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Sustainable Design and Synthesis of Shale Gas Processing and Chemical Manufacturing Processes

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In this work, we propose a superstructure of integrated shale gas processing and chemical manufacturing processes with 51,840 alternative possible process designs. The superstructure consists of eight sections, namely acid gas removal, dehydration, NGLs recovery, NGLs separation, hydrocarbons conversion, light olefins separation, C4 separation, and acid gas disposal. For the steam cracking reactions in the hydrocarbons conversion section, we optimize the product distributions of steam cracking of ethane, propane, *n*-butane, and *i*-butane. Extensive process simulations are performed for all the involved processes in the superstructure in order to collect high-fidelity process data and develop detailed process models for the technology/process alternatives in the superstructure. Next, we propose a multiobjective mixed-integer nonlinear programming (MINLP) superstructure optimization model with five groups of constraints, namely superstructure network configuration constraints, mass balance constraints, energy balance constraints, techno-economic evaluation constraints, environmental impact assessment constraints. Three objective functions are maximizing the net present value per GJ of raw shale gas, minimizing the global warming potential per GJ of raw shale gas, and minimizing the water footprint per GJ of raw shale gas, respectively. A tailored global optimization algorithm is applied to efficiently solve the resulting nonconvex MINLP problem. The application of the proposed framework is illustrated through a case study based on a Marcellus shale gas feed.

1. Introduction

The combination of hydraulic fracturing and horizontal drilling has led to a substantial increase in shale gas production, indicating the advent of a “shale revolution” (Brown and Yücel, 2013). With continued development of shale plays, the U.S. will soon become a net gas exporter (EIA, 2017). In response to the rapid expansion of shale gas production, additional facilities dedicated to shale gas processing and natural gas liquids (NGLs) upgrading must be designed and developed in the near term (Gao and You, 2015). An integrated shale gas processing and chemical manufacturing process simultaneously produces pipeline quality gas and a collection of value-added chemicals from wellhead shale gas (Cafaro and Grossmann, 2014). The value-added chemicals can bring in a higher profit margin than the NGL products from a conventional shale gas processing system (Fernandez et al., 2017), contributing to a better economic performance under relatively low energy prices (Julian-Duran et al., 2014). By sharing assorted utilities, process integration could lead to a higher energy efficiency, mitigating the environmental impacts associated with utility generation systems. To fully exploit these advantages, it is important to address the optimal process design of the integrated shale gas processing and chemical manufacturing process that should be both economically competitive and environmentally sustainable. In this work, we develop a comprehensive superstructure of integrated shale gas processing and chemical manufacturing processes with 51,840 alternative possible process designs. We optimize the product distributions of steam cracking of ethane, propane, *n*-butane, and *i*-butane and perform extensive process simulations for all the involved processes in the superstructure in order to collect high-fidelity process data. We propose a multiobjective mixed-integer nonlinear programming model to simultaneously maximize the net present value (NPV) per GJ of raw shale gas, minimize the global warming potential (GWP) per GJ of raw shale gas, and minimize the water footprint per GJ of raw shale gas. Heat integration as performed for each process alternative. The application of the proposed framework is illustrated through a case study based on a Marcellus shale gas feed.

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2. Process description

We develop the most comprehensive superstructure for shale gas processing and chemical manufacturing processes. As shown in Figure 1, there are eight sections in the superstructure, namely acid gas removal, dehydration, NGLs recovery (He and You, 2015), NGLs separation, hydrocarbons conversion (Yang and You, 2017a), light olefins separation (Yang and You, 2017b), C4 separation, and acid gas disposal (Gong and You, 2017). The raw shale gas from wellhead is first sent to the acid gas removal section to split hydrogen sulfide and carbon dioxide. The acid gas waste is then sent to the acid gas disposal section, and the sulfur content is captured before the remaining waste is emitted. The sweet gas becomes dry gas after the dehydration section. The dry gas is then separated by the NGLs recovery section to a pipeline gas product and a NGLs product. In the NGLs separation section, the mixture is split into ethane, propane, *n*-butane, *i*-butane, and natural gasoline. The first four hydrocarbons are sent to their corresponding processes in the hydrocarbons conversion section to form a spectrum of olefins. The effluents are then handled by a series of separation processes in the light olefins separation section. In additional to the light olefin products, the unreacted ethane and propane are recycled to the hydrocarbons conversion section, and the C4 mixture is sent to the C4 separation section. The unreacted C4 hydrocarbons are recycled to the hydrocarbons conversion section, while other C4 chemicals are fractionated to their corresponding products.

