Nonsmooth Distillation Models Robust to Convergence Errors: Numerical Methods and Topological Aspects

by

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Distillation is the most widely used (yet highly energy-intensive) industrial separation method and one of the most well-studied chemical engineering processes. However, engineers still often encounter distillation simulation errors while using state-of-the-art process software such as Aspen Plus and HYSYS. These errors preclude one from converging flowsheets with recycle streams and from successfully utilizing rigorous process optimization methods, which are both essential tasks in designing more energy-efficient and economically viable processes. In this thesis we address these challenges by developing nonsmooth (i.e., non-differentiable) distillation models and equation-solving methods that are robust to a wide range of convergence errors. As demonstrated by our results, nonsmooth functions are a powerful tool due to their ability to automatically switch between different terms, which allows us to describe and adapt to different modes of behavior of a system using a single model.

To investigate the "dry column" errors often encountered in Aspen Plus we developed a nonsmooth version of the MESH model, which can be solved with Newton-type methods using exact generalized derivatives obtained with automatic differentiation techniques. This model allows us to simulate distillation columns in which one or more stages operate with a single phase, either superheated vapor or subcooled liquid. By developing continuation methods to simulate the nonsmooth MESH model, we discovered a new class of degenerate bifurcations in distillation columns which are generally observed regardless of the mixture or parameter being varied. These bifurcations are characterized by infinitely-many, multiple steady states with dry/vaporless stages, and happen at the so-called critical parameter value associated with the first flow rate in the column reaching zero.

In order to describe the topological structure of these bifurcation curves in a rigorous fashion, we proved a piecewise-differentiable (PC^r) Rank Theorem that allows us to characterize nonsmooth curves and surfaces as PC^r manifolds, according to the theoretical framework introduced in this thesis. We also generalized a previous Lipschitz Rank Theorem and applied it to define Lipschitz embedded submanifolds. Further, we developed sufficient and practically verifiable conditions, in terms of the B-subdifferential generalized derivative, that can be applied to the PC^r MESH model function to theoretically predict the geometric behavior of its level sets that we observed numerically.

The nonsmooth MESH model overcomes dry column errors for specifications that lead to a feasible state with dry/vaporless stages. To address convergence failure due to column specifications being infeasible, which in general is unpredictable prior to simulation,

we developed a second class of nonsmooth, adaptive distillation models. Our modeling strategies return a feasible solution even when one or two specifications are infeasible, by automatically resetting the latter to ensure that all flow rates are within their imposed lower and upper bounds. Additionally, we developed a nonsmooth version of the inside-out algorithm to converge these nonsmooth models reliably from an *ab initio* starting point, even for highly non-ideal mixtures. With a series of test cases, we demonstrate that our distillation modeling methods outperform Aspen Plus due to their ability to converge both individual columns and flowsheets with recycle under infeasible or near-infeasible specifications, non-ideal thermodynamics, and poor initial guesses.

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