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Two objective Optimization of Toluene Removal from Water by Pervaporation under the influence of permeate side pressure build up

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Abstract

Pervaporation is one of the several techniques employed for the removal of volatile organic components (VOCs) from waste water. The model used in the present study for the removal of VOCs from a multi-component aqueous mixture is a single stage pervaporation process using PDMS membrane in a shell and tube module without recycling of permeate. In our previous study [1] of two objective optimization of minimization of the treatment cost and maximization of the percent removal using an evolutionary algorithm of Non Sorting Genetic Algorithm-II, we have neglected permeate side pressure build up effect. However, the fluxes of the species through PDMS membrane were high and neglecting the permeate side pressure build up leads to unrealistic study. Hence, the present study considers the permeate side pressure build up effect for two objective optimization of treatment cost versus percent removal of toluene with decision variables of the problem as : reynolds number of the feed, membrane thickness, radius of the tube, length of the tube, downstream pressure, number of tubes, and toluene concentration in the feed. Thickness of membrane and permeate side pressure are found to be the most important decision variables. The study also reveals that optimal length of the module is 1.2 m and maximum removal percent attainable is 25% only. Further, attractive tradeoffs are available between the treatment cost and toluene removal percent.

Keywords: *Pervaporation, shell and tube module design, permeate pressure, VOCs, multi-objective optimization*

1. INTRODUCTION:

Removal of VOCs from water has been a global concern. Pervaporation is a fast emerging clean and cleaning technology for the removal of volatile organic compounds (VOCs) from water. This membrane-based separation technique is a better alternative to conventional techniques in terms of energy saving. With the advent of newer materials, membrane preparatory techniques, and module designs one can overcome the disadvantages of low flux and low selectivity of a pervaporation process. Several attempts have been made to find a better PV membrane material that can be used for VOC removal from water. The selectivity of PDMS for VOCs relative to water is found to be high [2, 3]. Proper design of the module is very essential for the commercial success of any process. A hollow fiber module with a very high packing density can offer higher permeation [3-6] for VOC removal from aqueous solutions as compared to plate-and-frame and spiral-wound modules. For commercial success of any process the effect of operating variables on the performance of the process should be thoroughly understood and optimized.

In our previous study [1] single stage pervaporation process in a shell and tube module with PDMS membrane without recycling of permeate and permeate side pressure build up effect was studied using an evolutionary algorithm of Non Sorting Genetic Algorithm-II. In reality the vacuum falls off towards the entrance of the module as it is applied at the opposite end. This drop in driving force due to accumulating permeate on downstream side of the membrane is known as permeate side pressure build up. Downstream pressure [7] build up affects flux and selectivity. Not only the flux may decrease but also collection velocities (close to or above sonic velocities) may increase which effects the efficiency and ease of operation.

Hence, the present study considers the permeate side pressure build up effect for two objective optimization of treatment cost versus percent removal of toluene from water in a shell and tube type of module with PDMS membrane. The decision variables used are *Reynolds number of the feed, membrane thickness, radius of the tube, Length of the tube, downstream pressure, number of tubes, and toluene concentration in the feed.*

The feed is taken on shell side and permeate is collected on the bore side. The overall fluxes of toluene and water are modeled based on film theory and resistance in series model [8, 9] as shown in Fig 1.

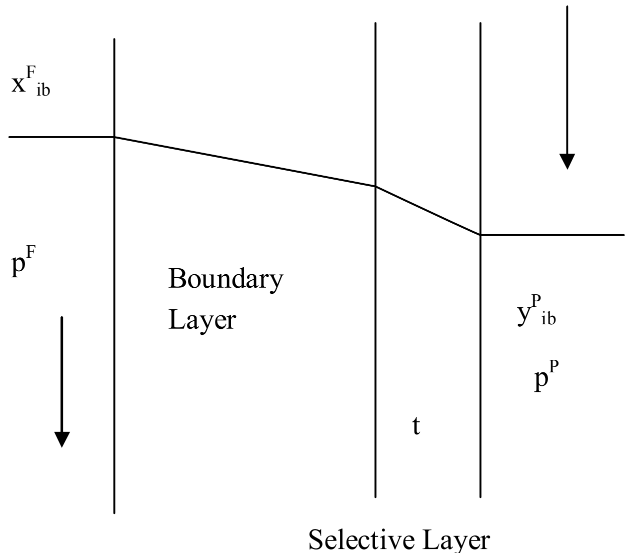

Fig-1: Schematic diagram of various resistances to mass transfer in Pervaporation process

2. NOTATIONS:

C	concentration (kmol/m^3)
C_P	specific heat
D	diffusion coefficient (m^2/s)
G	total permeate flow (kmol/s)
H	Henry's law constant (kPa m^3/mole)
L	length of the module (m)
N_{tube}	number of tubes
P	pressure (Pa)
p	downstream pressure (Pa)
PD	packing density
q	feed flow rate (m^3/s)
R_{tube}	radius of the tube (m)
Re	Reynolds Number
T	temperature (K)
t	membrane thickness (m)
x	retentate concentration mole fraction

Subscripts

app	applied
i, tol	toluene

3. THEORY & NUMERICAL SIMULATION:

The objective of the present work is to study two objective optimization of toluene separation from aqueous solutions in a shell and tube hollow fiber membrane module with permeate side pressure build up using NSGA-II to get Pareto optimal solutions.

