DOE Bioenergy Technologies Office (BETO) 2023 Project Peer Review

DE-EE0009943:

A closed loop upcycling of single-use plastic films to biodegradable polymers

April 5th, 2023

Plastics Deconstruction and Redesign

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Iowa State University

Project Overview

History

DE-FOA-0002473: Single-use plastic recycling funding opportunities / Topic Area 1: Novel approaches to recycling and upcycling films. The initial verification was completed.

Project Goal

The project will develop a plasma-biological hybrid technology to upcycle mixed single-use plastic film (SUPF) wastes into biodegradable polymers using a circular carbon approach.

Project Objectives

- Determine feedstock variability in material recovery facilities (MRF)-received plastic wastes and obtain decontaminated SUPFs comprising >95% of polyolefins
- Deconstruct the decontaminated SUPF wastes using a low-temperature CO_2 plasma to achieve > 90% oxygenated intermediate liquid (OIL).
- Synthesize polyhydroxyalkanoates (PHAs) from the OIL with >15% conversion efficiency and >85% product recovery.
- Determine the economic and environmental impacts.

Impact

Develop a novel technology to upcycle currently landfilled plastic wastes into environmentally friendly products using an energy and carbon-efficient approach.

Problem Statement

- Roughly half of our global annual plastic production is destinated for a single-use product. Over 91% of plastic waste is not recycled.
- Polyolefins account for nearly 2/3 of the total plastic production.
- About 60% of MRF bales consisted of single-resin films. Approximately 90% of the single-resin films are polyethylene (PE), whereas 10% are polypropene (PP).
- MRF-derived SUPFs are sent to landfills without recycling.
- Challenges in recycling SUPF wastes:
  - Heterogeneous composition with high contaminants
  - Difficulty of processing (e.g., clogging reactor, hard to shred)
  - Energy-intensive deconstruction
  - Low-value products with limited potential applications

Management

- Monthly team meetings with the DOE-AMO managers to update progress against milestones, discuss technical challenges and review project administration.
- Bi-weekly meetings between different tasks and institutional co-PIs to coordinate research efforts, identify potential problems, and develop solutions.
- Integrate TEA throughout the project; improve technical approaches based on the preliminary TEA results to mitigate potential risks in meeting the project targets.

Institute support
- Project finance
- Project management
- Patent application

PI: Xianglan Bai
- Coordinate report
- Management
- Facilitate communication
- Implement milestones

Industrial collaboration
- Feedstock information
- Evaluate products for commercialization

SUPF feedstock (Task 2)
ISU: Keith Vorst & Greg Curtzwiler

SUPF deconstruction (Task 3)
ISU: Xianglan Bai, Hui Hu
SNL: Nils Hansen, Lenny Sheps

