Determination of Optimal Conditions and Kinetic Rate Parameters in Continuous Flow Systems with Dynamic Inputs

by

Kosi C. Aroh

Submitted to the Department of Chemical Engineering
on December 20, 2018, in partial fulfillment of the
requirements for the degree of
Doctor of Philosophy

Abstract

The fourth industrial revolution is said to be brought about by digitization in the manufacturing sector. According to this understanding, the third industrial revolution which involved computers and automation will be further enhanced with smart and autonomous systems fueled by data and machine learning. At the research stage, an analogous story is being told in how automation and new technologies could revolutionize a chemistry laboratory. Flow chemistry is a technique that contrast with traditional batch chemistry in one aspect as a method that facilitates process automation and in small scales, delivers process improvements such as high heat and mass transfer rates. In addition to flow chemistry, analytical tools have also greatly improved and have become fully automated with potential for remote control.

Over the past decade, work utilizing optimization techniques to find optimal conditions in flow chemistry have become more prevalent. In addition, the scope of reactions performed in these systems have also increased. In the first part of this thesis, the construction of a platform capable of performing a wide range of these reactions on the lab scale is discussed. This platform was built with the capability of performing global optimizations using steady state experiments. The rest of the thesis concerns generating dynamic experiments in flow systems and using these conditions to gain more information about a reaction.

The ability to use dynamic experiments to accurately determine reaction kinetics is first detailed. Through these experiments we found that only two orthogonal experiments were needed to sample the experimental space. After this an algorithm that utilizes dynamic experiments for kinetic parameter estimation problems is described. The approach here was to use dynamic experiments to first quickly sample the design space to get a reasonable estimate of the kinetic parameters. Then steady state optimal design of experiments were used to fine tune these estimates. We observed that after initial orthogonal experiments only three more conditions were needed for accurate estimates of the multi-step reaction example.

In a similar fashion, an algorithm for reaction optimization that relies on dynamic experiments is also described. The approach here extended that of adaptive response

surface methodology where dynamic orthogonal experiments were performed in place of steady state experiments. When compared to steady state optimizations of multi-step reactions, a reduction by half in time needed to locate the optimum is observed. Finally, the potential issues that arise when using transient experiments in automated systems for reaction analysis are addressed. These issues include dispersion, sampling rate, reactor sizes and the rate of change of transients. These results demonstrate a way with which technological innovation could further revolutionize the chemistry laboratory. By combining machine learning, clouding computing and very efficient, high information experiments reaction data could be quickly collected, and the information gained could be maximized for future predictions or optimizations.

Thesis Supervisor: Klavs F. Jensen
Title: Warren K. Lewis Professor of Chemical Engineering