The process model is available elsewhere [10, 11] by simplifying species molar, mass and momentum balances. The set of non-linear ordinary differential equations describing the properties variation on either side (shell side and bore side) are solved numerically.

The cost model of the pervaporation process for optimization study are available elsewhere [12].

The two objective unconstrained optimization problem is described as below:
Min annual treatment cost (Re, t, R_{tube}, L, P^P, N_{tube}, x_{tol})
Max toluene removal fraction (Re, t, R_{tube}, L, P^P, N_{tube}, x_{tol})
subject to:

$$20 < Re < 7100$$
$$5x10^{-5} < t < 25x10^{-5}$$
$$67.5x10^{-5} < R_{tube} < 90x10^{-5}$$
$$0.6 < L < 1.2$$
$$2200 < P^P < 4100$$
$$10^4 < N_{tube} < 10^6$$
$$9.8x10^{-6} < x_{Tol} < 9.8x10^{-5}$$

The removal percent of toluene is defined as

$$\frac{G_{Tol}}{q_{Tol}C_{Tol}} * 100$$

The tubular membrane is simulated by solving the overall and component continuity equations element by element on both shell and tube side of the shell and tube module. The total flux of toluene is obtained by summing the flux at each individual element. The removal percent of the toluene is estimated according to the eq 1.

The operating and module parameters for the study are given in the Table 1 and properties of the feed components are given in the Table 2.

Table 1: Operating and module conditions for simulation.

x_{in}	*Re*	$T_{in}(K)$	P_{app} (Pa)	T_{ap} (K)	*t µm*	*PD*
9.8x10⁻⁶	*20*	*303*	*200*	*303*	*50*	*0.1*

Table 2: Properties of feed components*.

Compound	**Heat capacity (J/mol. k)**	**Heat of Vaporization (kJ/mol)**	**Henry's Constant (k.Pa.m3/mol)**	**Diffusion Coefficient x10⁻⁹ (m2/s)**
Toluene	157.44	37.56	0.7358	1.97
Water	34.25	43.76		
* Temperature 30⁰C				

The study made by Satyanarayana et. al. [8] clearly shows that the GA parameters do not have much of an effect on the optimum value obtained for this optimization problem. Therefore, fixed GA parameters were used for the present work. The parameter values used for the study are given in Table 3.

Table 3: Genetic Algorithm parameters.

maximum number of generations	: 240
maximum population size	: 100
random seed	: 0.001
crossover parameter	: 0.70
mutatation probability	: 0.0143
SBX operator	: 5.0

4. RESULTS AND DISCUSSION:

In the process of pervaporation as the permeate starts accumulating from one end of the module while the vacuum is being applied from the other end. The permeate side pressure decreases gradually from the entrance of the module and attains the applied pressure at the exit of the module. This pressure build up from exit of the module to the entrance is due to the accumulating permeate. The permeate side pressure build up towards the beginning of the module is shown in Fig 2. On the other hand the permeate velocity goes on increasing with accumulating permeate which is shown in Fig 3.

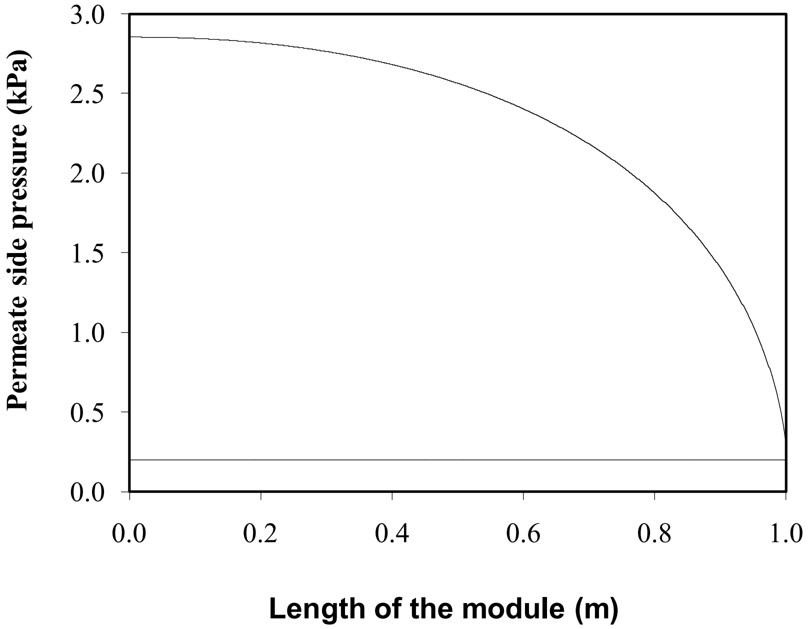

Fig- 2: Variation of permeate side pressure along the length of the module. P_{app} = 200 Pa, Re = 20, L = 1 m, L_{pot} = 0.1 m, t = 5 x 10^{-5} m, N_{tube} = 25x10^4

