Available at www.sciencedirect.com

ScienceDirect

journal homepage: www.elsevier.com/locate/he

Reduced superstructure solution of MINLP problem in refinery hydrogen management

M. Khajehpour, F. Farhadi, M.R. Pishvaie*

Chemical & Petroleum Engineering Department, Sharif University of Technology, # 593, Azadi Ave., Tehran, Iran

ARTICLE INFO

Article history:
Received 29 January 2009
Received in revised form
28 August 2009
Accepted 28 August 2009
Available online 7 October 2009

Keywords:
Optimization
Hydrogen management
Refinery
Superstructure
Reduced superstructure
Mixed integer nonlinear programming
Genetic algorithm

ABSTRACT

Minimizing Hydrogen waste into fuel gas within the H_2 network in a refinery is the objective function of an optimization problem in this paper. The superstructure obtained for a refinery wide concept, is first solved and validated for literature cases, then is reduced by heuristic rules, based on engineering judgment. The reduced superstructure contains all simulation procedures of pseudo-components definitions, fine tunings of all unit operations to reach actual operating conditions, reactions characterization, linear and nonlinear equalities and inequalities as system constraints. The set of governing equations are solved with Genetic Algorithm. Based on this optimization, in an Iranian refinery 22.6% reduction of H_2 production and a saving of 1.19 million $/year could be achieved.

1. Introduction

The world's energy demand, driven by the economic growth, is increasing at a high rate. The fossil fuels, as the most cost effective sources of energy production, are depleting. The viable alternatives should comply with the awareness of growing environmental concerns; among the criteria which make an alternative process-wise acceptable, one may name but a few: renewable, sustainable, efficient, cost effective and safe. Among politicians, environmentalists, and scientists an ongoing discussion is underway to accept and adopt the most suitable alternative process. The hydrogen has been expected as the most prominent resource [1]. Since the crude oil crisis waves of 70s and 90s, processes like electrolysis and coal gasification has emerged. But these processes are still under doubtful inspections for environmental or economical acceptance. Nevertheless any such decision should satisfy both "external" and "internal" interactions for "policy planning" and "managerial decision making" [2], while global warming and carbon-free nature of the process should be kept as a constraint.

As hydrogen can be found in nature only as compounds, a great amount of energy is needed to produce it [3]. Many authentic sources argue that hydrogen has the potential of overcoming its development obstacles including the global warming issue. They believe that hydrogen would be the fuel of future and it seems to solve the environmental issues as it looks to be a green energy carrier [4]. The hydrogen as a continuous and renewable energy source is one of the alternatives to succeed the current fossil fuels energy system.

* *Corresponding author*. Tel.: +98 21 66005819.
E-mail address: farhadi@sharif.edu (F. Farhadi).

0360-3199/$ – see front matter © 2009 Professor T. Nejat Veziroglu. Published by Elsevier Ltd. All rights reserved.
doi:10.1016/j.ijhydene.2009.08.086

Nomenclature

Symbols

C_P	heat capacity at constant pressure, J/kg °C
C_{Heat}	cost per unit of heat energy
F	flowrate, Kmol/h
H_2	Hydrogen
P	Pressure, Pa
T	Temperature, K
y	mole fraction of H_2
L	Lower bounds flow-rate which can be sent to new equipments
U	Upper flow-rate which can be sent to new equipments

Greek letters

ΔH^0	enthalpy of formation
ρ	density, kg/m^3
η	compressor efficiency
α	ratio of specific heats

On a long term basis, the substitution of fossil fuel by hydrogen will need a solid base large scale production process. The actual economically feasible process for large scale H_2 production is natural gas–steam reforming, mostly called steam–methane reforming (SMR) [5]. However in short-term time, the optimization and the improvement of existing hydrogen production technology is a necessity [6]. On the other hand, more strict environmental regulations and standards led refiners to increase the use of H_2. Thus, H_2 production, consumption and management in the industry, for a more efficient use of this valuable material, should be reconsidered thoroughly. Although the modification of each single process has merits, but the interaction between all integrated processes in a refinery, determines finally the system's performance.

One available option to ameliorate the H_2 distribution system is to increase its purity in one or more H_2 sources. The stream with higher H_2 purity will provide the system with more H_2 per unit flowrate. This surplus of H_2 will reduce the need of fresh H_2 production and increase the recycling [7].

