

# Application of Multi-Objective Optimization in the Design of SMB in Chemical Process Industry

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**Abstract**—A new optimization and design strategy, multi-objective optimization, is applied to improve the performance of Simulated Moving Bed (SMB) and its modification, the Varicol systems with or without reactions. The capabilities of the new approach are illustrated considering three different applications of SMB (and Varicol process), namely, chiral drug separation, production of high concentrated fructose syrup and synthesis of methyl acetate ester. The non-dominated sorting genetic algorithm (NSGA) is used in obtaining Pareto optimal solutions. It was found that significant improvement is possible for both SMB and Varicol process.

*Key Words* : Multi-objective optimization, Simulated moving bed, Varicol, Genetic algorithm, Pareto set

## INTRODUCTION

The simulated moving bed (SMB) introduced by UOP in 1960s (Broughton and Gerhold, 1961; Broughton, 1968) as a continuous chromatographic separation process, has been widely applied in chemical process industry for separation of hydrocarbons and sugar purification in the last few decades. By virtue of its superior separating power and relatively mild operating conditions, SMB has recently received a great deal of attention as a promising technique in the pharmaceutical industry for enantio-separations.

SMB is a practical way to implement the true moving bed process (TMB) in order to circumvent the problems caused by handling of solids. In SMB, the countercurrent movement of the fluid phase towards the solid phase is mimicked by switching the introduction and withdrawal ports periodically and simultaneously along a series of fixed columns in the direction of the fluid flow (Ray *et al.*, 1990, 1994). For ease of operation, the column is actually divided into sections or zones, which consist of a number of columns. The number of columns within each section and total number of columns are adjustable depending on the design of the system for any particular applications. One of the limitations of the SMB is that during much of the operation, the stationary phase in the column is either completely free of solutes or contains only product so that the separation capacity is reduced (Ludemann-Hombourger *et al.*, 2000). This is primarily due to synchronous switching of all the inlet and outlet ports. Recently, SMB

was modified into Varicol process (Ludemann-Hombourger *et al.*, 2000) for chiral separation by non-synchronously shifting the inlet and outlet ports during a global switching period. Therefore, the column configuration varies at different sub-time intervals in Varicol process, which provides more flexibility compared to traditional more rigid SMB process. SMB and Varicol can also be integrated to include reactions (Ray and Carr, 1995; Yu *et al.*, 2003), which can provide economic benefit for equilibrium limited reversible reactions, such as many hydrogenation, isomerization, and esterification reactions. In-situ separation of the products facilitates the reversible reaction to completion beyond thermodynamic equilibrium while obtaining products of very high purity.

The optimal design and operating parameters are essential to evaluate the economic potential of SMB and Varicol process and to successfully implement them on industrial scale. Although several studies (Dunnebier *et al.*, 2000; Biressi *et al.*, 2000; Azevedo and Rodrigues, 2001) have been reported on the optimization of SMB and Varicol with or without reactions, they only involved single objective optimization in terms of maximization of productivity, which is usually not sufficient for the real-life design of complex SMB systems, since the operating variables influence the productivity and other important objectives, such as product purity, eluent consumption, etc. usually in conflicting ways. This leads to unfavorable change in the second objective function whenever a desirable change in the first objective function is achieved. Therefore, the simultaneous optimization

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of multiple objective functions is very important for the design of SMB and Varicol process.

The principle of multi-criterion optimization with conflicting objectives is different from that of single objective optimization (Bhaskar *et al.*, 2000a). Instead of trying to find the best (global) design solution, the goal of multi-objective optimization is to obtain a set of equally good (non-dominating) solutions, which are known as Pareto optimal solutions. In a set of Pareto solutions, no solution can be considered better than any other solutions with respect to all objective functions. The choice of a solution over the other solutions requires additional knowledge of the problem, and often this knowledge is intuitive and non-quantifiable. However, by narrowing down the choices, the Pareto set does provide decision makers with useful guidance in selecting the desired operating conditions (called the preferred solution) from among the (restricted) set of Pareto optimal solutions, rather than from a much larger number of possibilities.

