CHEMICAL ENGINEERING TRANSACTIONS
VOL. 61, 2017
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.aidic.it/cet
Guest Editors: Petar S Varbanov, Rongxin Su, Hon Loong Lam, Xia Liu, Jiří J Klemeš

ISBN 978-88-95608-51-8; **ISSN** 2283-9216
DOI: 10.3303/CET1761284

Rigorous Design Optimisation for Combined Process of Raw Natural Gas Treating and CO_2 Compression using Surrogate Models

Kai Liu, Bingjian Zhang*, Qinglin Chen

School of Chemical Engineering and Technology, Guangdong Engineering Technology Research Center for Petrochemical Energy Conservation, Sun Yat-Sen University, Guangzhou, 510275, China
zhbingj@mail.sysu.edu.cn

Raw natural gas treating processes remove enormous CO_2 and consume a lot of energy. The CO_2 can be further compressed for storage or utilization to promote environmental protection. For the purpose of economic design of the combined process of raw natural gas treating and CO_2 compression, a new superstructure combining CO_2 removing and compression processes is presented. In the superstructure, the rich solvent is not regenerated in the distillation column but through a series of flash separators at different operating pressures and temperatures. The removed CO_2 with varying pressures is then compressed to meet the transportation pressure separately. The optimal process structure and operating conditions are determined by minimizing the total annual cost (TAC). To obtain a good trade-off between calculation accuracy and efficiency, a surrogate based optimization framework is presented to address the NLP problem. The complex unit operation models are represented by the Kriging surrogate models, which are built from training data generated via Aspen HYSYS. The surrogate models are then incorporated into the mathematical superstructure framework for optimization. The case study results indicate that the proposed process achieves a significant TAC decrease (9 % - 21 %).

1. Introduction

Natural gas is becoming more and more favourable because it's clean and environmental friendly. Raw natural gas often contains 4 – 50 % of CO_2. The CO_2 is separated from raw natural gas, and directly emits into the environment. To alleviate the greenhouse effect, it is essential to compress and transport the removed CO_2 to storage reservoirs or utilization sites (Tapia et al., 2017). Unlike flue gas in post-combustion CO_2 capture process, the pressure of raw natural gas is often lager than 1.5 MPa, while the gas mixture in power plants is about 0.1 MPa. The CO_2 is often captured by amine solvent with high pressure in the absorption column and separated from the amine with low pressure in the distillation column. Svensson et al. (2004) indicated that the CO_2 should be the liquid or supercritical/dense phase for efficient transport. The high-pressure rich solvent is decompressed for regeneration, while the removed low-pressure CO_2 is compressed for transportation, therefore, it is possible to use the high pressure of raw natural gas to reduce the power requirement of CO_2 compression.

A lot of research has been devoted to the optimal operation of the CO_2 removing process. Mores et al. (2011) proposed an equilibrium stage mathematical model to determine the best operating conditions of the CO_2 post-combustion process. The proposed model was based on an equilibrium model which was not as rigorous as a rate-based model, Cho et al. (2015) proposed a simulation-based optimization strategy to optimize the arrangement and operating conditions of the CO_2 removing process. Cho's model was more rigorous than the complete mathematical model because it integrated the rigorous commercial simulator into the optimization framework. Since the simulation calculation was less efficient than the equation-based model, the strategy considered only a number of possible modification.

Recently, surrogate models have become a popular method for process optimization. Caballero and Grossmann (2008) presented an algorithm to use surrogate models in modular flowsheet. Cozada and Sahinidis (2014) introduced an automated learning surrogate models for simulation-based optimization. Quirante et al. (2015)

Please cite this article as: Liu K., Zhang B., Chen Q., 2017, Rigorous design optimisation for combined process of raw natural gas treating and co2 compression using surrogate models, Chemical Engineering Transactions, 61, 1717-1722 DOI:10.3303/CET1761284

proposed a framework for rigorous design of distillation columns using surrogate model. Eason and Biegler (2016) proposed a trust region filter method for glass box/ black box optimization and the convergence of the method to KKT points was proved.