Two main methods are practiced for an efficient H_2 management system: graphical (mainly to search for the pinch point) and mathematical approach.

Alves analyses the refinery H_2 distribution by graphical targeting approach [7]. Hallalea describes the superstructure method applying to the system for finding the optimal solution for H_2 distribution [8].

The main disadvantage of graphical approach is that it considers solely the purity and flowrates of streams, while the pressure of sources and sinks also has to be considered. In the case that the source's outlet pressure is less than sink's inlet, a compressor should be used to satisfy the destination pressure. Compressor is one of the major investment expenses in a refinery. However, this method will give a theoretical solution which is not necessarily applicable in a real system. On the other hand, this approach will find the minimum use of H_2, while the commercial and environmental aspects have to be considered as well.

2. Hydrogen management in a typical refinery

There are several processes for H_2 production in the refining industry and many applications where it is consumed. Among most common production processes are steam methane reforming (SMR) and partial oxidation (POX). In SMR process, the most widely used H_2 production route, a mixture of steam and methane flows in a fixed bed catalytic reactor to produce a mixture of H_2, CO_2 and CO, a mixture known as synthesis gas. The water gas shift reaction (WGS) produces an additional amount of hydrogen in two phases of high and low temperature [4]. The H_2 is separated from the mixture by different physical, like Pressure Swing Adsorption (PSA), or chemical processes like CO_2 and CO's chemical absorption by solvents.

3. Hydroprocessing

The Hydroprocessing is the major H_2 consumer. Lighter hydrocarbon cuts are richer in hydrogen, which in turn affects the Gasoline, Kerosene, and Diesel cuts quality. Tough regulations are the cause of most hydrogen-demanding sink within the overall H_2 balance in a refinery with an acceptable environmental impact [9].

Three products criteria have to be considered by refiners: lighter, higher performance, and environmentally acceptable. Sulfur and Nitrogen, as well as undesirable hydrocarbon components like aromatics, are removed by hydroprocessing to meet product specifications and satisfy environmental regulations [9].

4. Imperfections of superstructure

The main difficulty within the superstructure method is its complexity in real large networks which may cause the problem not to be solved at all. Therefore the system becomes so complicated and large that the existing problem solving tools are either inadequate or they require long time to get results. Conventional methods have difficulties like large computation, long CPU time, and also encountering local optima which makes them incompatible. In this paper it is suggested to reduce the superstructure with a sense of process engineering before applying the optimization methods. Thus, it is vital to remove some improper complexities without losing the accuracy, in addition to use some simplifying assumptions.

5. Formulation of objective function

Considering all possible connections between sources and sinks is necessary before formulating the objective function.

The general Mixed Integer NonLinear Programming (MINLP) formulation can be stated as [10]:

$$\begin{aligned}&\text{Minimize } f(x,y) \text{ objective function}\\&\text{subject to } \quad h(x,y)=0\\&g(x,y)\leq 0\\&x\in X\subseteq\mathbb{R}^n\\&y\in Y:\text{integer}\end{aligned} \tag{1}$$

Here x represents a vector of n continuous variables (e.g., flows, pressures, compositions, temperatures, sizes of units), and y is a vector of integer variables (e.g., devices); $h(x, y)=0$ denote the m equality constraints (e.g., mass, energy balances); $g(x, y)\leq 0$ are the p inequality constraints (e.g., specifications on purities, environmental regulations, logical constraints); $f(x, y)$ is the objective function (e.g., annualized total cost, profit).

Minimizing fresh H_2 generation, operating cost or the total cost may be considered as target function. There are some classes of constraints imposed on the system, although the constraints are independent from the objective function type.

1. Sink requirements

Both the amount of gas fed and the H_2 purity (partial pressure) at the sink inlet must be kept constant.

$$\sum_i F_{i,j}=F_{\text{sink},i} \tag{2}$$

$$\sum_i F_{i,j}y_i=F_{\text{sink},j}Y_{\text{sink},j} \tag{3}$$

2. Source availability

The amount of gas available from each source must equal the total amount sent to the sinks and fuel system.

$$\sum_j F_{i,j}=F_{\text{sources},i} \tag{4}$$

3. Compressors (mass balance and capacity)

Because of their nature, compressors are considered as both sinks and sources, different from other hydrogen consumers [8]. The purity as well as the flowrate of compressor streams can be varied, while these parameters are constant in the H_2 consumers.

