

Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment



U.S. DEPARTMENT OF ENERGY

DOE Bioenergy Technologies Office (BETO) 2023 Project Peer Review BOTTLE 5 – **Redesign** & Modeling April 3, 2023

Technology Session Review Area: Plastics Deconstruction and Redesign

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Office of ENERGY EFFICIENCY & RENEWABLE ENERGY

BIOENERGY TECHNOLOGIES OFFICE ADVANCED MATERIALS & MANUFACTURING TECHNOLOGIES OFFICE

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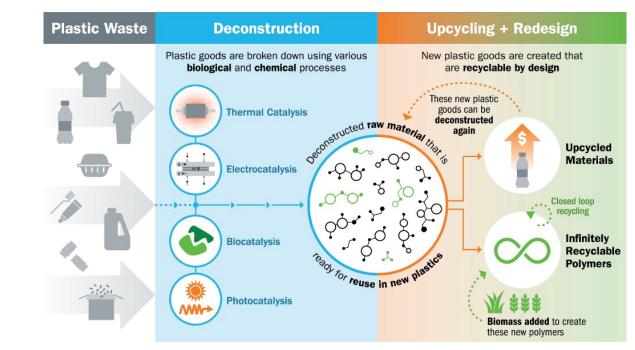
Overview: Redesign in BOTTLE

Goal of Redesign

- To create tomorrow's plastics to be recyclable-by-design (RBD), economically viable, and derived from bio-based sources and today's waste plastics
- Develop energy efficient processes to produce and recycle redesigned materials
- Redesign aligns with DOE's Strategy for Plastics Innovation

Redesign approaches

- RBD acrylic bioplastics with performance & recyclability advantages over incumbent acrylics
- RBD polymers that unify conflicting polymerizability, recyclability, and performance properties
- Biodegradable polyhydroxyalkanoates (PHAs) designed to exhibit properties of all types of polymer classes
- Redesigned PHAs that overcome three challenges facing traditional PHAs: mechanical toughness, melt-processability, and chemical circularity

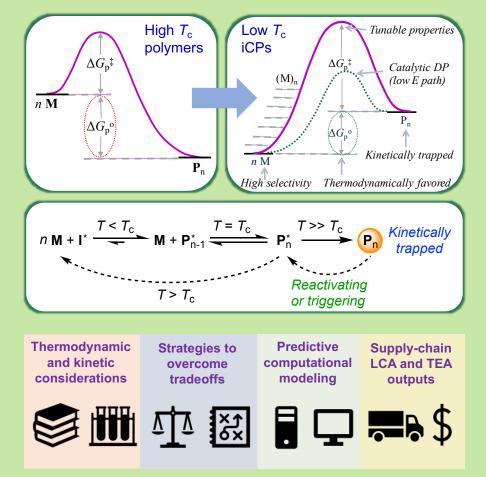




Approach: four design principles for circular polymers



Intrinsically Circular Polymers (iCPs)



Impact:

- Three key challenges for circular polymers Energy Input, Selectivity, and Recyclability/Performance tradeoffs — can be addressed by kinetically trapped, low-ceiling temperature (LCT) RBD polymers
- Kinetically trapped polymers can be designed to exhibit both closed-loop lifecycles and thermal and mechanical properties rivaling/exceeding commodity plastics

Risk:

Unable to produce RBD polymers at commodity prices and achieve ≥50% energy saving relative to today's materials

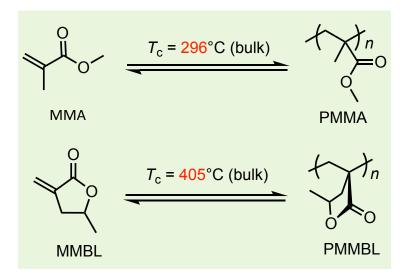
Mitigation:

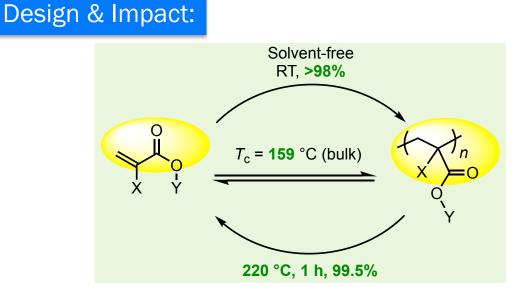
Building blocks for RBD polymers will likely be initially more expensive, but analysis can enable cheaper syntheses and full life cycle assessment of linear versus RBD polymers

Progress: RBD-LCT bio-based acrylic plastics

Current challenges :

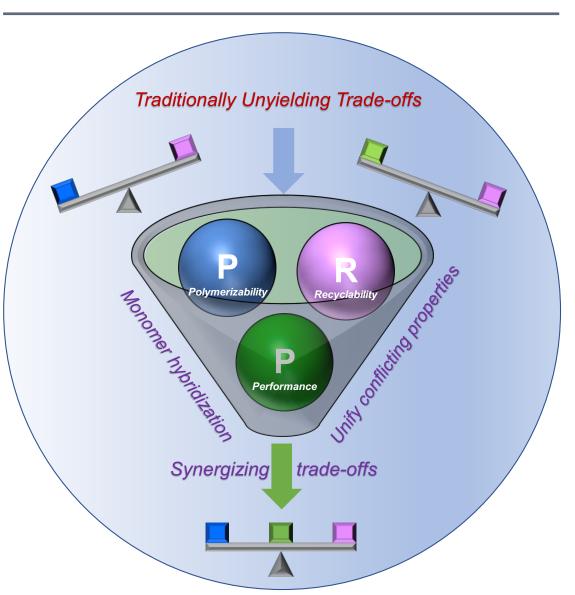
- PMMA and typical bio-based acrylics have high $T_{\rm c}$
- They require high E input (≥400°C) and suffer from competing decomposition reactions
- Can we design bio-based acrylic plastics with full recovery of monomer under mild conditions (<250°C)?