In this article, three case studies of SMB as well as Varicol process covering a wide range of possible industrial applications were presented to illustrate the capabilities of multi-objective optimization approach. The Pareto optimal solutions were obtained by using Non-dominated sorting genetic algorithm (NSGA) (Nandasana *et al.*, 2003). To the best of our knowledge, this is the first attempt to extend the concept of Varicol process to reactive systems, and compare the optimal solutions with that of reactive SMB. The objective of this paper is to provide a new more realistic approach towards the optimal design of SMB and Varicol systems.

## SIMULATED MOVING BED AND VARICOL PROCESSES

A schematic representation of a SMB system is illustrated in Fig. 1(a) that consists of a number of columns of uniform cross-section connected in a circular array, each of length  $L_{col}$  and packed with the ion exchange resin adsorbent. The two incoming streams (the feed and the eluent) and the two outgoing streams (the raffinate and the extract) divide the system into four sections namely  $P$ ,  $Q$ ,  $R$  and  $S$ , each of which comprising  $p$ ,  $q$ ,  $r$ , and  $s$  columns respectively. The flow rates in the section  $P$ ,  $Q$ ,  $R$ , and  $S$  were designated as  $Q_P$ ,  $Q_Q$ ,  $Q_R$ , and  $Q_S$  respectively. However, only four of the above eight flow rates are independent, as the remaining four are determined from the mass balance at points A, B, C, and D (see Fig. 1(a)). By suitably advancing the inlet and outlet ports, column by column, in the direction of the fluid flow at a pre-set switching time,  $t_s$ , the countercurrent movement of the solids can be mimicked. This

switching time is the key parameter, which defines the hypothetical solid-phase velocity. However, countercurrent separation of the components could be achieved only by appropriately specifying the internal flow rates in the columns and the switching time. Petroulas *et al.* (1985) defined for true countercurrent moving bed chromatographic reactor (CMCR) a parameter,  $\sigma_i$ , called relative carrying capacity of the solid relative to the fluid stream for any component  $i$  as

$$\sigma_i = \frac{1-\varepsilon}{\varepsilon} NK_i \frac{u_s}{u_g} = \delta_i \frac{u_s}{u_g}, \quad (1)$$

where  $u_s$  and  $u_g$  are respectively solid phase and fluid phase velocity. They showed that to achieve countercurrent separation between the two components, one must set  $\sigma$  greater than 1 for one and less than 1 for the other. Later, Fish *et al.* (1986) verified the above fact experimentally. Fish *et al.* (1986) also defined  $V_i$ , the net velocity at which component  $i$  travels (or the concentration front moves) within the column, which for linear isotherm is given by

$$V_i = \frac{u_g(1-\sigma_i)}{(1+\delta_i)}. \quad (2)$$

Therefore, when  $\sigma_i < 1$ ,  $V_i > 0$  (species move with the fluid phase), and when  $\sigma_i > 1$ ,  $V_i < 0$  (species move with the solid phase). When  $\sigma = 0$ , it represents fixed bed. Ray *et al.* (1994) re-defined the above parameter,  $\sigma$ , by replacing the solid-phase velocity,  $u_s$ , in CMCR by a hypothetical solid phase velocity,  $\zeta$ , defined as  $\zeta = L_{col}/t_s$ . They found, both theoretically (Ray *et al.*, 1994) and experimentally (Ray and Carr, 1995), that simulation of the countercurrent movement between two components can be achieved when redefined  $\sigma$ 's were set such that it is greater than 1 for one and less than 1 for the other component. Hence, if  $\sigma$  is set properly, the more strongly adsorbed component will move with the imaginary solid stream, and can be collected at the extract port (point D in Fig. 1(a)), while at the same time the less strongly adsorbed component will travel with the fluid stream, and can be collected at the raffinate port (point B in Fig. 1(a)). It is worth noting that in SMB the switching time and column configuration (the number of columns in each section) is decided a priori and is maintained constant during the entire operation.

