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A Simulation-Optimization Framework for Efficient CO_2 Capture Using Amine Absorption

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The control of CO_2 emission is becoming one of the most challenging environmental issues facing many countries today. Recent years has seen an increase in the reported use of process simulators to assess feasibility and design and troubleshoot of CO_2 capture using amine solution. We present a simulation-optimization framework comprising of HYSYS simulator and a jumping gene based multi-objective simulated annealing technique to evaluate the efficacy of CO_2 removal using DEA solution. The novelty of this approach is that, both simulation and optimization of the process are performed simultaneously in an automated fashion to fully explore the trade-off surface of CO_2 capture efficiency and operating cost. We show that for 80-93% of removal, the cost incurred is between $52 and 68 per tonne of CO_2 captured.

1. Introduction

Among the human-related activities, the process of generating electricity through combustion of fossil fuels has been the largest source of CO_2 emissions to the atmosphere. Among the available technologies for controlling CO_2 release, amine scrubbing process has been identified as the best available technology for post combustion CO_2 capture (Simmonds *et al*., 2003). The process involves absorption-reaction of CO_2 with an amine solution followed by regeneration of the amine. MEA (monoethanolamine) has been the preferred choice due to its high absorption efficiency (Øi, 2007). However, the energy requirement for MEA regeneration is the highest (Veawab *et al*., 2003). Furthermore, MEA is known to be very corrosive. Therefore, there is a considerable incentive for using an alternative solvent such as DEA (diethanolamine), which is comparable to MEA in terms of performance and cost.

2. Process Simulation Approach of Amine Process

Today, process simulator has become an essential tool in the repertoire of process engineers. This is because using process simulator, various process alternatives and

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variable modifications can be evaluated easily in a short time without the need for extensive experimentation or pilot plant testing. In the case of CO_2 capture process, because testing at large scale is expensive, it is therefore natural to employ process simulators to assess its feasibility.

Literature has been abundant with references to modelling, simulation and optimization of amine process using commercial process simulators. Singh *et al*. (2003) used HYSYS and Aspen Plus simulator to estimate the CO_2 capture costs from a coal based power plant. Their objective was to compare the performance of two technologies: MEA scrubbing and O_2/CO_2 recycle combustion process. Alie *et al*. (2005) applied flowsheet decomposition method for simulating key variables affecting MEA scrubbing process. Their simulation was performed using Aspen Plus. Øi (2007) applied HYSYS simulator to design a MEA based CO_2 removal from a combined cycle gas power plant. Both the power plant and the MEA process were simulated using the software package. Mofarahi *et al*. (2008) compared the performances of different amine solutions (MEA, DEA, MDEA and DGA) for design of CO_2 capture from the flue gas of a refinery gas turbine. They performed the simulation using their own column model written in MATLAB combined with the equilibrium parameters of CHEMCAD and Aspen Plus.

While the process simulators are useful, their capability is limited to predicting the behaviour of the process in response to changes in the process structure or operating variables. Their use for optimization is not a straightforward task as it demands for considerable manual and trial-and-error efforts by the user to adjust the variables to attain the objective function. In this paper, we propose a simulation-optimization framework for evaluating the CO_2 capture performance from a flue gas stream of power plant. The term "simulation" describes the use of simulator as tool for simulating the output of the process plant in response to changes in the input variables. The "optimization" signifies the application of mathematical optimization algorithm to find the most optimum variable settings to improve the objective functions that are not defined by explicit mathematical equations but a simulation model. Herein lies the novelty of our approach – the simulation and optimization of the process is performed simultaneously in an automated fashion for fully exploring the trade-off surface of the CO_2 capture efficiency and operating cost. In the next section, we discuss our proposed framework and apply it to solve an industrial case study.

2. Simulation-Optimization Framework

Detailed mechanisms of CO_2 absorption into an amine solvent are complex involving simultaneous mass transfer and reaction phenomena. Simulation using commercial process simulators such as HYSYS and Aspen Plus are advantageous as they offer good user-interface and reliable property packages for modeling and simulation of such complex process.

