Institute for Cooperative Upcycling of Plastics (iCOUP)
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Lead Institution: Ames Laboratory
Class: 2021 – 2024

Mission Statement*: To uncover macromolecular and catalytic phenomena at the interface of molecular-scale chemistry and mesoscale materials science to enable upcycling of energy-rich plastics.*

The iCOUP research team is investigating the catalytic conversions of hydrocarbon polymers into more valuable chemicals and materials. Plastics are essential in the global economy, as reflected by production of new polymers in 2019 surpassing 400 million tons that consumed the equivalent of 6–8% of the crude oil and natural gas produced worldwide. Almost half of the currently manufactured plastics are polyolefins (POs), including polyethylene (PE), polypropylene (PP) and polystyrene (PS), 80% of which are single-use products discarded into overflowing landfills, contributing to a global waste catastrophe with widespread environmental, economic, and health-related consequences. Polyolefin upcycling requires the ability to break inert bonds in long chains of chemically indistinguishable repeat units at regular spatial intervals, thereby converting waste into targeted, narrow distributions of molecules and materials with desirable properties and added value.

Inspired by nature's approaches to biopolymer deconstruction, we are creating abiotic multifunctional materials that target and cleave specific bonds in macromolecules through three mechanistic motifs shown in **Figure 1**. The first strategy will achieve selective cleavage of carbon-carbon bonds in POs via molecular-scale mechanisms (**Figure 1A**), while those in **Figure 1B,C** will emphasize processive and polymer-site directed mechanisms akin to those in enzyme-catalyzed conversions. The iCOUP team will utilize their best features to create new upcycling methods which enable precise cleavages in polymer chains.

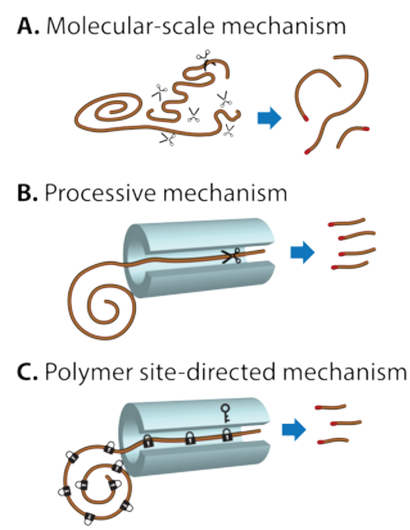

Figure 1. Three mechanistic motifs for polymer upcycling.

We will study how mesoscale architectures adapt conversions of small molecules, through these mechanistic motifs, to be useful for the upcycling of macromolecules. This scientific challenge will be addressed through studies focused upon catalytic sites, architectures, and polymers during deconstruction reactions in the following Objectives:

i. Discover new methods to transform intractable plastics into upcycled intermediates by breaking and functionalizing C–C and C–H bonds with molecular-scale selectivity;
ii. Design processive approaches to deconstruct polymers, leveraging molecular and macromolecular phenomena, to produce uniform, higher-value small molecules; and
iii. Construct next-generation POs containing sequences that facilitate end-of-life conversion.

The current state-of-the-art syntheses of heterogeneous catalysts revolve around the immediate molecular scale environment of reactive binding sites through generation of single sites, uniform nanoparticles, or ordered materials. The next and more daunting challenge is to create effective and selective catalytic architectures, operating at multiple length scales. The design and assembly of such multifunctional catalytic systems that favor selective upcycling pathways relevant to specific polymers will require advances in synthesis, theory, and spectroscopy with the following Objectives:

iv. Investigate synthetic and analytical methods for constructing and characterizing hierarchically-structured catalysts with spatially organized functional groups;
v. Develop population balance and microkinetic models to relate experimental signatures of polymer deconstruction with macromolecular upcycling mechanisms; and
vi. Predict how molecular-scale interactions and polymer conformational entropy govern the adsorption and mobility of polymers on surfaces (shown in Figure 2) and in mesoscale pores of catalysts.

By establishing the fundamental macromolecular phenomena germane to upcycling, our interdisciplinary team will create robust, selective inorganic catalysts and next-generation polymers that can be purposefully deconstructed and transformed into valuable, upcycled products. In a broader perspective, iCOUP's scientific advances create opportunities to depart from the current make-then-discard approach toward plastics and achieve a truly circular economy for these energy-rich resources.

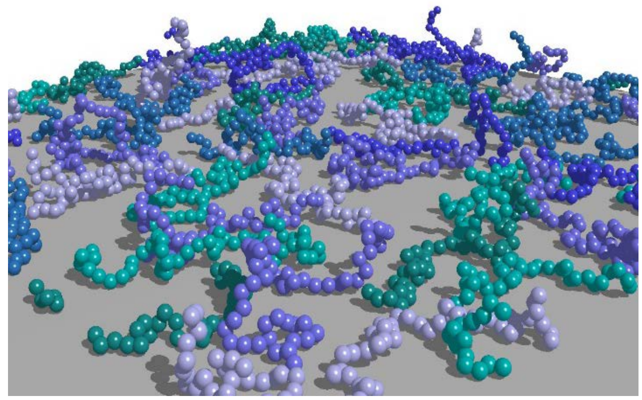

Figure 2. Typical conformation of polymers at a uniformly attractive surface, below the adsorption transition temperature.

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