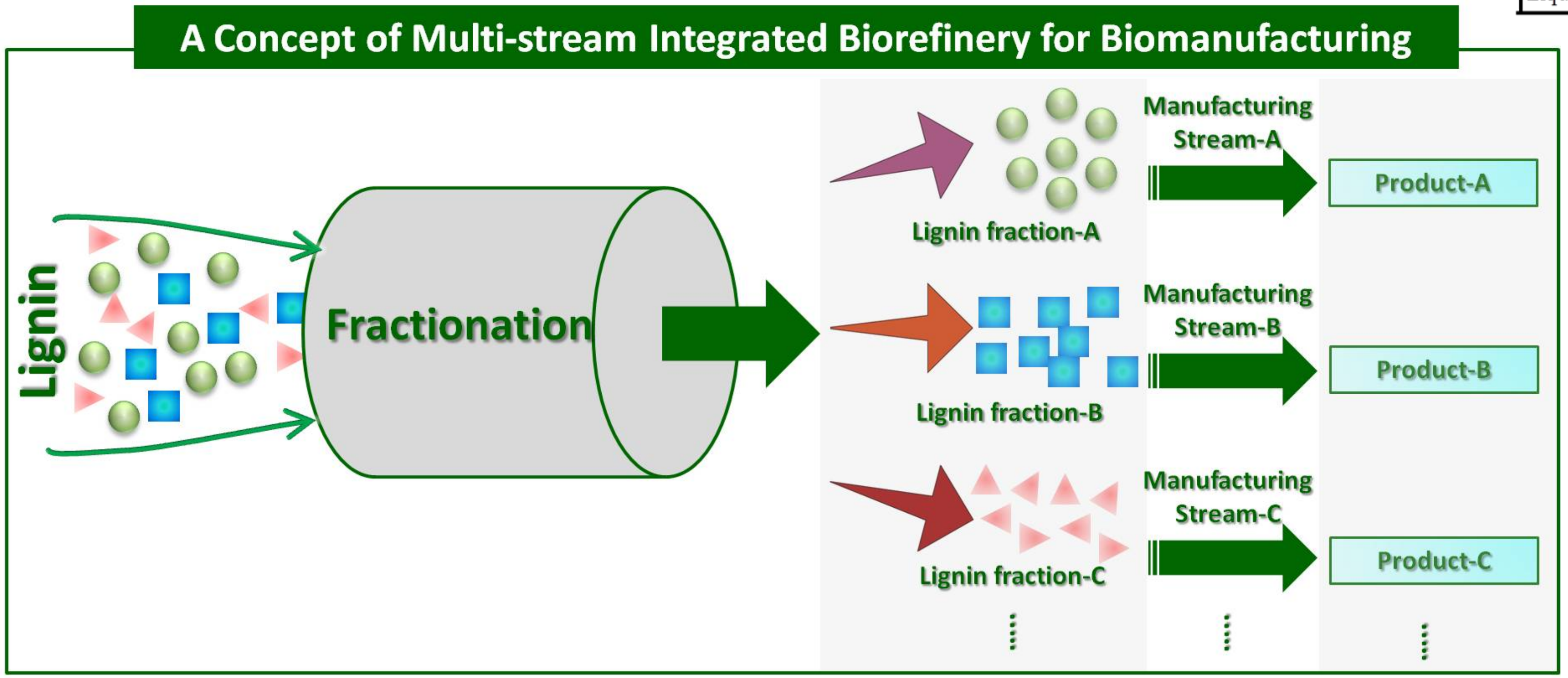
PRODUCT

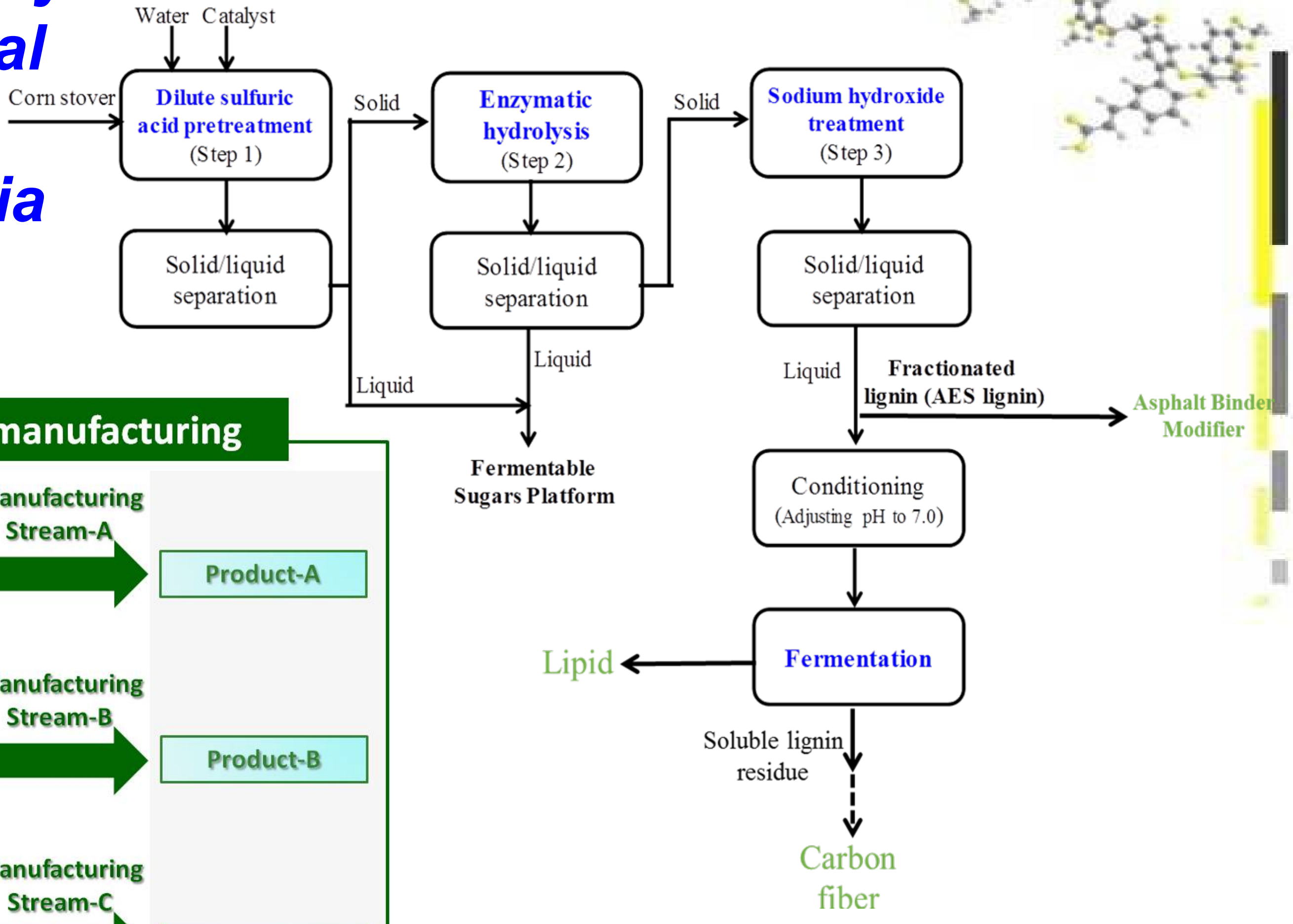
- Carbon Fiber
- Conversion to Lipid
- Conversion to PHA
- Nanoparticle
- Pavement Material
- Polymer Material

Represented studies:
Q. Li et al. *Green Chem.*, 19, 1628–1634
Z. Liu et al. *Green Chem.*, 21, 245-260
L. Lin et al. *Green Chem.*, 18, 5536-5547.
Q. Li et al., *iScience*, 23, 101405.
Q. Li et al., *J. Mat. Chem. A., 5, 12740-12746.*
S. Xie et al., *Adv. Sci.*, 6, 1801980.

***Concept:* Lignin with different chemistry should be suitable for different material productions**

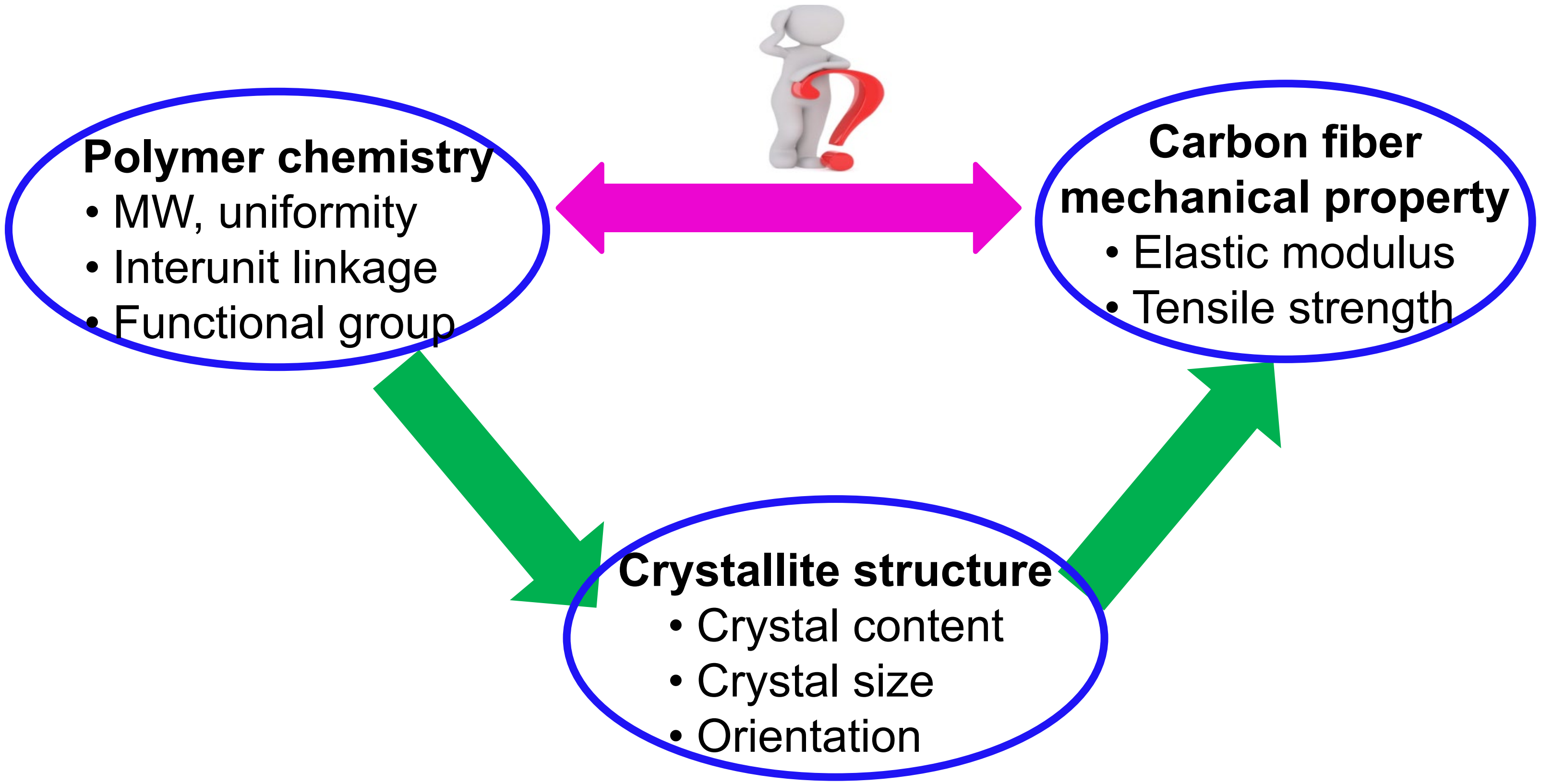
***Strategy:* Tuning polymer chemistry via lignin processing (fractionation)**





