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Improvement of Methanol Synthesis Process through a Novel Sorption-Enhanced Fluidized-bed Reactor , Part II: Multiobjective Optimization and Decision-making Method

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Abstract
In the first part (Part I) of this study, a novel fluidized bed reactor was modeled mathematically for methanol synthesis in the presence of in-situ water adsorbent named Sorption Enhanced Fluidized-bed Reactor (SE-FMR) is modeled, mathematically. Here, the non-dominated sorting genetic algorithm-II (NSGA-II) is applied for multi-objective optimization of this configuration. Inlet temperature of gas phase (T_g), temperature of saturated water (T_{shell}), total molar flow rate (F_t), diameter of solid adsorbent (d_s), mass adsorbent solid to mass catalyst ratio (M_{ratio}) and inlet pressure are selected as the decision variables. The production rate of methanol and selectivity is maximized as two objective functions. The Shannon's Entropy, LINMAP and TOPSIS methods as the three decision making approaches are applied to select the final solution of Pareto front. The optimization results approved by about 203.63 and 276.65 ton/day methanol production rate and CO_2 consumption, respectively, based on LINMAP methods compared with the conventional methanol configuration. The results recommend that consuming optimized-SE-FMR for improvement of methanol production could be feasible and beneficial.

Keywords
Multiobjective optimization; NSGA-II; Decision-making method; LINMAP; Pareto front.

1. Introduction

In the last two decades, multiobjective optimization of chemical reaction has become a major issue of concern. Methanol synthesis process is the best candidate for application of multiobjective optimization due to its nature of complexity. Multiple conflicting objectives are optimized for the popular of engineering design problem. These objectives are sometimes coupled and often in conflict. The multiobjective optimization problem is a set of solutions, namely 'Pareto optimum', and not only a single optimum solution. Pareto optimum front is the trade-off between multiple objectives (Ghiasi et al., 2010; Le Riche et al., 2003). They demonstrate when one or a small number of Pareto solutions are needed. A comprehensive traditional simplex method and a multiobjective optimization technique are more efficient than an evolutionary algorithm (EA) and vice versa (Deb, 2001). The non-dominated sorting genetic algorithm-II (NSGA-II) or the ruling NSGA is recognized as the famous evolutionary algorithm. It is noted that NSGA-II is able to maintain a satisfactory convergence of the non-dominated front and an appropriate spread of solutions (Deb et al., 2002).

Evolutionary algorithms (EAs) are easy, vigorous and self-determining as to gradient data, and apply a population-based procedure that allows obtaining many solutions in one stage. Evolutionary algorithms are devised to solve multiobjective problems, like PAES; (Sayin and Karabati, 1999), SPEA2; (Zitzler et al., 2001)) and MODE; (Babu and Anbarasu, 2005). A summary of the available studies run through EAs for multiobjective optimization is presented in Fonseca and Fleming (1995). In the sixty decade, Rosenberg first realized the ability of Genetic Algorithm in multiobjective optimization. Appling GA with particular value of fitness to each individual for multi-objective

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optimization is restricted. HLGA (Hajela and Lin's Genetic Algorithm) applied the technique of weighted-sum to implement this role (Hajela and Lin, 1992). The intricacy of defining suitable weights is the main drawbacks of this technique (Bingul, 2007). This problem is solved through the Vector Evaluated Genetic Algorithm (VEGA) (Schaffer, 1984). Purshouse and Fleming (2001) used a real code for Multiobjective Genetic Algorithm (MOGA) presented by Fonseca and Fleming Fonseca and Fleming (1995). Niched Pareto Genetic Algorithm (NPGA) is another model that applies a Pareto domination tournament (Horn et al., 1994). NSGA (Srinivas and Deb, 1994) and its modified form NSGA-II (Deb et al., 2002) are the new cases of GA based approaches applying Pareto solution to classify individual population. The Non-dominated sorting GA-II (NSGA-II) is applied in this article and further clarification is presented in Appendix A. Coello (1999) and Zitzler et al. (2000) have assessed the details of MOGA, VEGA, NPGA, NSGA method and their advantages and disadvantages. Zitzler et al. (2000) revealed that Non-dominated sorting genetic algorithm-II could yield the nearest results to the Pareto boundary in compassion with other GA-based multi-objective optimization procedures.