Mass balance:

$$\sum_j F_{\text{comp},j}=\sum_j F_{i,\text{comp}} \tag{5}$$

$$\sum_j F_{\text{comp},j}Y_{\text{comp}}=\sum_j F_{i,\text{comp}}y_i \tag{6}$$

Capacity of compressor:

$$\sum_i F_{i,\text{comp}}\leq\sum_i F_{\text{max, comp}} \tag{7}$$

Several streams may be mixed at the compressors suction, so the H_2 purity is not known and the problem would be a nonlinear programming.

4. The total mass balance for hydrogen on the whole system

$$F_{\text{fuel}}=F_{H_2\text{ plant}}+\sum_{i=1}F_{\text{source},i}-\sum_{j=1}F_{\text{sink},j} \tag{8}$$

5. Adding new facilities to the system

If adequate investment resources are readily available, new facilities, like compressors and absorbers might be added. Incorporating these facilities can be represented by using binary variables. For each binary variable two logical constraints have to be added as follow [11]:

$$\sum_{i=1}^{n}F_{i,\text{new equipment}}-e_{\text{new equipment}}\times U\leq 0 \tag{9}$$

$$\begin{aligned}&\sum_{i=1}^{n}F_{i,\text{new equipment}}-e_{\text{new equipment}}\times L\geq 0\\&e_{\text{new equipment}}\in\{0,1\}\end{aligned} \tag{10}$$

5.1. *Minimizing the expenses*

Further to minimize the amount of fresh H_2 generation one may include expenses minimization into the target function. The overall hydrogen usage cost is due to the following four cost components: production, distribution, storage and end-user costs [12].

Major costs of a H_2 system are its generation as utility, plus the electricity cost for compressors, minus the value of H_2 as feed gas [8]. It is assumed that the operating cost of a H_2 utility is directly proportional to the utility flowrate.

Stream inlet–outlet pressures and flowrates are determining the compressors needed power [8]:

$$\text{power}=\frac{C_p\times T}{n}\left(\left(\frac{P_{\text{out}}}{P_{\text{in}}}\right)^{\gamma-1/\gamma}-1\right)\times\frac{\rho_0}{\rho}\times F \tag{11}$$

For simplicity it is assumed that the fuel gas is a mixture of H_2 and methane with a heating value expressed by Eq. (12) [8]:

$$\text{Fuel value}=\left(y\times\Delta H^0_{C,H_2}+(1-y)\times\Delta H^0_{C,H_2}\right)\times C_{\text{Heat}} \tag{12}$$

6. Explanation of reduced or heuristic superstructure

Reducing the superstructure and examining the characteristic of variables before starting optimization procedure leads to eliminate excess or unrealistic variables from the formulation. This reduction is necessary for achieving feasible results faster. Indeed, number of variables of the superstructure would be eliminated with the technical and process engineering initiations.

In this case the following rules have to be considered for H_2 management in a refinery:

1. The material flows only from a higher pressure source to a lower pressure sink inlet. Assuming that new

compressors won't be added to the system, existing compressors should be used for the best performance.

2. A recycle stream is admitted for a reactor but not for other unit operations. Therefore, for the sources except reactors the recycling stream does not have sense and will be eliminated.
3. Some devices need make-up streams to meet the inlet specification. A stream from outlet of a device to its make-up is illegal and should be eliminated.
4. Sending a stream directly from a H_2 production source with high purity to the fuel sink is energy wasting.

Moreover, some variables specified as pressure, temperature or purity of some especial streams must be set to their operating value. These variables should be eliminated from the superstructure as well.

The nature of variables must be determined and the unrealistic variables should be removed from the system formulation before starting the optimization mathematical procedure.

7. Case study: H_2 network in a typical Iranian refinery

Two Steam Reformers are supplying the H_2 in North and South Plants of this 250,000 bpd Refinery. The major feed of these plants is the off-gas streams of Platforming unit with a H_2 content of 60% to 70%.

The major consumer of H_2 in the refinery is the Isomax unit for its hydrocracking reactions. H_2 production and consumption units are interconnected as shown in Fig. 1.

One possible technology for purifying the Hydrogen content in existing streams is PSA. This issue is considered with all operation associated problems like soot formation, filter need and related costs. These constraints may cause PSA exclusion in early stage process selection screening.