PHA synthesis (Task 4)
MU: Caixia Wan
Quasar: Yebo Li, Xumeng Ge

TEA & LCA (Task 5)
ISU: Mark Mba Wright

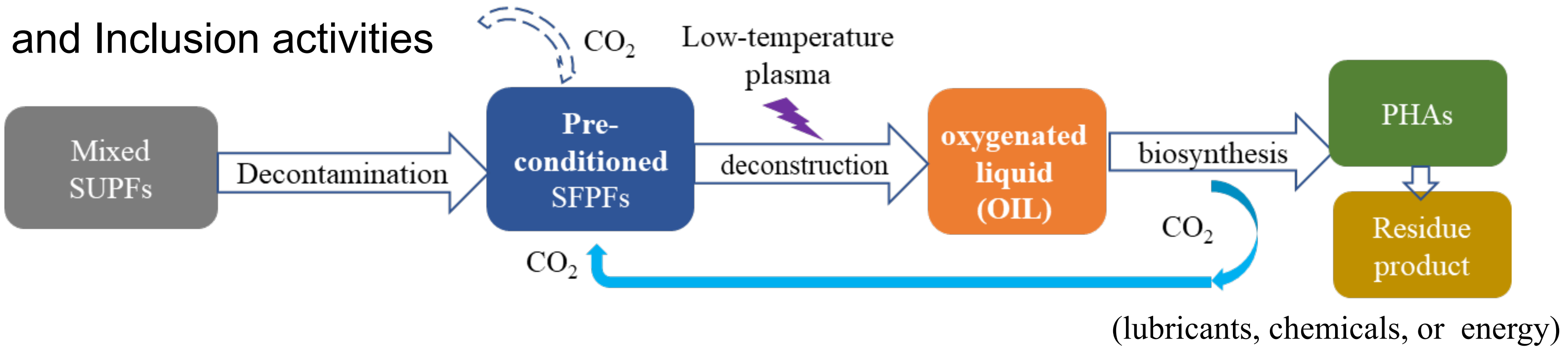
Approach

Tasks

1. Initial verification
2. Prepare decontaminated SUPF feedstock consisting of polyolefins, and determine waste feedstock variability
3. Develop plasma-based deconstruction of SUPF wastes using CO_2
4. Develop bioprocessing of SUPF-derived OIL to PHAs and characterize residues as a second product stream
5. Develop TEA & LCA modeling to evaluate economic and environmental impact, and aid process optimization
6. Diversity, Equity, and Inclusion activities

Risks/challenges identified

- Feedstock variability in MRFs
- Biological processing is not optimized



Approach

Initial verification milestone:
Demonstrate the ability to produce >90% of OIL on PE mass basis using CO_2 plasma; Demonstrate growth of PHA-accumulating microbes on formulated OILs and biological conversion of OIL to PHAs based on literature (completed).

Go/No-Go #1: PHA yield of 15% from OIL on a dry cell basis at a 1-liter scale using the optimized formula (due in 11/2023)

End of Project Goals:

- A SUPF decontamination process will be developed and validated to prepare the feedstock suitable for downstream biological conversion.
- An optimized plasma deconstruction process will be developed to convert the decontaminated SUPFs into clean OILs at >90wt% yield by utilizing CO_2.
- A fermentation process with mixed culture will be developed to achieve >15% OIL-to-PHAs conversion and >85% PHA recovery.
- TEA and LCA of the hybrid SUPF upcycling for different conversion scenarios will be obtained to meet the carbon, energy, and cost targets.

Impact

Technical impacts

- Innovate MRF plastic recovery technique to prepare decontaminated SUPF feedstock with significantly reduced heterogeneity and better processibility.
- Transformation of waste plastics into chemicals suitable for value-added applications.
- Improved metabolic engineering to produce high-value products from waste plastics.
- A new approach to convert CO_2 with improved efficiency.

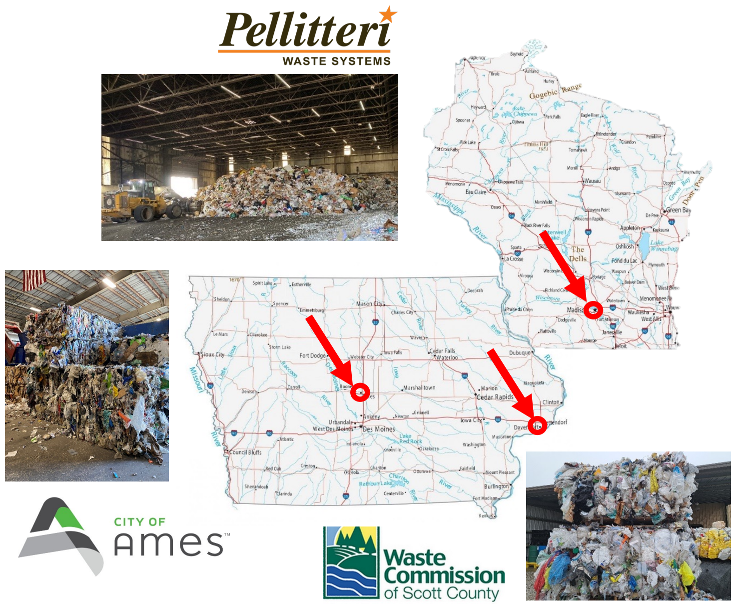
Broder impacts

- Convert wastes meant for landfills into biodegradable polymers and value-added products using an environmentally friendly approach.
- Waste plastic upcycling with concurrent CO_2 utilization.