The results of multiobjective optimization in sorption-enhanced fluidized bed reactor for methanol synthesis process is presented here. A robust optimization method (NSGA-II) is applied to determine the optimal operating conditions with trade-off relations for two objective functions, including maximum methanol production rate and selectivity in SE-FMR. The final solution of Pareto front is selected through three different decision-making approaches. The mathematical modeling and process description of SE-FMR is presented in Part I (Bayat, 2017).

2. Optimization

In this article, non-dominated sorting GA-II is coded through MATLAB programming for two case studies.

2.1. Case Study 1

2.1.1. Test problems

The test problem assessed here is the one is reported as difficult to be solved by general MO solvers (Deb et al., 2002) There is a great chance to make the efficiency of NSGA-II evident. Test problems, recognized as artificial landscapes, are useful in assessing the characteristics of optimization algorithms:

- Convergence velocity
- Accuracy
- Robustness
- General performance

The test problems applied to assess the algorithms for MOP are extracted from Deb (2001) Binh and Korn (1997) and Binh (1999).

In this article, two sets of test functions, one for unconstrained and the other for constrained, problems are applied to assess the NSGA-II performance and the proposed constrained handling method. The Algorithm is coded through MATLAB.

2.1.1.1. Unconstrained Test problems

2.1.1.1.1. Zitzler-Deb-Thiele's Test problems

Zitzler et al. introduced six sets out of multi-objective problems (ZDT1 to ZDT6) are based on a unique structure with different level of difficulties (Zitzler et al., 2000). The functions have two objectives with the aim of minimization:

$$ZDT: \begin{cases} \text{Minimize} \quad F = \{f_1(x), f_2(x)\} \\ f_1(x), \\ f_2(x) = g(x) \end{cases} \tag{1}$$

Here, four of the six test problems (ZDT1, ZDT2, ZDT3 and ZDT4) are implemented through NSGA-II algorithm. The ZDT1, ZDT2 and ZDT3 is a function with 30 variables and convex pareto-optimal set as follows:

$$ZDT1: \begin{cases} \text{Minimize} \quad F = (f_1(x), f_2(x)) \\ f_1(x) = x_1 \\ f_2(x) = g(x)\left(1 - \sqrt{f_1 / g(x)}\right) \\ g(x) = 1 + 9\sum_{i=2}^{n} \dfrac{x_i}{n-1} \end{cases} \tag{2}$$

$$ZDT2: \begin{cases} Minimize \quad F = (f_1(x), f_2(x)) \\ f_1(x) = x_1 \\ f_2(x) = g(x).\left(1 - \left({x_1}/{g(x)} \right)^2 \right) \\ g(x) = 1 + 9\sum_{i=2}^{n} \frac{x_i}{n-1} \end{cases} \quad (3)$$

$$ZDT3: \begin{cases} Minimize \quad F = (f_1(x), f_2(x)) \\ f_1(x) = x_1 \\ f_2(x) = g(x).\left(1 - \sqrt{{f_1}/{g(x)}}\right) - \left({f_1}/{g(x)}\right).\sin(10\pi f_1) \\ g(x) = 1 + 9\sum_{i=2}^{n} \frac{x_i}{n-1} \end{cases} \quad (4)$$

All the variables are limited within [0 and 1].
ZDT4 Test Function is a 10 variable problem presented as follows:

$$ZDT4: \begin{cases} Minimize \quad F = (f_1(x), f_2(x)) \\ f_1(x) = x_1 \\ f_2(x) = g(x).\left(1 - \sqrt{{f_1}/{g(x)}}\right) \\ g(x) = 1 + 10(n-1) + \sum_{i=2}^{n} \left(x_i^2 - 10\cos(4\pi x_i)\right) \end{cases} \quad (5)$$

All variables except x_1, which lies within [0 and 1] range, are limited within -5 and 5.