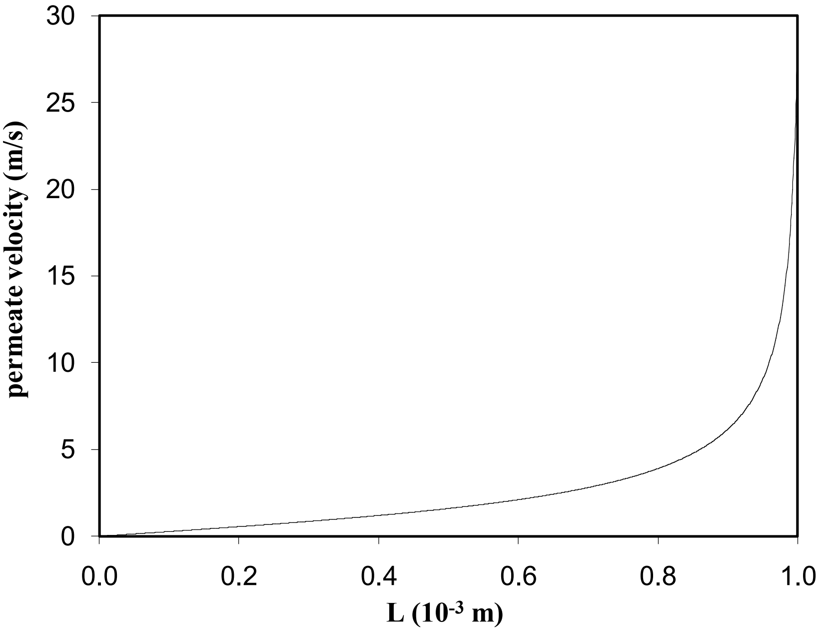

Fig 3: Variation of permeate collection velocity along the length of the module. P_{app} = 200 Pa, Re = 20, L = 1 m, L_{pot} = 0.1 m, t = 5 x 10^{-5} m, N_{tube} = 25 x 10^4

As we have stated earlier that the studies which do not consider the permeate side pressure build up effect can lead to non-realistic perceptions of the process. For example when permeate side pressure build up effect is not considered, the permeate pressure is same right from the entrance of the module to the exit of the module. Thus any amount of separation is achievable with single stage process (by increasing the length of the module) without recycle. Whereas when the buildup effect is considered, the increase of permeate side pressure towards the entrance of the module is limited to a maximum value that corresponds to the saturation vapor pressure of the feed. It is observed from simulation that the maximum length of the fiber (for the conditions mentioned in Fig 2) is 2.283 m only. Hence it may be impossible to attain a given removal percent of VOC by mere single stage without recycle. Further the separation efficiency of the pervaporation process and treatment costs are also non-realistic as the increase in downstream pressure can increase the selectivity, decrease the stage cut and increase the treatment cost.

The present study is presented in two parts; Pareto optimal study in objective function space and Pareto optimal study in decision variable space.

4.1 Pareto Optimal Study in Objective Function Space

Fig 4 represents the Pareto optimal study in objective function space. Fig 4 is the plot of removal percent versus treatment cost. The objective function space clearly shows that there exists tradeoff between the two objectives (i.e minimization of treatment cost and maximization of removal percent of toluene).

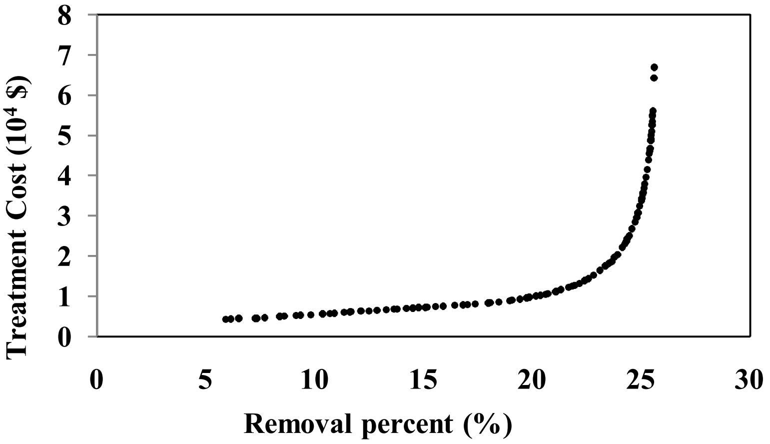

Fig- 4: Removal percent versus total cost Pareto solutions for removal of toluene from water.