Every unit involved in H_2 production–consumption is simulated. To do this all relevant petroleum fractions are first defined as pseudo-components, then the simulation is fine tuned to obtain close results to the actual operating conditions. H_2 consumption and production reactions have been incorporated within the simulation. Afterward, using this information along with system's constraints, in terms of the linear and nonlinear equalities and inequalities, the problem of H_2 management has been solved with Genetic Algorithm.

The reliability of Genetic Algorithm method has been validated by two cases from literature before applying this method for the case study of the considered refinery [13].

7.1. Optimizing the H_2 system in the refinery

The objective here is to minimize the amount of H_2 sent as fuel to the burners. The first subjective of importance of this objective function is the H_2 value. By the use of H_2 in hydrotreating and hydrocracking processes, like the Isomax unit, one can gain economic profits. On the other hand, if off-gas streams are sent to the fuel gas system, H_2 presence would cause some operating problems like corrosion. In addition, reduction of H_2 in the fuel gas has a significant commercial aspect, for heating value of H_2 is less than other components of the fuel gas while its cost is more.

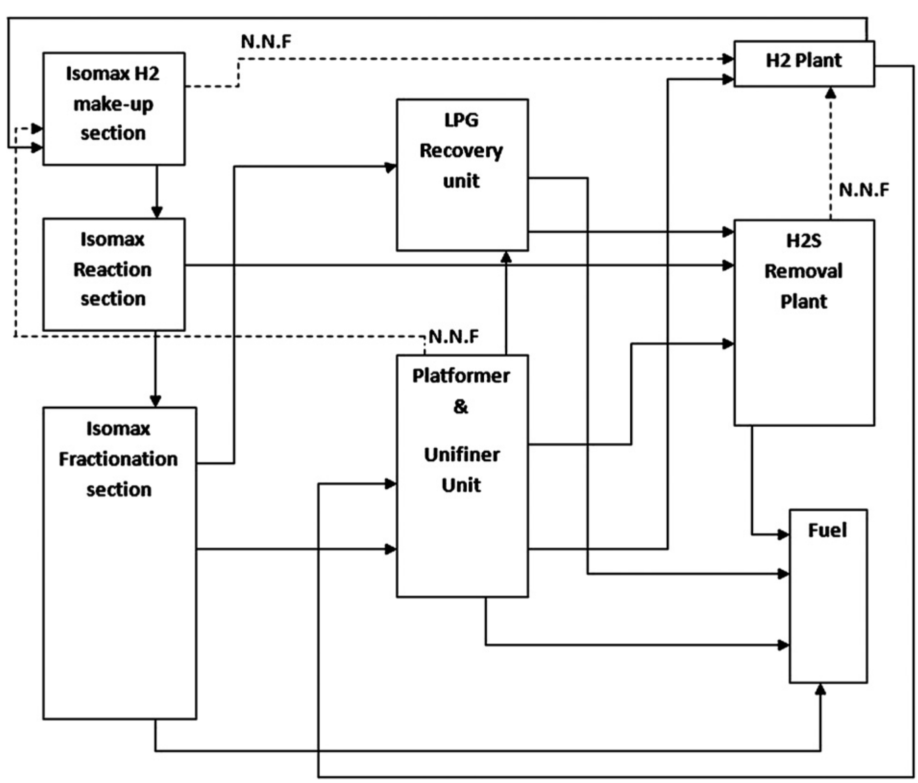

Fig. 1 – H_2 network interconnection in Tehran North Refinery.

Table 1 – Decision variables using reduced superstructure for the H_2 network at Tehran North Refinery.

Source/Sink	H_2 plant	Isomax comp.	Isomax reaction	Isomax fract.	LPG	CRU	Amine	Fuel
H_2 plant		x_1, y_1				x_2, y_1		
Isomax comp.	x_3, y_2		x_4, y_2			x_5, y_2		
Isomax reaction	x_6, y_3			x_7, y_4		x_8, y_3	x_9, y_5 x_{10}, y_3	
Isomax frac.					x_{11}, y_6 x_{12}, y_7	x_{13}, y_8		x_{14}, y_9
LPG unit							x_{15}, y_{10} x_{16}, y_{11}	x_{17}, y_{12} x_{18}, y_{13}
CRU unit	x_{19}, y_{14}	x_{20}, y_{14}			x_{21}, y_{15} x_{22}, y_{16}	x_{23}, y_{14}	x_{24}, y_{14}	x_{25}, y_{14}
Amine unit	x_{26}, y_{16}							x_{27}, y_{16} x_{28}, y_{17}

7.2. *Independent variables*

Flowrate between different units is represented by F_i,i = 1,2,…,62 while the mole percent of H_2 in each flow stream is also shown by y_i,i = 1,2,…,62.