Unlike the SMB unit as shown in Fig. 1(a) for an 8-column system, the Varicol system is based on non-simultaneous and unequal shift of the inlet and outlet ports. Figure 1(b) illustrates schematically the operational difference between 8-column four-subinterval Varicol systems with the 8-column SMB system for one switching period. The switching time

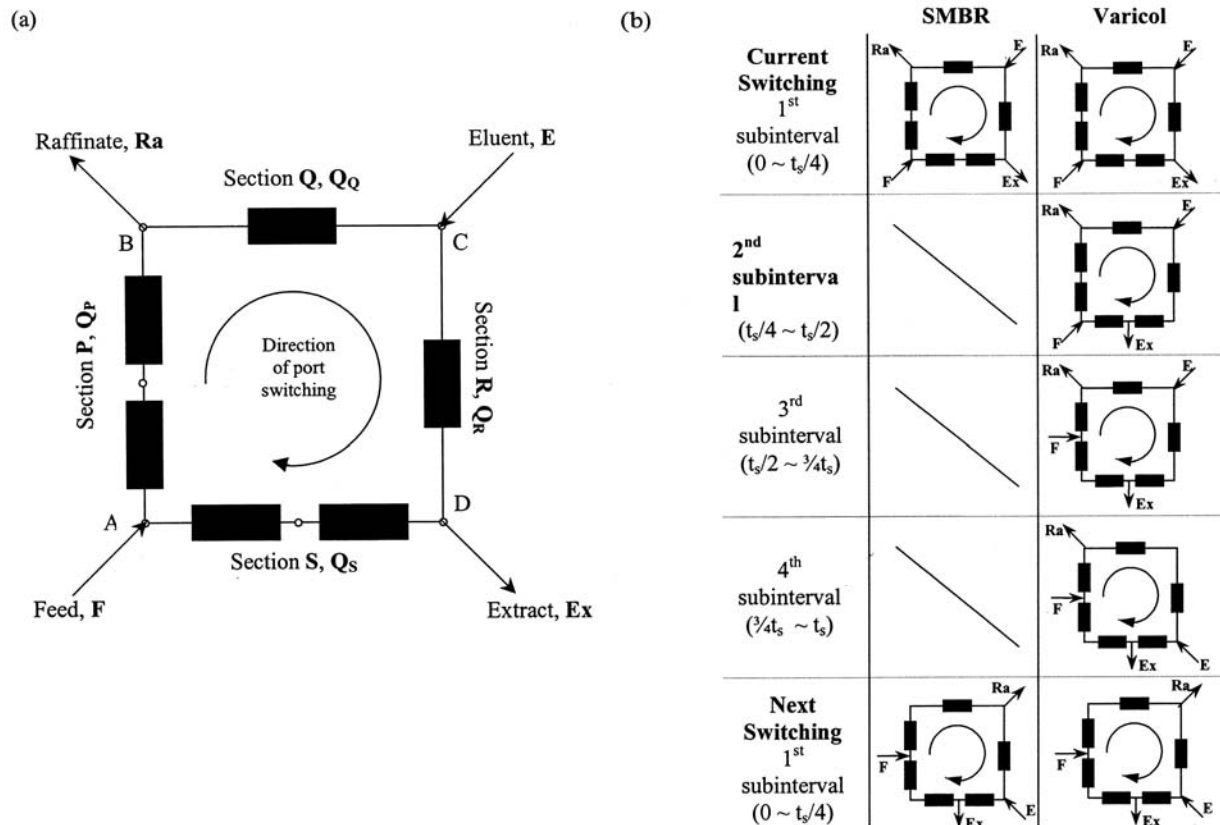


Fig. 1 (a) Schematic diagram of a 6-column SMBR system; (b) principle of operation of SMBR and 4-subinterval Varicol (port switching schedule). The inlets and outlets divide the entire system into four sections: *P*, *Q*, *R*, and *S* with respectively 2, 1, 1, and 2 number of columns. The flow rates in each section is given by  $Q_Q = (1 - \beta) Q_P$ ,  $Q_R = (1 - \beta + \gamma) Q_P$ , and  $Q_S = (1 - \alpha) Q_P$ , where  $\alpha$ ,  $\beta$ ,  $\gamma$  are given by  $F/Q_P$ ,  $Ra/Q_P$ ,  $E/Q_P$ .

$t_s$ , which is related directly to the solid flow rate in SMB, is also a key parameter in the Varicol process, although the relationship is not straightforward. In Varicol operation, a non-synchronous shift of the inlet and outlet ports is usually employed within a global switching period, which is again kept constant in time. Within a global switching period,  $t_s$ , the column configuration is allowed to alter for each quarter of  $t_s$  for a four-subinterval operation. For example, the column configuration for a typical sequence in a given cycle corresponding to Fig. 1(b) changes from 2/1/2/3 ( $0 \sim t_s/4$ ) to 3/1/1/3 ( $t_s/4 \sim t_s/2$ ) by shifting both the feed and the extract port by exactly one column in the backward direction, then to 3/1/2/2 ( $t_s/2 \sim t_s/3/4$ ) by shifting the extract port by one column in the forward direction, and finally to 2/2/2/2 ( $t_s/3/4 \sim t_s$ ) by shifting the raffinate port backward by one column. The configuration 2/1/2/3 explicates that there are two columns each in sections *P* and *R*, while one column in section *Q* and 3 columns in section *S* respectively. As a result, in a 4-subinterval Varicol process, there are four different column configurations for the four subintervals due to local switching during one global switching period. The number of columns in each zone varies with