Further, it may be inferred that for the given range of decision variables, when the removal percentage varies from 5.9 to 25.6, the total cost varies from \$ 4.37 x 10^5 to \$ 6.69 x 10^4 per year. Further all pareto solutions are found to lie in laminar region only near the lower bound of the Reynold number equal to 20. As per the previous study without permeate side pressure build up the percent removal varied from 72.6 to 73.8 and the treatment cost varied from \$2 .8x10^4 to \$ 1.07x10^7. Also as per the previous study the pareto optimal solutions were found both in laminar and turbulent regions. However the previous study was carried out for a retentate concentration of toluene less than or equal to 10% of that of the feed. The single stage removal of toluene was possible in previous case because the permeate side pressure build up effect is not considered. Thus the present study reveals the importance of considering the permeate side pressure build up effect in pervaporative removal.

4.2 Pareto Optimal Study in Decision Variable Space

Reynolds number of the feed, membrane thickness, radius of the tube, Length of the tube, downstream pressure, number of tubes, and toluene concentration in the feed are the decision variables chosen to study their effect on the Pareto optimal solutions. Their effect on the Pareto is given in the Fig 5.a to Fig 5.g. It is found that the optimal values of four of these seven decision variables are almost constant (*Reynolds number of the feed, radius of the tube, number of tubes, and toluene concentration in the feed*), all at their lower bounds, i.e., they are ineffective on the Pareto. On the other hand, *membrane thickness, Length of the tube, downstream pressure* are affecting the Pareto.

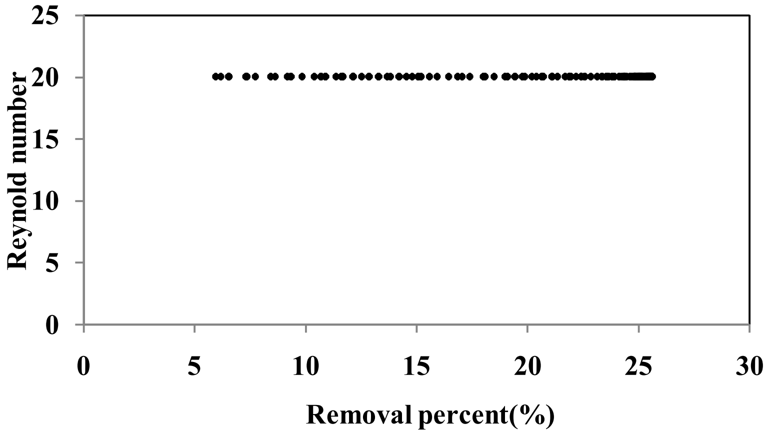

Fig- 5.a: Plot of removal percent versus Reynolds number of the feed.

The Reynolds number is varied between the lower limit 20 to 7100. It is clear that the Pareto solutions lye at the lower bound. This may be attributed to the fact that lower Reynolds number means lower volumetric flow rate on tube side and hence the removal percent is increased as per the eq 1. On the other hand, the decrease in Reynolds number decreases the organic flux by decreased mass transfer coefficient. Therefore the treatment cost is also favored. Hence Reynold number in pareto solutions is at its lower bound.