However, few researchers have studied the possibility to make full use of the high pressure of raw natural gas to reduce the CO_2 compression energy. The aim of this paper is to present a new combined process of raw natural gas treating and CO_2 compression to address the problem and propose a surrogate based optimization framework to optimize the process. The presented process consists of two major parts, the absorption column and the multi-stage flash and compression process, which acts like regeneration column in conventional process while offers more flexibility for amine regeneration. A superstructure is proposed to represent possible structures of the process. Kriging surrogate models are used to emulate the absorption column and the multi-stage flash and compression process using data generated by commercial simulator. The obtained surrogate models are incorporated into the mathematical superstructure for process structure and operation optimization.

The paper is organized as follows. The proposed combined process is described first, followed by the superstructure of the process. Next, a description surrogate models is presented, and the surrogate based optimization methodology model is proposed. Finally, an application of the proposed methodology is presented. The results of the application are discussed before conclusions and perspectives are provided.

2. Process superstructure and description

To utilize the pressure energy of the raw natural gas, a combined raw natural gas treating and CO_2 compression featured by multi-stage flash solvent regeneration process is presented. Figure 1 is the superstructure representing possible layouts of the process. In the superstructure, the regeneration column is replaced by the multi-stage flash process. There are k (k = 1, 2, 3… K) stages of flash and compression processes. Sour gas enters the absorption column at the bottom and exits the column from the top as sweet gas. The absorption column has two lean solvent feeds, one is at the top of the column denoted as Fl and the other is at the middle of the column denoted as Fs. The solvent inlet temperatures and operating pressure are fixed at 40 °C and 6,000 kPa. The rich amine solvent Fr exits the absorption column and enters into the flash SEP_k after heating in HE_k, and then the liquid from the SEP_k is split into three possible streams, Fn_k, Fl_k and Fs_k. Fn_k goes to the next stage, Fl_k is pumped in the PPL_k and mix with $Fl_{k'}$ from other flash processes to feed the absorption column at the top, and Fs_k is pumped in the PPS_k and mix with $Fs_{k'}$ from other flash processes to feed in the middle of the absorbing column. The gas from the SEP_k is cooled in CE_k and then enters decanter DEC_k to remove water, the rest of the gas will be compressed in series compressors COM_k, which consists of four compressors and coolers. Finally, the compressed CO_2 from each stage merges denoted as CC.

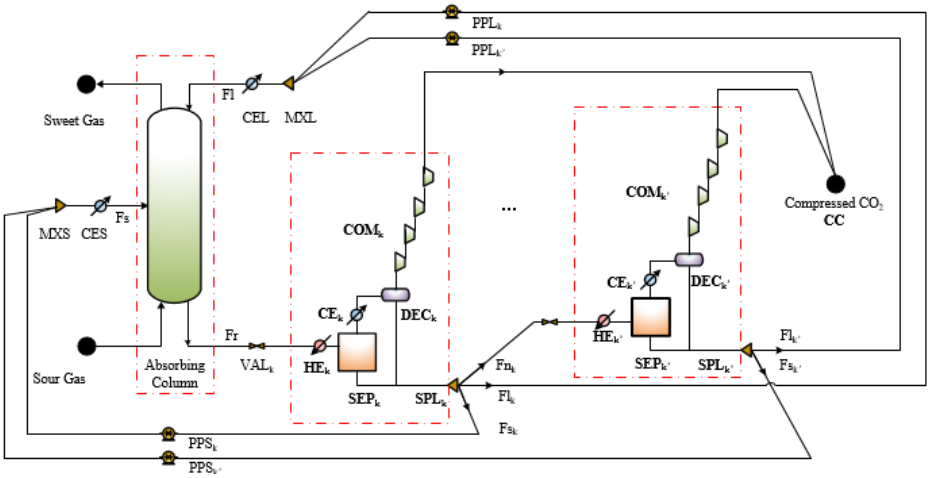

Figure 1: A process superstructure of the multi-stage CO_2 removing and compression

3. Surrogate models

Surrogate models are capable of constructing cheap-to-evaluate models to emulate the response of the expensive inexplicit black box functions. The Kriging interpolation is one of the most used surrogate models. The Kriging method views all the observed responses as if they are a stochastic process. The random field has a mean of 1μ (1 is an n×1 column vector of ones) and variance σ^2. The random variables are correlated with each other using the basis function expression

$$\mathrm{cor}\left[Y\left(x^{(i)}\right), Y\left(x^{(l)}\right)\right] = \exp\left(-\sum_{j=1}^{k} \theta_j \left| x_j^{(i)} - x_j^{(l)} \right|^{p_j}\right) \quad (1)$$