time within a global switching period, but the number of columns in each zone returns to the starting value at the end of the global switching period. In terms of the time average number of columns per zone this corresponds to 2.5/1.25/1.75/2.5 for the above example. Therefore, locations of input/output ports in a Varicol process are quite different from that of the SMB process. Note that in principle it is possible that a port may shift more than once during one global switching period, either forward or even in backward direction. As a result, Varicol process can have several column configurations, which endow more flexibility compared to a SMB process. SMB process can be regarded as a special case of the more flexible Varicol process without requiring any additional fixed cost.

## MULTIOBJECTIVE OPTIMIZATION

In earlier years, multi-objective optimization problems were usually solved using a single scalar objective function, which was a weighted-average of the several objectives ('scalarization' of the vector

objective function). This process allows a simpler algorithm to be used, but unfortunately, the solution obtained depends largely on the values assigned to the weighting factors used, which is done quite arbitrarily. An even more important disadvantage of the scalarization of the several objectives is that the algorithm may miss some optimal solutions, which can never be found, regardless of the weighting factors chosen. This happens if the non-convexity of the objective function gives rise to a duality gap (Deb, 1995). Several methods are available to solve multi-objective optimization problems, *e.g.*, the  $\epsilon$ -constraint method (Chankong and Haimes, 1983), goal attainment method (Fonseca and Fleming, 1998) and the non-dominated sorting genetic algorithm (NSGA) (Goldberg, 1989; Deb, 2001). In this study we use NSGA to obtain the Pareto set. This technique offers several advantages (Deb, 1995, 2001), as for example: (a) its efficiency is relatively insensitive to the shape of the Pareto optimal front, (b) problems with uncertainties, stochastics, and with discrete search spaces can be handled efficiently, (c) the 'spread' of the Pareto set obtained is excellent (in contrast, the efficiency of other optimization methods decides the spread of the solutions obtained), and (d) it involves a single application to obtain the entire Pareto set (in contrast to other methods, *e.g.*, the  $\epsilon$ -constraint method, which needs to be applied several times over).

