Review of Ph.D. Dissertation: "Algorithm Development for Reaction and Composition Characterization in Multicomponent Mixtures"

In industry, numerous technological processes involve multicomponent mixtures. Therefore, modelling techniques that utilise a multicomponent approach are essential for technological optimisation and development. For example, in the oil industry, the multicomponent approach is crucial because most product streams contain so many compounds that measuring the concentration of individual components is challenging. Recently, the development and optimisation of separation and chemical conversion in the oil industry have become key areas in transforming the industry towards sustainability. Hence, research and optimisation processes should be inexpensive and prompt. In modern research and development, mathematical modelling of processes is as important as experiments because it makes development processes faster and cheaper.

The dissertation by Omar Péter Hamadi focuses on the characterisation of multicomponent mixtures. This research covers three important fields: developing the descriptive capabilities of different lumping methodologies, exploring application domains for thermodynamic models, and introducing a new modelling approach based on gas chromatographic analysis. This dissertation presents new algorithms that support the research process from quality measurement during experiments to the identification of reaction parameters.

- Developing the descriptive capabilities of different lumping methodologies
    - Discrete Lumped Model Based Investigation of Hydrocracking of Sunflower Oil and Kerosene Mixture

      In this chapter, a mathematical model was fitted to experimental data. The aim was to develop a sequential fitting algorithm for identifying reaction rate constants across varying temperatures and pressures in a multicomponent system. The reaction models were developed with catalyst deactivation mechanisms. The algorithm handles models with numerous unknown parameters by fixing initial results with the smallest error, providing a robust foundation for further refinement. It iteratively adjusts parameters, minimising the error at each step to converge on accurate reaction rate constants. By systematically fixing and refining parameters, the algorithm effectively manages the uncertainty inherent in models with multiple unknowns. The results show that the accuracy of the models increases with their complexity.

    - Distributed parameter model-based continuous lumping approach: an application to a pilot-plant hydrocracking reactor

      This chapter evaluates lumped kinetic models used in the prediction of hydrocracking reactor performance. The results are based on experimental data of VGO hydrocracking. The chapter compares three types of model approaches with different selectivity distributions in hydrocracking reactions, highlighting the evolution in model accuracy and applicability. It discusses how lumped models are used to simplify complex reaction networks by grouping similar components together, thus reducing computational complexity. Suitable models are highlighted in the chapter, and potential further development of the models is also determined.

- Exploration of application domains for thermodynamic models through mixture of experts learning

  This chapter evaluates thermodynamic models in different domains in the case of hydrogen solubility using the mixture of experts learning approach. It introduces various thermodynamic models and their significance in predicting the behaviour of complex chemical systems. The chapter explains the mixture of experts learning technique, which involves combining multiple expert models to enhance predictive accuracy. The results show that the developed method explores how thermodynamic models should be combined and significantly improves the prediction performance of the models.

- New modelling approach based on gas chromatographic analysis
  - Retention time alignment of gas chromatographic data

    This chapter addresses the challenge of retention time (RT) drifts in gas chromatographic data analysis. The experimental data is generated from processes like plastic waste pyrolysis. RT drifts complicate the identification and quantification of compounds. The chapter proposes methodologies to align retention times accurately, ensuring consistent data interpretation. A modified K-means based algorithm is developed to automate the RT alignment process, minimising manual intervention and potential errors. The results show that the developed algorithm is appropriate for handling retention time drifts and can accurately trace how component concentrations change over time.

  - Improving Molecular Composition Estimates using Kovats Retention Index and Molecular Similarities

    This chapter focuses on enhancing the accuracy of molecular composition estimates by leveraging the Kovats retention index (KRI) and molecular similarity measures. The chapter describes a methodology that integrates the KRI with molecular similarity metrics to improve the identification and quantification of compounds in mixtures. Details are provided on the development of algorithms that utilise these combined metrics to refine molecular composition estimates. The chapter discusses the validation of these algorithms against experimental data from catalytic pyrolysis of plastic waste. It concludes that combining the Kovats retention index with molecular similarity measures provides a powerful approach to molecular composition analysis, offering substantial improvements in analytical accuracy and reliability.

Questions and notes:

Chapter 3: How can you develop the model equations to describe the effect the pressure change?

Chapter 4: Why identification reach α_2 upper limit of the M2 model? Do you tried to expand the range of identification.

Chapter 5: The presented method helps to identify the best thermodynamic model (TM) for process simulation. How can you identify the best TM if the required domain of the simulation is wide, and more than one TM is suggested based on the results?

Chapter 6, Figure 6.5-1: On right and left-hand side of figure there are sequence of points, these points could be homologous series? The developed method capable to find the homologous series?

Summary

The dissertation is well-organised with a clear structure, including an introduction, literature review, and separate chapters for the different topics. The tone of the document is formal and academic. Despite the complex topics, the wording is well-understandable.

This dissertation shows novel algorithms and approaches in the topics listed above. The chosen examples, such as hydrocracking of sunflower oil and waste plastic pyrolysis, are modern and relevant.

To summarise, with this dissertation work, Omar Péter Hamadi has proved he can conduct high-quality independent scientific research. I accept all the theses as new results. Therefore, I recommend accepting the thesis and awarding the Ph.D. degree.

Százhalombatta, 31. March 2025.

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