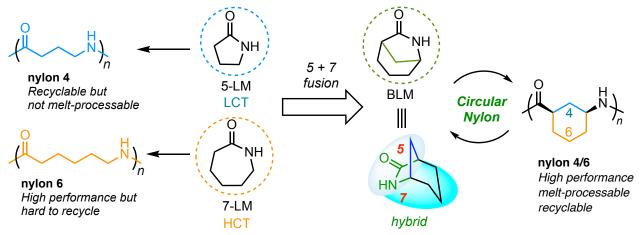
- Energy-efficient polymerization / recycling with quantitative yield, no solvent
- PMVL with M_n up to 100 kDa, $T_g = 184^{\circ}C$ (vs $T_g \sim 105^{\circ}C$ for PMMA)
- Depolymerizable under mild (220°C) conditions (vs. 400°C required for other acrylics)
- TEA showed that PMVL is cost competitive with PMMA, it breaks even with only 15% recycling, and the cost drops to only \$1/kg with ~90% polymer recovery after end use

Hybrid monomer design

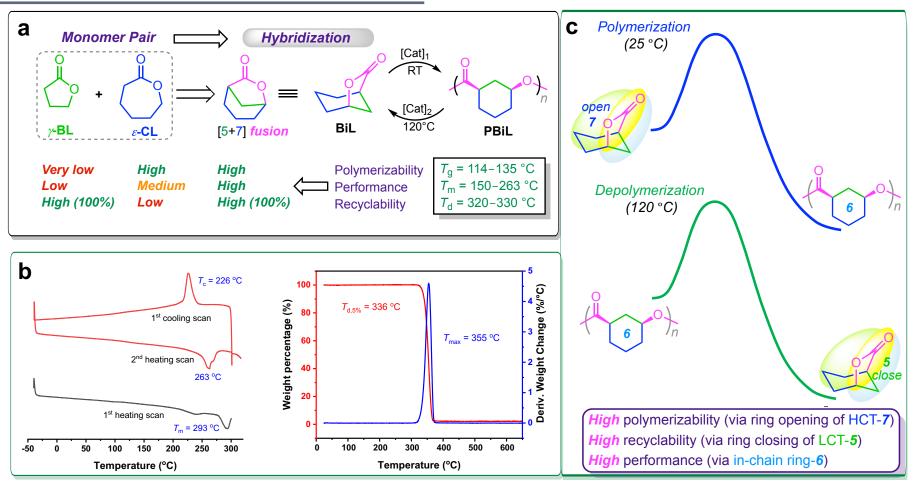


Impact:

- Hybrid monomer design can synergize property trade-offs
- Hybrid monomer design can both unify desired and conflicting properties in a single monomer structure and radically alter performance properties of the resulting circular polymers



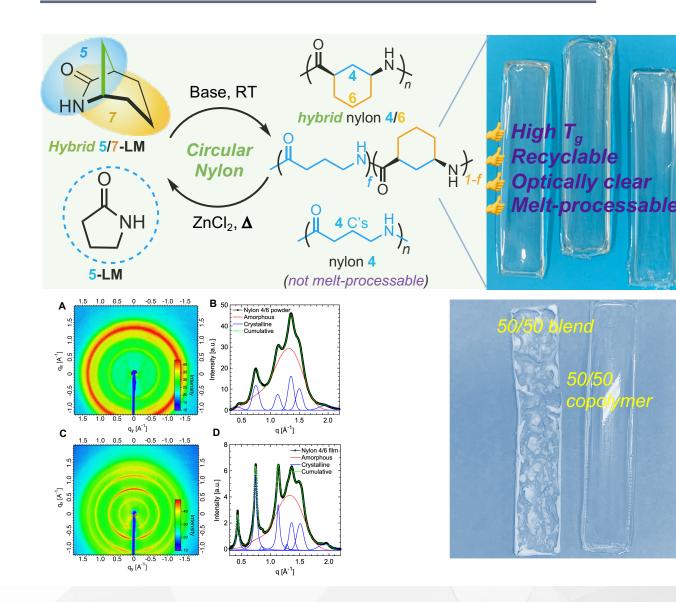
High-performance RBD polyester



- Impact:
- High-performance properties & full recyclability under mild conditions (120°C, bulk)
- Potentially biodegradable and compostable (aliphatic polyester)
- Promising candidate for PET replacement