Indeed, NSGA has been applied recently to optimize several processes of industrial importance in chemical engineering, including an industrial nylon-6 semi-batch reactor (Mitra *et al.*, 1998), a wiped-film polyester reactor (Bhaskar *et al.*, 2000b, 2001), PMMA film reactor (Zhou *et al.*, 2000), a steam reformer (Rajesh *et al.*, 2000), beer dialysis (Yuen *et al.*, 2000), cyclone separators (Ravi *et al.*, 2000), hydrogen plant (Rajesh *et al.*, 2001; Oh *et al.*, 2002), venture scrubber (Ravi *et al.*, 2002), MTBE synthesis (Zhang *et al.*, 2002) and styrene reactor (Yee *et al.*, 2003).

The method of optimization used in this work is very general, and can easily be applied to almost any other applications. In fact, in this paper we *illustrate the procedure* to be used, and present solutions of a few relatively *simple* optimization problems with two or three-objective functions for three important industrial application. A whole variety of other problems can, indeed, be formulated and solved, depending upon one's interest.

### Case 1: separation and purification of chiral drugs

Application of multiobjective optimization is particularly true in chiral separation using simulated moving bed (SMB) systems where purities of the

products are crucial and have to satisfy relatively narrow specifications. Since 1990s, SMB has drawn more and more attention for enantio-separation among drug producers, due to recent developments in chiral stationary phases (CSPs) and nonlinear chromatographic theory, as well as stringent drug administration policy. Even though the separating power of SMB is widely acknowledged, researchers are still trying to improve the performance of SMB process to decrease the operating cost.

Recently, Ludemann-Hobourger *et al.* (2000) developed a novel process, Varicol, which is based on a non-synchronous switch of the inlet and outlet ports. With illustrative purposes, they considered chiral (enantio) separation of 1,2,3,4-tetrahydro-1-naphthol racemate, using Chiralpak AD 20  $\mu\text{m}$  as CSP and *n*-heptane, 2-propanol and trifluoroacetic acid as eluent. They showed both experimentally and numerically, that Varicol is indeed superior to SMB in terms of product purity and productivity. They found out that similar purities could be achieved by a 5-column Varicol compared to a 6-column SMB process for the same productivity. They reported an 18.5% improvement in productivity for a 5-column Varicol system at almost the same eluent flow rate and product purity over a 5-column SMB process. However, this comparison was not definitive since no systematic optimization of the column configuration or operating conditions in the Varicol process was attempted.

In this work, the same chiral separation system reported by Ludemann-Hombourger *et al.* (2000) is considered as an illustrative example. Details of model equations can be obtained in Ludemann-Hombourger *et al.* (2000) and is not repeated here for brevity. We first compared our optimal results in terms of maximum feed flow rate (*i.e.*, capacity of the exiting SMB unit) determined by simple genetic algorithm (SGA) with those reported by Ludemann-Hombourger *et al.* (2000). It was found that about 6% and 10% increase in feed flow rate are possible for SMB and Varicol respectively. These comparisons, relative to single objective optimization show the reliability and efficiency of SGA in finding optimal solutions.

The multiobjective optimization study is aimed either at improving the separation quality (product purities) under same capital cost (fixed number and size of columns) and productivity (feed flow rate), or at reducing capital cost and/or eluent consumption and/or increasing productivity for the same product purity requirements. Furthermore, by comparing the performance of the Varicol process with an equivalent SMB process, this work tries to determine to what extent operation of a simulated moving bed system can be improved by applying non-synchronous switching with varying zone lengths.

