DEVELOPING STRATEGIES FOR POLYMER REDESIGN AND RECYCLING USING REACTION PATHWAY ANALYSIS

Linda J. Broadbelt

Department of Chemical and Biological Engineering
Northwestern University
broadbelt@northwestern.edu

The current lack of sustainability of and the limited portfolio of recycling processes for synthetic polymers have posed serious threats to the environment. Approximately 90% of plastics are produced via fossil fuels, and over 150 million tonnes of plastics have been discarded in the ocean. Annual production of plastics is expected to reach over 1 billion tons in 2050, but the current manufacturing, consumption, and disposal schemes of fossil-based polymers follow an unsustainable framework. Using reaction pathway analysis, we are pursuing a portfolio of strategies for redesign and recycling of polymers for sustainability.

Pyrolysis is a promising method for resource recovery from plastic waste that is compatible with current petrochemical infrastructure that thermally converts polymers in the absence of oxygen into valuable chemical feedstocks and monomer. To provide further insight into polymer pyrolysis, a greater understanding of the mechanistic and kinetic details of the underlying reaction network is needed. To handle the complexity of mechanistic modeling of polymer degradation, we have developed both continuum and kinetic Monte Carlo (kMC) models. We have applied these approaches to study the degradation of polystyrene, polypropylene, polyisoprene, polystyrene peroxide and binary mixtures, and we are able to capture diverse experimental measures, including yields of individual low molecular weight products, as a function of reaction conditions. In order to solve the large models that are created, values of the rate coefficients for $O(10^5)$ reactions are typically required. The approach that we have developed to specify rate coefficients is hierarchical, based on a combination of literature values, estimation methods, and computational chemistry.

Alternatively, redesign efforts focusing on polymers that can be reused and recycled to monomers can lead to sustainable solutions for the plastics waste problem. One pathway to success is to identify bioprivileged molecules, biology-derived chemical intermediates that can be efficiently converted to a diversity of chemical products, including both novel molecules and drop-in replacements, and molecules emanating from them that can be used as monomers leading to recyclable polymers. We have developed a framework for molecule discovery and reaction pathway design that is automated and flexible and can be used to screen for bioprivileged candidates and target molecules. The application to discovery of known and novel monomers for poly(hydroxyurethanes) that are derived from biobased molecules and lead to recyclable materials will be discussed, and computational methods to evaluate the recyclability of different polymers will be outlined.