With p_j often fixed at 2, the values of parameters μ, σ^2 and θ_j are estimated to maximize the likelihood of the obtained sample data:

$$L = \frac{1}{(2\pi\sigma^2)^{n/2}|\Psi|^{1/2}} \exp\left[-\frac{(y - 1\mu)^T \Psi^{-1} (y - 1\mu)}{2\sigma^2}\right] \quad (2)$$

Where L is the likelihood, and Ψ is an n×n correlation matrix of all the observed data.
The new prediction $\hat{y}$ at x should also be consistent with the observed data and the estimated parameters. The augmented correlation matrix $\tilde{\Psi}$ with the addition of $\hat{y}$ is calculated to replace Ψ. With all the parameters constant, the likelihood function is determined by the prediction $\hat{y}$. And $\hat{y}$ can calculated through maximizing the likelihood function. The mathematical details of the surrogate models can be found in the book by Forrester et al. (2008).
The proposed process consists of two complex chemical processes, the absorption column and the separation and compression processes. Kriging surrogate models are used to substitute the two processes. In this way, the complex physical and chemical equations will be omitted and a more tractable model is obtained. In this study, the loss of amine and water is ignored. For the absorber column, the input variables include solvent (excluding absorbed gases) flow rate F (kg/s), the split ratio of the regenerated solvent z, the CO_2 mole concentration of semi-lean solvent Cs, and the CO_2 mole concentration of lean solvent Cl. The output variables include the purified gas CO_2 mole fraction Cp, the rich solvent flow rate Fr (kg/s), the annualised capital cost of the absorption column Mc ($/y), and the temperature of the rich solvent Tr (°C). The surrogate model of the absorption column can be conceptually written as follows:

$$[Cp, Cr, Mc, Tr] = Kr(F, z, Cs, Cl) \quad (3)$$
$$5 \le F \le 20 \quad (4)$$
$$0 \le z \le 1 \quad (5)$$
$$0 \le Cs \le 0.05 \quad (6)$$
$$0 \le Cl \le 0.03 \quad (7)$$

For the CO_2 removing and the compression processes, the input variables include inlet rich solvent(excluding absorbed gases) flow rate Fr (kg/s), its CO_2 concentration Cr, the flash temperature Tf (°C) and the flash pressure Pf (kPa). The output variables include the CO_2 concentration of the outlet solvent Ct, the temperature of the outlet solvent Ts (°C), the operating cost Mo ($/y), and the annualised capital cost Ms ($/y). In each stage, the number of compressors is fixed at 4, and the compression ratios of all the compressors are equal. The surrogate model of the CO_2 removing and the compression processes can be conceptually written as follows:

$$[Ct, Tt, Mo, Ms] = Kr(Fr, Cr, Ts, Ps) \quad (8)$$
$$0 \le Fr \le 20 \quad (9)$$
$$0 \le Cr \le 0.05 \quad (10)$$
$$110 \le Ts \le 130 \quad (11)$$
$$150 \le Ps \le 900 \quad (12)$$

4. Optimisation framework

Figure 2 shows the surrogate based optimization framework that is used to optimize the proposed process, as described below.

1) In the preparation step, s (s = 1, 2, 3 … S) models will be selected to be surrogated, and the input variables and output variables will be identified for each model alongside their bounds.
2) In the sampling step, a design of experiments will be conducted to obtain n pairs of initial sampling points X_s for surrogate model s. Latin Hypercube sampling method will be applied in this paper to make the sample points space-filling.
3) In the data collection step, HYSYS is used to generate data pairs $[X_s, Y_s]$ according to the sampling. Notably, the normalization of the variables is necessary to avoid having large values dominate the results of the calculation.
4) In the building step, the kriging surrogate models will be used to build the mapping between the input and output variables, $\hat{Y}_s = f_s(X_s)$
5) In the formulation process, the surrogate models will be integrated with the rest of the process equations to form an NLP model. The number of flash stages starts with k = 1.
6) In the optimization step, the NLP model will be solved within MATLAB, the minimum TAC is $TAC_{min,k}$, and

the optimal input and output variables for each surrogates are $[x^*_{s,k}, \hat{y}^*_{s,k}]$. Compare the Kriging model predicted results $\hat{y}^*_k$ with the simulation results y^*_k at the optimal point x^*_k. If the relative error smaller than tolerance, go to the next step; if not, update the surrogate models with the point $[x^*_{s,k}, y^*_{s,k}]$ and go back to step 4.