Optically clear, melt-processable, recyclable hybrid nylon



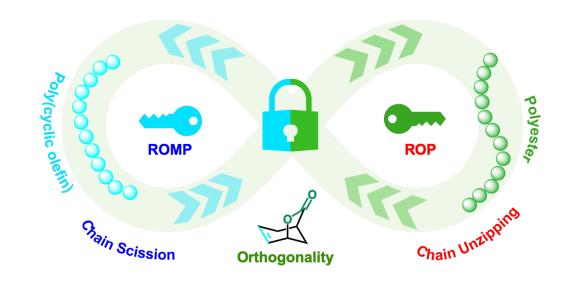


Impact:

- Synergistically combine full recyclability of nylon 4 with high-performance of nylon 6 into hybrid nylon 4/6
- Nylon 4/6 overcomes non-melt-processability of nylon 4 and poor recyclability of nylon 6
- Nanocystalline domains allow for optical clarity

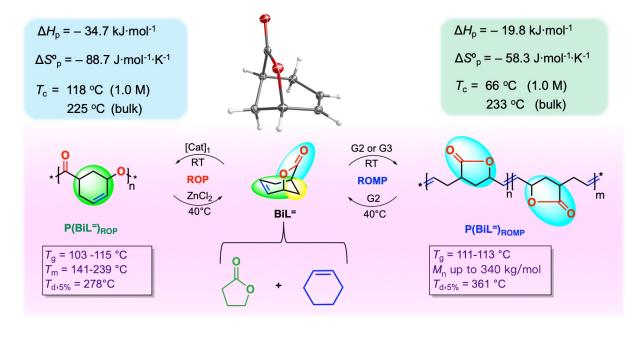
Closing one monomer – two polymer loop

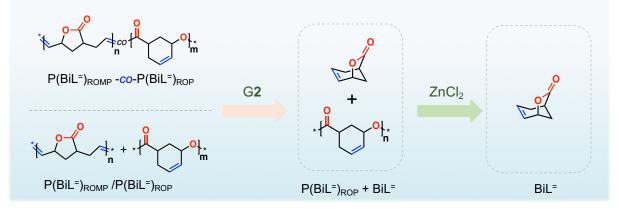




Impact:

- One monomer for two different classes of polymers
- Orthogonal production of polyester and polyolefin
- Recycling of polyester, polyolefin, their copolymer, or physical blend, all leads to the same, single monomer
- High *T*_g, *T*_m, & barrier material (PET replacement)

