ResIn: Responsible Innovation for Highly Recyclable Plastics

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Industrial Partners: Dow Chemical Company, Michelin, Fenner Precision

Objectives: We propose to develop a responsible innovation approach that marries computational-based approaches with experiments to design materials that achieve high polymer recyclability and benign degradation products at end-of-life (EOL). We will target the polyurethane (PU) family of polymers, which ranks sixth in worldwide production at 36 billion pounds produced in 2016 and a 6% annual growth rate. PU is not recycled at any significant level when made of linear chains (thermoplastics) or crosslinked networks (thermosets). SOA is 0% recovered monomers for PU.

The NU-ANL team, in collaboration with its industrial partners, will focus its efforts on responsible recycling of biobased PU-like materials, namely biobased polyhydroxyurethanes (PHUs) and polythiourethanes (PTUs), that offer the possibility of recovering value and improving sustainability in two ways: (a) recovery of monomer from spent materials, whether thermoplastics or thermosets, and (b) reprocessability of spent networks with full recovery of crosslink density and associated properties after reprocessing. Our approach is designed to meet the monomer recovery challenge as well as put thermosets on par with thermoplastics, where melt-state reprocessing of spent polymer into recycled high-value products that meet original use guidelines is the most energy efficient and responsible method of recycling.

Approach: The overall design framework that is proposed begins with biomass-derived intermediates as starting molecules, to which a computational framework for reaction pathway design will be applied to generate potential monomers. These monomers will be used to synthesize PHU and PTU that can be reversibly thermally reprocessed (chemical recycling) for their original use. Experimental studies and kinetic Monte Carlo (kMC) simulations will be used to design conditions for optimal chemical recycling and assess extent of degradation of properties upon repeated chemical recycling. Experimental design and kMC simulations of monomer recovery for PHU and PTU will also be done, exploring conditions for high monomer yield. Environmental and economic analyses will be conducted in concert with these studies. Risk assessment for environmental performance will be used to understand the potential impacts of disposal pathways of the plastics and exposure analysis of the polymers and their monomers. Testing for EOL properties will involve both engineered and natural environments. Finally, life cycle analysis (LCA) and technoeconomic analysis (TEA) will be done to assess alternatives, explore economic feasibility, and evaluate sustainability.

Impact: The project will have impact on multiple fronts. We will achieve chemical recyclability of 25% monomer recovery from PHUs and PTUs. We will develop modeling and analysis tools to guide the production and recycling of PHUs and PTUs that are at least 50% biomass-derived, benign at EOL, are cost effective, and are less energy-, greenhouse-gas, and water-intensive than baseline PUs. An additional focus is to develop PHUs and PTUs that exhibit equivalent or improved properties compared to conventional PUs. Further outcomes of the project are publicly available tools for PHU and PTU design for recyclability, performance and benignity that can be evolved to cover other types of polymers, and publicly available cost and environmental assessment tools for recyclable biobased polymers. A critical outcome is the demonstration of successful design of a market-relevant polymer while incorporating key performance requirements and EOL considerations that can be replicated for other polymer types.