The variation range for each variable is considered between 80% and 120% of its operating value. In the H_2 Plant, the H_2 content of outlet stream varies between 0.92 and 0.97, but for other units, the H_2 concentration has not a specific fixed value.

7.3. *Reduced (heuristic) superstructure*

Based on engineering judgment and the above heuristic, more than half of the variables of superstructure would be eliminated with such technical and process engineering arguments. There would be 28 variable flowrates and 17 variables of H_2 purity in different streams. Values of purities for some streams are dictated by the process. Finally, 45 variables remain to consider for the optimization problem, as shown in Table 1.

7.4. *Equality constraints*

The total and component mass balances of H_2 for each unit constitute the equality constraints. Each unit has been studied and relevant equations are derived.

7.5. *Objective function*

The objective function is to minimize the waste of H_2 into the fuel system:

$$\mathrm{Obj}\dot{\mathrm{F}}\mathrm{un} = \sum_k F_k y_k \quad k = \{34, 43, 44, 53, 61, 62\}$$

In this paper "Genetic Algorithm" – GA toolbox of Matlab – is used for solving the problem of optimization which is based on superstructure's model. Plots of Genetic Algorithm progress for this problem are shown in Fig. 2.

7.6. *Objective function's value*

Objective function is calculated by GA for the actual operating and for the optimized conditions. The relevant values are shown in Table 2.

The optimization shows that the fresh H_2 production rate could be reduced by 22.6% leading a 1.191e6 $/year saving.

In this study the cost of H_2 is the main concern for objective function formulation. While the cost has its own merits, other criteria such as maintenance, space limitations, piping restrictions and extra power requirements are also important to consider. These could be represented as constraints, but for managerial decision making other process limitations like inclusion of PSA unit will completely change the formulation as well as the results.

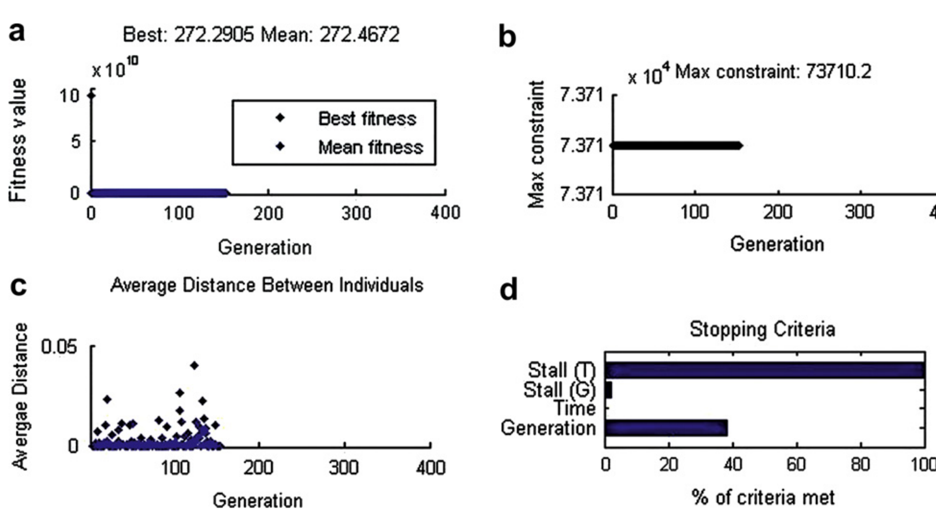

Fig. 2 – Plots of Genetic Algorithm progress for the problem of optimization of the hydrogen network at North Refinery: (a) the changes in best and average value for objective function, (b) the constraint's maximum, (c) varying distances between individual values in each generation, and (d) the stopping criteria for evaluations of the algorithm.

Table 2 – Values for the objective function.