We considered two typical multi-objective optimization problems of interest in this application, which are (a) the simultaneous maximization of purity of raffinate and extract streams at fixed capital and operating costs, and (b) the simultaneous maximization of productivity and minimization of eluent consumption for fixed product purity of both extract and raffinate streams. Figure 2 shows Pareto optimal solution for the first case where one cannot improve purity of raffinate without sacrificing purity of extract stream. A 5-column 4-subinterval Varicol performs better than an equivalent 5-column SMB, especially when high purities for both product streams are desirable. However, the maximum attainable purities in a 5-column Varicol are less than that could be obtained in a 6-column SMB, which contains 20% more stationary phase. Figure 3 shows the results for the second case when desired purity of both product streams is greater than 99%. It can be observed that in both SMB and Varicol process the eluent consumption must be increased in order to increase the feed flow rate (capacity) for a particular purity specification, and Varicol consumes less eluent than SMB for the same feed flow rate or for the same eluent consumption, Varicol can treat more feed. By performing multi-objective optimization, we not only provide decision makers with useful guidelines in selecting desired design and operating parameters but also deepen the understanding of complex SMB and Varicol process.

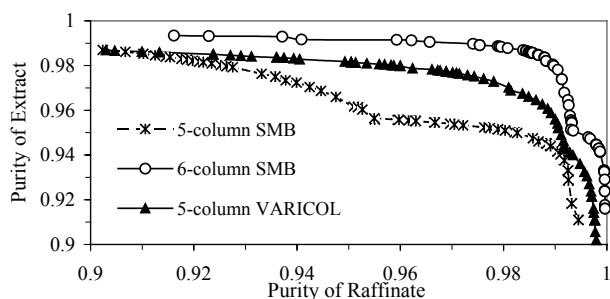


Fig. 2. Pareto optimal solutions for maximization of purity of raffinate and extract streams (case 1: separation and purification of chiral drugs).

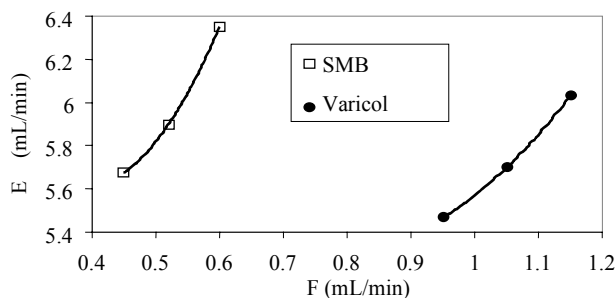


Fig. 3. Pareto optimal solutions for maximization of feed and minimization of eluent (case 1: separation and purification of chiral drugs).

## Case 2: production of high concentrated fructose syrup

There are many bio-separation problems for which SMB has been employed. The most known industrial application is the separation of sugar, in particular, the isolation of fructose (which is of interest in the food industry as sweetener) from a mixture of glucose and fructose solution. Both Glucose and Fructose have linear isotherms over a wide concentration range, and hence are an excellent example for the development of design stage SMB units. The separation is performed using ion-exchange resins with warm water as the eluent. The preferred implementation consists of using polystyrene cation-exchange resins in the calcium form in which the fructose forms a complex with the calcium ions and is retarded, while the glucose and the other oligosaccharides are eluted with the eluent.