2.1 HYSYS simulation of CO_2 removal by amine absorption process

The basic principle of CO_2 removal process involves contacting the flue gas stream and the amine solution in a counter-current direction - 'lean' amine flows from the top while gas stream from the bottom. The reaction between CO_2 and amine (NRH_2) is exothermic forming a water soluble salt according to the following expression:

$$CO_2\,(g) \;?\; CO_2\,(\text{absorbed}) \tag{1}$$

$$CO_2 + NRH_2 \;?\; RH_2^{+}NCOO^{-} \tag{2}$$

$$RH_2^{+}NCOO^{-} + NRH_2 \;?\; RH_2NCOO^{-}NRH_2^{+} \tag{3}$$

High removal of CO_2 is possible and depends on the absorption and reaction rates as well as the equilibrium conditions. The 'rich' amine exiting the bottom of the absorber is sent to a stripper column for regeneration. Prior to entering the stripper column, this 'rich' amine is preheated in a heat exchanger by the 'lean' amine leaving the stripper. In the stripper, the reaction is reversed with CO_2, having been liberated from the amine solvent, leaves through the top of the column. This CO_2 stream is high in purity and can be sent to a storage or used for other purposes. The 'lean' amine is then cooled and recycled back to the absorber.

We have used HYSYS simulator as the modelling and simulation tool. The procedure starts with specifying amines property package as the thermodynamic model. For this, we have selected Li-Mather electrolyte model (Li and Mather, 1994) as the equilibrium model. Next, we construct the flowsheet of the process through "drag-and-drop" of different unit operations and connecting them altogether. The unit operation for the absorber is a tray column (with 10 stages) operating under atmospheric pressure. The solver model for this absorber is HYSIM Inside-Out algorithm with fixed damping factor. For the stripper, a distillation column (12 stages) with reboiler and condenser is used. The solver model for the stripper is based on HYSIM Inside-Out algorithm with adaptive damping factor. Next, the simulation is tested for convergence. The models for both columns are highly nonlinear – all the equations including thermodynamic equilibria, gas and liquid mass balances and chemical reactions need to be solved simultaneously at each stage of the column. To ensure flowsheet convergence, several trial-and-errors are performed for specifying the column parameters.

2.2 Multi-objective simulated annealing optimization

The next step is to optimize the process. In this case, only variables which incur insignificant capital cost are selected as the optimization variables - these are amine flowrate, concentration, and reboiler temperature of the stripper. While the current version of HYSYS is equipped with an optimizer module, we did not use this functionality of HYSYS but instead applied multi-objective stochastic optimization algorithm. This is due to the fact that the optimizer module of HYSYS is designed for solving single objective problems, while in our approach two objective functions are to be simultaneously optimized - maximization of CO_2 capture and minimization of cost. While methods are available to convert multi-objective into single objective function, they are limited in their ability to explore the trade-off surface (Smith *et al*., 2004).

We have applied a multi-objective simulated annealing – jumping genes algorithm for optimization. The basic principle of simulated annealing is derived from the statistical mechanics of annealing of metals. The goal of the annealing is to reach the global minimum state of energy through random search method. The advantage of this algorithm lies in its ability to escape from local optima by uphill moves. The algorithm has recently been extended to multi-objective optimization problems. The concept of jumping genes (or transposons) in multi-objective simulated annealing was introduced by Sankararao and Gupta (2007). They found that by replacing a segment of binaries (genes) with randomly generated jumping genes (transposon), the optimal solution would converge more rapidly due to higher genetic diversity. The jumping genes technique has been applied to the multi-objective simulated annealing (MOSA) of Suppapitnarm *et al*. (2000). The basic algorithm can be summarized as follows:

(1) Start by specifying the solution vector x, whose elements are the three decision variables of the process, i.e., amine flowrate, concentration, and reboiler temperature. Specify also the minimum x_{min} and maximum x_{max} values for each of the variables. To initialize, the variable values from the base-case design $x?$ are used as the solution vector.