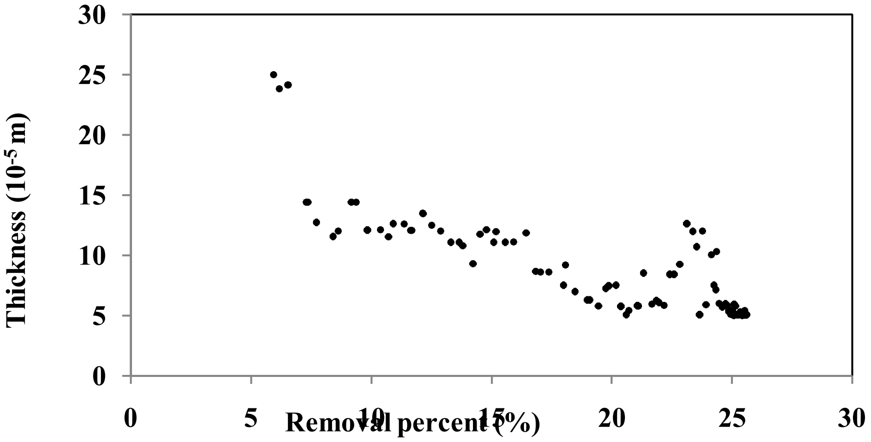

Fig- 5.b: Plot of removal percent versus thickness of the fiber

Thickness of the fiber is varied from $5x10^{-5}$ to $25x10^{-5}$ m. Study reveals that very few solutions are possible at the upper bound of the variable. These solutions correspond to lower end of the pareto solutions in objective function space, which means both percent removal and treatment cost are low. At these points the length of module is at its lower bound, the downstream pressure is at its upper bound. The poor driving force over short length of module results in lesser stage cut as well as degree of toluene removal. Thus both treatment cost and removal percent are low. Whereas at the other end of thickness pareto both driving force is high due to continuously decreasing down stream pressure and amount of permeate removed is high due to length at its higher bound. Thus these solutions correspond to solutions of high percentage removal and high treatment cost.

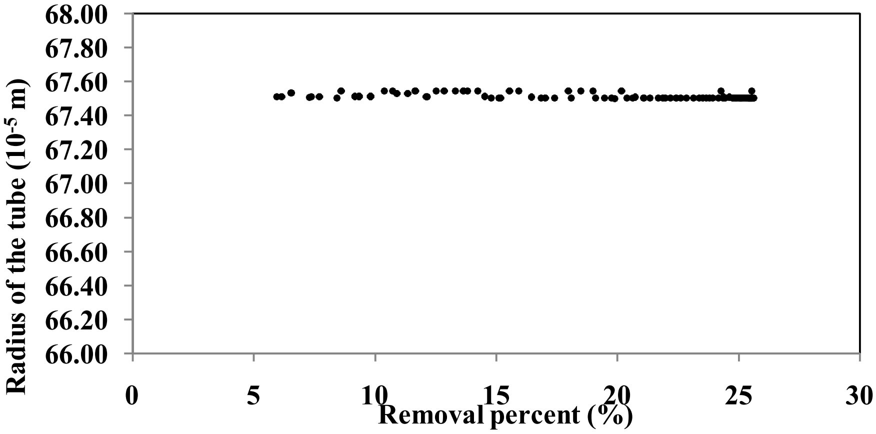

Fig- 5.c: Plot of removal percent versus radius of the tube

The radius of the tube is varied from $67.5x10^{-5}$ to 90 x 10^{-5} m and the Pareto falls at lower bound of radius. It is no surprising because decreased radius of tube gives decreased area of mass transfer which decreases the stage cut and increases selectivity. Therefore for favoring both the objectives of treatment cost and percent removal of toluene the pareto solutions fall at the lower bound only.

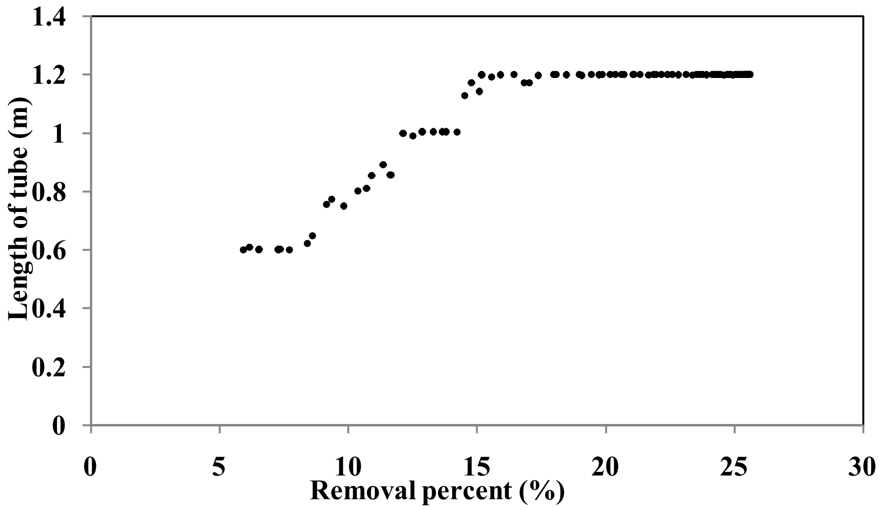

Fig- 5.d: Plot of removal percent versus length of the tube

The length of the tube is varied from 0.6 to 1.2 m. It is clear from the plot that the solutions vary from lower bound to upper bound of the variable. Hence length is a significant variable in decision variable space. In the previous study no bounds were placed on length of the module. It was because any length of the module is feasible when permeate side pressure build up effect is not considered. However module length in the range of 0.6 m to 1.2 is realistic and the present study accommodates the length of the module.

