Institute for Cooperative Upcycling of Plastics (iCOUP)
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Mission Statement*: To establish the molecular and macromolecular scientific principles governing deconstruction and reconstruction of polymers that enable sustainable manufacturing from energy-rich plastics.*

The iCOUP team is investigating the selective catalytic conversions of hydrocarbon polymers into valuable chemicals and new recyclable materials to establish new transformative manufacturing science principles that enable a circular economy for plastics. Plastics are essential in the global economy, as reflected by production of new polymers surpassing 400 million tons in 2023, for applications that touch all aspects of modern life. Many of the advantages of plastics are diminished by environmental impacts of their initial manufacture and end-of-life. Manufacturing of plastics consumed the equivalent of 6–8% of the crude oil and natural gas produced worldwide. Approximately 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. The 'upcycling' conversion of used polyolefins into new chemicals with defined end-of-life, including new feedstocks for chemically recyclable polymers, could avoid the negative impacts of the conventional plastic economy. 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.

In general, selective conversions of macromolecules require catalytic sites to react with polymers on length and time scales suited to molecules and they require that the architecture supporting the catalytic site also influences the polymer chains' behavior on macromolecular length and time scales. Nature's approaches to selective biopolymer conversions, including DNA synthesis and repair as well as cellulose deconstruction, achieve precision using enzymes that operate under the constraints of these multiscale phenomena. Inspired by nature, we are creating catalytic materials and chemically recyclable polymers that address molecular and macromolecular length scales to enable circular transformative manufacturing. Our approaches involve processive mechanisms in which the catalyst creates a template to influence the conformations of adsorbed polymer chains (**Fig 1A-B**) to break polymer backbone linkages or install new reactive groups at specific positions. We will use these functionalized species, both mesomonomers and macromolecules, to create chemically recyclable polymers with high-quality mechanical properties (**Fig 1C**).

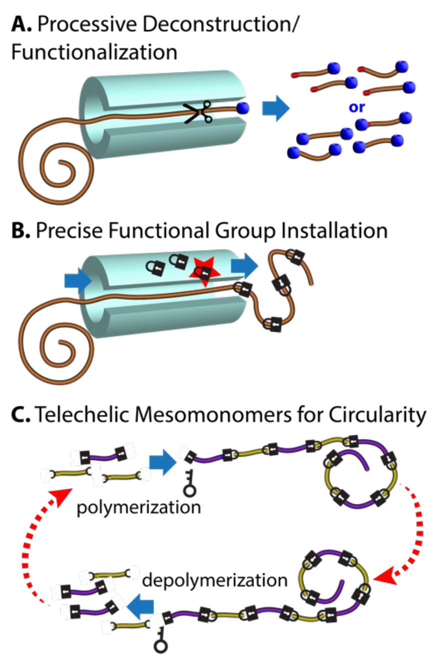

Fig. 1. Three approaches for selective deconstruction or manufacture of plastic materials.

We will study how materials with mesoscale architectures govern conversions of long polymer chains, while adapting catalytic principles from molecular chemistry to access desired functionality, through the following Objectives:

(1) Invent catalytic reactions for breaking and functionalizing C–C and C–H bonds to transform polymers into upcycled mesomolecular intermediates.
(2) Install catalytic active sites for C–C functionalization within hierarchical support architectures to produce uniformly functionalized, value-added chemicals from discarded polymers.

The iCOUP team will adapt these new transformations for the synthesis of telechelic mesomonomers and recyclable polyolefin-like materials, while establishing the new chemical and materials science principles that provide recyclable plastic materials with desirable mechanical properties.

(3) Synthesize telechelic mesomonomers from discarded plastics, leveraging length-selective cleavage and functionalization to install reactive groups at both chain ends.
(4) Reassemble these telechelic mesomonomers into recyclable-by-design polymers and establish their structure-thermomechanical property relationships.

These transformations, as well as the mechanical properties of the resulting plastics, are driven by intermolecular interactions and size-dependent dynamics of populations of hydrocarbon polymer chains (**Fig 2**). We will study these phenomena to establish the mechanisms that control polymer upcycling reactions and re-polymerizations.

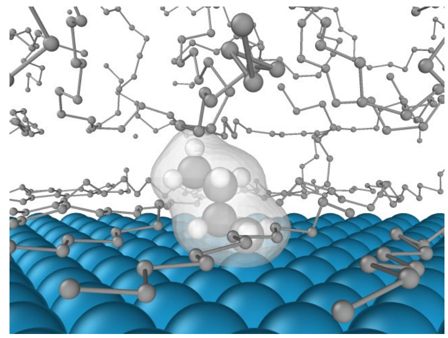
Figure 2. Model of long and short chains reacting on metal surfaces.

(5) Design unique mechanistic motifs that drive selectivity across length and time scales.
(6) Measure polymer-surface interactions and dynamics to provide quantitative multiscale thermochemical properties and kinetics underpinning polymer conversions.

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, repolymerized, 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.

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