(2) Initialize the Pareto set with the triplet [$x?$, $f_{capture}$ ($x?$), f_{cost} ($x?$)] as its element. Here, $f_{capture}$ is the CO_2 capture efficiency and f_{cost} is the process operating cost.

(3) Specify an initial annealing temperature T, which is a control parameter. Initially T is set to a large number $T_{initial}$ and then gradually reduced. Specify also the initial size of step change $ST_{initial}$.

(4) For the three decision variables, a constant parameter f_b is first determined with $f_b < 3$. A random number is next generated to identify the location of a decision variable p as one end of the jumping genes that lies between 0 and 3 - f_b. The other end of the jumping genes is set as $p + f_b$.

(5) Perform random perturbation to generate a new solution vector $x??$ in the neighborhood of $x?$,
$$x?? = x? + \mathrm{R}.ST_{initial} \quad (4)$$
where R is the random number between -1 and 1.

(6) Subsequent to the perturbation of the decision variables, the following jumping genes operation is performed:
$$x_i??? = x_i?? + \mathrm{r}\,(x_{i,\,max} - x_{i,\,min}) \quad (5)$$
where $i = p$ to $p + f_b$ is the length of jumping genes, r is the random number between 0 and 1 and $x_{i,\,min}$ and $x_{i,\,max}$ are the minimum and maximum values for the respective variables. Using the candidate solution vector, simulate the process to obtain new objective values $f_{capture}$ ($x???$) and f_{cost} ($x???$).

(7) Compare the objective values of $x???$ with all solutions in the Pareto set. A solution is deemed as non-dominated, if no other solution is superior when all objectives are considered – in this case, higher capture efficiency and lower operating cost. If $x???$ dominates any element of the Pareto set, replace that element with $x???$. If $x???$ is Pareto-optimal with all the elements in the set, then include it in the set.

(8) If $x???$ is dominated, then temporarily accept it as the current solution vector with a probability P defined as
$$P = \min\{1, \exp(-?S_{capture}/\mathrm{T}).\exp(-?S_{cost}/\mathrm{T})\} \quad (6)$$
where
$$?S_{capture} = f_{capture}\,(x???) - f_{capture}\,(x?) \quad (7)$$
$$?S_{cost} = f_{cost}\,(x???) - f_{cost}\,(x?) \quad (8)$$
At the same time, a random number P_{rand} is generated between 0 and 1.

(9) $x???$ is accepted as the new solution only when $P > P_{rand}$; otherwise the earlier solution vector $x?$ is retained.

(10) As T is set as high at the start of search, P is close to 1 and any feasible random move would be accepted. For convergence, periodically the annealing temperature T is reduced using a reduction factor R_T. The step change size is periodically reduced also.

(11) To escape from local optima, periodically, $x?$ is replaced with a selected solution from the Pareto set.

(12) Steps (4) to (11) are repeated for a predefined number of iterations N_{Total} to obtain the complete Pareto solution set.

The above algorithm has been coded in G2 software. A bridge between G2 and HYSYS has been developed using G2-ActiveXLink and the HYSYS-Browser components for two-way automatic data transfer.

3. Case Study: Gas Power Plant

We have tested the framework on an industrial case study. Flue gas stream from a gas power plant is to be washed with DEA solvent. The basic flowsheet of this process is shown in Figure 1 while its operating condition is presented in Table 1. Simulated annealing with 5000 iterations was run. Figure 2 shows the Pareto optimization results for this process. As seen in the figure, an increase in CO_2 removal requires increased

operating cost. In this case, for 80-93% of CO_2 capture, the operating cost incurred is between $52 and 68 per tonne, excluding the costs of transport and sequestration.

