

Synthesis and depolymerization of CO₂-derived polyhydroxyurethane

Wonki Lim¹, wonki0123@kaist.ac.kr, **Cafer T. Yavuz**², **Sheng Li**¹. (1) Chemical and biomolecular engineering, Korea Advanced Institute of Science and Technology, Daejeon, Korea (the Republic of) (2) Chemistry, King Abdullah University of Science and Technology, Thuwal, Makkah, Saudi Arabia

Polyhydroxyurethane (PHU) is a non-isocyanate polyurethane synthesized through the step-growth polymerization of bis(cyclic carbonate) and diamine. In this contribution, we report a CO₂-derived route to prepare PHU and subsequent efforts to chemically recycle the polymer. We utilized an imidazolinium network polymer to catalyze the cycloaddition of CO₂ to epoxide and successfully obtained bis(cyclic carbonate) monomers at high conversion efficiency. The CO₂-derived monomers were then polymerized in combination with diamines to yield PHU polymers of reasonably high molecular weights. We further explored the feasibility of reversing the aminolysis reaction to chemically depolymerize PHU. Different types of amines were examined to determine their effectiveness at breaking down the PHU chains. We found that both primary and secondary amines were capable of cleaving the urethane linkage to produce oligomers of reduced chain lengths. Furthermore, when hexamethylenediamine was employed, the recovered oligomers contained amino groups at both chain ends and thus could be repolymerized with additional bis(cyclic carbonates) to generate PHU. Our findings highlight the feasibility and efficacy of an aminolysis-mediated PHU polymerization and recycling strategy, offering a sustainable pathway for PHU materials from synthesis to end-of-life management.