2.1.1.2. Constrained Test Functions

A two objective test problem with two variable proposed by Tanaka et al. (1995) is implemented in the NSGA-II code as follows:

$$TNK: \begin{cases} Minimize \quad f_1(x) = x_1 \\ Minimize \quad f_2(x) = x_2 \\ C_1(x) = x_1^2 + x_2^2 - 1 - 0.1\cos\left(16\arctan \frac{x_1}{x_1}\right) \geq 0 \\ C_2(x) = (x_1 - 0.5)^2 + (x_2 - 0.5)^2 \leq 0.5 \\ 0 \leq x_1 \ , \ x_2 \leq \pi \end{cases}$$

$$x_1 \geq 0 \quad x_2 \geq \pi \quad (6)$$

2.2. Second case study

Here, the sorption enhanced fluidized bed methanol reactor (SE-FMR), modeled in Part I, which is optimized. The following operating parameters are applied as the decision variables.

- The inlet temperature of gas phase (T_g)
- Temperature of saturated water (T_{shell})
- Total molar flow rate (F_t)
- Adsorbent solid diameter (d_s)
- Adsorbent solid to catalyst mass ratio (M_{ratio})
- The inlet pressure of gas phase

The constraints for multiobjective optimization are as follows:

$$495 < T_g < 530 \quad K \quad (7)$$

$$0.5 < F_t < 0.7 \quad mol/s \quad (8)$$

$$1\times10^{-4} < d'_s < 6\times10^{-4} \quad m \quad (9)$$

$$510 < T_{Shell} < 535 \quad K \quad (10)$$

$$0.1 < M_{Ratio} < 0.6 \quad (11)$$

$$50 < P_t < 100 \quad bar \quad (12)$$

Because of $CuO/ZnO/Al_2O_3$ the catalyst can be deactivated at higher temperatures than 543K, this temperature is considered as a constraint for catalytic bed temperature.
The unwanted optimization results can be detached through a penalty function technique. Methanol production rate (Eq. (13)) and selectivity (Eq. (14)) are selected as two the objectives of this study.

Maximize:

$$OF_1 = (Methanol\ production\ rate) - 10^8 \sum_{i=1}^{N} G_i^2 \quad (13)$$

Maximize:

$$OF_2 = \frac{F_{CH3OH,out}}{F_{CO,out} + F_{CO_2,out}} - 10^8 \sum_{i=1}^{N} G_i^2 \quad (14)$$

where,

$$G = \max(0, T - 543) \quad (15)$$

2.3. Decision-making in the multi-objective optimization

Every non-dominated solution on a Pareto distribution of multiobjective optimization problems can be selected as the best result, while, only one point should be chosen. There exist many decision-making methods for multiobjective optimization problems, which can be applied to make a choice of ultimate best solution (Liu, 2014).

In multiobjective optimization function, the objectives almost have unrelated dimension. The nondimensionalization of these objectives are made by Linear, Euclidian, and fuzzy approaches. Here, the fuzzy approach is applied for nondimensionalization of these objectives.

2.3.1. LINMAP method

In this method, the Euclidian metric of the ideal point from every solution on the Pareto curve is computed through Eq. (16) as:

$$d_i^+ = \sqrt{\sum_{j=1}^{n} \left(F_{ij} - F_j^{Ideal}\right)^2} \quad (16)$$

where, n is the objective number and subscript of attitude *i* for each point on the Pareto solution. In Eq. (16), F_j^{Ideal} is the ideal value of j^{th} objective. The ultimate result is calculated as

$$i_{final} = i \in \min\left(d_i^+\right) \qquad i = 1,2,3,\ldots,m$$

2.3.2. TOPSIS method

The ideal solution of LINMAP decision-making method is inserted into non-ideal solution as:

$$d_i^- = \sqrt{\sum_{j=1}^{n} \left(F_{ij} - F_j^{Non-Ideal}\right)^2} \quad (17)$$

and, Y_i parameter is defined as follows,

$$Y_i = \frac{d_i^-}{d_i^+ + d_i^-} \quad (18)$$

In TOPSIS decision-making method, the point with highest Y_i is preferred (Etghani et al., 2013).