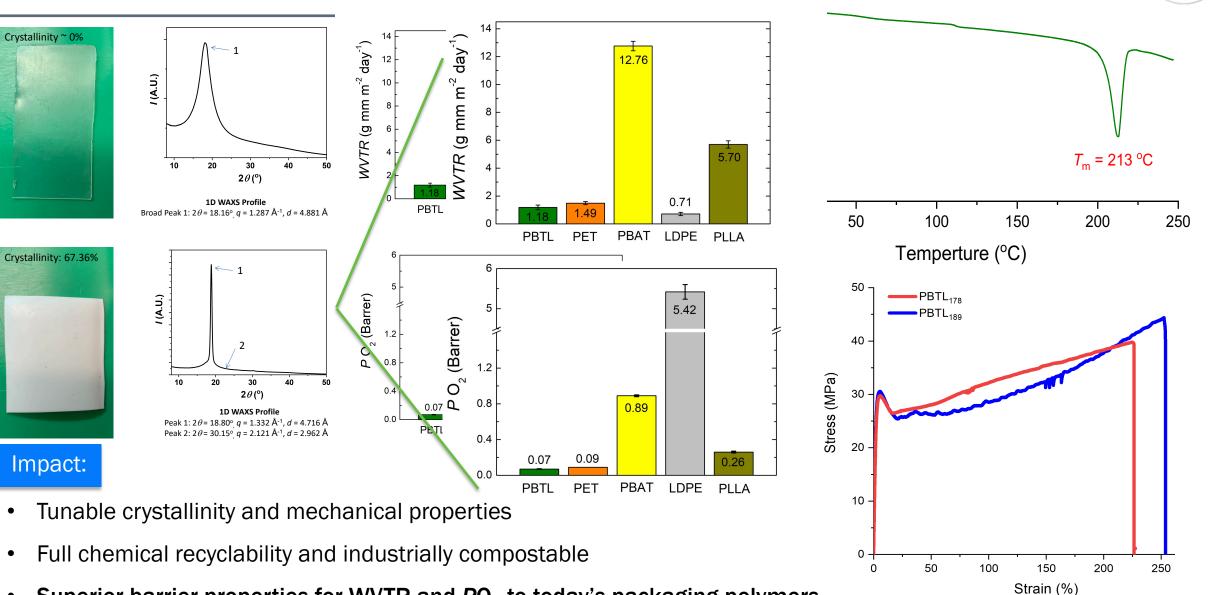




RBD polymer with superior barrier properties

Superior barrier properties for WVTR and PO₂ to today's packaging polymers

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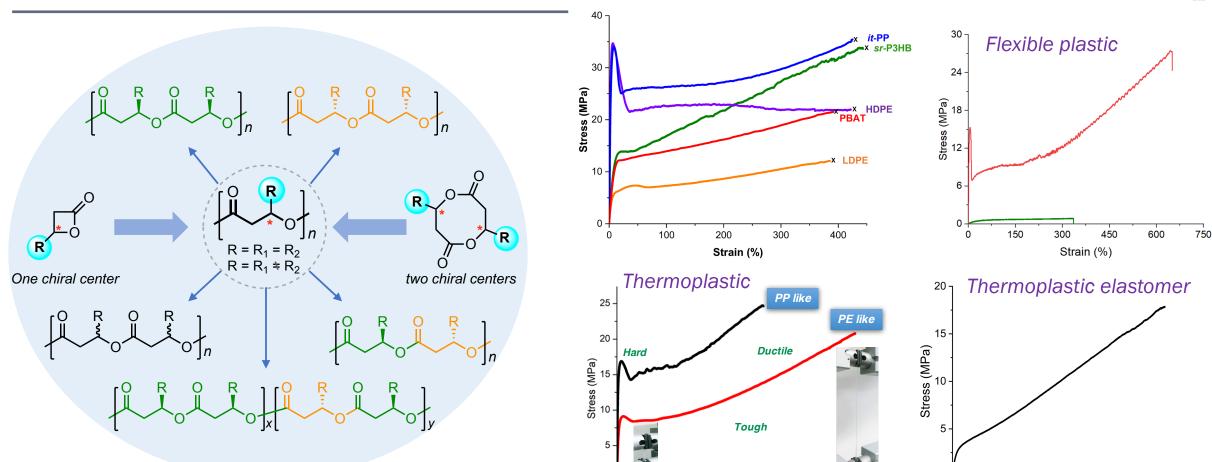


C. Shi et al. in preparation

90

Chemo-catalytic route to designer PHAs with tunable properties





100

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200

300

Strain (%)

400

500

600

- Impact:
- Fast kinetics in synthesis (minutes to hours)
- Scalability and large scope of monomers
- Designer PHAs with properties of all polymer classes
 - Promising alternatives to LDPE, HDPE, PP, SBS

0

100

200

300

Strain (%)

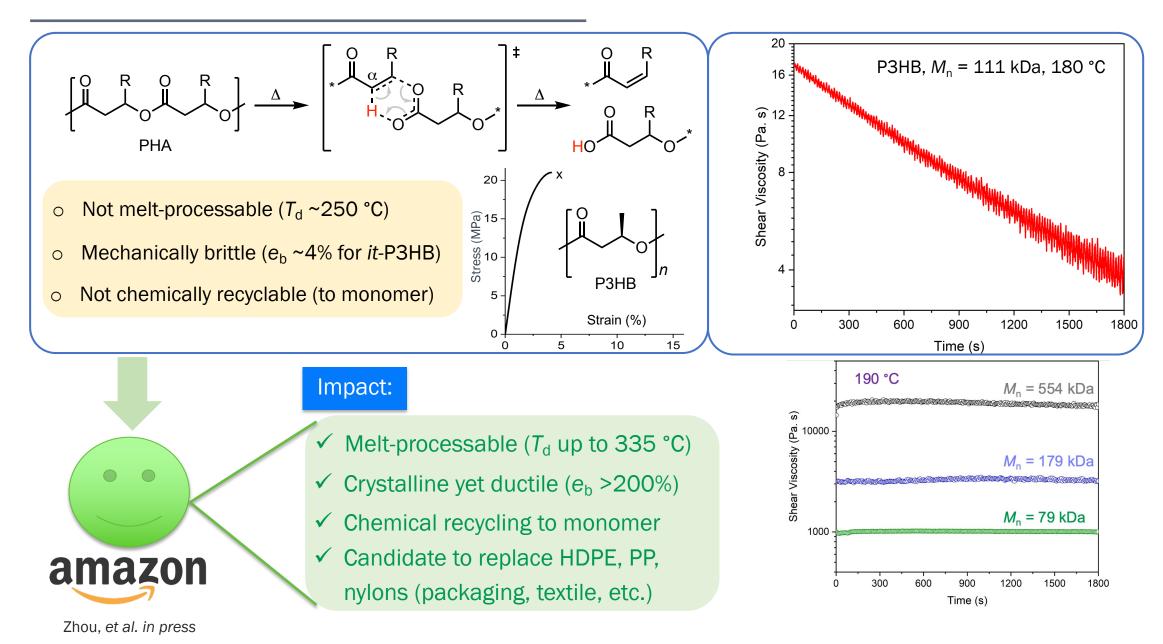
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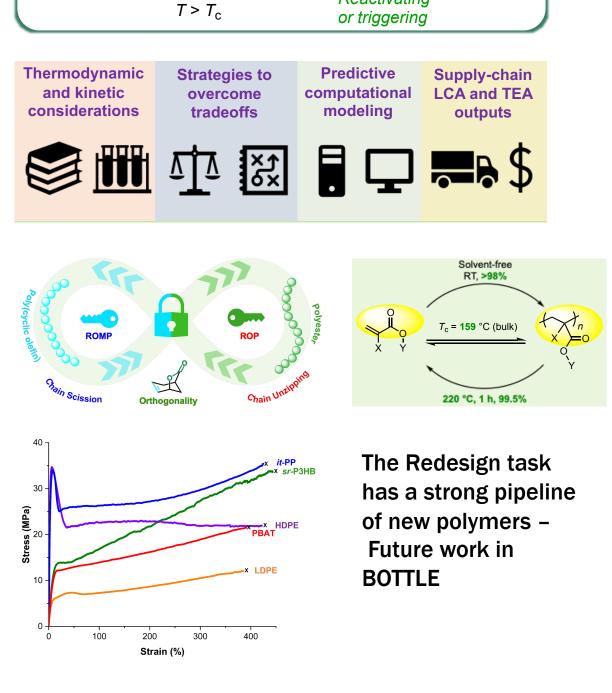
Redesigned PHAs with toughness, melt-processability & circularity





