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Pseudo-transient models for multiscale, multiresolution simulation and optimization of intensified reaction/separation/recycle processes: Framework and a dimethyl ether production case study

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ABSTRACT

Including detailed models of processing equipment in the process flowsheet model is required in the case of "non-standard" unit operations, including, e.g., intensified equipment and unconventional reactor designs. We provide a unified multiscale framework for including such models in equation-oriented process flowsheet modeling, simulation and optimization. Relying on the reaction/separation/recycle process prototype, we propose a multiresolution paradigm, whereby detailed, distributed-parameter representations of reacting systems and rigorous models of (intensified) separation units are embedded in process flowsheet models. We develop an equation-oriented modeling approach based on a pseudo-transient reformulation of the balance equations, enabling the reliable and robust simulation of the process flowsheet. We also describe a companion design optimization routine. We illustrate these concepts with an extensive case study on dimethyl ether production using an intensified process featuring a dividing-wall distillation column and an adiabatic packed bed reactor with intermediate quenching.

1. Introduction

Ever increasing global competition and regulatory constraints are driving the chemical and petrochemical industries to improve the economic and environmental performance of their processes. Specifically, manufacturers strive to maximize the production of product per unit of operating expenses (i.e., raw materials and utilities) while simultaneously minimizing capital expenses. Additionally, strict demand, product quality, safety, environmental, and process operating constraints must be met. To this end, process engineering seeks to find the optimal plant design, including both operating conditions such as pressures, temperatures, and flow rates, and unit designs such as reactor volumes or the number of stages in a distillation column.

Historically, process design has been carried out using sequential-modular process simulators, wherein a process flowsheet is created from a library of common unit operations, and connectivity among units is established. The flowsheet model is then solved iteratively, i.e., the individual unit operations are solved in sequence, and the recycle loops are updated until the flowsheet has been solved (Biegler et al., 1997). It has been recognized that the combination of a sequential-modular flowsheet simulator and the empirical procedure of independently optimizing each design variable typically results in a suboptimal design, motivating the use and development of equation-oriented process simulators (Biegler, 2010; Dowling et al., 2016; Zebian et al., 2012) coupled with optimization tools.

Equation-oriented simulators solve all model equations simultaneously (Biegler, 2010; Dowling and Biegler, 2015) and are preferred for flowsheet design and optimization calculations because analytical gradients are available via automatic or analytical differentiation. Derivatives can then be employed in state-of-the-art optimization algorithms (e.g., sequential quadratic programming, interior point methods) that rely on accurate calculations of the Jacobian and Hessian matrices to iteratively improve the process design.

Equation-oriented simulators allow engineers considerable flexibility to represent "unconventional" or non-standard process equipment models in the flowsheet. For example, the reactor unit operations available in many commercial flowsheet software packages include stoichiometric reactors, equilibrium reactors, stirred tank reactors, and (simplified) plug flow reactors. There is, however, no immediate way to incorporate (and accurately and completely describe) intensified, mulfunctional designs, such as membrane reactors or heat exchanger reactors. Similarly, intensified separation equipment such as dividing-wall columns must be described using ensembles of interconnected distillation columns. Furthermore, equation-oriented modeling affords flexibility in

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capturing the multiscale phenomena occurring in the aforementioned devices, e.g., diffusion into the catalyst pellets, detailed reaction kinetics, adsorption, membrane diffusion, heat transfer, flow distribution. Accuracy in predicting the performance of such intensified equipment is crucial for ensuring its widespread adoption by industry, especially considering that operating ranges may be diminished compared to conventional setups (Nikačević et al., 2012). Moreover, increasing the level of detail used in describing such equipment in flowsheet models allows for all of the relevant design decisions (e.g., reactor volumes, catalyst pellet sizes, cooling jacket volumes) to be considered simultaneously, such that the process design is optimized in a holistic manner.

While the need for such multi-scale flowsheet modeling, simulation and optimization has been clearly recognized, the literature reporting the inclusion of detailed, first-principles models of process equipment in process flowsheet modeling and optimization is limited. Existing efforts fall into two main categories:

1. *Surrogate modeling*, whereby simplified models are developed from scenario data generated by simulations of a more complex model, and *reduced-order modeling*, whereby low-order models are derived from full-order, high dimensional models using model reduction techniques (e.g., singular perturbation arguments, Baldea and Daoutidis, 2012). Here, we recall the work of Dowling et al. (2016), who report the optimization of an oxy-combustion process with surrogate boiler model. A reduced-order 1D/3D boiler model and a trust-region method were used to carry out the derivative-free optimization calculations for the boiler simultaneously with the equation-oriented process flowsheet. Other works in the oxy-combustion literature have attempted similar flowsheet optimization approaches using surrogate models for the boiler (Edge et al., 2012, 2013; Fei et al., 2015). Several other works employed surrogate models to approximate the input-output behavior of detailed unit models and embed these in process flowsheets, including (Caballero and Grossmann, 2008) where a rigorous non-isothermal plug flow reactor model is used, and (Teske, 2014) where a detailed rate-based distillation column is used.