Q. Li et al. Lignin Carbon Fiber: the Path for Quality. Tappi J. 16, 107-108.

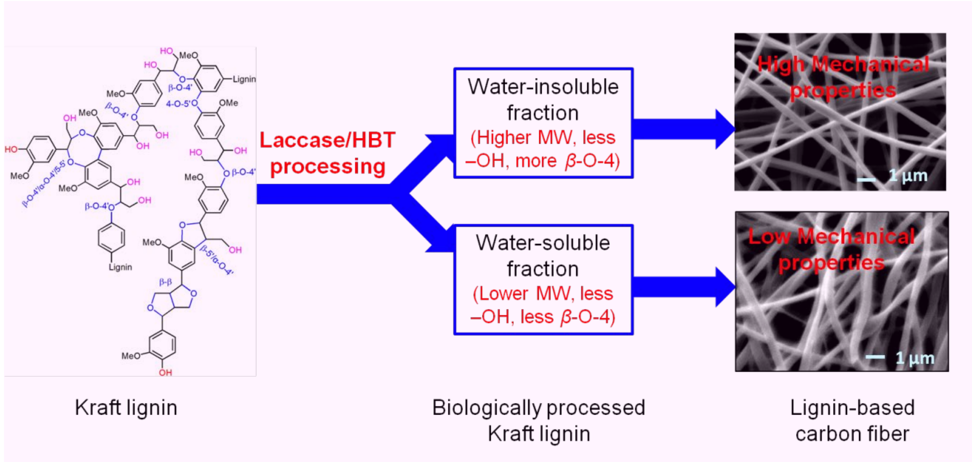
Hypothesis: Fundamental Structure-Function Relationships in Lignin Carbon Fiber

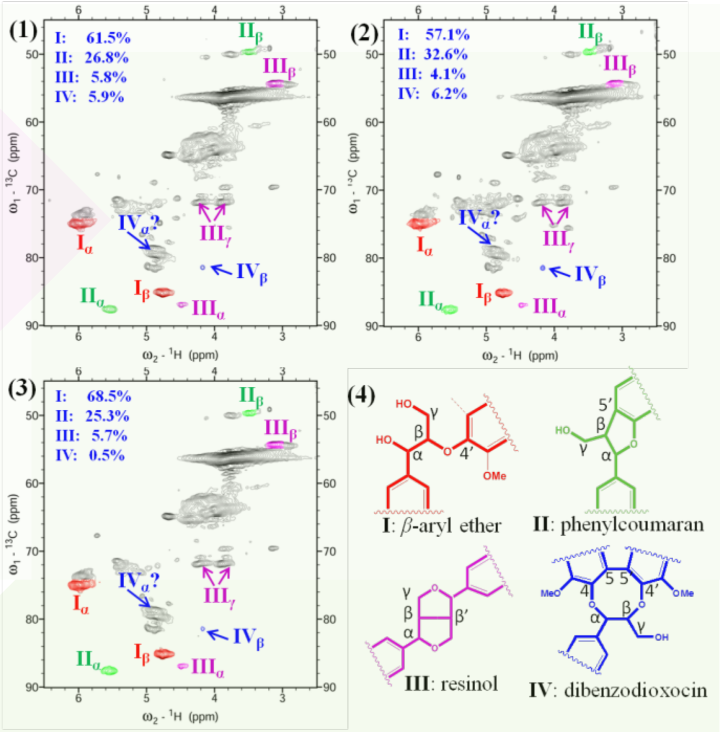


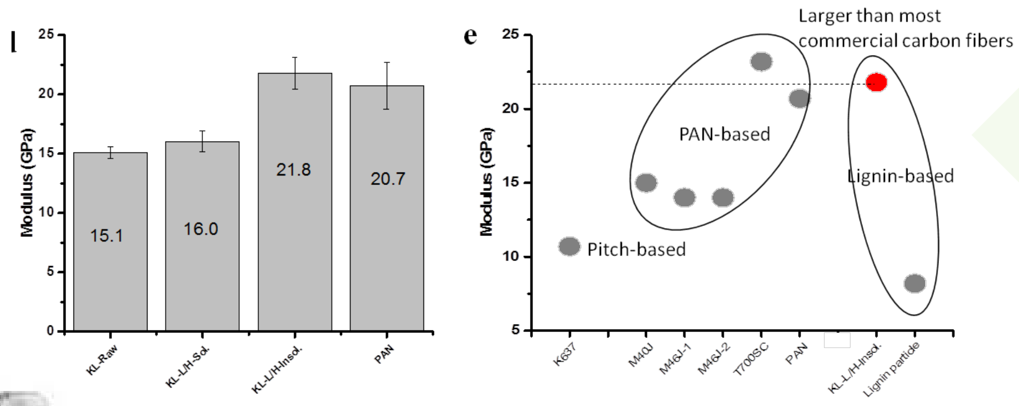
Processing/Feedstock—Polymer chemistry—Microstructures—CF properties relationship??

Processing Development Progresses: Fractionation

(1) Enzymatic: MW, linkage, -OH



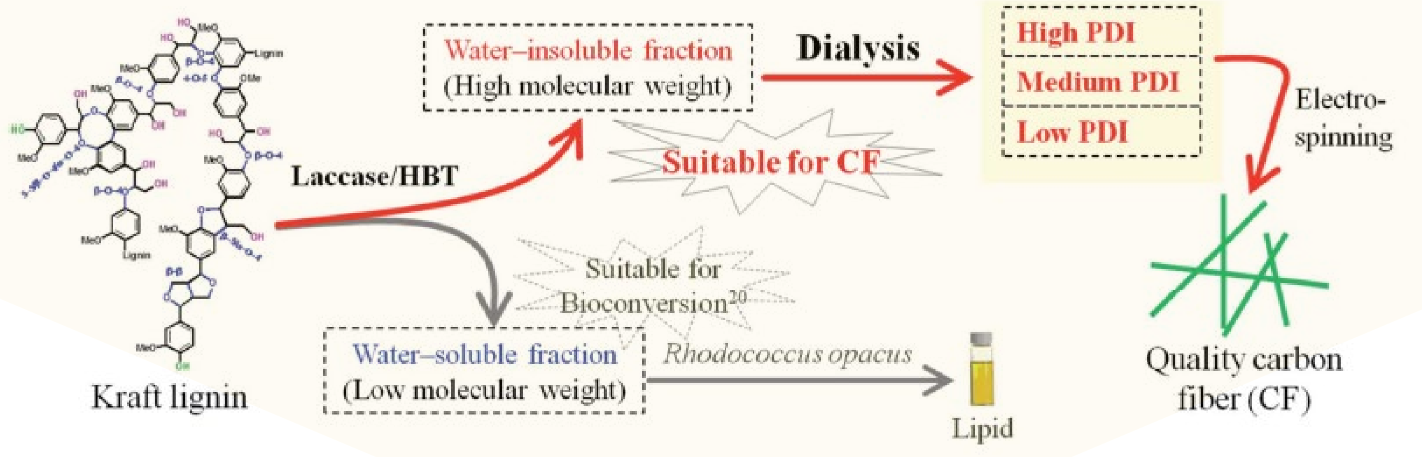


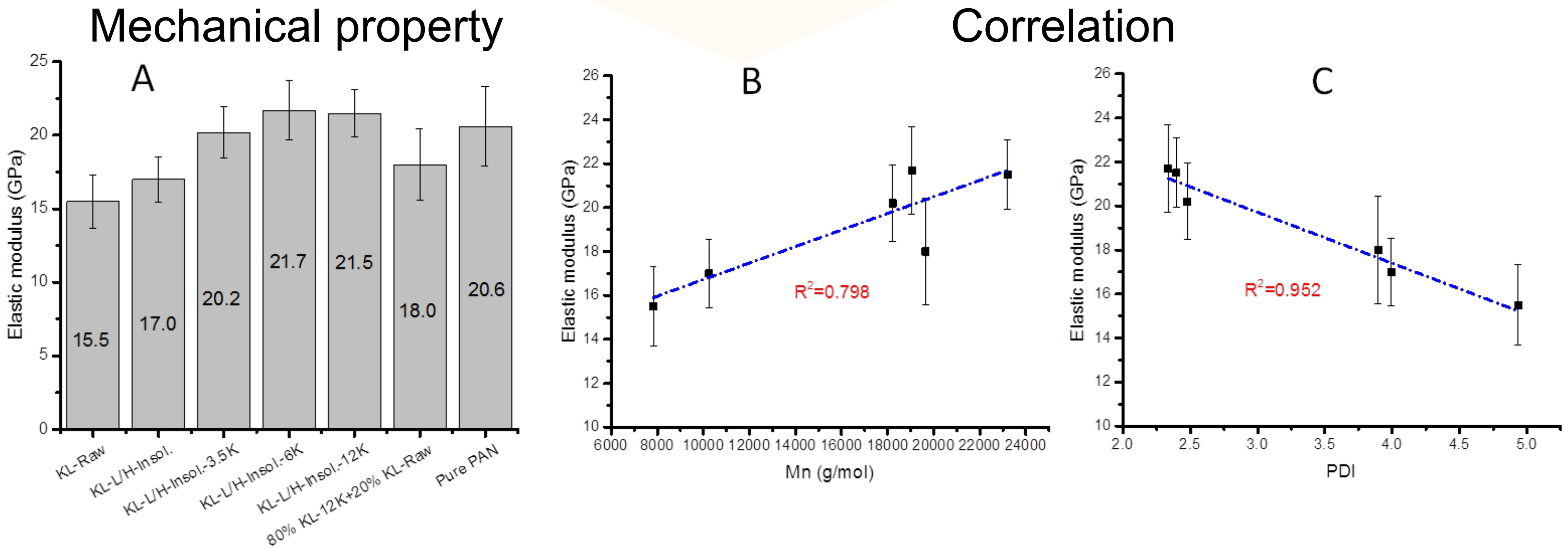


Mechanical properties

Q. Li et al. *Green Chem.*, 19, 1628–1634

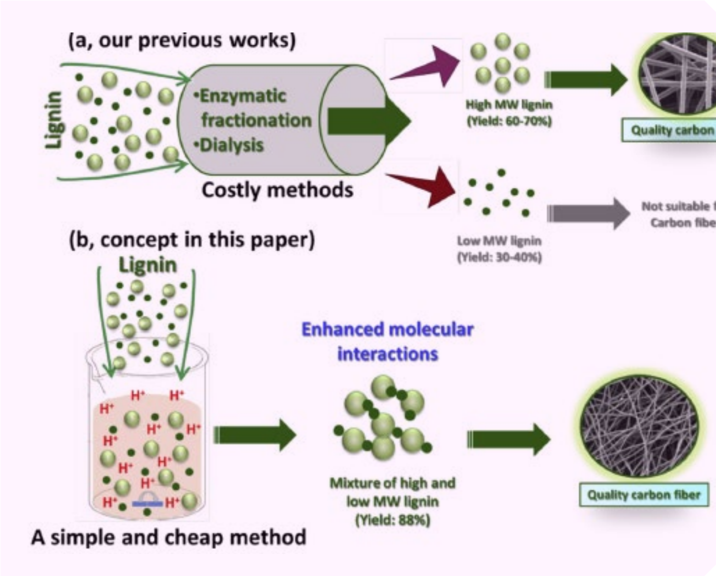
Polymer Uniformity as a Key Factor for Carbon Fiber Quality