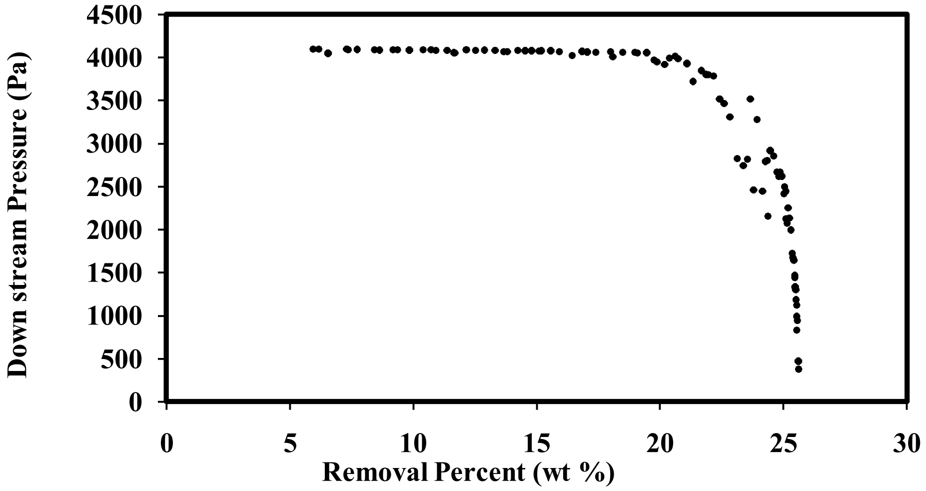

Fig- 5.e: Plot of removal percent versus downstream pressure

The applied down stream pressure varied from 378 to 4098 Pa. It can be seen that the applied down stream pressure is constant up to a removal percent of nearly 20. Whereas the other decision variables length and thickness are variying. The length of the module is increasing and the thickness is decreasing. Hence study reveals that applied down stream pressure is insignificant variable in this range and the other two variables are controlling the pareto solutions. In the other part of the pareto the downstream pressure is decreasing, thickness is low and simultaneously length of the module is high. The decreasing pressure increases driving force for mass transfer, the decreasing thickness decreases the resistance for mass transfer therefore the percent removal of toluene is favoured. The decreasing thickness and higher bound of length both increase stage cut therefore the treatment cost is also high.

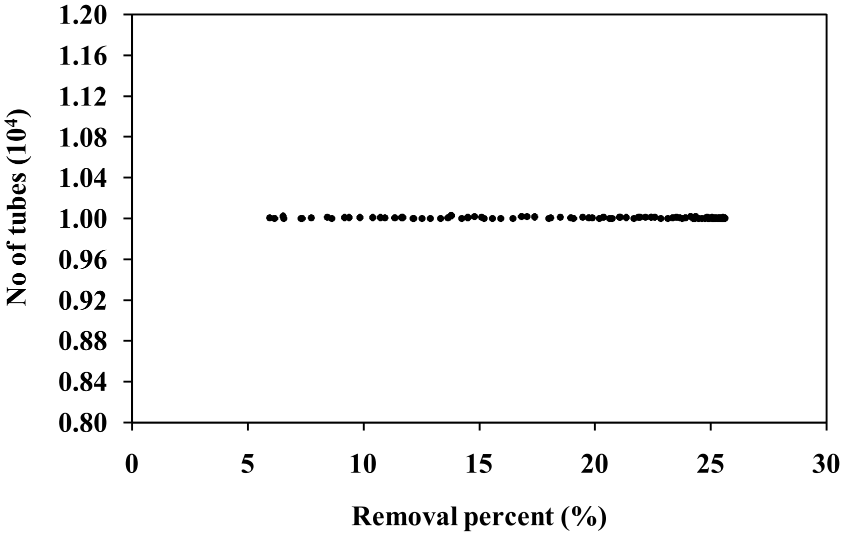

Fig- 5.f: Plot of removal percent versus number of tubes.

The number of tubes are varied from 10^4 to 10^6. As the number of tubes decreases the capital cost decreases and thereby the treatment cost is favoured. Hence the lower bound of number of tubes is picked for any percent removal.

The total membrane area A_T influences the Pareto set quite significantly. A_T, in turn, can be changed using any one or more of the four decision variables (R_{tube}, t, L, N_{tube}). However it is observed that t and L are most sensitive variables and complement each other to contribute for the removal percent as changes in the total membrane area are being achieved in this case by changing t and L, with the other variables (R_{tubed} and N_{tube}) at constant values.