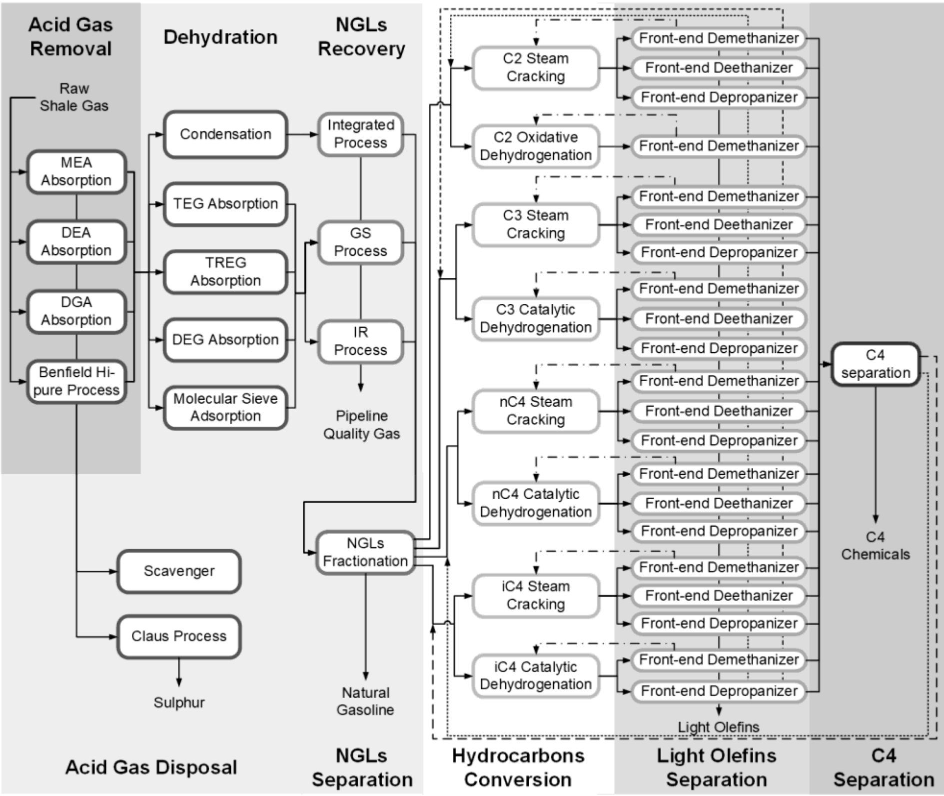

Figure 1: Superstructure for shale gas processing and chemical manufacturing processes

3. Model formulation and tailored solution strategy

The product distribution of a chemical reaction can vary drastically under different operating conditions. To identify the optimal performance of a process that involves chemical reactions, the product distributions of the chemical reactions should be optimized to enhance the yields of more profitable products in a superstructure optimization problem (Pitchaimuthu et al., 2017). Steam cracking of ethane, propane, *n*-butane, and *i*-butane are major reactions in the superstructure with kinetics models from the literature (Sundaram and Froment, 1977). We develop four ODE-constrained dynamic optimization models for steam cracking of these hydrocarbons, respectively. The product distribution optimization models are coded and solved in MATLAB.

max   Profit of selling hydrocarbon products
s.t.   Kinetic model based ODE constraints; Boundary conditions; Economic evaluation constraints

Based on the optimal product distributions of steam cracking reactions, we develop 53 simulation models in ASPEN PLUS for different alternative designs in the superstructure. The simulation results are then used to calculate key parameters such as inlet compositions and split fractions of the major unit operations in the process (Ahmetović et al., 2017). Given comprehensive data for the mass and energy balance relationship (You and Wang, 2011), a life cycle optimization model is formulated (Yue et al., 2013), to determine the optimal process design of the integrated shale gas processing and chemical manufacturing process (Garcia and You, 2015). The functional unit is processing 1 GJ raw shale gas and the system boundary covers four life cycle stages from cradle to gate (Gebreslassie et al., 2013a), namely shale gas extraction, utility production, shale gas processing and chemical manufacturing, and wastewater treatment. We focus on global warming potential (GWP) and water footprint in life cycle impact assessment (Gebreslassie et al., 2013). The techno-economic analysis accounts for capital expenditure and operating expenditure of the integrated process (Wang et al., 2013). The proposed model employs functional unit based fractional objective functions (Yue et al., 2013), because the performance of different process systems with distinct final products can be compared in a fair manner (Zhang et al., 2014).