Impact

- 4 design principles to guide circular polymer design
- Bio-acrylic that exhibits performance-advantaged properties and enhanced recyclability over incumbent PMMA
- Hybrid monomer design leads to RBD polymers with closed-loop lifecycles and excellent properties
- Chemo-catalytic PHAs can be designed to exhibit properties of all polymer classes
- Redesigned PHAs overcome the 3 challenges with conventional PHAs: mechanical toughness, meltprocessability, and chemical circularity
- Field-leading studies published in high-visibility outlets and established a comprehensive IP portfolio
- Materials are being used in industrial projects



In preparation

Amy A. Cuthbertson, Clarissa L. Lincoln, Joel Miscall, Lisa Stanley, David T. Moore, Brenna A. Black, Nicholas A. Rorrer, Gregg T. Beckham, Characterization of polymer properties and identification of additives in commercial research plastics, in preparation.

Shaik Afzal, Avantika Singh, Scott R. Nicholson, Taylor Uekert, Eric C.D. Tan, Abhijit Dutta, Robert M. Baldwin, Gregg T. Beckham, Techno-economic analysis of mixed plastic waste gasification for the production of methanol and hydrogen, in preparation for *Energy Env. Sci.*

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Julie E. Rorrer, Amani M. Ebrahim, Ydna Questell-Santiago, Clara Troyano-Valls, Arun S. Asundi, Simon R. Bare, Christopher J. Tassone, Gregg T. Beckham, Yuriy Román-Leshkov, Selective hydrogenolysis of polyethylene and polypropylene waste to liquid hydrocarbons over bifunctional Ru/acid catalysts, in preparation.

Geetanjali Yadav, Avantika Singh, Abhijit Dutta, Scott R. Nicholson, Kylee Harris, Calvin Mukarakate, Joshua A. Schaidle, Cody J. Wrasman, Yuriy Román-Leshkov, Robert M. Baldwin, Gregg T. Beckham, Techno-economic analysis of pyrolysis of mixed plastics waste, in preparation for *Energy Env. Sci.*

Submitted

Houqian Li, Robert D. Allen, Xianglan Bai, Gregg T. Beckham, et al., Expanding plastics recycling technologies: chemical aspects, technology status and challenges, submitted to ACS SusChemEng.

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Richard K. Brizendine, Erika Erickson, Stefan J. Haugen, Kelsey J. Ramirez, Joel Miscall, Davinia Salvachúa, Andrew R. Pickford, Margaret J. Sobkowicz-Kline, John E. McGeehan, Gregg T. Beckham, Effect of particle size and substrate crystallinity on enzymatic depolymerization of poly(ethylene terephthalate), in review at ACS SusChemEng.

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In revision

Anelia Milbrandt, Kamyria Coney, Alex Baggett, Gregg T. Beckham, Quantity, distribution, market value, and energy value of plastic waste in the United States, in revision at Resources, Conservation, and Recycling.

Allison Z. Werner, Rita Clare, Thomas D. Mand, et. al, Tandem chemical deconstruction and biological upcycling of poly(ethylene terephthalate) to b-ketoadipic acid by Pseudomonas putida KT2440, In revision at Metabolic Engineering.

In press

Scott R. Nicholson, Julie E. Rorrer, Avantika Singh, Mikhail O. Konev, Nicholas A. Rorrer, Alberta C. Carpenter, Alan J. Jacobsen, Yuriy Román-Leshkov, Gregg T. Beckham, The critical role of process analysis in chemical recycling and upcycling of waste plastics, in press at Ann. Rev. Chem. Biomolec. Eng.

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Taylor Uekert, Avantika Singh, Jason S. DesVeaux, Tapajyoti Ghosh, Arpit Bhatt, Geetanjali Yadav, Shaik Afzal, Julien Walzberg, Katrina M. Knauer, Scott R. Nicholson, Gregg T. Beckham, and Alberta C. Carpenter, "Technical, economic, and environmental comparison of closed-loop recycling technologies for common plastics," ACS Sustainable Chemistry & Engineering (2023) 11, 3, 965–978.

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Guido Zichittella, Amani M. Ebrahim, Jie Zhu, Anna E. Brenner, Griffin Drake, Gregg T. Beckham, Simon R. Bare, Julie E. Rorrer, and Yuriy Román-Leshkov, Hydrogenolysis of polyethylene and polypropylyne into propane over cobalt-based catalysts, JACS Au. (2022) 2, 10, 2259–2268.

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Julie E. Rorrer, Amani M. Ebrahim, Ydna Questell-Santiago, Jie Zhu, Clara Troyano-Valls, Arun S. Asundi, Anna E. Brenner, Simon R. Bare, Christopher J. Tassone, Gregg T. Beckham, and Yuriy Román-Leshkov, The role of bifunctional Ru/acid catalysts in selective hydrocracking of polyethylene and polypropylene waste to liquid hydrocarbons, *ACS Catal*. (2022) 12, 22, 13969–13979.