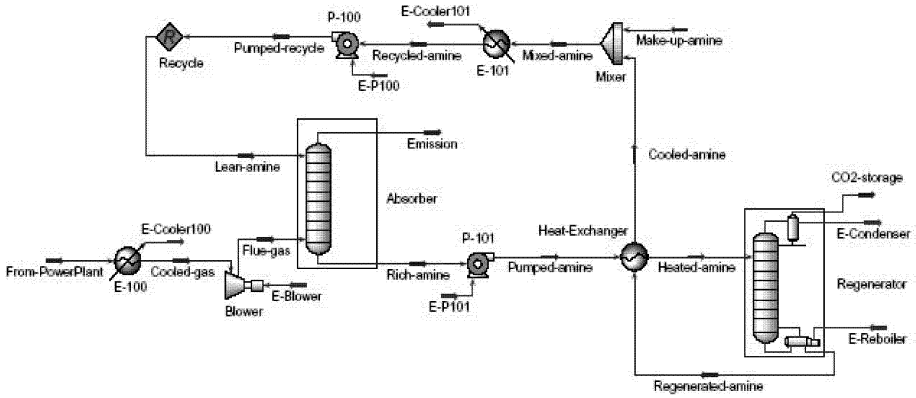

Figure 1. Flowsheet of amine process

Table 1. Amine process information

	Operating value		**Operating Value**
Amine temperature (°C)	40	Flue gas rate (kmol/h)	59,000
Amine pressure (kPa)	110	N_2 (mol%)	69.96
No. stages in absorber	10	CO_2 (mol%)	9.74
No. stages in stripper	14	O_2 (mol%)	13.79
CO_2 purity (mol%)	98	H_2O (mol%)	6.51
		Gas temperature (°C)	55
		Gas pressure (kPa)	110
	Decision variables		**Cost**
Amine rate (kmol/h)	300,000-350,000	Make-up amine ($/kg)	0.4
DEA concentration (wt%)	0.25-0.35	Electricity ($/kWh)	0.07
Reboiler temperature (°C)	118-121	Cooler ($/kWh)	0.0035
		Condenser ($/kWh)	0.006
		Reboiler ($/kWh)	0.032

4. Conclusions

We have described a simulation-optimization framework for evaluating CO_2 capture process using DEA solvent. The framework has been developed by integrating HYSYS simulator with a jumping gene based multi-objective simulated annealing technique and applied to CO_2 capture process from a gas power plant. It has been shown to be capable of generating Pareto optimal solution involving CO_2 capture efficiency and operating cost. Such Pareto set will form the basis for comparison with other amine technology. Thus, our future work will include using other amine solutions as well as mixture of amines. Application to coal or fuel based power plant will also be part of future investigation.

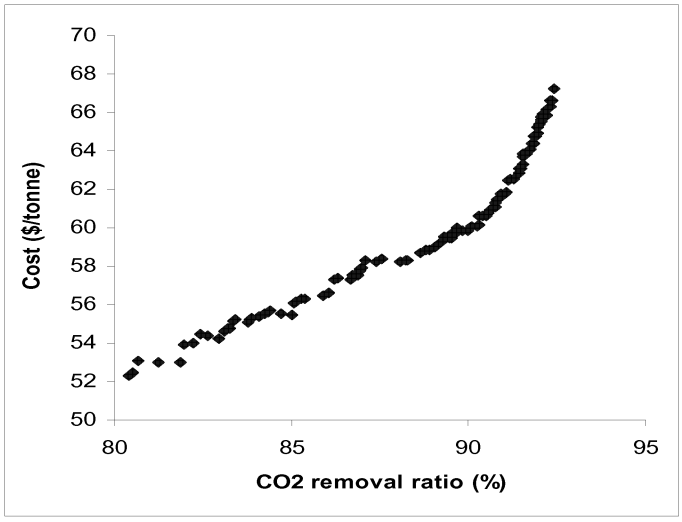

Figure 2. Pareto optimization results for amine process

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