Results dissemination

- A pending patent filed by Iowa State University.
- A Presidential Sympodium Presentation at the 2022 ACS Fall Meeting
- Journal article in progress

Progress Task 2. Preparation of clean SUPFs and characterizations

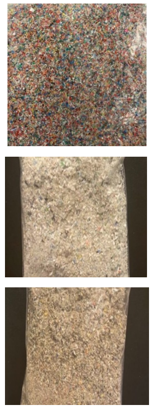


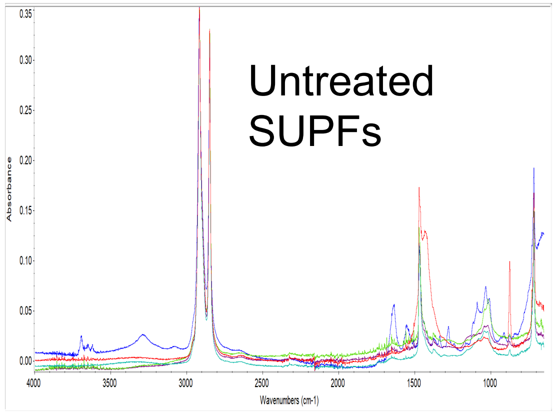
Waste materials from three different MRFs were sourced, sorted, and decontaminated.

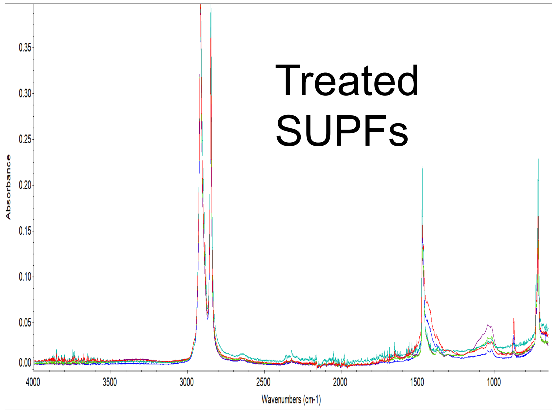
- Characterization was conducted using FTIR, DSC, and ICP-OES.
- Virgin resin (LDPE, LLDPE, and HDPE) and decontaminated samples from three MRFs were prepared.

Pretreatment process:

- Collection
- Sorting
- Densification
- Shredding
- Friction wash
- (Ultrasonic wash)





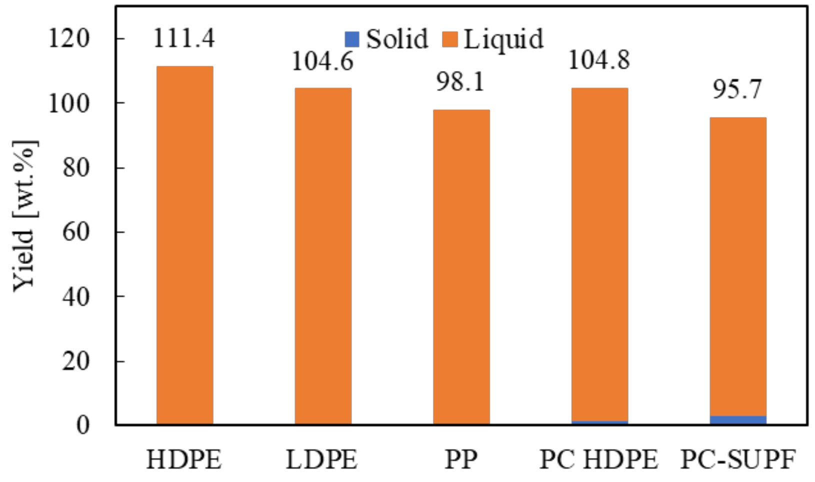


Milestone 2.1.1: (M6) MRF-received SUPFs material is sorted and decontaminated. The SUPF materials before and after the film treatment are characterized and quantified. (**Completed**)

Milestone 2.1.2: (M18) 5 kg of decontaminated SUPF containing > 95% of PE and PP. (**in-progress**)