7) For one-stage flash process, set k = k+1, and go back to step 5; for process with k (k > 1) stages of flash processes, compare the TAC_k with the TAC_{k-1}. If TAC_k greater than the TAC_{k-1}, end the process, the optimal number of flash stages will be k-1, and the minimum TAC is TAC_{k-1}; if not, set k = k+1 and go back to step 5.

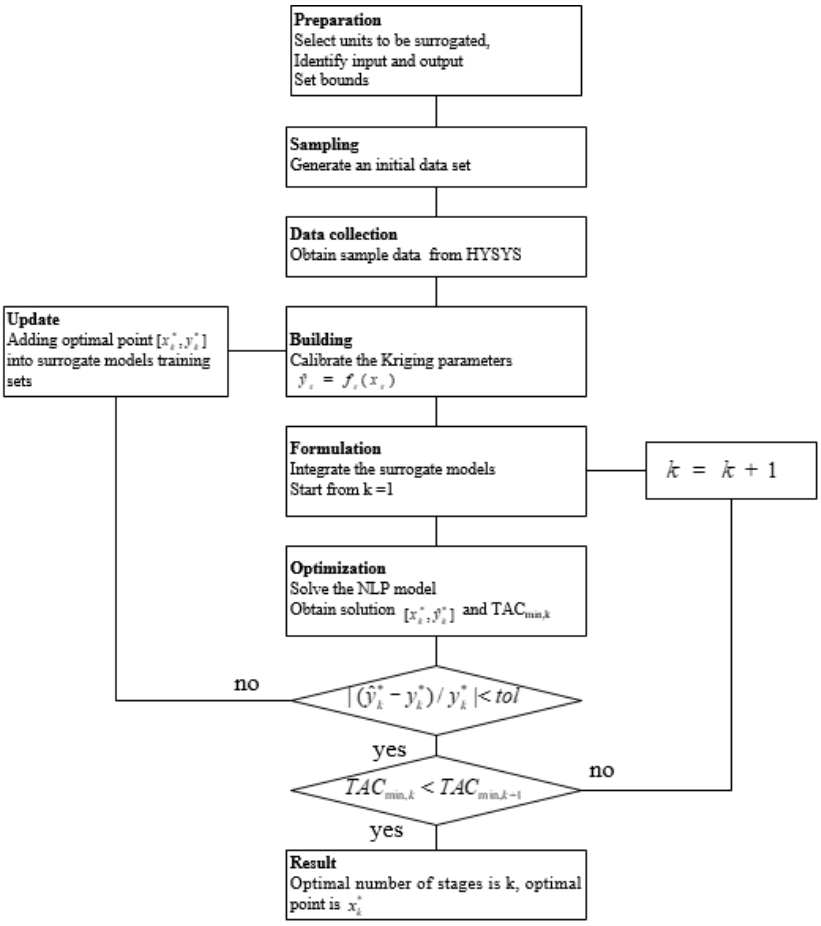

Figure 2: Flowchart of the optimization framework

5. Case study

The raw natural gas composed of 85 % CH_4, 5 % C_2H_6 and 15 % CO_2, and the inlet pressure of the raw natural gas is 6,000 kPa. The specification for CO_2 mole fraction in the purified natural gas is ranging from 1 % to 3 %. Figures 3 and 4 show the relative errors in testing points for separation and compression surrogate model and the absorption column surrogate model. The relative errors are all less than 3 %, which indicates the effectiveness of the surrogate models.

In Figure 5, the line indicates the TAC of the CO_2 removing and compression process, the dotted line indicates the TAC of the conventional process using distillation column to regenerate the rich amine solvent. The bars indicate the optimal number of stages for the new process. As illustrated, the new process has lower total annual cost under given conditions. When the sweet gas CO_2 requirement is beyond 2.6 %, one-stage process is preferred, otherwise, two-stage process is favoured.

Figure 6 is the optimal process structure with 1 % CO_2 in sweet gas. The optimal number of stages is two, and the minimal TAC is 1.85 M USD. The feed to the middle of the absorption column is omitted. The total flow rate of amine solvent is 7.32 kg/s, all the lean solvent from the first stage of flash process goes to the second stage and finally enters the absorption column at the top. The pressure and temperature in the first flash stage is 488 kPa and 130 °C. The pressure and temperature in the second stage is 150 kPa and 121 °C.