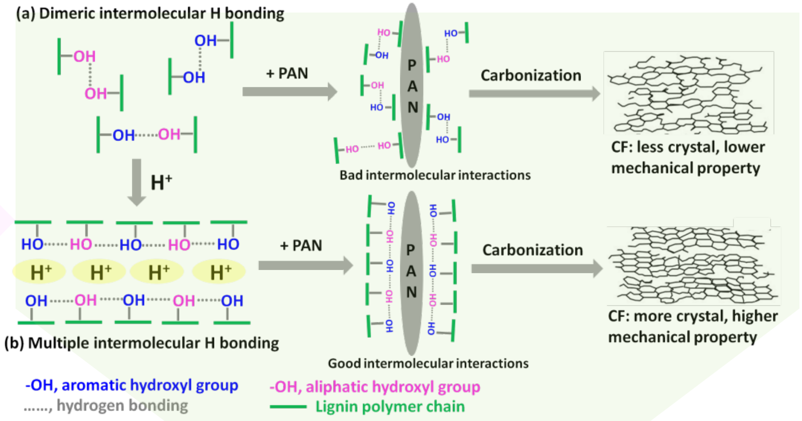




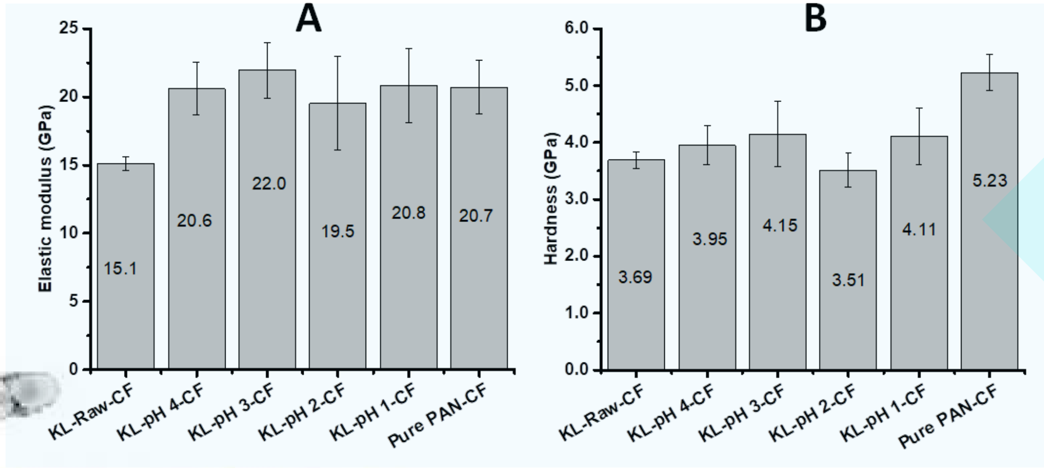
Q. Li et al. *J. Mater. Chem. A*, 5 (25), 12740-12746

The Impact of –OH, H-bonding on Carbon Fiber Properties

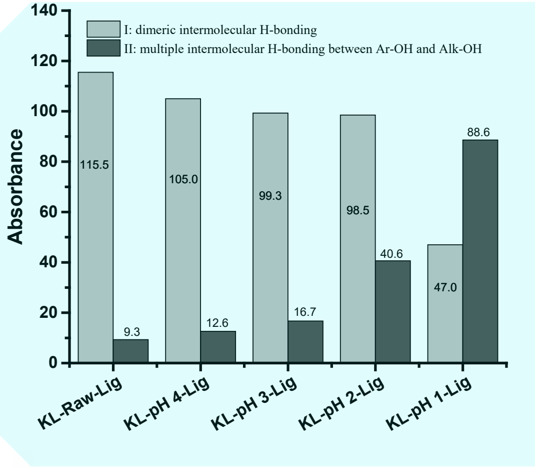




Mechanical properties

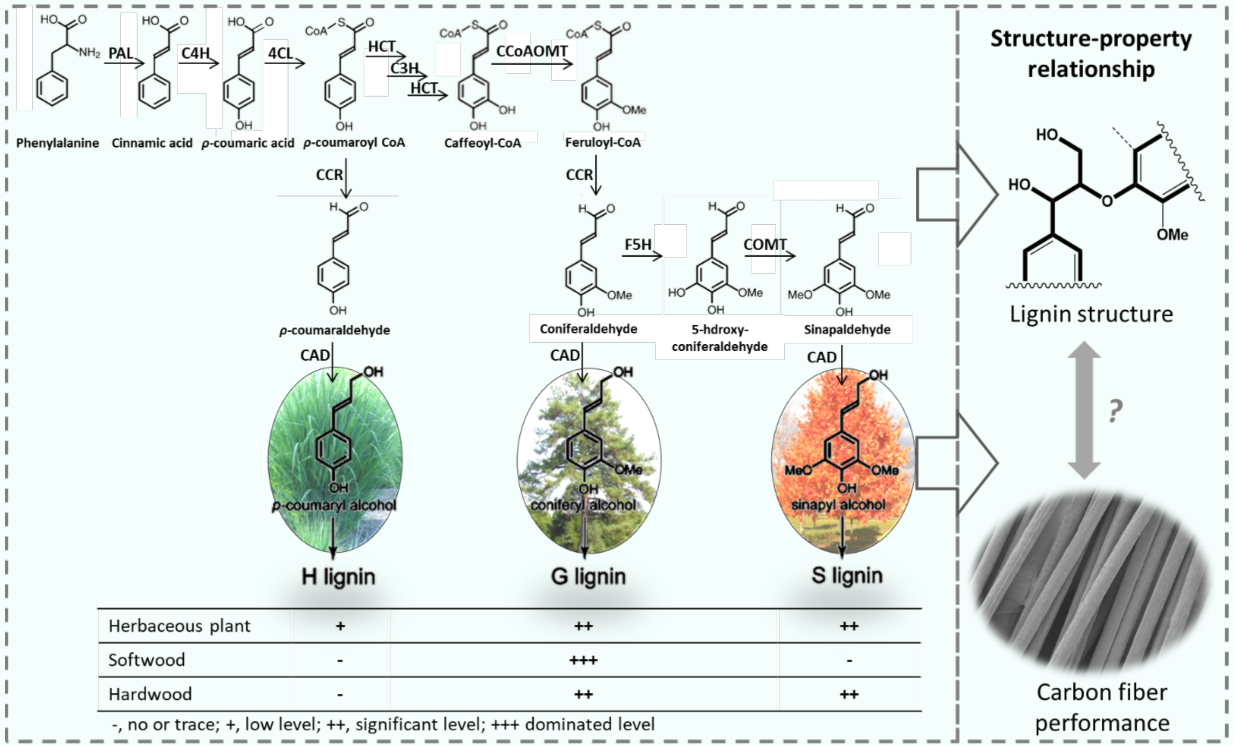


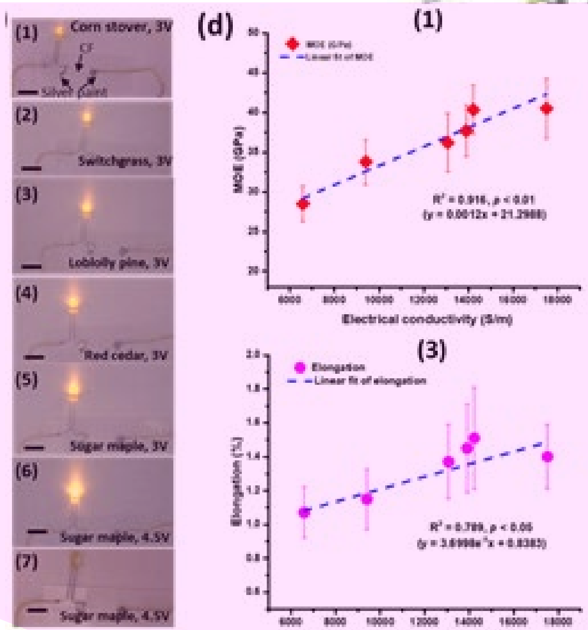
H-bonding system

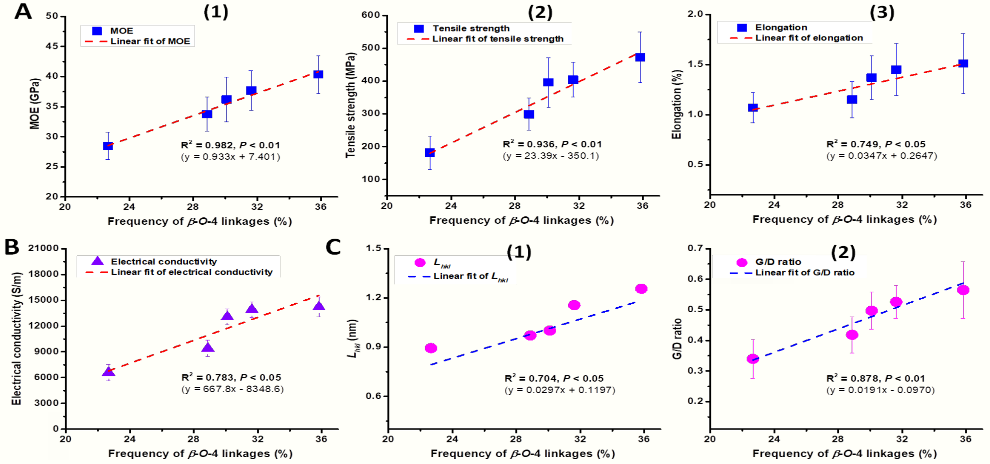


Q. Li et al. *Carbon,* 139, 500-511.

Linkage Profile Impacting Carbon Fiber Performance



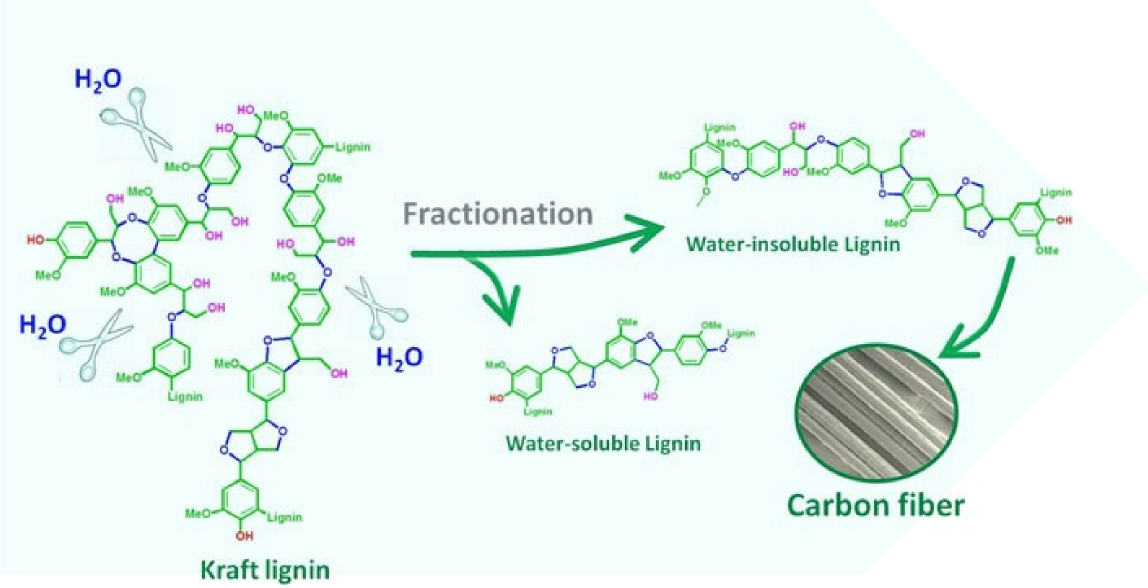


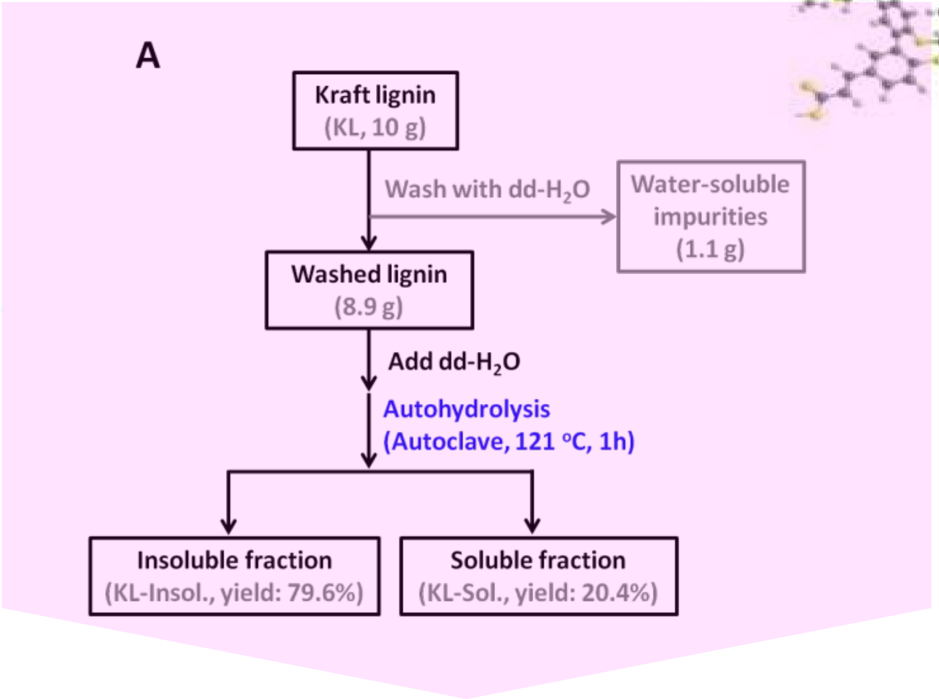


The study finds that linear linkages will enhance carbon fiber quality.