Progress Task 3. SUPF deconstruction to OIL

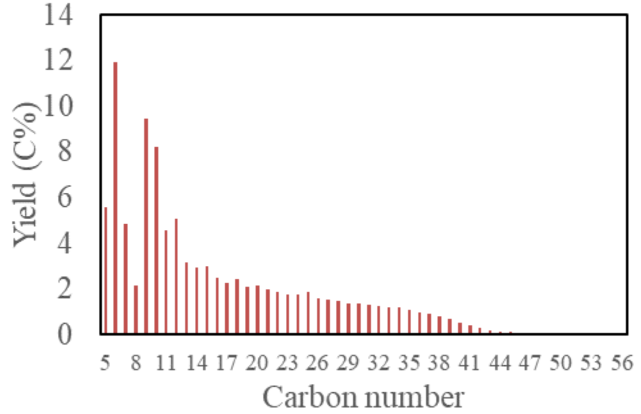
OIL yield based on starting mass of plastics



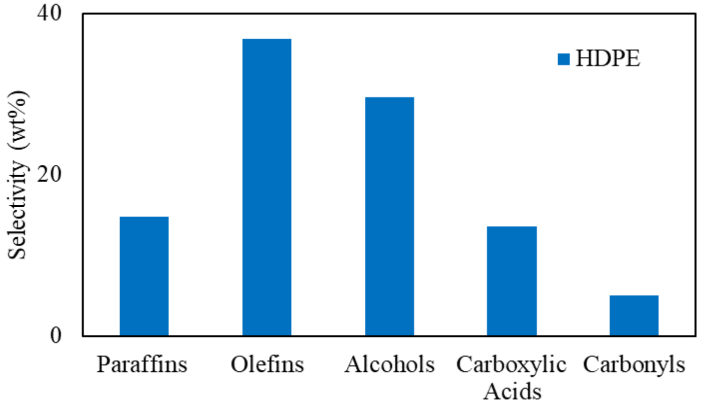
PC-HDPE

PC-SUPF

OIL product distribution



OIL functional groups



(Manuscript data)

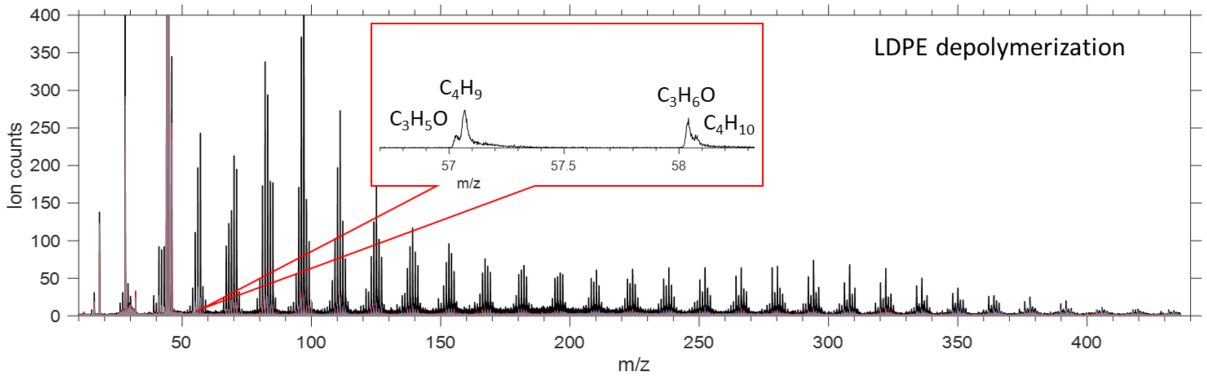
Plasma-based conversion of virgin polyolefins and mixed waste polyolefins to OILs using CO_2.