max Functional unit based net present value
min Functional unit based GWP
min Functional unit based water footprint
s.t. Superstructure network configuration constraints; Mass balance constraints;
Energy balance constraints; Techno-economic evaluation constraints;
Environmental impact assessment constraints

The multiobjective MINLP problem consists of both integer and continuous variables as well as multiple nonlinear nonconvex functions. Due to the combinatorial nature and nonconvexity, global optimization of this MINLP problem can be computationally intractable. To tackle the computational challenge, we employ a tailored global optimization algorithm that integrates the inexact parametric algorithm (Zhong and You, 2014) and the branch-and-refine algorithm (You and Grossmann, 2011). The computational challenge stemming from the fractional objective functions is tackled by the inexact parametric algorithm (Gong and You, 2014). Instead of solving the original MINLP problem directly, we introduce an auxiliary parameter r and an auxiliary parametric problem $F(r)$. The original optimal solution is identical to the optimal solution of the auxiliary parametric problem with the parameter r^* such that $F(r^*) = 0$. In each iteration of the inexact parametric algorithm, we need to globally optimize an MINLP problem $F(r)$ with separable concave terms in the objective function. To efficiently solve these MINLP problems, we replace the nonlinear terms with successive piecewise linear approximation functions and solve the relaxed mixed-integer linear programming (MILP) problems iteratively.

4. Case study

The superstructure optimization model is applied to handling a raw shale gas feed of 200 million standard cubic feet per day from Marcellus shale (Kidnay et al., 2011). The raw shale gas feed is collected from about 200 wells after being preprocessed on wellsites (Holditch, 2012). All computational experiments are performed on a DELL OPTIPLEX 7040 desktop with Intel(R) Core(TM) i7-6700 CPU @ 3.40GHz and 32 GB RAM. The superstructure optimization problem and its solution procedure are coded in GAMS (Rosenthal, 2016) with CPLEX 12.7. used as the MILP solver. The relative optimality tolerance is set as 10^{-6}.

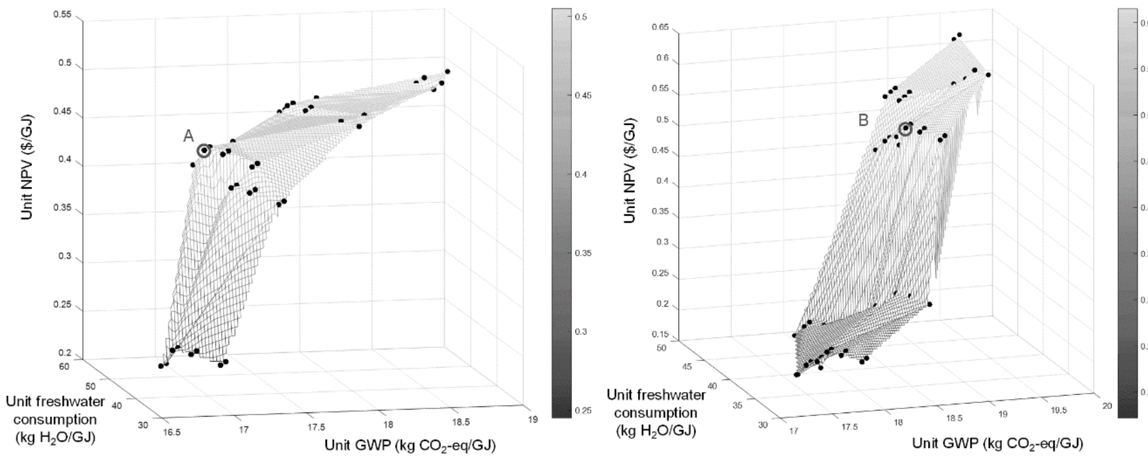

Figure 2: Pareto-optimal surfaces with product distributions from the literature (left) and from optimization (right)

The optimal solutions of this multiobjective optimization problem can be plotted as a 3D Pareto-optimal surfaces in Figure 2. The unit NPV, the unit GWP, and the unit water footprint of the good-choice solution A are \$0.46/GJ (corresponding NPV: \$0.83 billion), 16.99 kg CO_2-eq/GJ, and 38.92 kg H_2O/GJ, respectively. In contrast, the unit NPV, the unit GWP, and the unit water footprint of the good-choice optimal solution B are \$0.53/GJ (corresponding NPV: \$0.96 billion), 18.71 kg CO_2-eq/GJ, and 40.42 kg H_2O/GJ, respectively. Therefore, the optimal product distributions of steam cracking lead to better economic performance but worse environmental performance than the product distributions of steam cracking taken from the literature.