	Unit	Values	
		Operation	Optimized
Obj. Fun. (molar flow)	Kmol/h	139.74	61.793
Hourly cost	$/hr	243.13	107.51
Yearly cost	$/year	2.13 e6	9.41 e5
Production	Kmol/h	1763.61	1365.31

8. Discussion

In order to achieve the most realistic case for the best distribution network of H_2 in a system, it is inevitable to consider all commercial aspects as well as constraints. A more extended formulation which considers all expenses such as piping would be indisputably more generalist.

New recommended methods for MINLP problem solution such as OAGO-F [10] could be applied with appropriate modifications in the system and the control of variables in order to alleviate the objective function.

Other variables, beside the pressure, may affect H_2 production or consumption. For example, catalytic reactors within their normal operating ranges are not pressure sensitive. Very severe pressure surges may damage the catalyst and cause its inactivation. On the other hand, raising the H_2 purity in the feed of one reactor, and hence an increase of H_2 partial pressure, may lead to improve the conversion, catalyst's lifetime and yield. To obtain the impact of such changes, it is necessary to know the details of catalysts kinetic behavior, reactor's operation and also other commercial details of the refinery.

There are several other issues, like environmental regulations and commercial aspects that have to be considered in the optimization. One important example is CO_2 which has some commercial value and is subject to Kyoto protocol regulations. Thus, it is reasonable to consider its production, consumption and capture in refineries.

9. Conclusion

Using reduced superstructure – or heuristic superstructure – it is possible to get the feasible results faster and more justifiable. This means that based on experience and engineering judgment, the structure will be reduced in size and the undesired variables will be eliminated by heuristic rules. The million $/year saving could be achieved without any new equipment addition to the plant and just by tuning process parameters. Further saving can be attained while considering other criteria within the objective function.

Acknowledgment

The authors would like to thank Tehran Refinery Management for providing some of their operational data.

REFERENCES

[1] Zhao BF, Zhang Xiaodong, Sun Li, Meng Guangfan, Chen Lei, Xiaolu Yi. Hydrogen production from biomass combining pyrolysis and the secondary decomposition. International Journal of Hydrogen Energy 2009;. doi:10.1016/j.ijhydene.2009.04.011.

[2] Ridolfi R, Sciubbab E, Tiezzic E. A multi-criteria assessment of six energy conversion processes for H_2 production. International Journal of Hydrogen Energy 2009;34:5080–90.

[3] Tolga Balta M, Dincer I, Hepbasli A. Thermodynamic assessment of geothermal energy use in hydrogen production. International Journal of Hydrogen Energy 2009; 34:2925–39.

[4] Pilavachi PA, Chatzipanagi Anatoli I, Spyropoulou Antonia I. Evaluation of hydrogen production methods using the Analytic Hierarchy Process. International Journal of Hydrogen Energy 2009;34:5294–303.

[5] Ozalp N. Energy and material flow models for hydrogen production in the U.S chemical industry. International Journal of Hydrogen Energy 2008;33:5020–34.

[6] Tugnoli A, Landucci G, Cozzani V. Sustainability assessment of hydrogen production by steam reforming. International Journal of Hydrogen Energy 2008;33:4345–57.

[7] Alves JJ, Towler GP. Analysis of refinery H_2 distribution systems. Industrial and Engineering Chemistry Research 2002;41:5759–69.

[8] Hallalea N, Liub F. Refinery H_2 management for clean fuels Production. Advances in Environmental Research 2001;6: 81–98.

[9] Haun EC, Anderson RF, Kauff DA, Miller GQ, Stocker J. The efficient refinery H_2 management in 1990s. UOP. available at, http://www.uop.com; 1990.

[10] Floudas CA. Nonlinear and mixed-integer optimization-fundamentals and applications. Oxford University Press; 1995.

[11] Raeesi B., H_2 network optimization with ant colony algorithm. M.Sc. thesis, Mechanical Engineering Department, Sharif University of Tech.; 2006.

[12] Schoots K, Freioli F, Kramer GJ, van der Zwaan BCC. Learning curves for hydrogen production technology: an assessment of observed cost reductions. International Journal of Hydrogen Energy 2008;33:2630–45.

[13] Khajehpour M. Nonlinear optimization for the hydrogen at Refinery. M.Sc. thesis, Chemical and Petroleum Engineering Department, Sharif University of Tech.; 2009.