Therefore,

$$i_{final} = i \in \max\left(Y_i\right) \qquad i = 1,2,3,\ldots,m$$

2.3.3. Shannon's entropy decision-making method

The weights of each objective is calculated for Shannon's entropy method (Guisado et al., 2005). Considering P_{ij} in decision matrix F_{ij} with 'n' number of each member and *m* objectives, the element of this matrix for j^{th} objective is calculated as:

$$P_{ij} = \frac{F_{ij}}{\sum_{i=1}^{n} F_{ij}} \qquad i = 1,2,\ldots,n \qquad j = 1,2,\ldots,m \quad (19)$$

Shannon's entropy is calculated as:

$$SE_j = -\frac{1}{\ln(n)} \sum_{i=1}^{n} P_{ij} \ln\left(P_{ij}\right) \quad (20)$$

where, the weight of jth objective is formulated as:

$$w_j = \frac{1 - SE_j}{\sum_{j=1}^{m} \left(1 - SE_j\right)} \quad (21)$$

$$X_i = P_{ij} \times w_j \quad (22)$$

The solution with maximum X_i is considered as an ultimate solution of Shannon's Entropy decision-making approach.

3. Results and Analysis

The real Pareto optimal solutions with the optimization results of constrained (ZDT) and unconstrained (TNK) test problem are optimized through NSGA-II algorithm, Fig. 1 (a-e). The discontinuous area on the real Pareto solution is perfectly enclosed through the NSGA-II algorithm, observed in these figures.