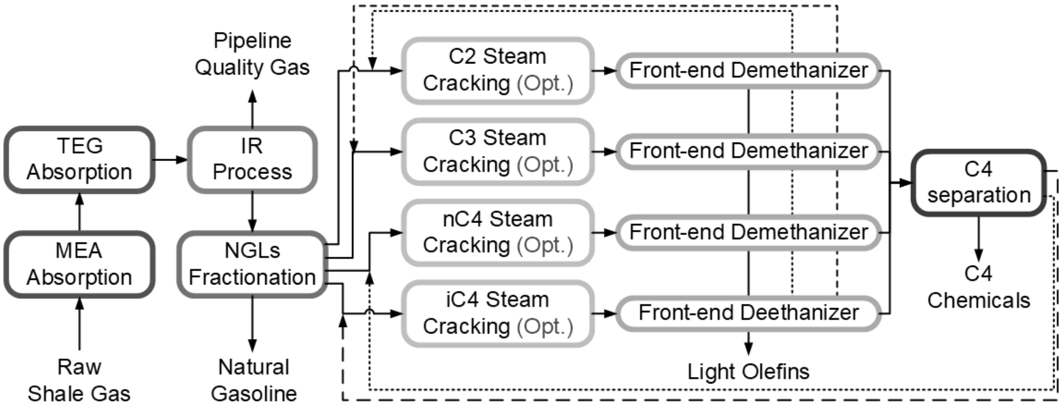

Figure 3: Technologies/processes selected by solutions A and B

Both solutions A and B consider a triethylene glycol (TEG) absorption process in the dehydration section (see Figure 3). Demonstrating a moderate GWP and a moderate water footprint rate simultaneously, the TEG absorption process becomes a balanced and preferred option when the unit GWP and the unit water footprint are minimized simultaneously. In the hydrocarbons conversion section, only steam cracking processes are selected by the good-choice solutions. However, the other technologies/processes in hydrocarbons conversion section can be favourable if only one objective function is considered. For example, to maximize the unit NPV, the optimal process flowsheet is equipped with catalytic dehydrogenation of propane and *n*-butane, because the product distributions of these reactions are more profitable than those of the corresponding steam cracking reactions.

To better understand the optimal process designs, we present the cost, GWP, and freshwater consumption breakdown of the good-choice optimal solutions A and B in Figure 4. The dominant contributor to the total annualized cost is the feedstocks cost and around 97% of the feedstock cost is spent in purchasing the raw shale gas. The GWP can be classified by the sources of emissions. For both optimal solution A and optimal solution B, the largest share of GWP comes from feedstocks extraction. Moreover, since producing heating utilities is relatively more energy-intensive than producing cooling utilities and electricity, heating utilities production causes more GWP than the other two utilities-related contributors. The GWP associated with heating utilities production in the optimal solution B is higher than that of the optimal solution A. Therefore, future development of the integrated shale gas processing and chemical manufacturing process can focus on improving the energy utilitzaiton efficiency to reduce the total GWP. Among the considered sources of freshwater consumption, cooling utilities consume much more water than others, because a substantial amount of water evaporates to reduce the temperature of the remaining cooling water. To ensure the cooling quality, makeup water is added to the cooling system. Given that cooling is needed for all the reactors and distillation columns, makeup water becomes the dominant consumer of water within the entire system. The water footprint for utilities production in the optimal process design of the optimal solution A is smaller than that of the optimal solution B, but the direct water consumption in the optimal process design of optimal solution A is higher than that of the optimal solution B. To save more water, the efficiency of the cooling water system should be improved.

Figure 5 present how the optimal economic objective function values of the most economically competitive optimal solutions change when the market related parameters are allowed to deviate 20% from their current values. It is noted that for the product distributions of steam cracking taken from the literature, the most influential market related parameter is the price of raw shale gas. A 20% increase in the price of raw shale gas reduces 29% of the highest NPV, while a 20% decline raises 29% of the highest NPV. The prices of feedstocks and utilities are in a negative correlation with the highest NPV, while the prices of products are in a positive correlation with the highest NPV. One exception is the price of electricity, because the optimal design generates surplus electricity and treats electricity as a product instead of a feedstock/utility. For the optimal product distributions of steam cracking, the price of raw shale gas causes the largest change in the highest NPV. Moreover, the influence of the price of pipeline quality gas and the price of ethylene is less significant, while the influence of the price of 1,3-butadiene is more pronounced.