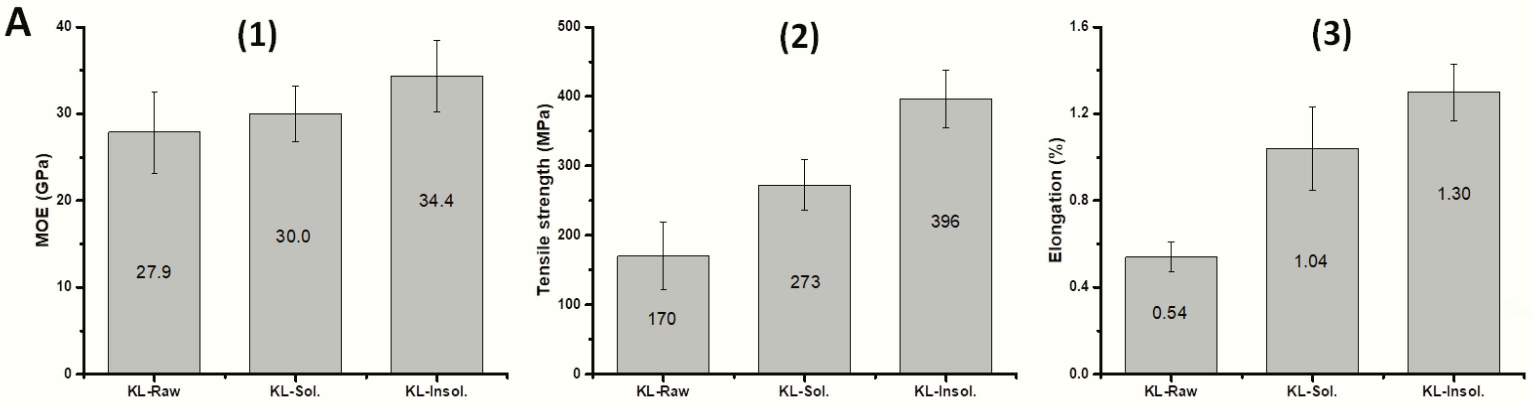
Q. Li et al. *Green Chemstry, Accepted*

Develop a Solvent-free Fractionation



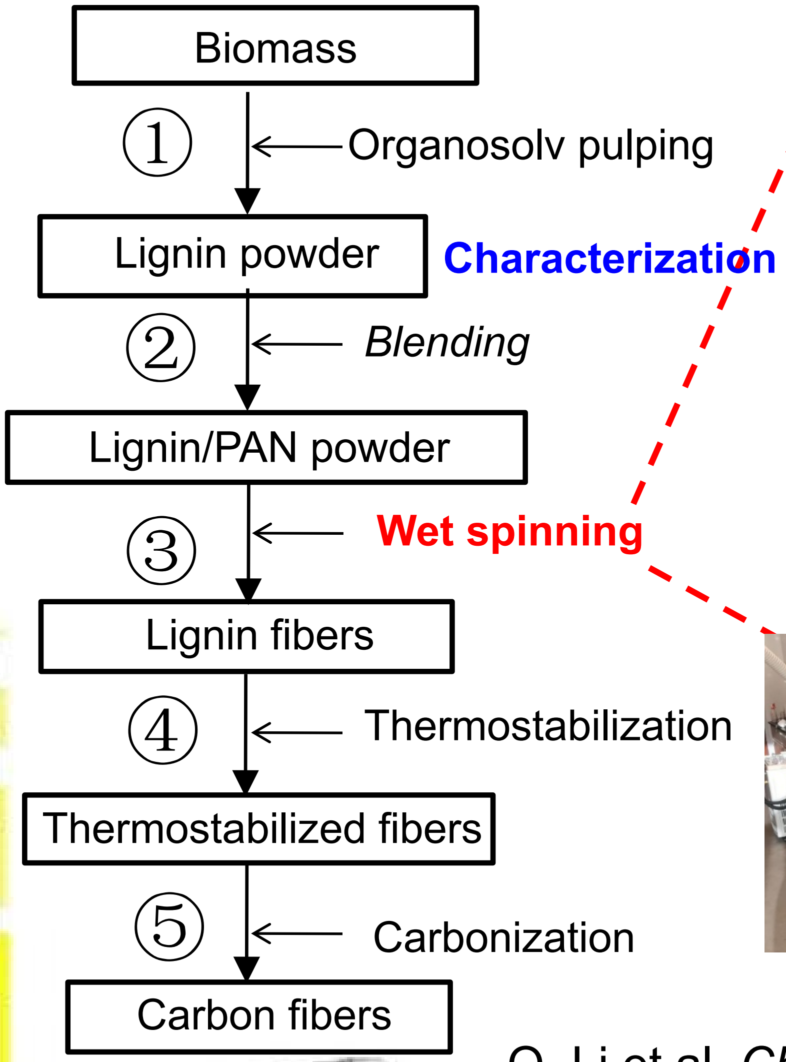


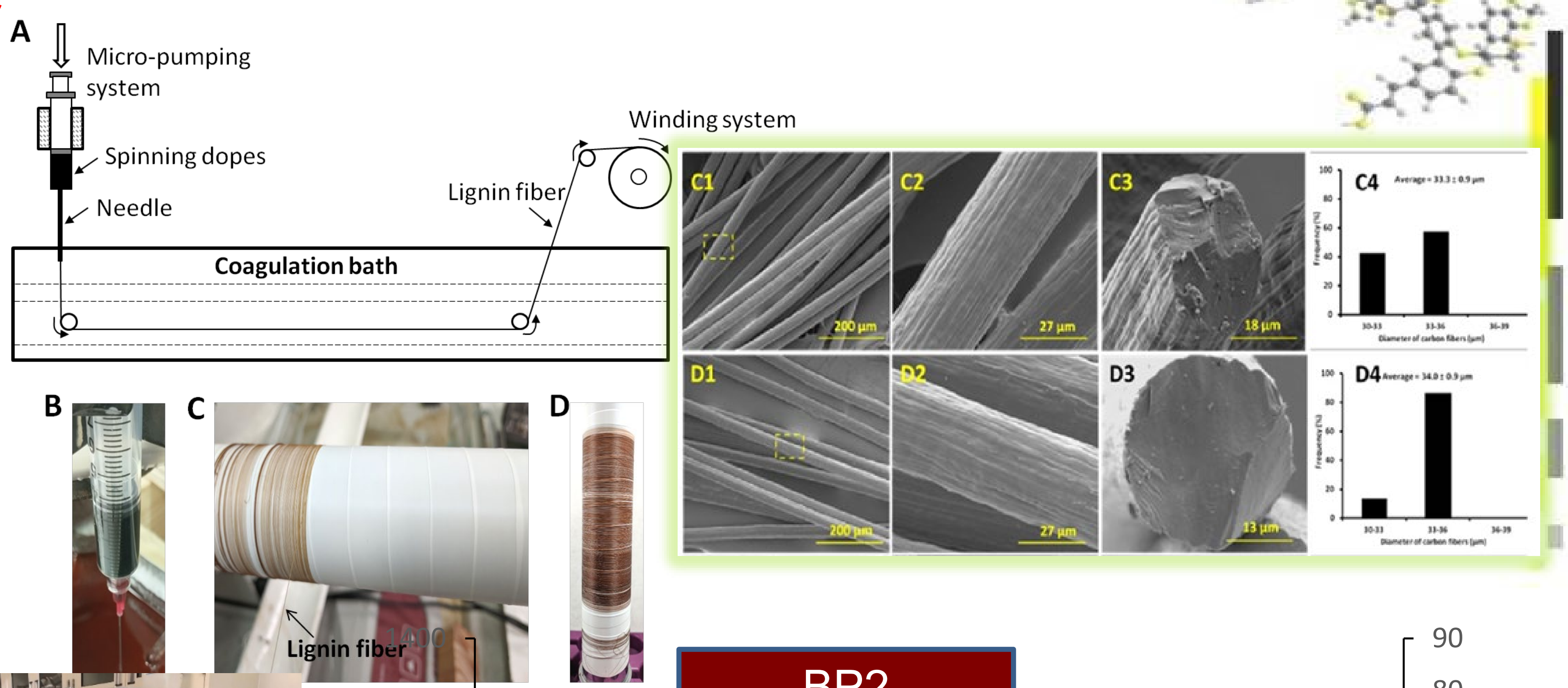
Mechanical properties

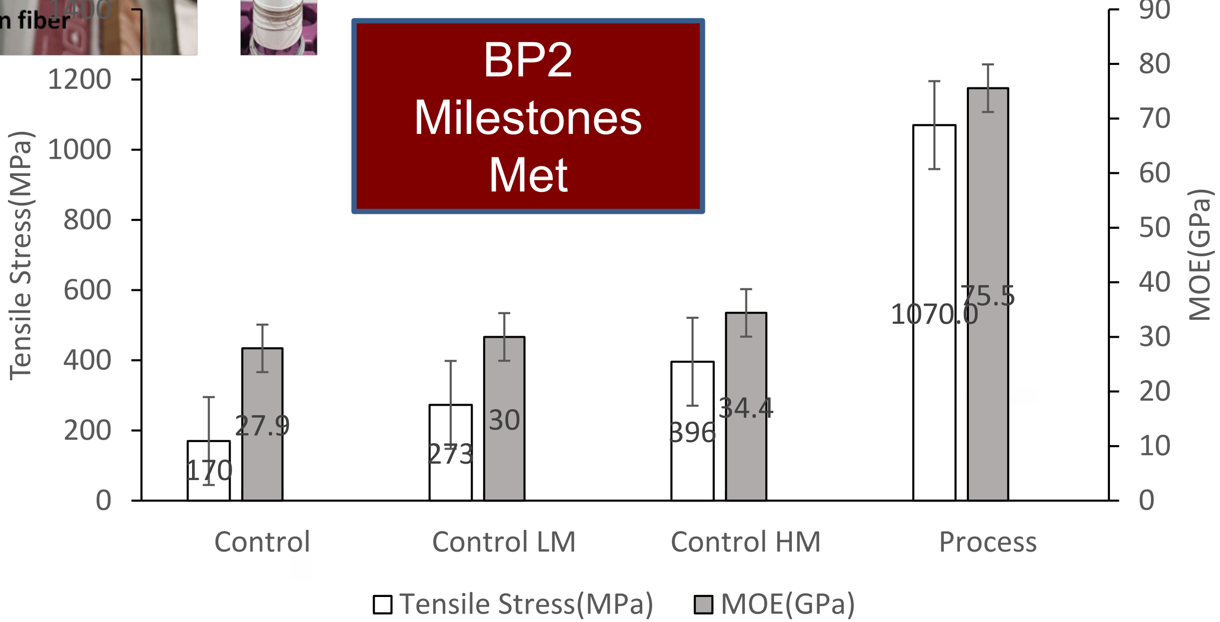


Q. Li et al. *ChemSusChem*, 12, 3249-3256

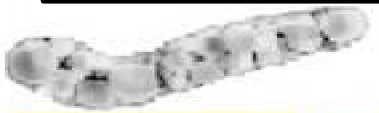
Customized wet spinning system





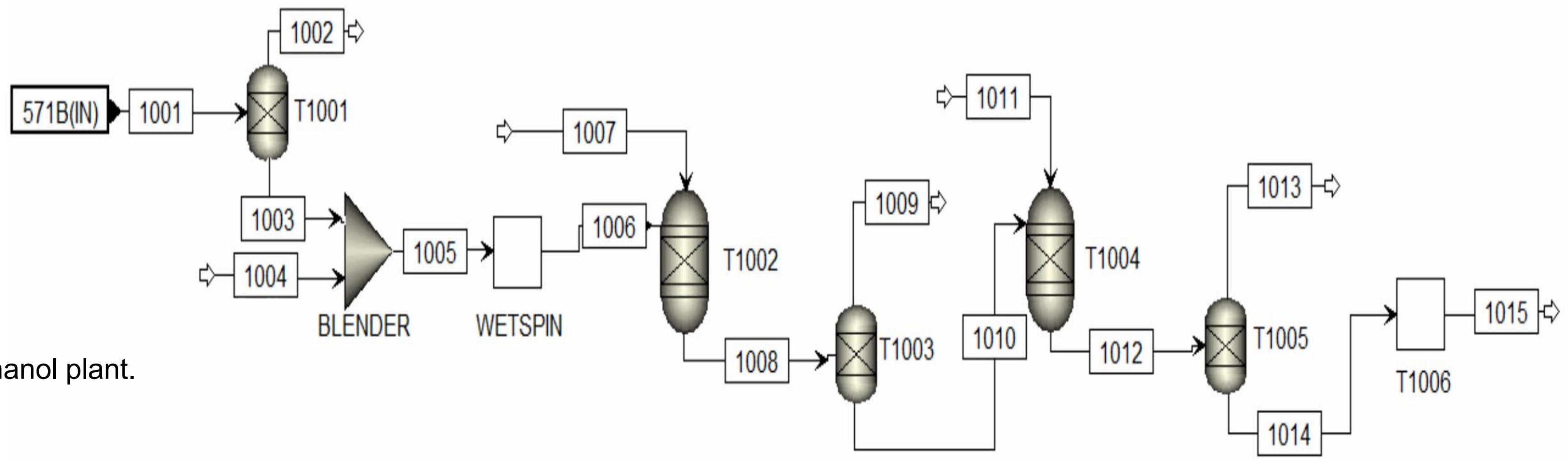


BP2
Milestones
Met



Q. Li et al. *Chem. Commun.*, 2019, **55**, 12655-12658.

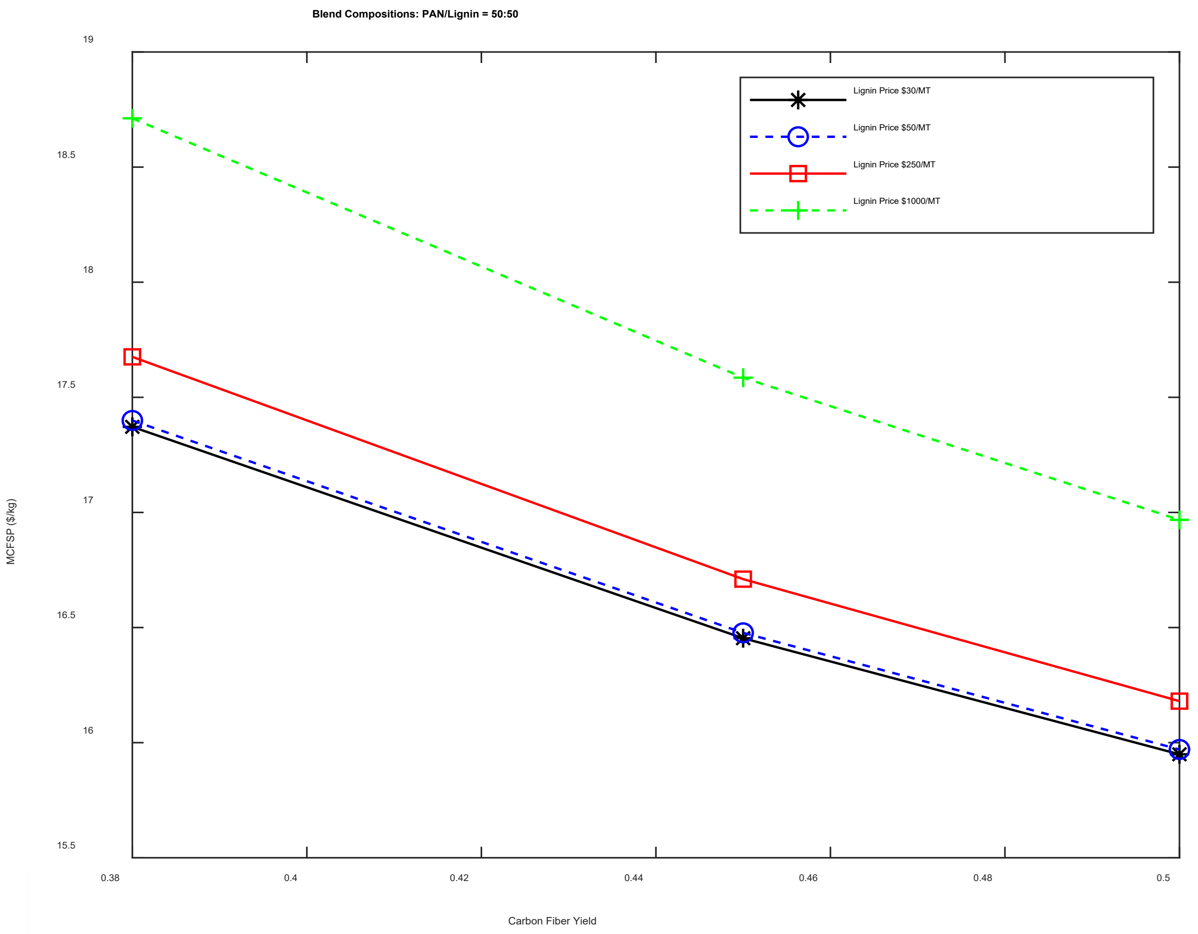