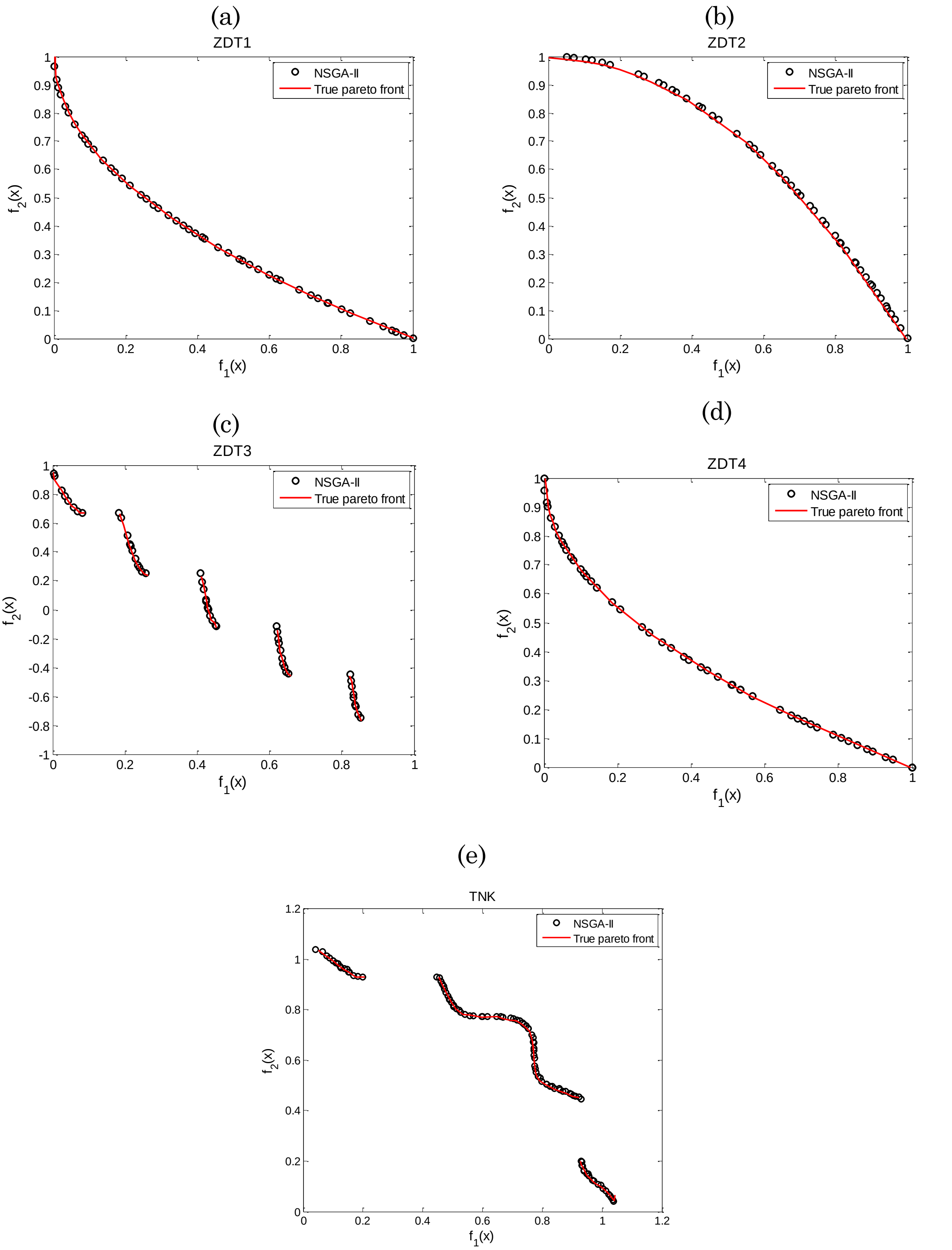


Figure. 1. Pareto fronts applying the NSGA-II algorithm and real solution of the (a-d) ZDT and (e) TNK test problem.

Methanol production rate and selectivity as incompatible objectives are shown in Fig. 2, where, the final optimum solutions of LINMAP, TOPSIS and Shanon's Entropy methods are exposed.

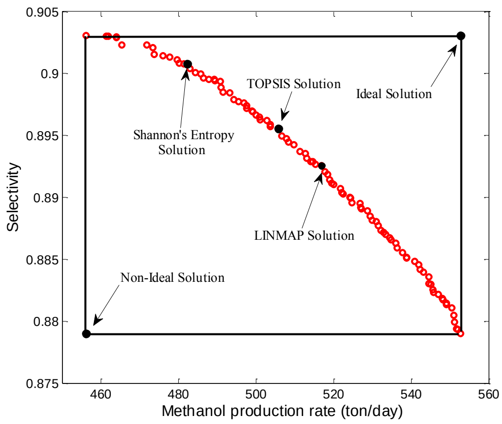


Figure. 2. Pareto-optimal fronts in objective space.

One index, Eq. (12) is introduced for different decision making approaches to calculate the deviation of various methods from the ideal and non-ideal value as follows (Ahmadi et al., 2013): This index is expressed as:

$$d = \frac{d_1}{d_1 + d_2} \tag{22}$$

where:

$$d_1 = \sqrt{\sum_{j=1}^{n} \left(F_j - F_j^{Ideal}\right)^2} \tag{23}$$

$$d_2 = \sqrt{\sum_{j=1}^{n} \left(F_j - F_j^{Non-Ideal}\right)^2} \tag{24}$$

Nearness to the ideal solution is shown by smallest value of deviation index; thus, each decision making method is shown by a deviation index value. The more appropriate method is recognized by the minimum value of deviation index. The values of deviation index for LINMAP, TOPSIS and Shannon's entropy method are tabulated in Table 1, where, the deviation index is minimized at LINMAP decision-making method.