   The evident advantages of surrogate modeling are the reduction in problem size, as well as the option of using model functional forms that lend themselves more easily to use in optimization calculations. On the other hand, the validity of surrogate models is confined to the domain spanned by the data used in their construction, and extrapolation presents obvious risks. These issues are to some extent avoided by using reduced-order models derived from a physics-based full-order system representation. Nevertheless, in many cases, the pathway for deriving such models is not obvious, and its implementation may require significant domain expertise.
2. A small number of literature works report the use of *rigorous first principles equation-oriented models*, predominantly for chemical reactors, in the flowsheet design optimization problem directly. These include the papers by Rodriguez et al. (2010), where a detailed multitubular reactor model is included in the optimal design of a propylene oxide process, and Recker et al. (2015), where a detailed multitubular differential sidestream reactor model is included in the optimal design of an ethyl tert-buthyl ether process.

   The use of such models is clearly desirable, but comes at the cost of high model dimensionality as well as model stiffness/ill conditioning, factors that create significant challenges for numerical simulation and optimization.

The literature overview provided above suggests that incorporating detailed physical models of process equipment (including intensified subsystems) into an equation-oriented process flowsheet model (potentially featuring numerous other "conventional" unit operations) is a significant challenge (Chen and Grossmann, 2016; Recker et al., 2015). Solving the process model in an equation-oriented simulator requires a method for solving a large system of nonlinear (and potentially discontinuous) algebraic equations. Newton-type solvers are typically used due to their super-linear convergence properties in the vicinity of the solution; however, Newton-type solvers are only locally convergent and require an initial guess that is "close" (in a norm sense) to the solution in order to converge. Sequential refinement or bootstrapping techniques are often implemented to initialize large process flowsheet models for optimization (Dowling and Biegler, 2015; Rodriguez et al., 2010; Recker et al., 2015). Nonetheless, these methods are difficult to use even for experienced engineers. Other methods for initialization have been proposed, including: homotopy continuation (Paloschi, 1998; Malinen and Tanskanen, 2010; Rahimian et al., 2011; Fletcher and Morton, 2000; Pantelides and Urban, 2004), interval Newton methods (Schnepper and Stadtherr, 1996; Maier et al., 2004; Gau and Stadtherr, 2002; Lin et al., 2006; Lin and Stadtherr, 2004), terrain methods (Lucia and Feng, 2002, 2004), and global-optimization-based approaches (Stuber et al., 2010; Maranas and Floudas, 1995), all of which have demonstrated limited success at the flowsheet level.

In our recent work, we proposed a transparent and reliable approach for equation-oriented flowsheet modeling, simulation and optimization (Pattison and Baldea, 2014). It is based on converting a subset of the algebraic model equations of unit operations to differential equations in a fictitious time variable, thereby creating a pseudo-transient model of the process that is statically equivalent (i.e., has the same steady state solution) to the original algebraic model. For simulation purposes, initial conditions replace the initial guess for a Newton-type method, and an integration over a fictitious time variable is carried out using a variable step integrator until steady state is reached (or the model is within the basin of convergence for a Newton solver). A library of models for common unit operations was created, and more complicated equipment models such as dividing-wall distillation column models (Pattison et al., 2015), and multistream heat exchanger models were later developed (Pattison and Baldea, 2015).

In this work, we extend the pseudo-transient flowsheet modeling, simulation, and optimization framework to include detailed models of process equipment, and provide the capability to account for phenomena occurring over multiple spatial dimensions. Specifically, we propose a pseudo-transient framework for modeling multiphase reactor units with an arbitrary degree of complexity. We show that the models can be easily embedded in a pseudo-transient reaction/separation/recycle process flowsheet model which also includes rigorous (although not necessarily multiscale) models of the separation systems and recycle streams, thereby creating a *multiresolution flowsheet model*. We apply these new concepts to the design optimization of an integrated/intensified dimethyl ether production process featuring a quenched, adiabatic, packed-bed catalytic reactor and a dividing-wall distillation column. Through the case study, we show that the optimal design of the dividing wall column is highly dependent on the level of detail included in the reactor model, amply justifying the need to utilize detailed, multi-scale models of unit operations and process equipment for the design of integrated/intensified processes.

2. Background: pseudo-transient process modeling

The steady-state behavior of a unit operation or process flowsheet can be expressed as a (typically large-scale and highly-nonlinear) set of n algebraic equations:

$$\mathbf{f}_{ss}(\mathbf{x}, \mathbf{z}, \theta) = 0 \tag{1}$$