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Multi-objective Optimization of a Rectisol® Process

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Abstract

This work focuses on the design, simulation and optimization of a Rectisol®-based process tailored for the selective removal of H_2S and CO_2 from gasification derived synthesis gas. Such task is quite challenging due to the need of addressing simultaneously the process design, energy integration and utility design. The paper, starting from a Rectisol® configuration recently proposed by the authors, describes the models and the solution strategy used to carry out the multi-objective optimization with respect to exergy consumption, CO_2 capture level and capital cost.

Keywords: Rectisol, CO_2 Capture, Numerical Optimization, PGS-COM, Pareto frontier

1. Introduction

Coal to Liquids (CTL) as well as Integrated Gasification Combined Cycle (IGCC) plants take advantage from the conversion of a cheap, fossil fuel like coal into a clean synthetic gas, mainly composed of hydrogen, carbon monoxide, carbon dioxide and other minor species, either to produce Liquid Fuels or Electricity as output. In such plants, sulfur-containing compounds are among the most critical contaminants, and they should be separated from the raw syngas not only to cope with emissions regulations but, in the specific case of CTL, also to avoid any potential detrimental impact on the catalyst of the downstream chemical synthesis process. Moreover, in a near-future characterized by restrictions on CO_2 emissions, Carbon Capture and Storage (CCS) becomes a standard feature of acid gas removal processes.

As reported by Koss (2006), the Rectisol® process (Weiss, 1988), licensed by Linde and Lurgi-Air Liquide companies, represents the Acid Gas Removal (AGR) benchmark for syngas purification with more than 85 units currently operating worldwide. Even though there is a significant industrial know-how about the Rectisol® process, very few data and documents are available in literature. Indeed, to the best of our knowledge, just Sun and Smith (2012; 2013) published a detailed simulation model of the process. In any case, all the available studies are based on a given set of operating conditions rather than on an optimized design. Moreover, there are no studies dealing with the optimal design of the Rectisol® process targeted for CCS application.

The goals of this study are: (i) develop and efficiently solve a detailed model of a Rectisol®-like absorption process suitable to be used as AGR in a CTL and IGCC plant; (ii) identify and include a strategy to simultaneously perform the process heat integration and the selection and design of the utilities; (iii) formulate and solve the multi-objective optimization problem with respect to the three conflicting objectives, maximum CO_2 capture level, minimum exergy consumption, and minimum capital cost.

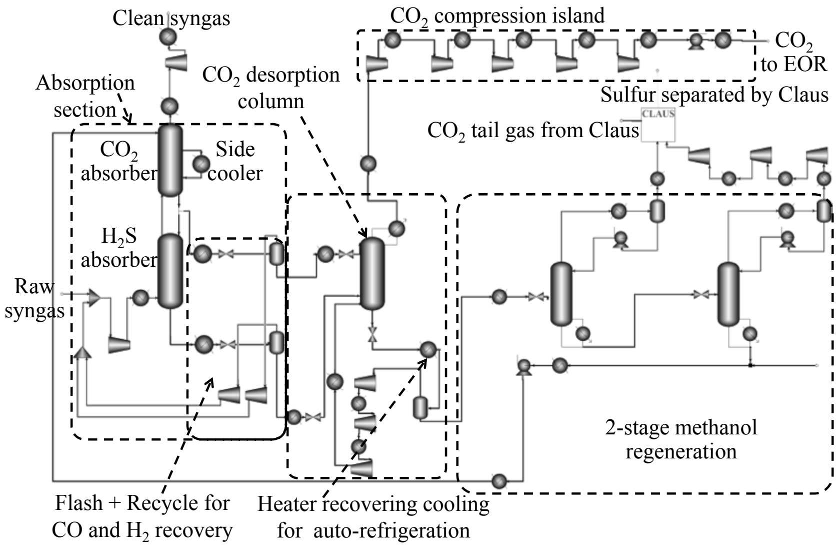

Figure 1. Process Flow Diagram of the Rectisol®-based AGR process to be optimized.