Analysis of key parameters like the temperature of gas phase, mole fraction of components and methanol production rate are selected to address the fundamental issues by applying the optimum parameter of LINMAP decision-making methods.

As observed in Table 1, the methanol production rate of OSE-FMR increase about 203.63 and 93.36 ton/day in relation to that of conventional methanol reactor (CMR) and (SE-FMR), respectively. Selectivity is enhanced about 3.95 % in OSE-FMR in comparison with non-optimized configurations. Water adsorption from the reaction zone, through the adsorbent solid particles, leads to a reduction in water production in the reaction region; consequently, the hydrogenation of CO_2 reaction changes to the right side and enhances selectivity and productivity of methanol.

The CH_3OH and H_2O production rate in optimized and non-optimized SE-FMR are compared in Fig. (3). AS observed, a difference between the outputs of optimized and non-optimized SE-FMR is evident, due to water adsorption from the reaction side. These figures prove the influence of optimized parameter on enhancing reactant conversion.

Table 1. Comparison among obtained optimal solutions

Optimization algorithm		NSGA-II		
		TOPSIS	**LINMAP**	**Shannon's Entropy**
Design variables	T_g(K)	505.615	**504.542**	506.851
	d_s(μm)	157.782	**158.972**	153.144
	M_{ratio}	0.429	**0.423**	0.429
	T_{shell}(K)	512.786	**512.642**	512.588
	F_t (mol/s)	0.579	**0.595**	0.548
	P_t (bar)	96.821	**96.885**	96.28
Objectives	OF_1 (methanol production rate)	505.73	**516.898**	482.354
	OF_2 (Selectivity)	0.896	**0.893**	0.901
Deviation Index		0.4875	**0.3718**	0.7296

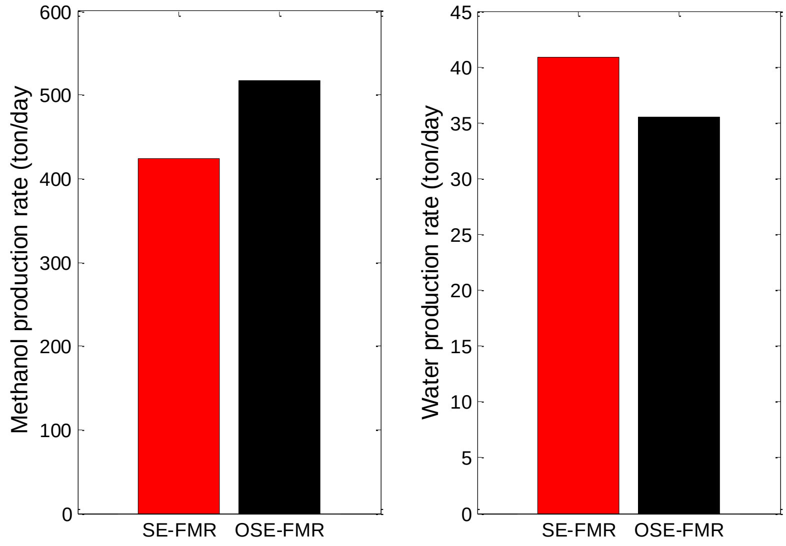
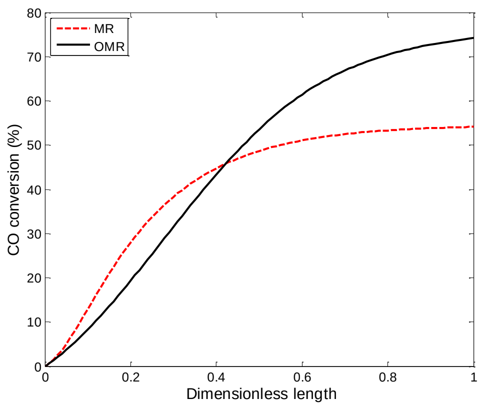


Figure. 3. Output of CH_3OH and H_2O production rate at the reactor axis between optimized and non-optimized SE-FMR.