Andrea H. Westlie, Eugene Y.-X. Chen, Chris M. Holland, Shannon S. Stahl, Meredith Doyle, Scott R. Trenor, Katrina M. Knauer, Polyolefin innovations towards circularity and sustainable alternatives, *Macromolecular Rapid Communications* (2022) 43, 2200492.

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Andrea H. Westlie, Ethan C. Quinn, Celine R. Parker, Eugene Y.-X. Chen, Synthetic biodegradable polyhydroxyalkanoates (PHAs): recent advances and future challenges, *Prog. Polym. Sci.* (2022), 134, 101608.

Erika Erickson, Japheth E. Gado, Luisana Avilán, et al., Sourcing thermotolerant poly(ethylene terephthalate) hydrolase scaffolds from natural diversity, *Nat Comm.* (2022) 13, 7850.

Robin M. Cywar, Nicholas A. Rorrer, Heather B. Mayes, Anjani K. Maurya, Christopher J. Tassone, Gregg T. Beckham, Eugene Y.-X. Chen, Redesigned hybrid nylons with optical clarity and chemical recyclability, *J. Am. Chem. Soc.* (2022) 144, 5366–5376.

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Coralie Jehanno, Jill W. Alty, Martijin, Steven De Meester, Andrew P. Dove, Eugene Y.-X Chen, Frank A. Leibfarth, Haritz Sardon, Critical advances and future opportunities in upcycling commodity polymers, *Nature* (2022), 603, 803–814.

William M. Kincannon, Michael Zahn, Rita Clare, Jessica Lusty Beech, Ari Romberg, James Larson, Brian Bothner, Gregg T. Beckham, John E. McGeehan, and Jennifer L. DuBois, Biochemical and structural characterization of an aromatic ring-hydroxylating dioxygenase for terephthalic acid catabolism. *Proc. Natl. Acad. Sci.* (2022) 19(13):e2121426119.

Jessica Lusty-Beech, Rita Clare, William M. Kincannon, Erika Erickson, John E. McGeehan, Gregg T. Beckham, Jennifer L. DuBois, A flexible kinetic assay efficiently sorts prospective biocatalysts for PET plastic subunit hydrolysis. *RSC Adv.* (2022) 12, 8119-8130.

Changxia Shi, Ryan W. Clarke, Michael L. McGraw, Eugene Y.-X., Closing 'one monomer-two polymers-one monomer' loop via orthogonal (de)polymerization of a lactone/olefin hybrid, J. Am. Chem. Soc. (2022) 144, 2264–2275.

Erika Erickson, Thomas Shakespeare, Felecia Bratti, Bonnie L. Buss, Rosie Graham, McKenzie Hawkins, Gerhard König, William E. Michener, Joel Miscall, Kelsey J. Ramirez, Nicholas A. Rorrer, Michael Zahn, Andrew R. Pickford, John E. McGeehan, Gregg T. Beckham, "Comparative PETase performance as a function of reaction conditions, substrate properties, and product accumulation," *ChemSusChem* (2022) 15, e202101932.

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Changxia Shi, Liam T. Reilly, Vi Sai Phani Kumar, Matthew W. Coile, Scott R. Nicholson, Linda J. Broadbelt, Gregg T. Beckham, Eugene Y.-X Chen, Design principles for intrinsically circular polymers with tunable properties, Chem (2021), 7, 2896–2912.

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Elani Lacovidou, Richard Geyer, Julia Kalow, James Palardy, Jennifer Dunn, Timothy Hoellein, Eugene Y.-X. Chen, Toward a circular economy for plastics, One Earth (2021), 4, 591–594 (Featured as Voices article).

Lucas D. Ellis, Nicholas A. Rorrer, Kevin P. Sullivan, et al. Chemical and biological catalysis for plastics recycling and upcycling. Nature Catal. (2021) 4, 539–556.

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Changxia Shi, Zi-Chen Li, Lucia Caporaso, Luigi Cavallo, Laura Falivene, Eugene Y.-X., Hybrid monomer design for unifying conflicting polymerizability, recyclability, and performance properties, Chem (2021), 7, 670–685.

Patents and patent applications

- Synergistic dual cure for rapid manufacturing of thermoset material, 22-71: U.S. provisional patent application 63/414,238
- Base-mediated method for the recycling of epoxy resin-carbon fiber composites, 22-130: U.S. provisional patent application 63/418,874
- Renewable bio-advantaged plasticizer generated by reductive cross coupling of lignin-derived aromatics, 22-124: U.S. provisional patent application 63/379,217
- Process for sequential acetolysis-autoxidation of plastic streams, 22-107: U.S. provisional patent application 63/383,293
- Methods and systems for dye removal from polymer textiles, 22-106: U.S. provisional patent application 63/384,137
- Biodegradable elastomeric thermosets from microbially-produced polyhydroxyalkanoates, 19-104: U.S. provisional patent application 63/386,011
- Light-driven C-C bond cleavage enabled by polyoxometalate photocatalysts, 21-95: U.S. provisional patent application forthcoming