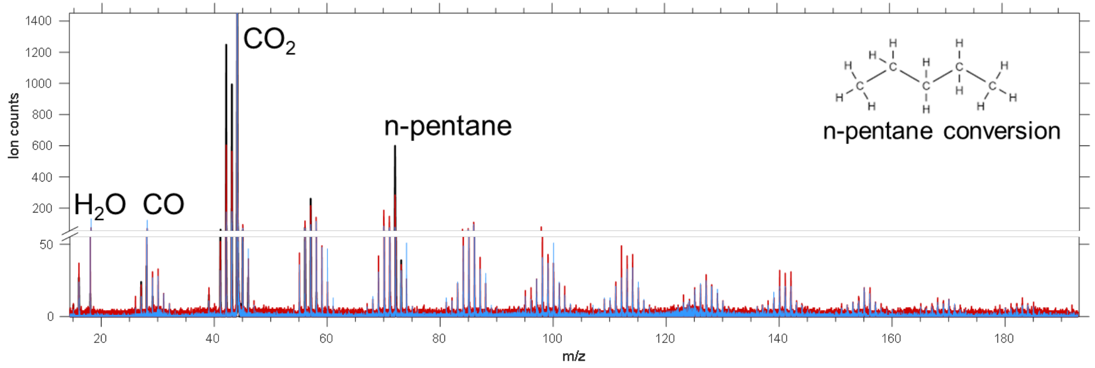
- OIL yields >90% per initial plastic mass.
- OIL characterizations by HT-GC/MS, NMR, elemental analysis, and GPC to confirm oxygenated functional groups and reduced molecular weights.
- Comparable OIL yields from virgin and mixed waste plastics.

Milestone 3.1.1: (M12) OIL produced from mixed SUPF using a bench scale reactor and products are fully characterized (**in progress**).

Progress Task 3. SUPF deconstruction to OIL

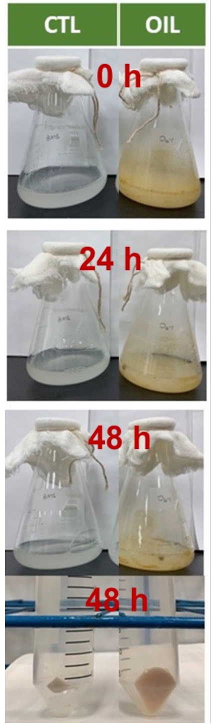
- **Operando characterization of the plastic conversion for comparison with offline product analysis**.
  - Rapid detection of volatile products by TOF mass spectrometry with < 1 min averaging
  - Online detection of condensable products by tandem (MS-MS) mass spectrometry
  - High-throughput optimization of process conditions
  - Model compounds and isotopic studies
- **Two different types of plasma reactors are designed.**





Milestone 3.1.2: (M15) OIL composition is analyzed in Operando during the reactions; condensed and vapor-phase product distributions are compared to those from offline analysis to discover key parameters or reactions determining product selectivity (**in progress**).

Progress Task 4. SUPF-derived OIL to PHAs



OIL vs. CTL (Basal salt medium)

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Emulsion

⇩

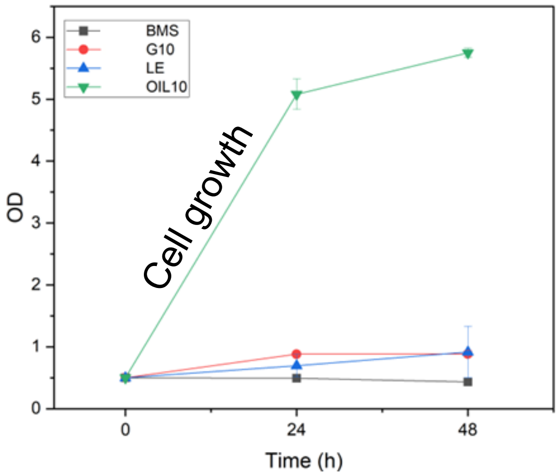
Fermentation

⇩

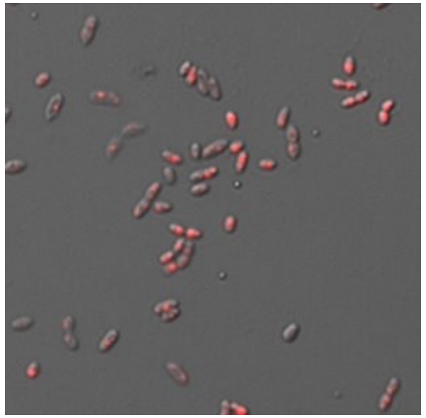
Cell pellets

PHA synthesis using OILs produced from virgin PE and waste PE

- Using OILs as the sole carbon source.
- No potential fermentation inhibitors were observed.
- PHA accumulation in cell biomass.
- PHA characterization showed medium-chain length (MCL) PHAs
- Different PHA extraction strategies are identified.