2. Rectisol® process configuration

Rectisol® is a quite flexible process which can be tailored to clean the raw syngas in order to meet the requirements of various type of downstream processes. Depending on the syngas route envisaged, and on the end-use of the side-product streams, namely CO_2 and H_2S concentrated flows, the process layout and the operating conditions may differ significantly (Weiss, 1988). In this paper we focus on the Rectisol® scheme recently proposed by Gatti et al. (2013), configured for the deep purification of a CTL syngas. The process, whose flowsheet is reported in Figure 1, is designed for producing the following outputs, whose specifications are reported in Table 1, together with the feedstock and input stream properties: a purified syngas stream suitable for Co-based Fischer-Tropsch synthesis; a CO_2-rich dense phase stream suitable for Enhanced Oil Recovery (EOR); an H_2S-rich stream suitable for a Claus process.

This novel Rectisol®-based version differs from Linde's patented Rectisol® (Weiss, 1988) for the following features: (i) the CO_2 desorption section instead of including a flash regeneration plus a rectifying column consists of a single desorption column exploiting CO_2 flashing, H_2S reabsorption and auto-refrigeration (in the sense that a significant fraction of the cooling duty is recovered within the process by flashing the bottom liquid stream of the CO_2desorber); (ii) the methanol regeneration section is split in two stages in order to minimize the exergy consumption of the reboiler.

3. Optimization framework

It's worth emphasizing that Gatti et al. (2013) developed the above-mentioned novel process configuration by applying pinch analysis rules and heuristic design criteria (based on simple rules of thumbs). In this paper the authors present a further improvement of such novel process achieved by the application of systematic heat integration tools and numerical optimization algorithms.

Table 1. Boundary conditions, assumptions and specifications of the process.

Inlet Stream Properties		Outlet H_2S rich stream to Claus Conditions	
Raw dried partly shifted syngas produced by GE gasifier fed with Illinois #6 coal		H_2S/CO_2 molar ratio	≥ 1/2
Composition	Mole %	Destination of CO_2 in tail gas	Recycled back as pure CO_2 stream
CO_2	28.02 %	Process assumptions	
H_2S (including COS)	1.27 %	Pressure loss $\Delta P/P_{in}$	2 %
CO	23.44 %	Polytropic efficiency of syngas and CO_2 compressors	84 %
N_2 (including other inerts)	0.40 %	Isentropic efficiency of expanders	88 %
H_2	46.87 %	Polytropic efficiency of refrigerator compressors	82 %
Total Molar Flow Rate	5.404 kmol/s	Mechanical/electric efficiency of the driver	92 %
Total Mass Flow Rate	110.2 kg/s	Utility assumptions	
Temperature	30 °C	Refrigeration cycle	Cascade Ethane/Ammonia
Pressure	35 bar	Cooling water	Closed loop between 15 and 25 °C
Outlet CO_2 Conditions		Steam for reboiling	Saturated steam at 0.5/1.5/3/10 bar
Destination: Enhanced Oil Recovery		$\Delta T_{min}/2$ for reboiler utility	10 °C
State: Supercritical dense at 150 bar		Outlet Syngas Conditions	
Temperature	25 °C	Temperature	25 °C
CO_2 molar concentration	> 97 %	Pressure	30 bar
H_2S molar concentration	< 150 ppmv	H_2S molar content	< 50 ppbv

3.1. Problem formulation and optimization approach

The optimization problem can be formulated as follows: for given inlet raw syngas thermodynamic conditions, determine the process design, heat integration and utility system design which minimizes the overall exergy penalty while satisfying a set of technological and environmental constraints reported in Table 1.

To tackle this problem, a robust black-box approach was adopted in which:

A. a derivative-free black-box algorithm optimizes the main process and utility design variables, namely seven stream temperatures (four of the process and three of the refrigeration cycle), five pressures, the mass flow rate of the solvent, the heat duty of the reboiler of the atmospheric regenerator, the split fraction of methanol sent to the H_2S absorber, the minimum approach temperature ΔT_{min}, and the number of trays of the absorber and desorber columns (19 optimization variables);
B. for given design variables listed in step A, the process is solved by a sequential flowsheeting software (Aspen Plus® V7.3);
C. for given utility design variables listed in step A, the process heat integration and the design of the utilities are simultaneously optimized;
D. the capital cost of the overall system is computed.