- Hydrogenolysis of Polyethylene and Polypropylene into Propane over Cobalt-Based Catalysts, 22-81: U.S. provisional patent application 63/340,322
- Catalysts for Depolymerizing Plastics, 20-22: 17/370,244
- Plastic Degrading Fusion Proteins and Methods of Using the Same, 20-86: PCT/US21/31610
- Polymer Degrading Enzymes, 21-88: PCT/US22/25624.
- Dissolution Purification and Recovery for Polymeric Recycling, 22-16: 63/307,676
- Method to Produce Branched-Chain Polyhydroxyalkanoates and Branched Chain 3-Hydroxyacids from Glucose, 21-63A: 63/321,207.
- Upcycling Mixed Waste Plastic Through Chemical Depolymerization and Biological Funneling, 20-123: PCT/US21/63725.
- Genetically engineered Pseudomonas strains capable of metabolizing ethylene glycol, 17-26: 11,021,721
- Engineered Pseudomonas for the Deconstruction of Polymers, 18-76: 17/055,626
- Microorganisms Engineered for Muconate Production, 20-48: 17/184,580

Patents and patent applications

- Polymers from bio-derived dicarboxylic acids (BKA to nylon), 17-48: 10,662,289
- Polymers and methods of making the same (PET formulated with adipic/muconic acids), 17-55A: 17/205,232
- Monomers, Polymers and Methods of Making the Same (Bio-plastic ABS), 18-69: 16/583,471
- Bio-derived biphenyl compounds (Polycarbonates), 18-81: 16/791,873
- Bioderived monomers as replacements in petroleum-based polymers and copolymers (novel bio-based plasticizers), 19-38: 16/790,093
- Conversion of dicarboxylic acids to monomers and plasticizers, 19-41A: 16/995,338
- Bio-derived Epoxide Triazine Networks and Methods of Making the Same, 20-26: 17/324,222
- Bio-derived Epoxy-Anhydride Thermoset Polymers for Wind Turbine Blades and Anti-Static Coatings, 20-59: 17/494,514
- Plastic waste derived polymers and resins and methods of making the same (PET upcycled to 3D printing materials). 20-37: 17/371,421
- Mixed Waste Plastics Compatibilizers for Asphalt (filed by ASU), 21-53: 63/148,423

Patents and patent applications

- Bioderived Benzoxazines, 20-130: 17/690,131
- Novel Routes to Bis-furan Diacids, Dialcohols and Diamines, US 9840485
- Improved Industrial Production of Isotactic Polylactides (PLA), US 10174161
- Chemically Recyclable Polymers to Combat Single-Use Plastics, PCT Patent Pending: WO 2021/113325
- Synthesis of Crystalline Polymers from Cyclic Diolides, US Utility Patent Pending: US 2019/0211144
- Novel Compounds and Methods for Upgrading Biomass to Produce Premium Biofuels, US Utility Patent: US 9469626 B2, US Utility Patent: US 9828354 B2
- High-Speed, Stereoselective Polymerization for Renewable Bio-derived Plastics, US Utility Patent: US 9309332

Selective Hydrogenolysis of Polyolefin Waste to Liquid Hydrocarbons over Bifuncational Ru/Acid Catalysts, AIChE National Conference, November 15, 2022.

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Development of non-model microbes as chassis organisms for bioconversion. Presented at the AIChE Annual Meeting, November 2022.

Redesigning Polymers to Leverage a Circular Economy, Chemical Engineering, Purdue University, November 2022.

Bio-based Polymers with Performance & Recyclability Advantages, Braskem, virtual seminar, November 2022.

Design Principles and Chemocatalytic Methods for Circular Polymers and Biodegradable Plastics, BASF Lecture in Organic Chemistry, November 2022.

Developments in Advanced Recycling, TA Instruments Webinar, October 2022.

Design of Polyolefin-like Polyesters with Closed-loop Lifecycles, ACS WRM Polymer Symposium, October 2022.

Adopting a sustainable plastics supply chain, RISE 2022, September 2022.

Redesigning plastics to be recyclable-by-design, RISE 2022, September 2022.

Advances in lignin and plastics conversion, VITO, September 2022.

Decoding the mechanism of autoxidation deconstruction reaction of plastics by in-situ simultaneous SAXS and WAXS," XVIII International Small-Angle Scattering Conference (SAS2022), September 2022.

Design of functionalized polyolefins and polyolefin-like polyesters with close-loop chemical recycling, ACS Advances in Polyolefins, September 2022.

Using synthetic biology to solve challenges in plastic waste and renewable chemical production, Biological Sciences Departmental Seminar, September 2022.

Advancing the catalytic upcycling of waste polyolefin plastics, Beckman Foundation Regional Symposium, August 2022.

Using redesigned iron catalysts to bring aromatic subunits to a common intermediate, SIMB 2022, August 2022

Techno-economic analysis and life cycle assessment for catalytic fast pyrolysis of mixed plastic waste, BioEnergy TRP Meeting, National Renewable Energy Laboratory, August 2022.

Bio-based, recyclable-by-design polymers, ACS National Meeting, August 2022

Techno-Economic analysis and life cycle assessment of mixed waste plastics via pyrolysis and gasification, ACS Fall Conference, August 2022.

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Bio-based acrylic plastics with performance and recyclability advantages, ACS Symposium: Green Polymer Chemistry and Sustainability, ACS National Meeting, August 2022.