3.1. Synthesis gas components conversion

The conversion of CO and H_2 components together with the optimized SE-FMR in comparison with the ones in non-optimized configuration are shown in Fig. 4 (a) and (b). An increase in H_2 and CO conversions in these reactors follows the same trends. Hydrogen and carbon monoxide conversions in OSE-FMR are higher than those of the SE-FMR due to in-situ water removal by adsorbents. These results are related to the favorable effect of consuming more water adsorbents in optimized configuration, through which water production rate decreases. More conversion of hydrogen and carbon monoxide leads to in an important (almost the best) methanol production rate Fig. 4.

(a)



(b)

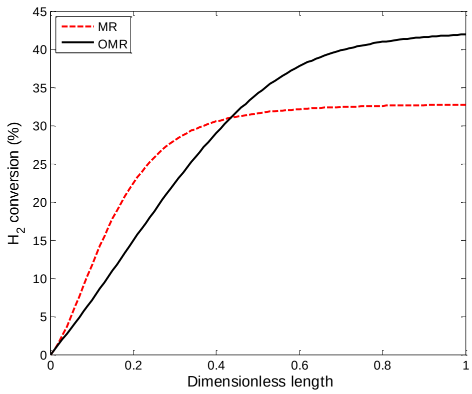


Figure. 4. Comparison of (a) CO and (b) H_2 conversion between optimized and non-optimized SE-FMR along the reaction side of reactor

3.2. CO_2 Trapping

The key advantage of OSE-FMR in relation to CMR is the CO_2 entrapment. The CO_2 removal rate for the OSE-FRM and CMR is shown in Fig. (5), where, As it can be seen, by applying OSE-FRM instead of CMR, 272.65 ton.day^{-1} of CO_2 can be eliminated. At this stage, it is better to substitute conventional fixed bed reactor with the optimized SE-FMR.

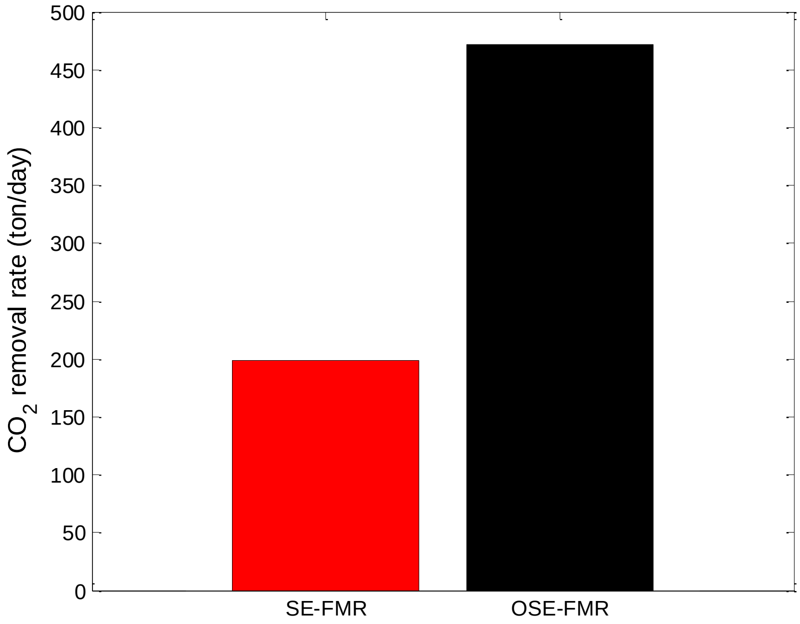


Figure. 5. Comparison of CO_2 removal rate at the reactor axis between OSE-FMR and CMR.

