

Experimental and Numerical Evaluation of Organic Compounds Decomposition in Continuous Supercritical Water Reactor

Igor Novosselov, Brian Pinkard, David Gorman, Kartik Tiwari, Per Reinhall,
John Kramlich

Mechanical Engineering Department, University of Washington, Seattle, WA 98195, USA

* ivn@uw.edu

Supercritical water hydrolysis of organic compounds is an attractive process for a variety of applications, including neutralization of toxic compounds, gasification of solid fuels, and chemical synthesis. A literature review shows a wide variation in chemical kinetic rates, potentially due to reactor geometries and catalytic wall effects. A modular, continuous flow, supercritical water reactor is designed to study the hydrolysis rates of organic compounds. This laboratory scale reactor accommodates a broad range of temperatures, pressures, residence times, and reagents to develop large chemical kinetic data sets. The reactor is equipped with an immersion Raman probe for *in-situ* analysis of chemical species compositions, which allows for rapid data collection and analysis. The reagent is introduced into the supercritical water via a mixing section, optimized using a numerical modeling study of jets in the cross-flow. A series of three-dimensional Computational Fluid Dynamics (CFD) simulations of the flow in the mixing section are performed. Kinetic theory is used to calculate the properties of the fluids in the supercritical state and is evaluated against the available literature. The CFD and nondimensional analysis of the momentum flux ratio of the jets to the bulk cross-flow show that mixing can be characterized by three distinct regions: (i) under-penetrating jets, (ii) jets forming counter-rotating vortices, and (iii) impinging jets. Mixing is fastest for impinging jets scenarios due to the increased turbulence in the mixing section. A dimensionless parametric analysis establishes guidance for the mixer design for this and other reactor designs. Rapid mixing limits the formation of char and phenolic compounds and establishes a clear reaction starting point for rate calculations. The decomposition of methanol is simulated by CFD using first-order chemical kinetic rates from the literature. The numerical modeling sheds insight into the complex physiochemical processes of organic compound decompositions in the supercritical environment.

An experimental study of glucose and methanol hydrolysis shows the ability to monitor the decomposition process *in-situ*, which can be related to reaction yields and to study the intermediate species formed during the hydrothermal process. Raman data are also used to determine chemical kinetic rate parameters in the near-critical and supercritical regions. Future experiments will facilitate the development of an extensive database to determine the chemical kinetic rate parameters and pathways. The study of model compounds enables the development and validation of detailed chemical kinetic mechanisms in supercritical water and the evaluation of the catalyst's role in the reactions. The eventual goal of this experimental and numerical study is to provide the guidance needed for process optimization and to scale up to industrial applications.