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Design principles and chemocatalytic methods for intrinsically circular polymers and biodegradable plastics, ACS Presidential Event: Series-Enabling Circular Economy via Polymer Molecular Recycling, ACS National Meeting, August 2022.

Techno-economic, life-cycle, and socioeconomic impact analysis of enzymatic recycling of poly(ethylene terephthalate), ACS Fall Conference, August 2022.

Kinetic Monte Carlo-based tool to unravel solvolysis chemistry of step-growth polymers, National Meeting of the American Chemical Society, August 2022.

Tracking in situ structural changes in Ru, Mo and Co-based hydrogenolysis catalysts for polyolefin deconstruction under mild temperature using in situ/operando X-ray absorption spectroscopy, ACS Fall Meeting: Polymer Upcycling Symposium, August 2022.

High throughput test tools for industrially relevant microbial chassis, SIMB 2022, August 2022.

Circular polymers and biodegradable plastics, Circular Polymers and Biodegradable Plastics International Research Training Group, University of Muenster, July 2022.

Engineering P450s to alleviate a bottleneck to lignin demethylation, Intl. Conference on Porphyrins and Phthalocyanines, July 2022.

Difficult to recycle plastics, Sustainable Packaging Coalition Engage Meeting, July 2022.

Selective chemical recycling of mixed plastics waste, Polymer Physics Gordon Research Conference, July 2022.

Plastics recycling and upcycling in the BOTTLE Consortium, NASEM Committee on Repurposing Plastic Waste, July 2022.

Developing strategies for polymer redesign and recycling using reaction pathway analysis, Gordon Research Conference on Polymer Physics, July 2022.

Multi-Material Flexible Packaging Coalition SPC, February 2022.

Development of chemical recycling approaches for plastic waste (via webinar), BASF, March 18th, 2022

Development of chemical recycling approaches for plastic waste, Enzyclic Consortium (via webinar), January 2022

Development of chemical recycling approaches for plastic waste, UIUC, December 2021

Design Principles and Synthetic Methodologies for Circular Polymers with Intrinsic Recyclability and Tunable Properties, Pacifichem Conference, December 2021

New building blocks for performance-advantaged renewable and recyclable polymers, Pacifichem (via webinar), December 2021

Discovery and characterization of PET degrading enzymes, University of Rochester microplastics workgroup seminar series, December 2021.

Design Principles and Synthetic Methodologies for Intrinsically Circular Polymers and Biodegradable Plastics, Columbia University, November 2021

Selective Hydrogenolysis of Polyethylene and Polypropylene to Liquid Alkanes over Tunable Ruthenium-Based Heterogeneous Catalysts, 2021 AIChE National Conference, Boston, MA, November 2021.

Plastics recycling and upcycling, ACS Converge (via webinar), October 2021

Genetic tools and microbial engineering for biological production of sustainable fuels and chemicals, Presented to Weekly Seminar for DOE CCI/SULI Students. October 2021

Heterogeneous Catalytic Deconstruction and Upcycling of Waste Polyolefins, Biodesign Institute at Arizona State University, SM3 Seminar Series, October 2021.

Domestication of diverse non-model microbes for plastics upcycling and sustainable fuel and chemical production, Biological Sciences Departmental Seminar, Michigan Technical University. October 2021.

Catalysis for valorization of lignin and plastics, Great Plains Catalysis Society (via webinar), June 2021

The critical role of economic and environmental analysis to guide research in lignin valorization and plastics upcycling, Keynote Invited Lecture, ACS Green Chemistry and Engineering (via webinar), June 2021

Towards Intrinsically Circular Thermoplastics and Reprocessable Thermosets, Dow Chemical Company, virtual seminar, May 2021

Recent progress in performance-advantaged bioproducts and plastics upcycling, Arizona State University (via webinar), April 2021

Recent adventures in biomass conversion and plastics upcycling, Rutgers University (via webinar), April 2021

Recent adventures in biological plastics upcycling, MIX-UP Consortium (via webinar), April 2021

Framing challenges and opportunities for chemical recycling of waste plastics, ACS Presidential Symposium on Chemistry and the Future of Plastics (via webinar), April 2021

Recent updates in plastics upcycling from the BOTTLE Consortium, ExxonMobil Research and Engineering, April 2021

Design Principles and Synthetic Methodologies for Circular Polymers and Biodegradable Plastics, KAUST, Physical Science and Engineering Division, virtual seminar, April 2021

Heme and non heme iron enzymes and renewable carbon, University of San Antonio Texas, April 2021

A flexible kinetic assay efficiently sorts potential biocatalysts for BHET hydrolysis, Symposium on Biomaterials, Fuels, and Chemicals, April 2021

BETO 2021 Peer Review, virtual, March 2021

Design Principles for Circular Plastics with Tunable Properties, CellPress LabLinks: The Circular Plastics Economy: Linking Across Scales, virtual event with 440 registered attendees. March 2021.

Process analysis for enzymatic PET recycling, Global Research and Innovation on Plastics annual meeting (via webinar), March 2021

Polyolefin upcycling in the BOTTLE Consortium, Annual SPE meeting (via webinar), February 2021

Biological processes for lignin and plastics conversion, University of California Riverside (via webinar), January 2021