Process Flow Diagram of Carbon fiber production
The Technoeconomic Model

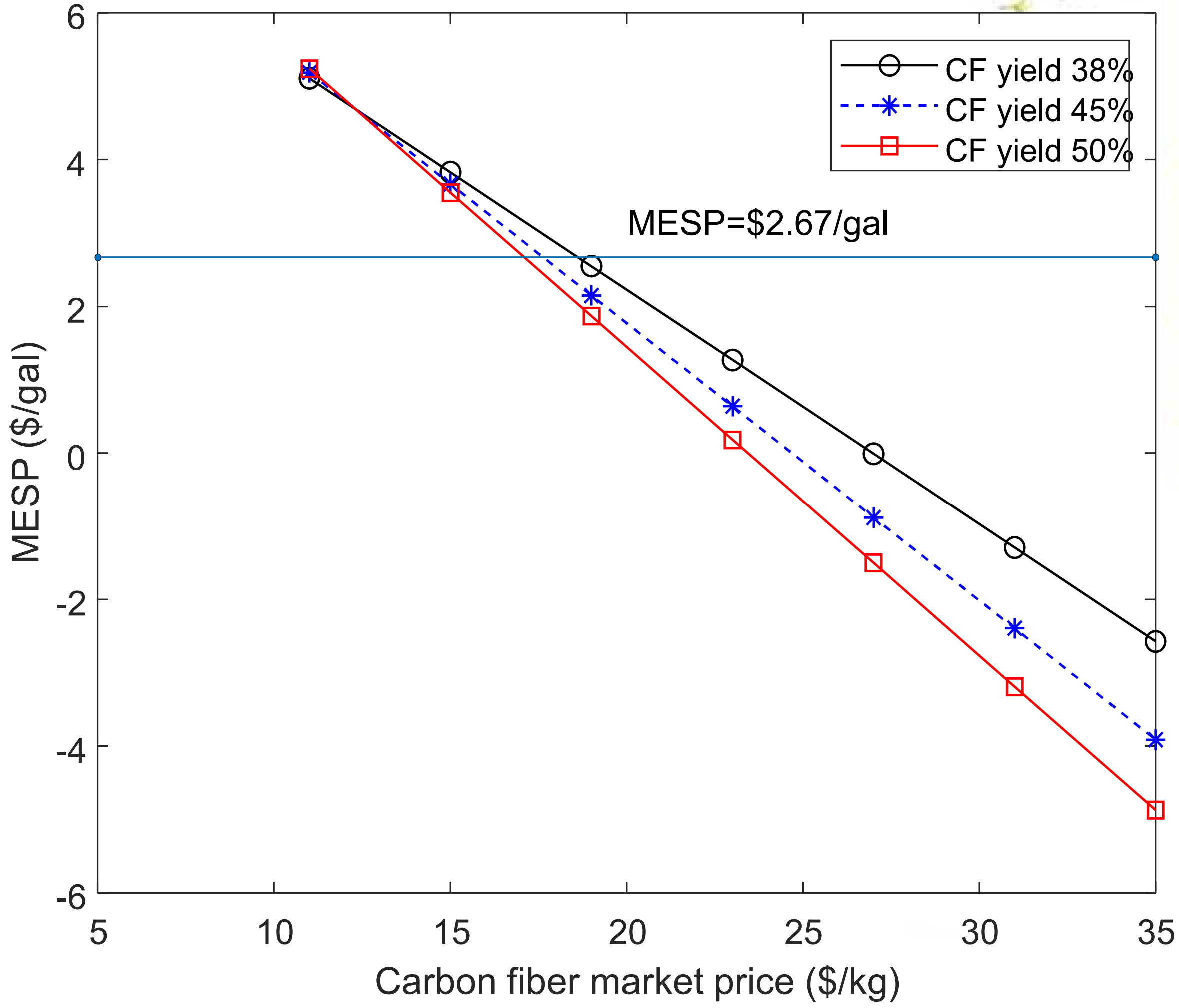


Material streams:
1001 –lignin from lignocellulosic ethanol plant.
1002 – impurities from raw lignin
1003 – purified lignin
1004 – PAN
1005 – lignin/PAN dope
1006 – lignin/PAN fiber
1007 – air
1008 – stabilized fiber + gases
1009 – gases
1010 – stabilized fiber
1011 – N2
1012 – carbon fiber + N2 and some waste gases generated
1013 – N2 and some waste gases generated
1014 – carbon fiber
1015 – surface-treated carbon fiber (product)