(LE: Lecithin; G10: Glucose 10 g/L; O10: OIL 10 g/L)



(draft paper data)

⇨

Waste PE-based MCL-PHAs

Hydroxyhexanoic acid (HH, C6)

Hydroxyoctanoic acid (HO, C8)

Hydroxydecanoic acid (HD, C10)

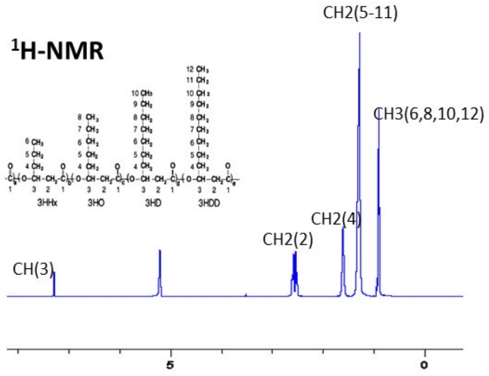
Hydroxydodecanoic acid (HDD, C12)

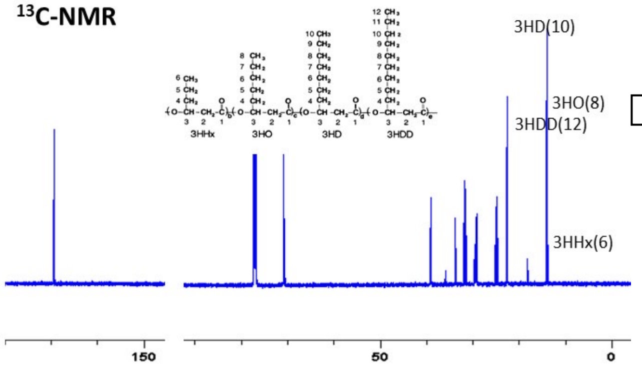
Progress Task 4. SUPF-derived OIL to PHAs

Model compounds	Strain 1	Strain 2
Hydrocarbons (C6, C10, C12)	+	-
Fatty alcohols (C2-C5)	++	+
Palmitic acid	+++	++
Stearic acid	+++	++

- Model compounds to determine fermentability of OIL composition.
- Stains utilizing OIL compositions were identified.
- Spent broth and residues after OIL fermentation were analyzed to identify utilized compositions.
- The residues as a secondary product stream were evaluated. The residue had a much narrower molecular distribution than OIL.

