Chemical Engineering Dissertation Defense

Computational Frameworks for Circular Reaction Network Development and Optimization-Based Decision Support in Sustainable Chemical Systems

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Abstract

Sustainable chemical and material systems require computational frameworks to identify, evaluate, and refine circular pathways before production and recovery strategies are fixed. This dissertation develops computational frameworks for the design and optimization of circular chemical reaction networks (CCRN) and for decision support based on optimization. The work advances circularity evaluation from isolated end-of-life assessment to an integrated network optimization problem encompassing feedstocks, chemical transformations, product use, recovery routes, emissions, and end-of-life transformations. By integrating multi-objective optimization, hierarchical screening, and process systems engineering principles, the methodology evaluates economic and greenhouse gas (GHG) trade-offs under different modeling assumptions.

The foundational CCRN framework is established using representative systems. The application to methanol production illustrates how baseline-referenced multi-objective optimization evaluates candidate pathways across the chemistry, reaction, and process levels, demonstrating that carbon-recovery performance depends on reaction yields, energy requirements, system-boundary assumptions, and life-cycle contributions. To address polymer circularity, solid waste recovery, and infrastructure constraints, the framework is extended to polyethylene terephthalate (PET). Incorporating capital-inclusive economic modeling, scenario constraints, and Pareto-front network interpretation, the PET case provides strategic guidance for prioritizing recycling motifs while clarifying cost--GHG trade-offs.

Effective decision support requires physically grounded refinement and broader pathway coverage. A multidisciplinary workflow is introduced to establish bidirectional interaction between system-level screening and chemical-level refinement. This workflow integrates CCRN-based evaluation with experimental data, molecular dynamics simulations, and kinetic analysis to iteratively validate and refine candidate pathways. To address limitations in manual network construction, artificial intelligence-assisted methodologies are being developed. By combining large-language-model-assisted reaction retrieval with retrosynthetic tools and algorithmic post-processing, the framework expands candidate reaction spaces while supporting stoichiometric consistency, technology-readiness interpretation, and network standardization.

As CCRNs expand, exhaustive pathway enumeration within complex binary topology spaces becomes challenging. A hybrid quantum--classical topology-exploration framework is introduced to address this challenge. Quantum-assisted sampling proposes candidate network topologies, while classical mathematical programming determines process scales, evaluates feasibility and objectives, and refines Pareto solutions. Implemented within a reusable software workflow, these components establish a computational foundation for sustainable-by-design analysis of circular chemical pathways by integrating pathway generation, system-level optimization, multidisciplinary refinement, topology exploration, and software-enabled decision support.

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