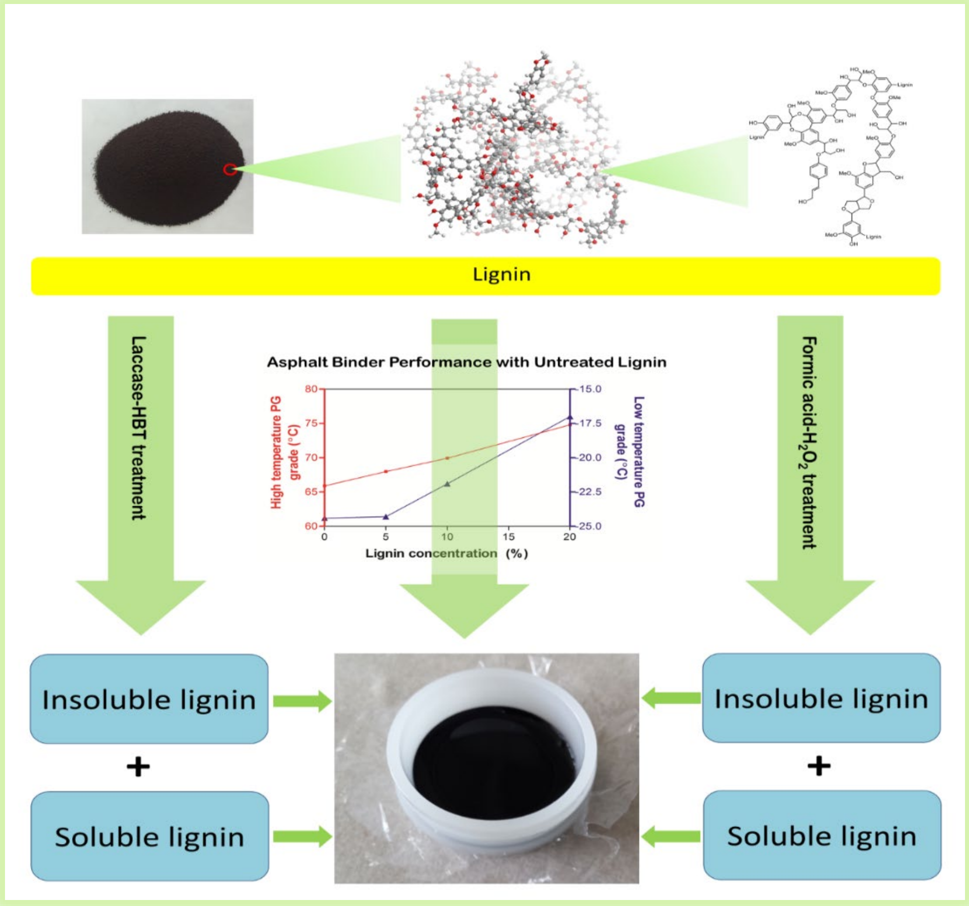
Major Equipment:
T1001 – Lignin separation/purification unit
BLENDER
WETSPIN – wet spinning equipment
T1002 – oxidation/stabilization oven
T1003 – gases/fiber separator
T1004- carbonation furnace
T1005 – carbon/N2 separator
T1006 – surface treatment subprocess

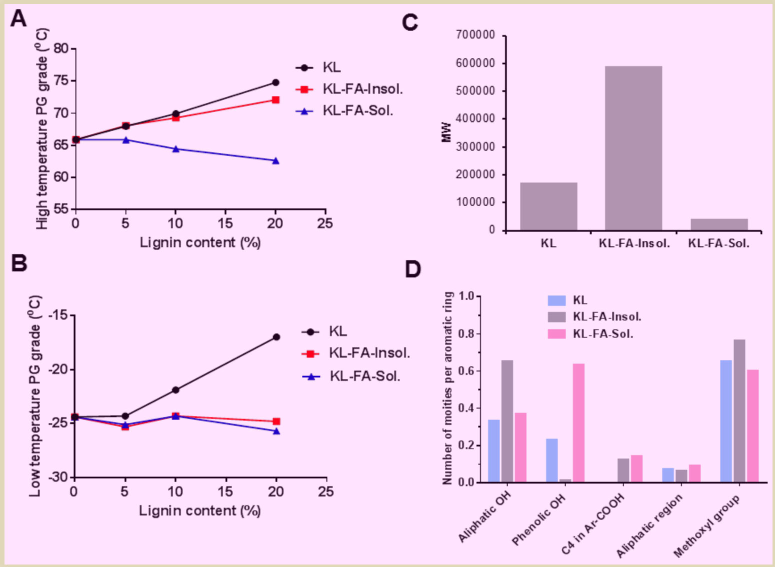
Effects of carbon fiber yield on MCFSP and MESP

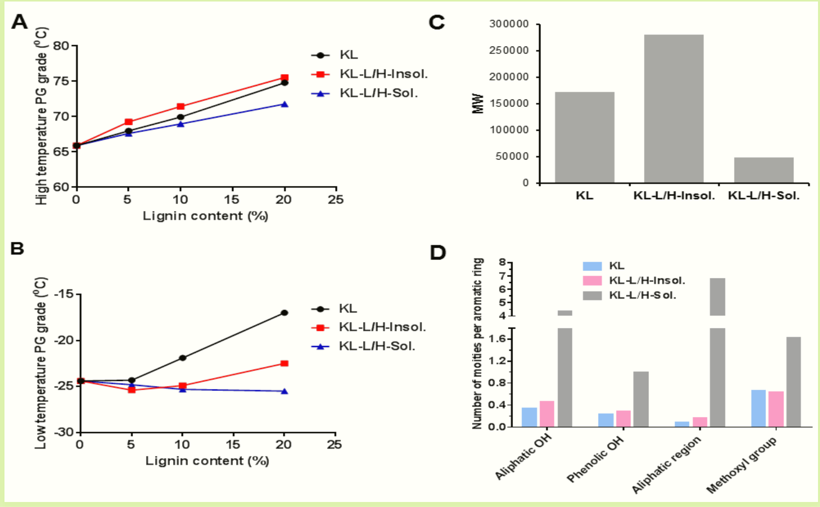




Fractionated Lignin as Unique Asphalt Binder Modifier



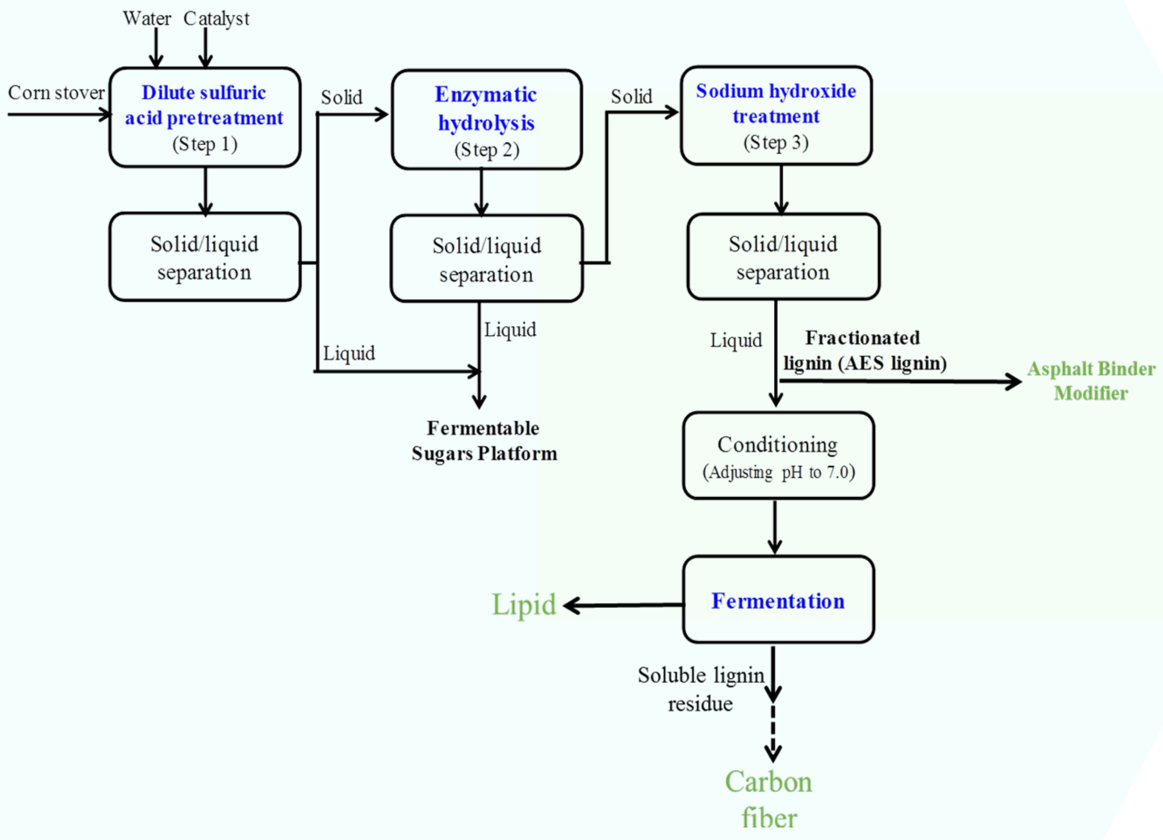


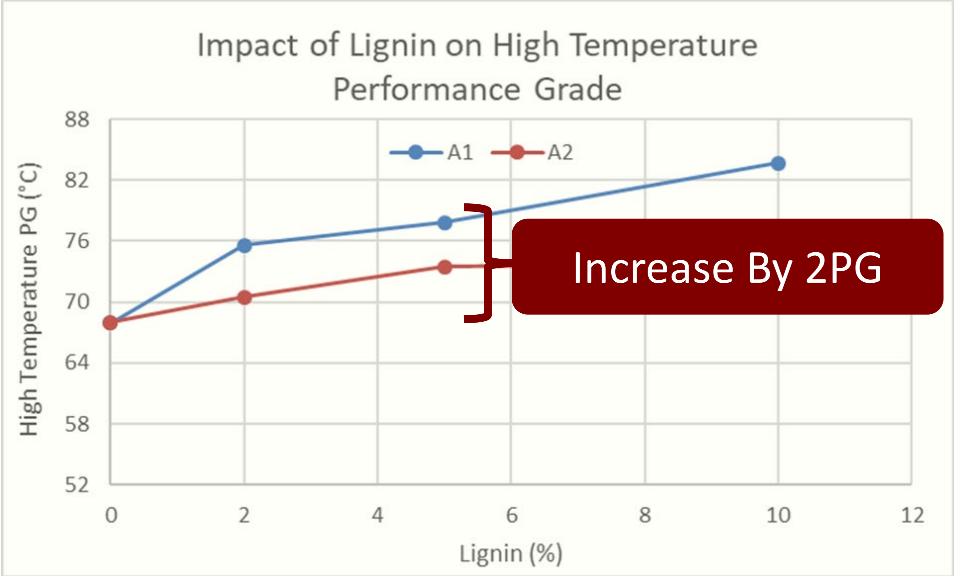


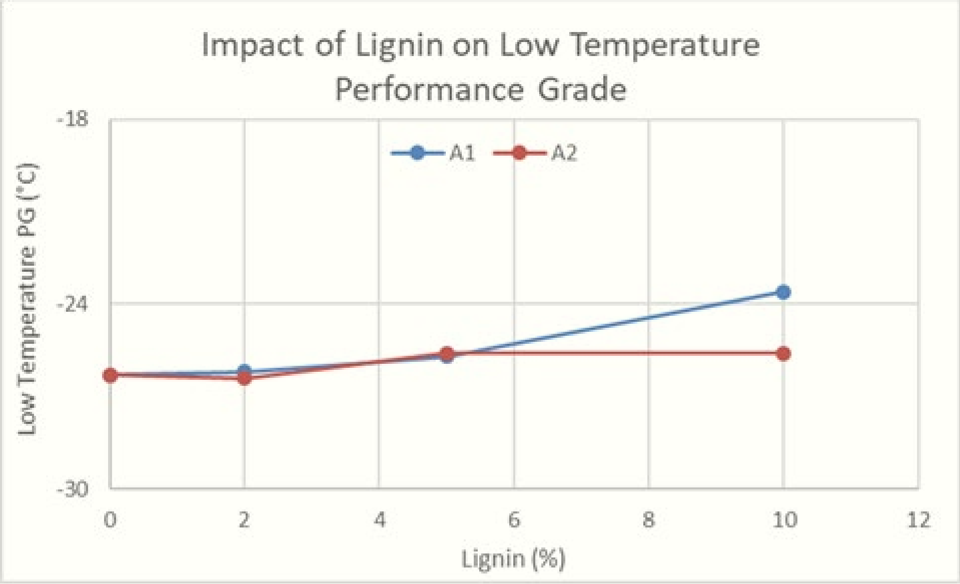
It is possible to improve high temperature performance without compromising low temperature performance.