Palmitic acid-based mcl-PHAs





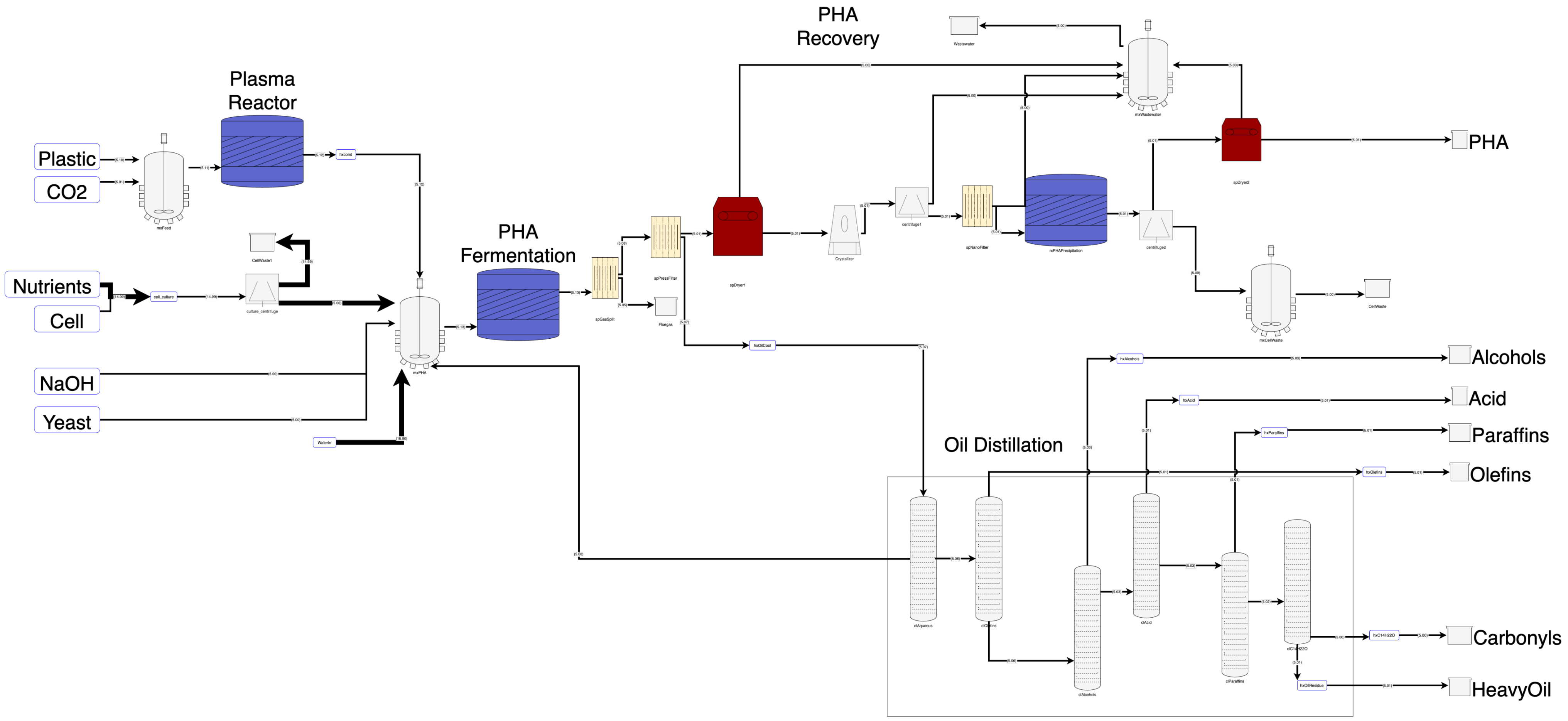
Copolymers composition (assume 95% purity):

HH (%)	HO (%)	HD (%)	HDD (%)
5.80	36.67	36.20	16.44

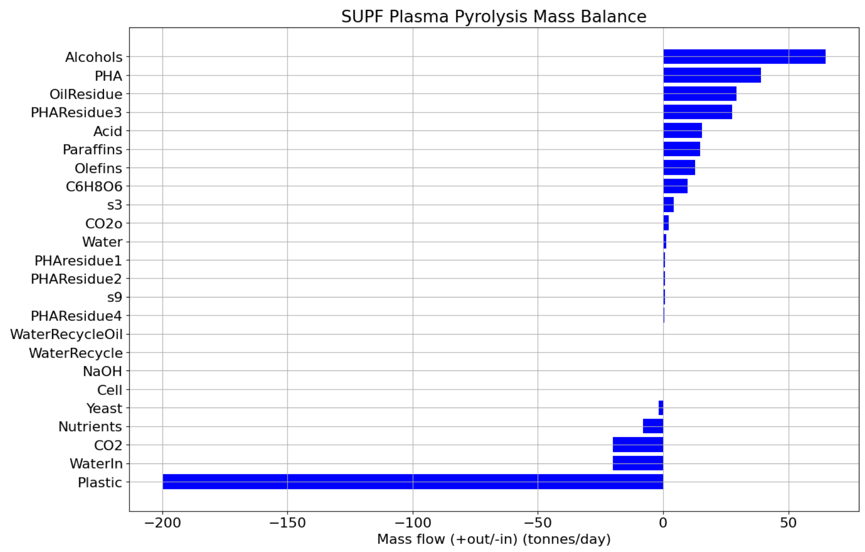
Milestone 4.1.1 (M15) Fermentable component functional groups in OILs for microbial growth and PHA accumulation are identified (**In progress**).