4. Conclusion

In this part, The second part out of two, a sorption-enhanced process in fluidized bed reactor, modeled in Part I, is optimized through NSGA-II algorithm. The optimization results reveal that 18 and 3.95% increase in the methanol productivity and selectivity in OSE-FMR rather than SE-FMR, respectively. This optimized reactor improves the CO_2 entrapment about 272.65 ton/day. It is predicted that applying this OSE-FMR in methanol synthesis improvement would be feasible and beneficial.

Appendix A: Non-Dominated Sorting Genetic Algorithm-II (NSGA-II)

One of the best and efficient manners in for solving multi-objective optimization problem is applying NSGA-II algorithm introduced by Deb studies (Deb, 2001; Deb et al., 2002), desirable, as follows:

Step 1: Develop a random population size N according to the ranges of variable.

Step 2: Sent the initial population according to non-dominated fast sorting criterion

Step 3: Calculate the distance of crowding for every element belonging to a front. The Euclidean distance between two neighboring solutions is applied to measure the crowding distance of a solution as a similar front in the objective space

Step 4: Measure the crowding distance for the arrangement of every solution, and apply the tournament operator for selection between two solutions. The solution with the lower grade is selected while the two solutions are from different fronts. If they are one of the same front, the solution with the higher crowding distance value is selected

Step 5: Implement crossover as well as mutation operator to generate the children solutions

Step 6: Merge parent and offspring populations into a population of size 2N, where N is the initial population size

Step 7: Select the best N components of the combined population.

Step 8: Repeat steps 2-7 until the termination criterion (desired number of generations) is met

The steps involved in the NSGA-II algorithm are flowcharted in Fig. (6) and the parameters of NSGA-II algorithm applied in the optimization of both parts (studies) are tabulated in Table 2.

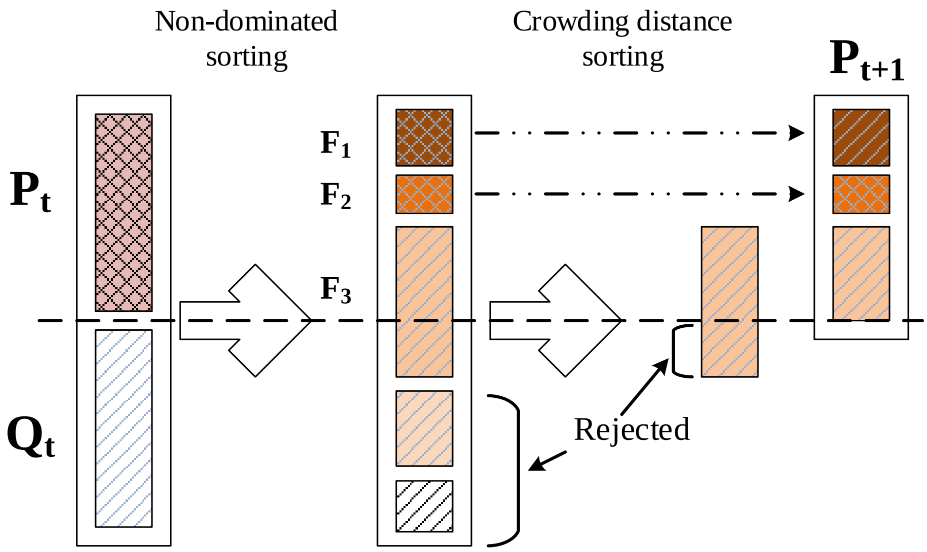


Figure. 6. Schematic diagram of NSGA-II algorithm

Table 2. Main parameters of NSGA -II algorithm for both case study

Parameter	First Case study					Second Case study
	ZDT1	ZDT2	ZDT3	ZDT4	TNK	
Number of decision variables	30	30	30	10	2	6
Number of objectives	2	2	2	2	2	2
Population size	50	50	50	50	100	100
Selection strategy	Binary tournament					
Crossover type	Simulated binary crossover					
Mutation type	Polynomial mutation					
Crossover probability	0.9					
Mutation probability	0.05					
Generations	300					

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