LABORATORY FOR CHEMICAL TECHNOLOGY
Technologiepark 125, 9052 Gent, Belgium

Coach	**Supervisors**	**Funding**
Kevin De Ras	Prof. Kevin Van Geem, Prof. Joris Thybaut,	FWO

Automatic kinetic model generation: a novel modeling approach for liquid-phase processes

Aim

This thesis aims to investigate the extension of automatic kinetic model generation from gas-phase processes to liquid-phase processes.

Justification

The liquid-phase oxidation of cyclohexane is the most important industrial process for the production of cyclohexanol and cyclohexanone with a global production capacity around 6 Mton per year. Cyclohexanol and cyclohexanone form both precursors for the production of ε-caprolactam and adipic acid, which are the building blocks for synthesis of nylon-6 and nylon-6,6 as indicated in Figure 1.

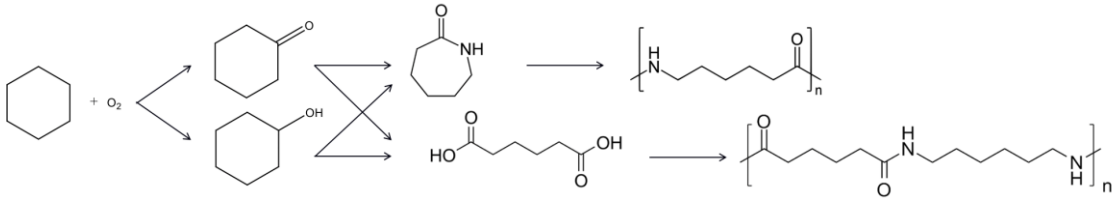

Figure 1. Production chain starting with the oxidation of cyclohexane to eventually form nylon-6 and nylon-6,6.

The first step, i.e., the oxidation, is a relative difficult one, since the desired products are intermediates in a complex network of free-radical chain reactions. In addition, cyclohexanol and cyclohexanone are more reactive towards oxidation than cyclohexane, causing the process to be carried out at low conversion to keep the selectivity acceptable. Subsequent oxidation reactions result in a number of undesired byproducts, such as dicarboxylic acids and hydroxy aldehydes.

Computer-aided model development for the gas-phase oxidation of cyclohexane is nowadays feasible due to an increase in computational resources and fundamental knowledge. However, this is in sharp contrast to this liquid-phase process for which experimental results still cannot be explained. An extension of the in-house automatic model generation framework (Genesys) to the liquid phase is intended (ALKIMO). The research encompasses both modeling and experimental aspects. A microkinetic model for the liquid-phase oxidation of cyclohexane will be developed with Genesys based on known reaction families. Accordingly, thermodynamic data and kinetic parameters are determined by on-the-fly fast estimation techniques, such as GAVs and neural networks, as well as high-level quantum chemical calculations.

Program

- Assessment of different techniques (ab initio calculations, GAVs, artificial neural networks, …) to accurately determine thermodynamic and kinetic parameters for species in the liquid phase.
- Development of a database with liquid-phase parameters from quantum chemical calculations.
- Develop a detailed kinetic model for the liquid-phase oxidation of cyclohexane with ALKIMO.
- Validation of the kinetic model with new experimental data obtained from the University of Wisconsin-Madison (USA).