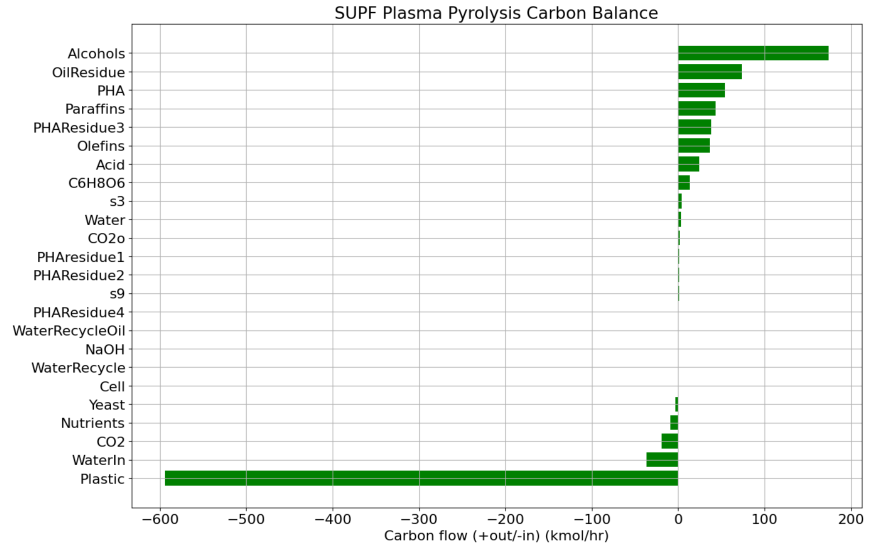
Progress Task 5. TEA & LCA

Develop a process model for the integrated upcycling system.



Progress Task 5. TEA & LCA





- Calculated mass and carbon balance.
- Three product scenarios are considered.

Scenario 1: Only PHAs are sold, and the fermentation residue is either combusted internally or discarded.

Scenario 2: The residue is a lubricant oil-type product.

Scenario 3: The residue compounds are separated and sold individually at market values.

Milestone 5.1.1 (M9) Completed mass and energy balances for a commercial-scale model of plasma-based plastics upcycling to PHA (**completed**).
Milestone 5.1.2 (M18) Established baseline minimum polymer-selling price with >6% lower value than commercial plastics (**in progress**)

Progress Task 6. Diversity, Equity, and Inclusion activities

- 6.1 Promote and develop scholars
- **Milestone 6.1.1** (M9) One underrepresented minority graduate student is recruited (**completed**).
- 6.2 Enhance DEI through institutional diversity program
- **Milestone 6.2.1** (M12) Two underrepresented minority undergraduate students are recruited at ISU (**completed**).
- **Milestone 6.2.2** (M12) One student hired for internship at co-op (**in progress**)
- **Milestone 6.2.2** (M12) One student hired for internship at national laboratory (**in progress**)

Summary

- Waste materials from three MRFs were processed.
- SUPF pretreatment process produced clean and decontaminated polyolefin streams.
- >90% of OILs were produced from HDPE, LDPE, PP, and mixed waste polyolefins.
- Operando characterization was developed for the plasma conversion.
- Mixed polyolefin wastes-derived OIL was fermented to produce middle-chain lengths PHAs.
- Process models were developed to calculate mass and carbon balance.
- DEIP activities incorporated into the project.
- Project milestones are on time.

Quad Chart Overview

Timeline

Start date: 06/01/2022
Ending date: 05/31/2025

	FY22 Costed	Total Award
DOE Funding	$6,122.21	$2,480,106
Project Cost Share *	$1,946.99	$631,000

Project Partners

- University of Missouri
- Sandia National Laboratories
- Quasar Energy Group

Project Goal

Develop a plasma-biological hybrid technology to upcycle mixed SUPF wastes using a circular carbon approach.

End of Project Milestones

- decontaminated SUPFs comprising >95% of PE and PP.
- produce > 90% OIL from the mixed SUPF wastes.
- PHAs from the OIL with >15% conversion efficiency and >85% product recovery.

Funding Mechanism

- DE-FOA-0002473: Single-use plastics recycling (SUPR) funding opportunities/ Topic Area 1: Novel approaches to recycling and upcycle films