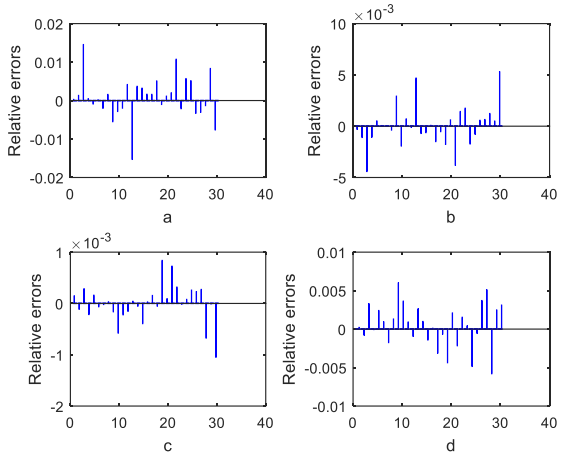

Figure 3: Relative errors in the tested points for absorption column (a: CO_2 contents in sweet gas, b: CO_2 contents in rich solvent, c: capital cost for absorption column, d: temperature of rich solvent)

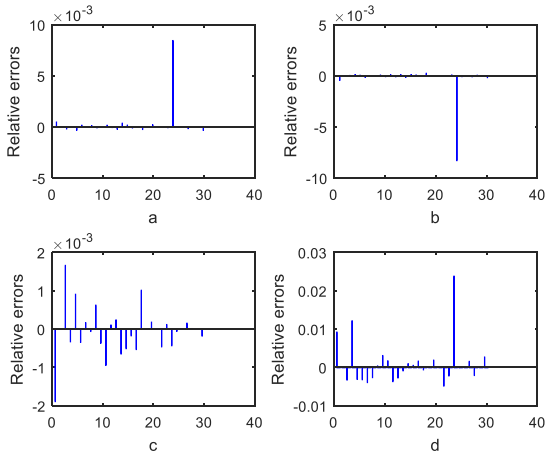

Figure 4: Relative errors in the tested points for CO_2 capture and compression process (a: CO_2 content in lean gas, b: temperature of lean solvent, c: operating cost for flash process, d: capital cost for flash process)

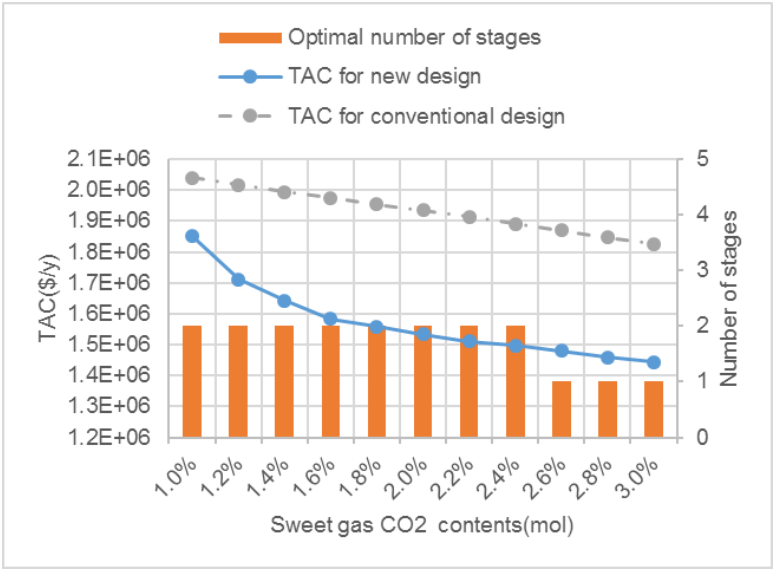

Figure 5: The optimal TAC and number of stages in the variety of sweet gas CO_2 contents