The performance of an existing SMB setup for glucose-fructose separation reported by Azevedo and Rodrigues (2001) was improved by determining the optimal operating parameters as well as column length. Once again, details of model equations are not repeated here for brevity and can be obtained in Azevedo and Rodrigues (2001). Figure 4 compares the experimental results for a 12-column SMB pilot plant unit with that of the optimal solutions obtained in this work. When the column length is the same as that in the reported experimental setup (0.30 m), not much improvement in the productivity of both glucose and fructose is possible for the existing set-up. However, for a given productivity of fructose ( $PR_F$ ) and glucose ( $PR_G$ ), the purity of fructose ( $Pur_F$ ) and glucose ( $Pur_G$ ) improved significantly as against the experimentally reported results. For example, a 2% increase in  $Pur_F$  was observed at a given  $PR_F$  of  $\sim 6.15 \text{ kg/m}^3 \text{ solid/h}$ . Similarly, a 4.2% increase in  $Pur_G$  was observed for a given  $PR_G$  of  $\sim 6.65 \text{ kg/m}^3 \text{ solid/h}$ . When the length of each column was optimized for the existing set-up, significant improvement in productivity of both the streams was obtained. The optimal length obtained was between (0.20-0.21 m) drastically reducing total adsorbent volume while improving the overall performance. For example, the improvement in  $PR_F$  as against the experimental results at a given  $Pur_F$  (97%) was about 38%, while that for  $PR_G$  at a  $Pur_G$  (88%) was about 40%. When total number of columns was optimized, the optimum number of columns obtained was 8 with optimum length of each column as 0.23 m while further improving the performance of the system. The optimum column distribution obtained was 2/1/2/3 compared to 3/3/3/3 used in the experimental work. When the same was optimized for 4-subinterval Varicol operation, a further improvement in performance was possible as shown in Fig. 4.

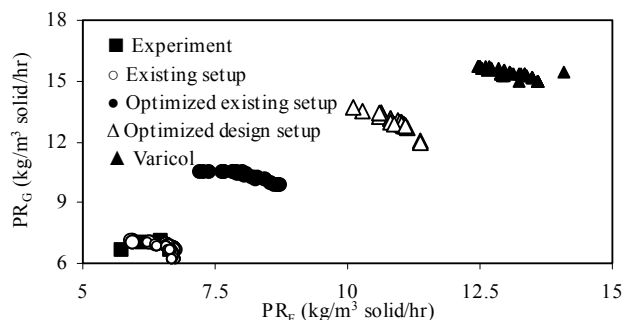


Fig. 4. Pareto optimal solutions for glucose-fructose separation (case 2: production of high concentrated fructose syrup).

### Case 3: synthesis of methyl acetate ester

SMB and its modification also provide opportunities for coupling reactions, which allow higher conversion for equilibrium-limited reversible reactions by on-site separation of the products, which leads to better yield and selectivity compared with typical fixed-bed processes. Additionally, the combination of two unit operations in one single apparatus reduces capital and operating cost. However, such integration of chemical reaction and separation complicates the process design and plant operation. The optimal design and operating parameters, such as switching time (and sequence), flow rates in each section, length of each column and its distribution, are therefore essential to evaluate the economic potential of the process and to successfully implement the reactive SMB and Varicol processes on industrial scale.

In this case, multi-objective optimization has been performed for the synthesis of methyl acetate (MeOAc) ester in reactive SMB and Varicol. The modeling, simulation and experimental study of SMBR for the synthesis of methyl acetate (MeOAc) have been carried out details of which are reported elsewhere (Yu *et al.*, 2003). A rigorous numerical model was developed to describe the dynamic behavior of SMBR for the synthesis of MeOAc, and comparing the experimental results with model predictions validated the model. To the best of our knowledge, this is the first attempt to extend the concept of Varicol process to reactive systems, and compare the optimal solutions with that of reactive SMB. A triple objective optimization of both reactive SMB and Varicol were performed with respect to simultaneous maximization of purity and yield of methyl acetate together with minimization of eluent consumption. For the convenience of analysis, the Pareto optimal solutions were plotted in two dimensions, that is, yield of methyl acetate and eluent flow rate against purity of methyl acetate. Figure 5(a) shows that purity of MeOAc decreases as yield of