S. Xie, et al. *ACS Sustainable Chemistry & Engineering*, 2017, 5 (4), 2817–2823

Fractionated Lignin Adds Significant Value to Biorefinery

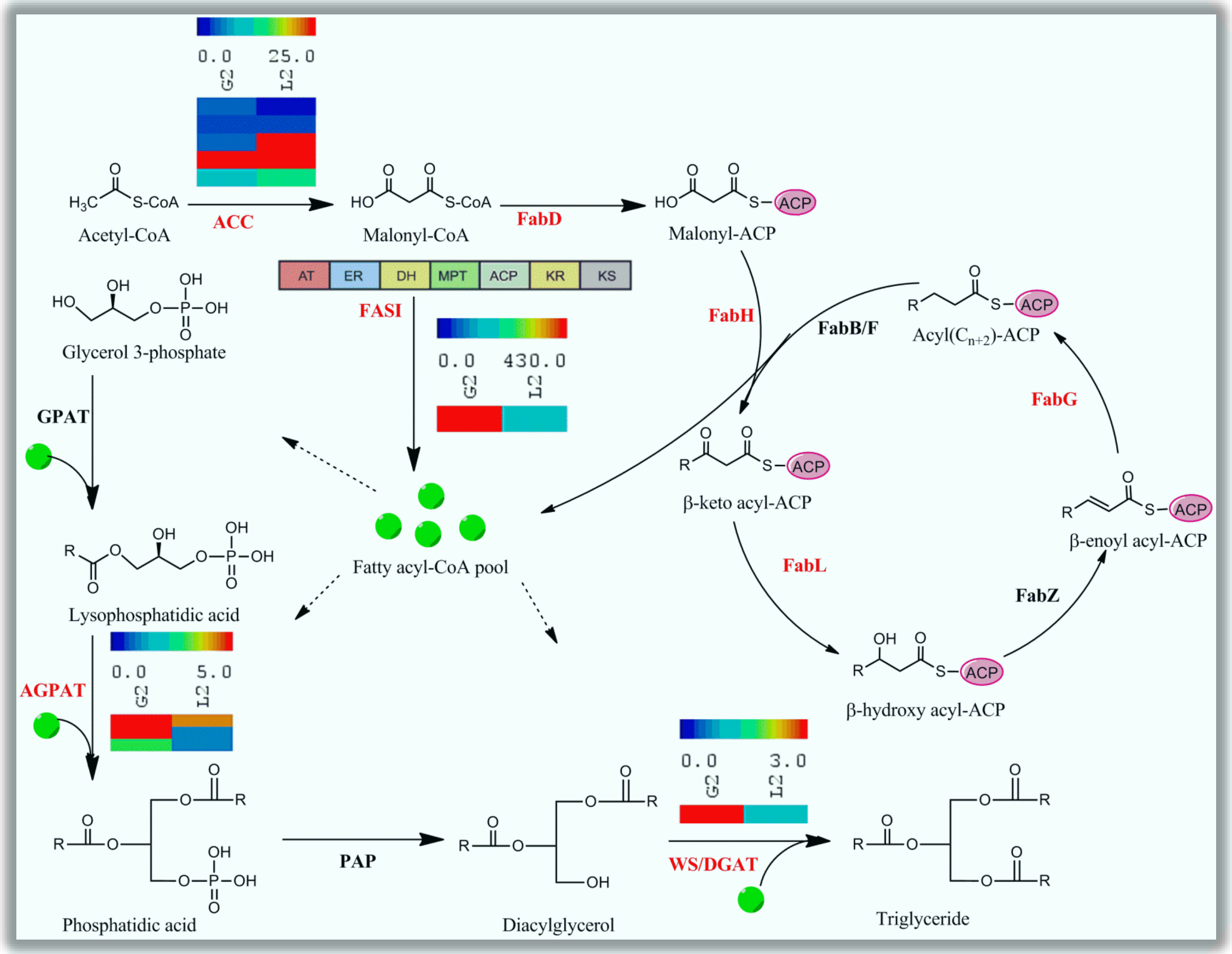


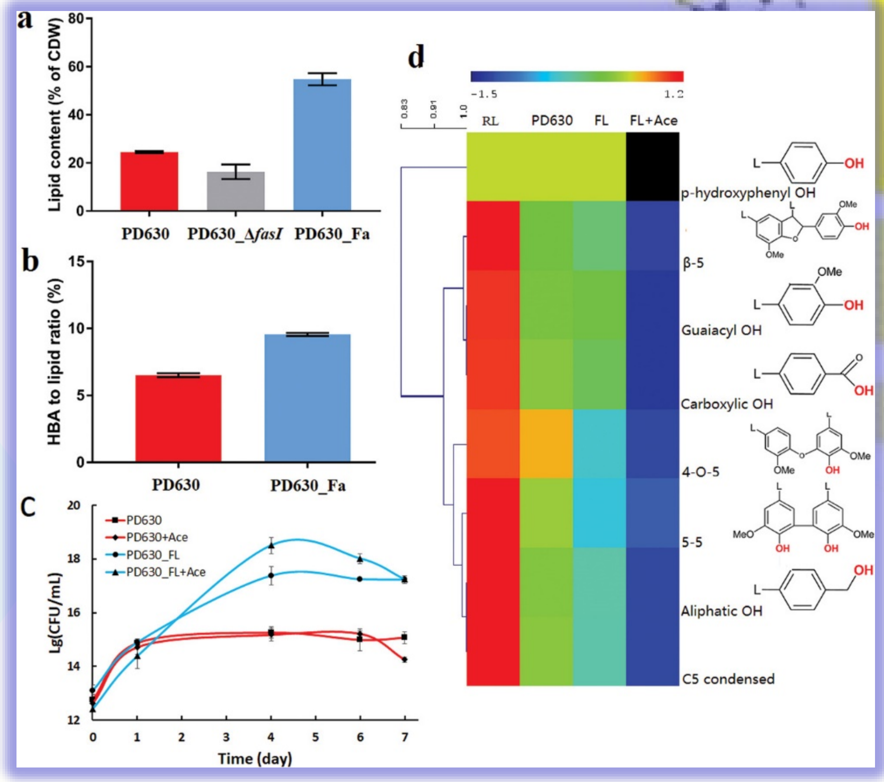




A 2 PG Grade Difference will translate into $2,000 market value of lignin. An ASPEN model is being built to evaluate the impact on MESP and $/GGE.

Systems Biology-Guided Design of Efficient Lignin to Lipid Conversion

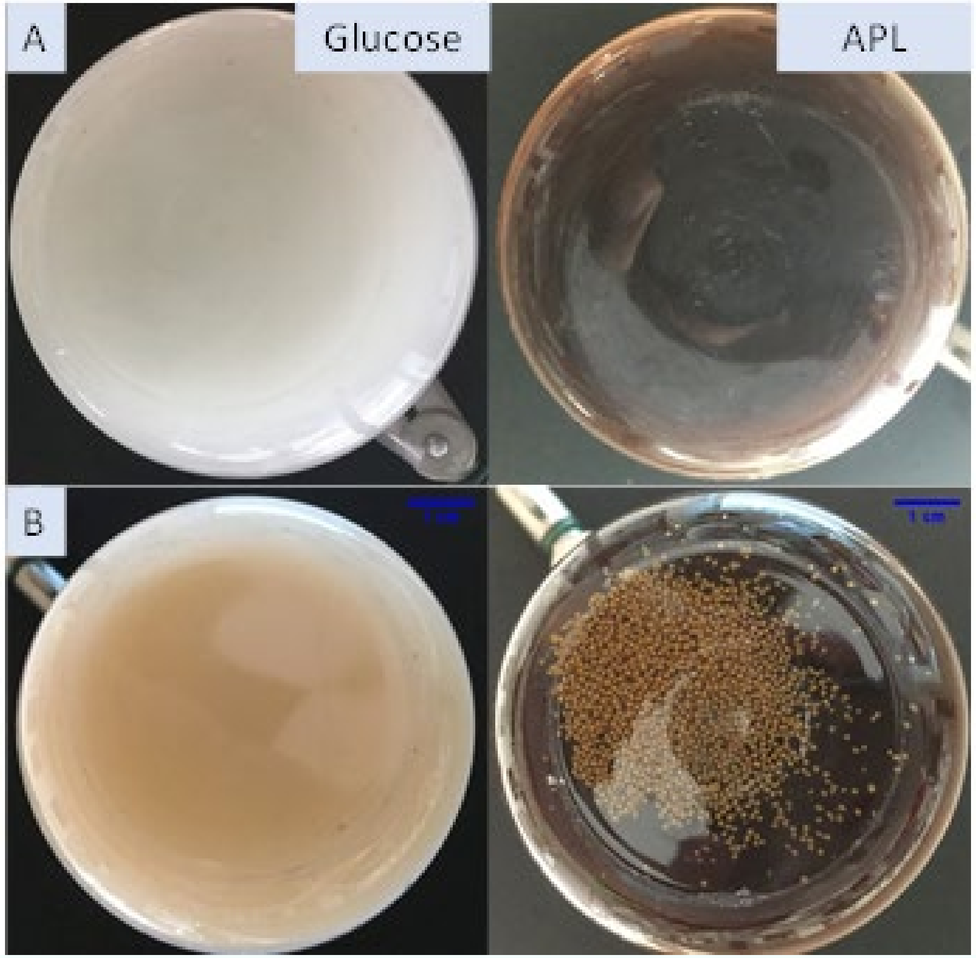


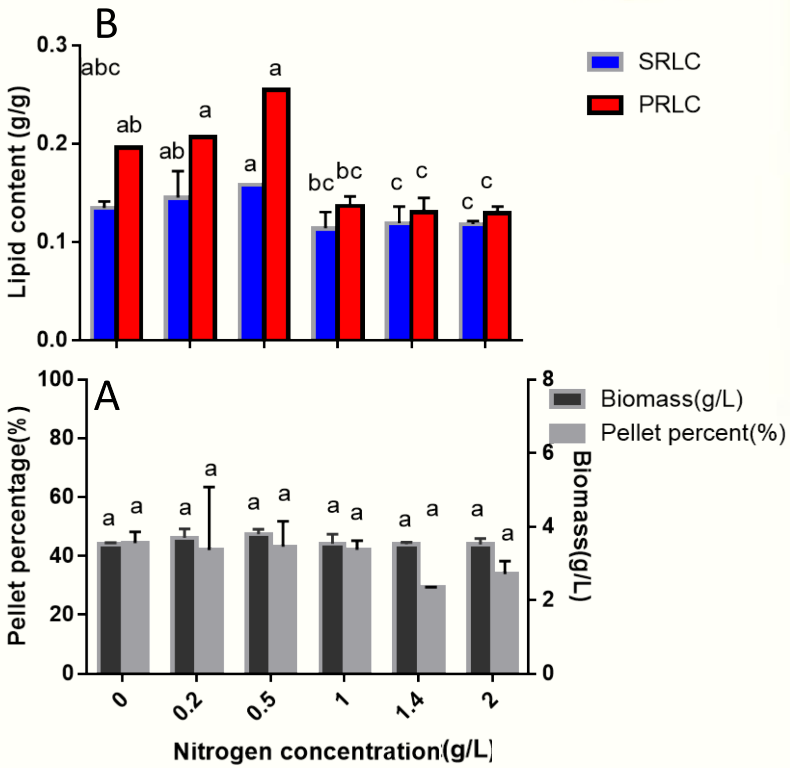


The microbial engineering and fractionation technology advancements has enabled >1,000 times increase cell growth.

QS. Xie et al., *Adv. Sci.*, 6, 1801980.

Linkage Profile Impacting Carbon Fiber Performance





A unique palletization technology was developed for *R. opacus* fermentation of lignin. Such a technology has increased lipid accumulation at the low nitrogen conditions and simplified the harvest.