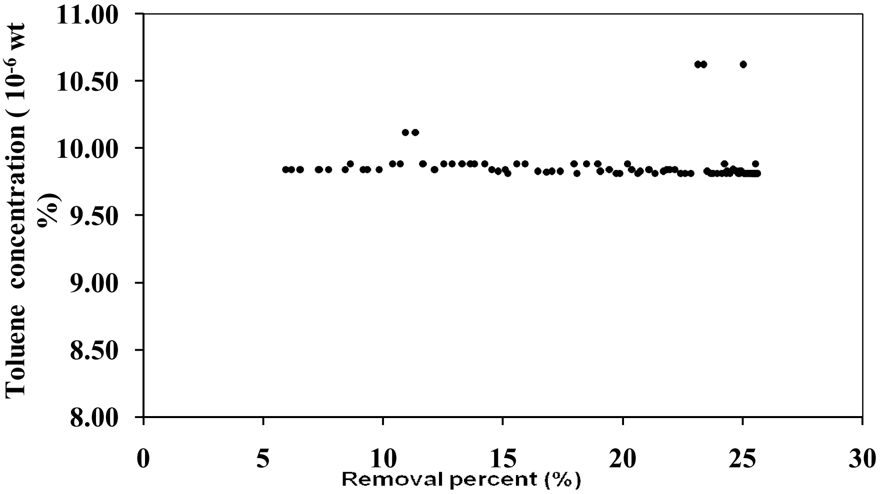

Fig- 5.g: Plot of removal percent versus concentrations of toluene

The concentrations of toluene is varied from 9.8 $x10^{-6}$ to $9.8x10^{-5}$ (wt %). The figures clearly indicate that concentration of toluene is insignificant in the Pareto.

4.3 Sensitivity analysis of the Pareto set to constant values of Re, R_{tube}, N_{tube}, x

The sensitivity of treatment cost with respect to the different variables is studied. Sensitivity analysis is carried out with a base case II picked up from the above Pareto optimal solutions. From the decision variable space it is clear that the Re, R_{tube}, N_{tube}, x are nearly constant, whereas t, L, p are found to vary over their entire range. Hence the reference cases III to X are obtained by varying the Re, R_{tube}, N_{tube}, x. These variables' values as presented in Table 4 are varied one at a time near the base case values to obtain Pareto solutions.

Table 4: Sensitivity analysis

	Decision variable	***Reference***	***Effect of Re***		***Effect of R_{tube}***		***Effect of N_{tube}***		***Effect of X_{Tol}***	
Problem no	***I***	***II***	***III***	***IV***	***V***	***VI***	***VII***	***VIII***	***IX***	***X***
Re	20-7100	20	18	22						
10^5 R_{tube} (m)	67.5-90	67.5			65.5	69.5				
10^4 N_{tube}	1-100	1					1.1	1.2		
10^6 X_{Tol} (wt%)	9.8 -98	9.8							10	10.2

for all cases I to X :
t = [$5x10^{-5}$ $25x10^{-5}$]
L= [0.6 1.2]
P = [2200 4100]

For number of tubes and toluene concentration the base case value are at their minimum values, hence the sensitivity Paretos are obtained on higher side only.

This sensitivity Pareto solutions shall be useful in identifying feasible designs which are optimal while designing the pervaporation process for the removal of VOC from waste water. These plots quantify the tradeoffs available between the removal percent and the treatment cost.

These diagrams are also useful to an engineer designing a Pervaporation module and will enable him to select feasible designs. Suppose if pervaporation module of higher radius is to be used in the process then the design engineers can know what will be the treatment cost, length and thickness of the fibers used for a given target of the removal percent.

These Pareto solutions are useful in reducing the designer's choices and thereby making the design options less cumbersome.