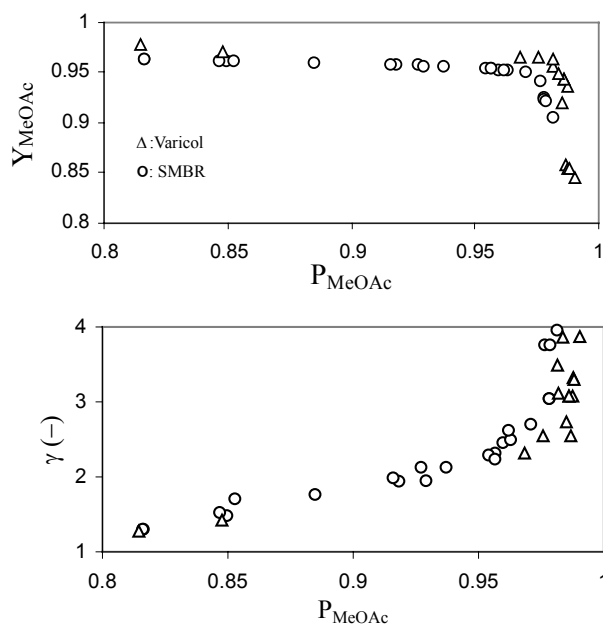


Fig. 5. Pareto optimal solution for the synthesis of methyl acetate (case 3).

MeOAc increases for both reactive SMB and Varicol and the performance of reactive Varicol is better than reactive SMB in terms of higher yield for the same purity requirement. From Fig. 5(b), it can be observed that the increase of purity of MeOAc is at the cost of eluent consumption for both reactive SMB and Varicol and the eluent consumption is less in reactive Varicol than that in reactive SMB for the same purity requirement, especially in the high purity region. The better performance of reactive Varicol results in its flexibility in column distribution, leading to better utilization of adsorbent.

## CONCLUSION

Simulated Moving Bed (SMB) systems are used in industry for separations that are either impossible or difficult using traditional separation techniques. By virtue of its superior separating power, SMB has become one of the most popular techniques finding its application in petrochemical and sugar industries, and of late, there has been a drastically increased interest in SMB in pharmaceutical industry for enantio-separations. SMB systems can also be integrated to include reactions, which can provide economic benefit for equilibrium limited reversible reactions. In-situ separation of the products facilitates the reversible reaction to completion beyond thermodynamic equilibrium and at the same time obtaining products of high purity. Recently, a new concept of non-synchronous switching instead of the synchronous one used in the traditional SMB technology is

developed. The more flexible modified process (Varicol) was found to perform better than the rigid SMB system. However, the successful operation and implementation of separative and reactive SMB (as well as Varicol) processes on industrial scales will necessitate one to determine the optimal design parameters and operating conditions based on multiple objectives.

In this paper, case studies of application of multi-objective optimization to evaluate the performance of a Simulated Moving Bed (SMB) system and its modification, Varicol process, for three industrially important systems, namely, chiral drug separation, production of concentrated fructose syrup and synthesis of methyl acetate ester have been carried out using artificial intelligence based robust optimization method non-dominated sorting genetic algorithm (NSGA). It was found that significant improvement is possible for both SMB and Varicol process.

## NOMENCLATURE

$C$	concentration, mol/L
$K$	equilibrium constant
$L$	length of column, m
$P$	purity
$p$	number of columns in section $P$
$q$	concentration in the solid phase, mol/L; number of columns in section $Q$
$Q$	volumetric flow rate, m <sup>3</sup> /s
$R$	reaction rate, mol/min/L
$r$	number of columns in section $R$
$S$	selectivity
$s$	number of columns in section $S$
$T$	temperature, K
$t$	time, s
$u$	superficial velocity, m/s
$X$	conversion
$Y$	yield
$z$	axial coordinate, m

## Greek symbols

$\alpha$	fraction of feed
$\beta$	fraction of raffinate withdrawn
$\gamma$	fraction eluent
$\delta$	phase ratio
$\varepsilon$	void fraction
$\zeta$	pseudo solid phase velocity
$\sigma$	relative carrying capacity
$\phi$	section

## Subscripts

$o$	initial, inlet
$e$	equilibrium

$f$	feed, forward
$g$	gas, carrier
$i$	component $i$
$j$	column number
$N$	number, switching period
$s$	solid, switching

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