Within such approach, every black-box function evaluation includes steps B, C and D, and its outputs are the overall (process + utilities) exergy consumption (EXCON), CO_2Capture Level (CO_2CL) and capital cost (CAPEX). Compared to an equation oriented approach, this approach allows the use of different (specifically developed) algorithms for each step, and reduces the number of variables to be handled at each step, and thus increases the procedure robustness. The major drawback of this approach is the significant computational time required to compute the black-box function (steps B, C and D), between 5 and 15 s per evaluation, which poses a limit on the maximum number of objective function evaluations. In addition, the black-box output functions (exergy consumption, CO_2CL, and capital cost) have the following features:

- nonlinearity and multimodality, due to the nonlinearities of the process model,
- non-smoothness and discontinuity, due to the heat integration technique (i.e., integer variables associated to the selection of the best utilities, and non-differentiable points associated to the activation of pinch points),
- numerical noise, due to the numerical issues originated by the solution of the multiple recycle loops and absorption columns,
- the objective function value may not be defined in some points, due to the possible convergence failure of the process flowsheet,
- the feasible region turns out to be very small compared to the box defined by the bounds on the variables and a not-connected set.

Thus, the problem must be tackled with a robust derivative-free multi-objective algorithm. Among the several multi-objective evolutionary algorithms, we selected the Non-dominated Sorting Genetic Algorithm II (NSGA-II) by Deb et al. (2002) because it is quite effective on black-box problems (see Custodio et al., 2011), well-proven, and readily available within the MATLAB Global Optimization Toolbox (MathWorks, 2013). The major disadvantage of such algorithm, clearly shown by our computational experiments, is the lack of an intensification method capable of further refining the search around the non-dominated solutions. For this reason, a "push-Pareto" step is added to further improve the non-dominated solutions by applying the PGS-COM (Particle Generating Set – Complex algorithm) presented by Martelli and Amaldi (2014), a single objective direct-search algorithm specifically developed for constrained non-smooth problems.

3.2. Thermodynamic model and simulation assumptions

Due to the non-ideality of the physical transformations occurring within the process and because of the presence of many material recycle loops, particular attention was given to the definition of the flowsheet and its convergence features. We implemented and simulated a 0-D steady-state model of the process in Aspen Plus®, adopting the PC-SAFT equation of state, that, as described in Gatti et al. (2013), reproduces properly the Vapor Liquid Equilibria as well as the volumetric and thermal properties of the mixtures involved in the Rectisol®. Further details about the model are in Gatti et al. (2013).

3.3. Heat integration and utility design strategy

Once the process flowsheet is solved, the heat integration and the utilities are optimized with the algorithm proposed by Maréchal and Kalitventzeff (1998).The most significant exergy penalties of the process are: (i) the electric energy required to drive the process compressors and pumps, (ii) the electric energy required by the refrigeration cycle to supply the cooling duty needed by the process, (iii) the mechanical equivalent of the steam hypothetically extracted from the steam turbine for the reboiler, (iv) the chemical exergy associated to the co-captured fuel species (sent together with CO_2 to EOR). Among the utilities, a key impact is originated by the refrigeration cycle, whose design is customized to the T-Q profile of the process. The selected scheme is a state-of-the-art ammonia/ethane cascade cycle, featuring an evaporation level for each fluid. According to the method of Maréchal and Kalitventzeff (1998), given the set of utility systems and fixed the design variables optimized in step A, the "Problem Table Algorithm" is generalized into a Mixed Integer Linear Program (MILP) whose variables involve the selection of the utility systems and the mass flow rates of fluid at each utility temperature level. The objective function is the overall exergy consumption of the utilities. The MILP is implemented and solved with GLPK (GNU v4.47).

3.4. Multi-objective optimization

Instead of tackling a complex three-objective optimization problem with the conflicting objectives CO_2CL vs. EXCON vs. CAPEX, we noted the possibility of simplifying the problem into a set of bi-objective ones with respect to CO_2CL and EXCON. We noted that only the ΔT_{min} variable creates conflict between EXCON and CAPEX. Indeed, for fixed values of ΔT_{min} and CO_2CL, since the costs of the main equipment units depend on the power consumption (e.g., compressors), the higher is EXCON and the higher is CAPEX. As a result, for fixed values of ΔT_{min} and CO_2CL, EXCON and CAPEX are not-conflicting objectives. For this reason we converted the original three-objective problem into a set of bi-objective ones (CO_2CL vs EXCON) with fixed values of ΔT_{min}. In order to span the CAPEX space, we repeated the bi-objective optimization for three different values of ΔT_{min} (3, 5 and 10 K).