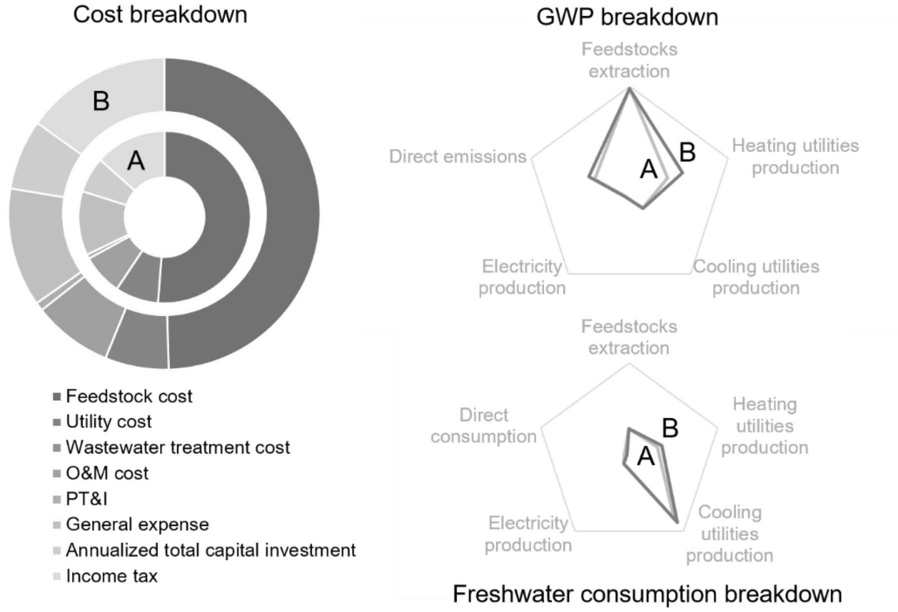

Figure 4: Cost, GWP, and freshwater consumption breakdown of good-choice optimal solution A and B

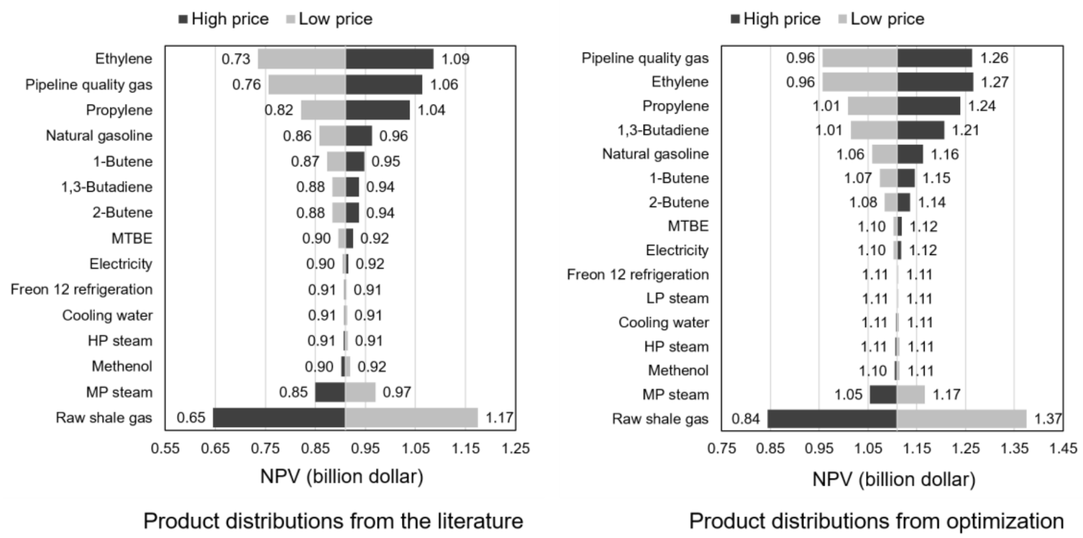

Figure 5: Sensitivity analysis results for the most economically competitive optimal solutions

5. Conclusions

We proposed a novel process synthesis framework that combines product distribution optimization of chemical reactions and superstructure optimizaiton of the process flowsheet. The proposed framework was illustrated by an application to an integrated shale gas processing and chemical manufacturing process. Two good-choice solutions were identified on the resulting Pareto-optimal surfaces with balanced performance.

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