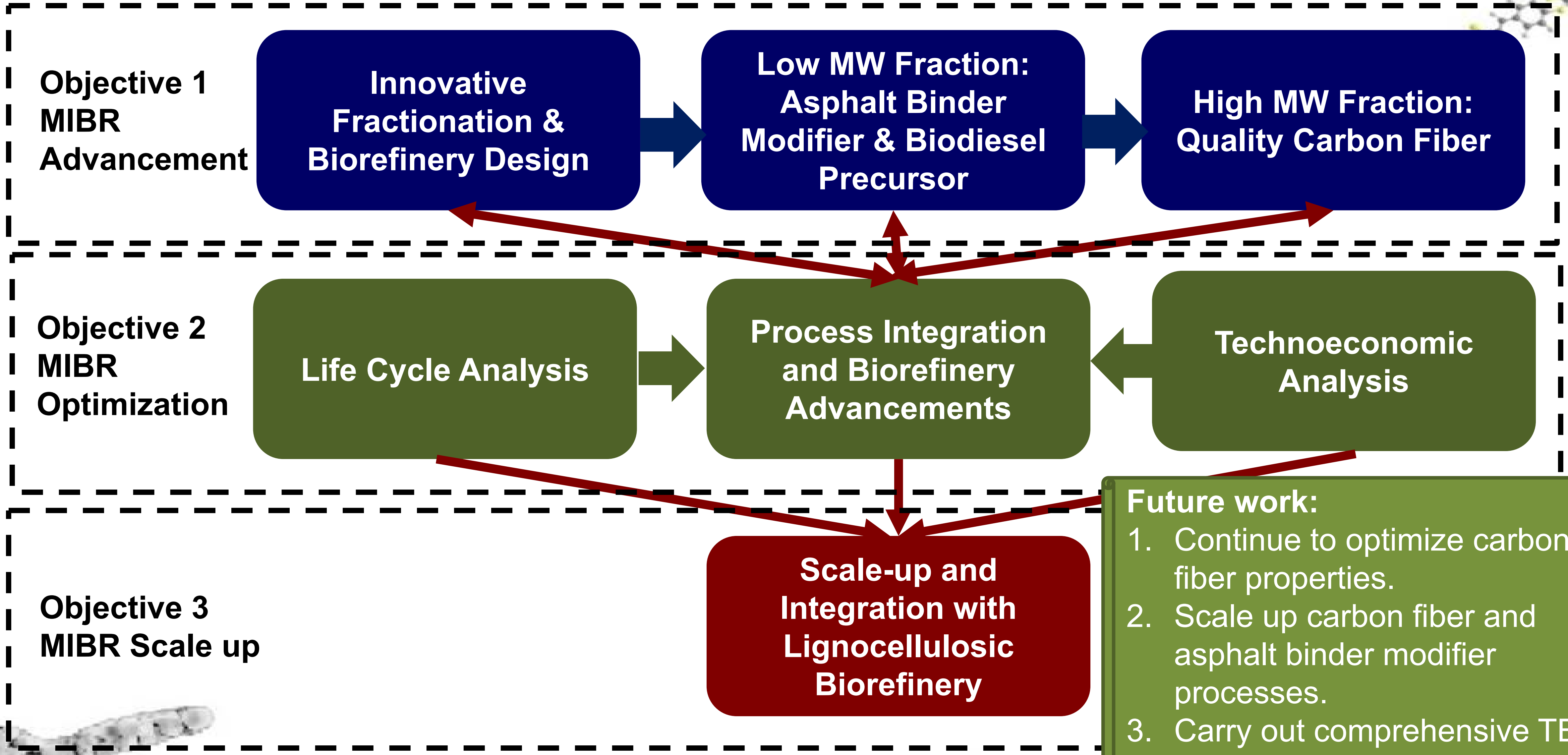
B. Xu et al. Drafted

Progresses and Outcomes Summary

Time Point		Benchmark		End of BP2		End of the Project	
Product	**Metrics**	Milestones	Actual	Milestones	Actual	Milestones	Actual
Carbon Fiber	**MOE**	20GPa	21 Gpa	50GPa	75GPa	100GPa	
	Tensile	100MPa	~200MPa	1GPa	1.07GPa	2GPa	
Asphalt Binder Modifier	**Rutting Temp Incr.**	7°C	1PG 7°C	10°C	2PG 15°C	10°C	2PG 15°C
	Low temp	Same	Same	Same	Same	Same	Same
Lipid for Biodiesel	**Titer**	10g/L	10g/L	15g/L	12g/L	25g/L	
	Conversion	30%	30%	30%	30%	40%	
Economic Outcome	**MESP[1]**	N.A.		N.A.	Depends	N.A.	
	~$/GGE[2]	N.A.		N.A.	~$3/GGE	~$3/GGE	

We have met the milestones to down select carbon fiber and asphalt binder modifier for scale up.

Future Work



Future work:

1. Continue to optimize carbon fiber properties.
2. Scale up carbon fiber and asphalt binder modifier processes.
3. Carry out comprehensive TEA and LCA.

Summary

The project will leapfrog the technologies to enable multi-stream integrated biorefinery (MIBR).

1. Overview – The develop and integrate multiple value-added bioproduct streams to enable multi-stream integrated biorefinery (MIBR) to reduce MESP and improve sustainability and cost-effectiveness of lignocellulosic biorefinery.
2. Management
   - S.M.A.R.T Milestones, and Go/No-Go milestones at the end of each BP.
   - Constant engagement with commercial partners.
   - TEA and LCA guide the process advancement for commercial relevance.
   - Two PCT patents filed.
3. Approach
   - Rigorous management approach to enforce milestones.
   - MIBR Development by Optimizing and Advancing Each Product Process
   - MIBR Integration and Optimization
   - MIBR Scale-up, TEA and LCA to guide the technology advancements and commercialization.
4. Impact
   - The project is directly addressing MYPP goals.
   - Aspen Plus model significant potential of carbon fiber to reduce MESP
   - The low-cost carbon fiber could improve the efficiency of energy sector substantially.
   - The asphalt binder modifier can enhance the infrastructure resilience to global climate changes.
5. Technical Accomplishments/Progress/Results
   - The project has met all BP2 milestones.
   - The project has delivered solutions to reduce MESP significantly
   - The project has led to profound scientific discoveries, guiding future process development
   - We will continue the scale up of two out of three streams according to the Go/No-Go milesones.

Quad Chart Overview

Timeline

- Project start date: 09/01/2018
- Project end date: 06/31/2022

	FY20 Costed	Total Award
DOE Funding	$610,916	$2,236,211
Project Cost Share	$330,254	$559.056

Project Partners*

- University of Tennessee, Knoxville/Oak Ridge National Lab
- Washington State University
- Texas Transportation Institute
- ICM inc. (as initial stage partner).

Project Goal

The project will leapfrog the technologies to enable multi-stream integrated biorefinery (MIBR), which will improve the economics and sustainability of lignocellulosic biorefinery and reduces MESP and $/GGE.

End of Project Milestone

At the end of the project, we will deliver integrated biorefinery to produce carbon fiber at MOE of 100GPa and tensile strength of 2GPa, along with asphalt binder modifier with 1PG increase of high temperature performance without compromising low temperature performance. We will select two product streams out of three streams in BP2. Currently, we set to select asphalt binder modifier and carbon fiber as next step focus.

Funding Mechanism

DE-FOA-0001689; Topic Area 2: High value products from waste/or other undervalued streams in an integrated biorefinery.

2018.

Acknowledgement

Project Management:

Joshua Messner   Marykate O'Brien   Jay Fitzgerald

CoPIs:

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Dr. Fujie Zhou
Dr. Brandon Emme
Dr. Bin Yang
Dr. Bruce McCarl

Texas A&M Transportation Institute

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OAK RIDGE National Laboratory

U.S. DEPARTMENT OF ENERGY | Energy Efficiency & Renewable Energy

Responses to Reviewers' Previous Comments

- We appreciate the reviewers to acknowledge that the project 'is scientifically sound and well planned', 'highly relevant to the BETO MYP goals', and has a 'good team' for execution. We also appreciate the reviewers' recognition of potential 'serious commercial interest' and being 'supportive of' BETO's goals in a 'unique way'. We agreed with the reviewer that value-added products from waste is the near-term path for lignocellulosic biorefinery. The same principle applies to corn ethanol, where DDGS enables the economics. We also agree with the reviewer that the integration with current biorefinery processes should be explored. In fact, we are using waste from AFEX, alkaline, and acid pretreated biomass. One of the advancement of our technology is to develop a plug-in process to treat acid-pretreated biomass with alkaline to dissolute lignin, and this dissolution fraction will be used to neutralize acid stream, forming a lignin stream. The alkaline will have to be used to neutralize the acid stream, anyway. The plug-in process can be readily integrated with current biorefinery design without significant infrastructure changes. A full ASPEN model has been built for the process in the project DE EE 0007104 and proven that it did not add upstream cost significantly, yet resulting in various value-added streams. The details has been submitted for publication. We agree with the reviewer and will continue to integrate current pretreatment technologies, instead of creating new processes, to achieve immediate, tangible and transformative impact on biorefinery industry.

Responses to Reviewers' Previous Comments

- The reviewers also highlighted 'the specific target of reducing the cost of ethanol (by $0.50/GGE)'. In fact, we have built a complete ASPEN model for carbon fiber and is working on one for asphalt binder modifier. The TEA models allow us to identify the economic drivers and impact quantitatively.
- We appreciated that the reviewer's recognition of ICM as a strong industrial partner. We actually enjoyed working with ICM very much. Even though ICM are not working with experimentally due to the re-structuring, they are still serving as an advisor for commercial development. We are also engaging with other biorefinery companies like POET. The project actually attracts significant interest from Venture Capitals and carbon fiber start-ups, too. We are engaging with them, aiming to commercialize the technologies.
- We appreciate that the reviewer recognizes the 'qualified and organized' team. Regarding to the process, we have explored the processing of AFEX, acid, and alkaline pretreatment-derived lignin into carbon fiber, lipid conversion, and asphalt binder modifier. Moreover and as aforementioned, our optimized process is a slightly modified acid pretreatment, where the alkaline to neutralize acid soluble stream were first used to treat solid fraction to dissolute more lignin. This allows synergistic improvement of carbohydrate conversion and lignin utilization. Full ASPEN model has been built to evaluate the process and identify the economic drivers.

Responses to Reviewers' Previous Comments

- We agree with the reviewer that the three different product paths makes this project more complicated than a traditional project. This is why we down-select to two different products at the end of BP2. In fact, some of the most profitable corn ethanol wet-milling biorefineries also produce multiple products, such as starch, corn oil, and syrup. The multiple-product approach is critical to avoiding market saturation and maximizing the economic return. As aforementioned, the pretreatment and fractionation technologies will be based on a modification of current pretreatment without changes in infrastructure, which will allow us to quickly scale up the project within three years. We agree with the limited budget and resources very much, but we are leveraging the resources from existing EERE projects and institutional resources. During the past two years, we have built two full ASPEN models for carbon fiber and our process. We are working on the third one for asphalt binder modifier.

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13. Zhi-Hua Liu, Rosemary K Le, Matyas Kosa, Bin Yang, ***Joshua S. Yuan****, Arthur J Ragauskas, Identifying and creating pathways to improve biological lignin valorization, *Renewable and Sustainable Energy Reviews,* 2019, 105, 349-362.
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The project has led to 28 publications with a total impact factor at 190.

Patent and Commercialization

- **The project has led to two patent applications.**

1. J.S. Yuan, et al., “Conversion of lignin into bioplastics and lipid fuels”, PCT/US2016/024579, WO 2016154631 A1 – The PCT patent is at US and EU application stage.
2. J.S. Yuan et al., “Lignin fractionation and fabrication for quality carbon fiber”, PCT/US2019/019620 – This is a PCT application.

- **Commercialization efforts -- We have actively engaged with two industries.**

1. For lignocellulosic biorefineries, we have worked closely with ICM inc. We also had dialogue with POET for lignin utilization.
2. For carbon fiber industry and pavement industry, we are engaging with Venture Capital and start ups for commercialization.