4. Optimization results

Given the significant computational time required by each function evaluation, we had to limit the number of function evaluations to approximately 6,000. Even though we used a 12-core computer featuring 2.8 GHz/core and executed both optimization algorithms (NSGA-II and PGS-COM) in parallel computing, the total computational time is close to 55 h. We decided to spend half of the available evaluations (3,000) to cover as much as possible the search space with the multi-objective algorithm, and the remaining ones to improve the most interesting non-dominated solutions with the single-objective optimizer. Figure 2 reports the Pareto frontiers generated by the multi-objective genetic solver on the left and after the subsequent application of the "Push-Pareto" algorithm on the right. The graph on the right highlights the improvement made by the PGS-COM algorithm. The relative improvement between the non-dominated points of the bi-objective and the corresponding "pushed" points varies in a range between 3 % and 10 % and is larger for the cases with ΔT_{min} = 10 K, meaning that this frontier was farther from Pareto-optimality than the others.

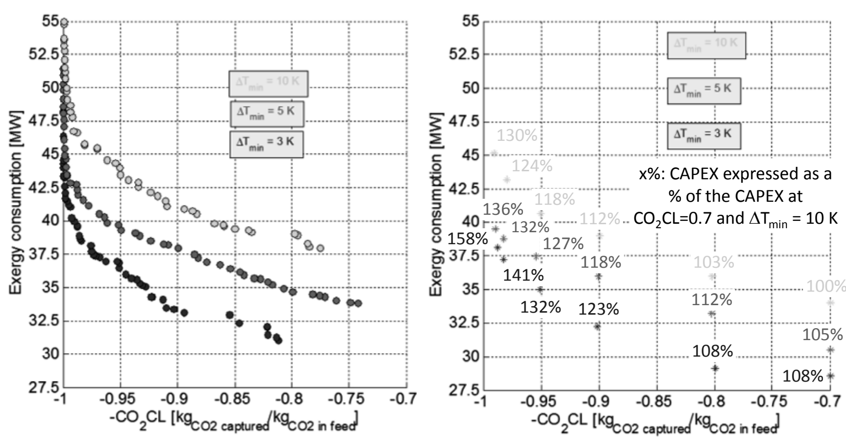

Figure 2. Left) Pareto frontiers for each ΔT_{min} considered after 30 generations of the bi-objective solver for a population of 100 individuals. Right) Trajectory of the Pareto frontiers after the application of PSG-COM and related CAPEX expressed as a percentage of a reference solution.

All of the frontiers share the same shape: the trend is almost linear between 70 % and 90 % CO_2 capture (with the exception of the curve related to the smallest ΔT_{min} which does not show a constant steep in the range); between 90 and 98% the linearity disappears and the slope tends to increase monotonically, whereas in the very high CO_2CL range above 98 % the exergy required tends to increase dramatically due to the finite solubility of CO_2 in methanol. So, it may not be economically justified to go beyond a CO_2 capture limit of 98 % - 99 %.

In order to assess the effectiveness of the multi-objective optimization, it is interesting to compare its best solution at ΔT_{min} = 10 K with the reference scheme proposed by Gatti et al. (2013), whose ΔT_{min} assumption where somehow in between the case with ΔT_{min} = 5 K and the one with ΔT_{min} = 10 K. The herein optimized configuration gives a specific exergy consumption of 662 kJ/kg of CO_2 captured whereas the reference one requires 755 kJ/kg of CO_2 captured, resulting into a 12% saving of exergy consumption.

5. Conclusions

This paper proposes a methodology for the multi-objective optimization of a novel Rectisol®-based process designed for CCS. The process and the utility systems are optimized with a black-box approach including a detailed process model as well as a heat integration & utility selection technique. The solution quality is improved by applying PGS-COM, a recently proposed direct-search method. Despite the large number of variables and the relatively small number of function evaluations allowed by the black-box solution time, the resulting Pareto frontier covers a wide range of CO_2 capture levels and shows a significant improvement with respect to the solution previously found by the authors on the basis of well-known design criteria.

Further research will focus on the application of such procedure to other pre-combustion CO_2 capture processes.

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