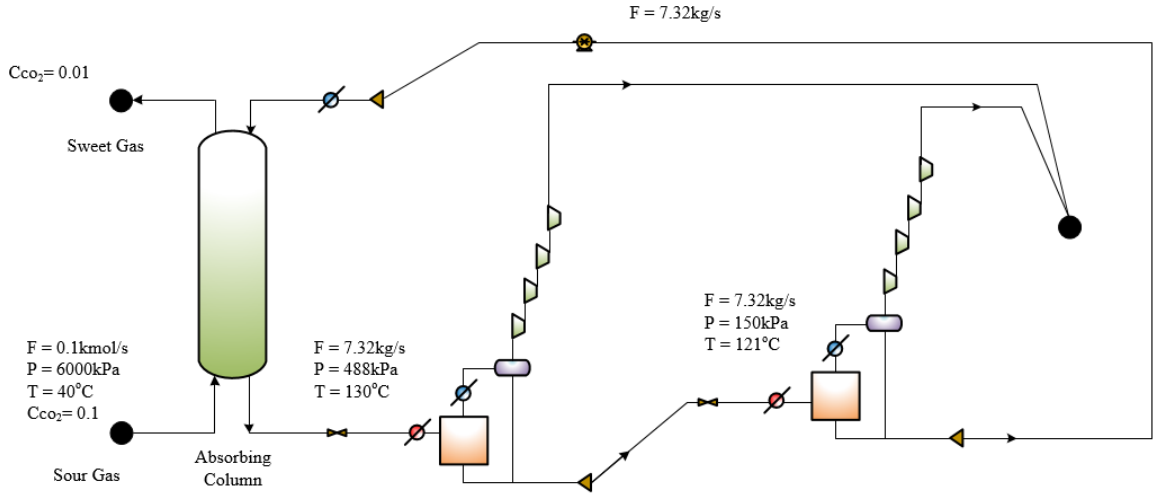

Figure 6: The optimal process structure and operating conditions with 1 % CO_2 in sweet gas

6. Conclusion

In this work, a superstructure for multi-stage CO_2 removing and compression process in natural gas refining process is presented. To obtain a better trade-off between model accuracy and solving efficiency, a surrogate-based optimization approach is proposed to address the problem. In addition, case study is carried out to optimize the process in a variety of different sweet gas purity requirements. The new process indicates significant TAC decrease (9 % to 21 %). In addition, the presented process can also offer flexible operation for natural gas refining plants when sweet gas CO_2 requirement is changing. According to the calculation results, when the sweet gas CO_2 requirement is beyond 2.6 %, one-stage process is preferred, otherwise, two-stage process is favoured. In summary, the proposed approach can be applied to explore novel process design and optimize operating parameters for complex chemical processes in an efficient way.

Acknowledgments

This research is supported by the National Natural Science Foundation of China (No. 21276288, 21376277), and the project of Guangdong Provincial Natural Science Foundation of China (No. 2015A030313112).

References

Caballero J.A., Grossmann I.E., 2008. An Algorithm for the Use of Surrogate Models in Modular Flowsheet Optimization. AIChE J. 54, 2633–2650.

Cho H., Binns M., Min K.J., Kim J.K., 2015. Automated Process Design of Acid Gas Removal Units in Natural Gas Processing. Comput. Chem. Eng. 83, 97–109.

Cozad A., Sahinidis N. V., Miller D.C., 2014. Learning Surrogate Models for Simulation-Based Optimization. AIChE J. 60, 2211–2227.

Eason J.P., Biegler L.T., 2016. A Trust Region Filter Method for Glass Box/Black Box Optimization. AIChE J. 62, 3124–3136.

Forrester A.I.J., Sbester A., Keane A.J., 2008. Engineering Design via Surrogate Modelling. John Wiley & Sons, Ltd, Chichester, UK.

Mores P., Scenna N., Mussati S., 2011. Post-Combustion CO_2 Capture Process: Equilibrium Stage Mathematical Model of The Chemical Absorption Of CO_2 into Monoethanolamine (MEA) Aqueous Solution. Chem. Eng. Res. Des. 89, 1587–1599.

Quirante N., Javaloyes J., Caballero J., 2015. Rigorous Design of Distillation Columns using Surrogate Models Based on Kriging Interpolation. AIChE J. 61, 2169–2187.

Svensson R., Odenberger M., Johnsson F., Stromberg L., 2004. Transportation Systems for CO_2 — Application to Carbon Capture and Storage. Energy Conversion and Management, 45(15), 2343-2353.

Tapia J.F., Promentilla M.A., Tseng M.L., Tan R.R., 2017, Selection of CO_2 Utilization Options in Carbon Capture, Utilization and Storage (CCUS) Systems using Analytic Hierarchy Process - Data Envelopment Analysis (AHP-DEA) Approach. Chemical Engineering Transactions, 56, 469-474