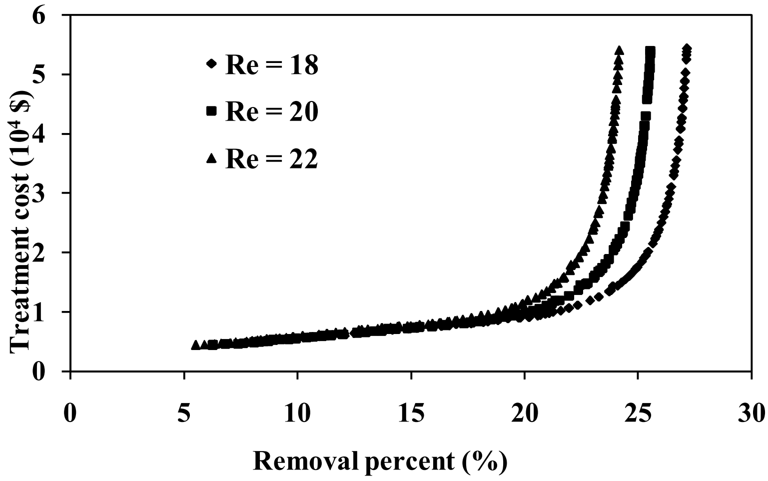

Fig 6-a: Effect of Re on the Pareto set

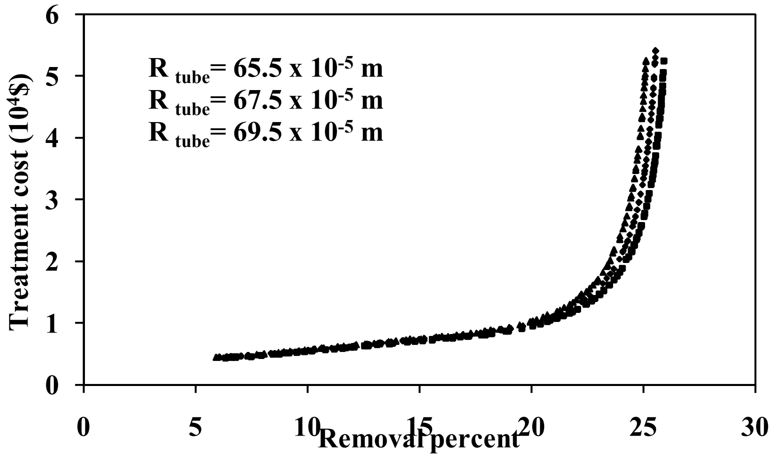

Fig 6-b: Effect of R_{tube} on the Pareto set

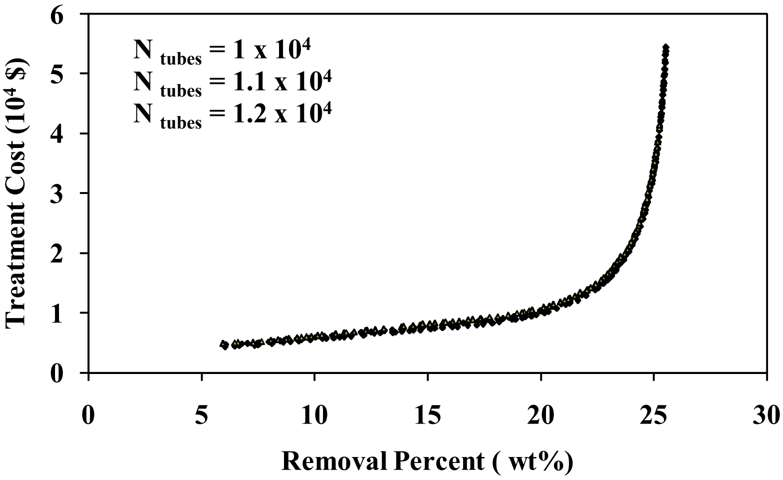

Fig 6-c: Effect of N_{tube} on the Pareto set

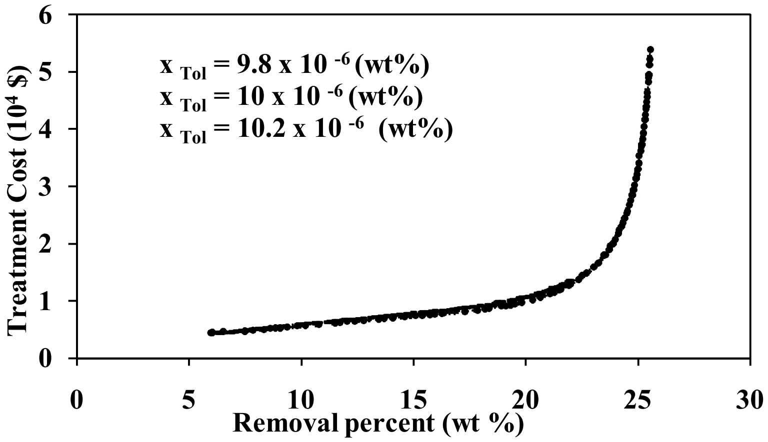

Fig 6-d: Effect of X_{tol} on the Pareto set

From Fig 6.a it is clear that treatment cost marginally increases with increase in Reynolds number. From Fig 6.b to 6.d it is apparent that the change in tube radius, number of tubes and toluene feed concentration has only minor effect on Treatment cost.

However the present study generalizes the removal of organic pollutants in waste water for single stage pervaporation process with permeate pressure build-up without recycle in a shell and tube type module.

5. CONCLUSIONS:

The two objective optimization study of simultaneous minimization of treatment cost versus maximization of percent removal of VOC (toluene) is carried out using evolutionary algorithm of real coded NSGA-II for removal of VOCs from waste water in a shell and tube type module with PDMS membrane by taking into consideration permeate side pressure build-up. The optimization study is carried out based on previously derived mathematical model. It is concluded that attractive tradeoffs are available between the removal percent of toluene and annual treatment cost. It is found that the optimal values of four of these seven decision variables are almost constant (*reynolds number of the feed, radius of the tube, number of tubes, and toluene concentration in the feed*), all at their lower bounds, i.e., they are ineffective on the Pareto. On the other hand, *membrane thickness, length of the tube and downstream pressure* are affecting the Pareto. An optimum module length of 1.2 m is recommended based on present study. The maximum removal percent attainable is only 25% and hence it is suggested to use hybrid process to improve separation efficiency.

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