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# Research on the Energy Saving and Environmental Protection Characteristics of a New Integrated Process of Distillation-Membrane Separation

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In view of the disadvantages of various types of industrial close-boiling liquid mixtures and azeotropic as well as high energy consumption and easy pollution caused by traditional separation methods, the integrated distillation-membrane separation process has obvious economic advantages of energy saving and consumption reduction. The systematic and economic research is helpful to realize the win-win of environment and economy. In this paper, a comprehensive solution strategy for the integrated distillation-membrane separation process based on intelligent genetic programming (GP) is proposed. According to the characteristics of the material system and separation requirements, the most suitable membrane type and material can be matched, and then the most economical and environmentally friendly new distillation-membrane separation process can be calculated quickly. On this basis, the hot issues related to chemical industry are further discussed, including energy saving and consumption reduction, carbon or other pollutant discharge control, etc. At the same time, the improvement of the separation model and the cost model are predicted for different membrane types. The significant influence of the above key factors on the system and economy of the integration process is discussed. The modification measures of membrane materials are proposed to predict whether they have industrialization potential and finally promote the common development of membrane and integration technology effectively.

## 1. Introduction

As contemporary social needs change from a single material demand to multiple needs, the requirements for the upgrading of the chemical industry are getting higher and higher. The society has proposed more changes to the chemical industry and enterprises in terms of energy consumption, pollution, green and sustainable development. In recent years, the development of membranes has been very rapid. However, membrane separation technology has limitations in terms of processing capacity, membrane life, etc. There are certain limitations to using only permeation gasification or steam infiltration to separate mixtures. Complementary advantages of other separation technologies can be considered to achieve the purpose of improving the overall economics of the process (Sun and Jiang, 2011).

Muhammad et al. (2019) proposed a system framework that can efficiently design a distillation separation process. Ahadi et al. (2018) and others tried a new distillation-membrane module design method. The influence of baffle structure on the performance of three modules of liquid-gap membrane distillation was studied. Roth et al. (2013) optimized and analyzed mixed processes by using evolutionary algorithm and non-equilibrium model. Schwantes et al. (2018) proposed a technical design and economic analysis of membrane distillation (MD) suitable for the zero liquid discharge process chain, and compared it with the cost of mechanical vapor compression (MVC) for the same application. The results show that the cost-effectiveness of MD is much higher than MVC. Khalid et al. (2019) concluded that when mixed with different schemes, the combination of distillation and membrane separation technology can effectively reduce the cost of bioethanol.

Although great progress has been made in the integration of distillation-membrane, there are still many problems waiting to be solved in the field of calculation of distillation-membrane integration processes. For example, for most azeotropic and near-azeotropic systems, there is no universally applicable calculation method. There are many possibilities for integrating the distillation-membrane process. Under the condition of multiple integration,

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the calculation and optimization of each logistics parameter also needs to be performed, and the optimization of multi-objective problems become very difficult. This makes it very urgent to develop a general algorithm to achieve a fast and flexible solution to the corresponding optimal distillation-membrane separation integration process. Aiming at these problems, a genetic programming algorithm is proposed in this paper. GP is a multi-objective optimization algorithm, which has wide applicability in multiple disciplines. Compared with other algorithms, GP has many outstanding advantages. For example, the biggest difference between GP and genetic algorithm is that GP is no need to define the superstructure in advance. It is more conducive to the comprehensive optimization of chemical integration process with multiple variables and high integration.
In this paper, on the basis of the previous research, the comprehensive solution strategy of distillation-membrane separation for industrial azeotropic system separation is further studied to meet the separation requirements of different azeotropic systems (Wang et al., 2008). According to the composition of the raw materials of different systems and the characteristics of different membrane materials, it can quickly and accurately find the best distillation-membrane separation integration process (Wang and Li, 2010). Under the Visual Studio platform (VS), a set of optimized GP algorithms suitable for the integration process of distillation membrane is written in C++ language. Taking the benzene-cyclohexane azeotrope system and polyurethane membrane as a example, the optimal process of the system and the optimal flow rate and data under the corresponding process conditions are obtained. The theory of membrane performance is extended and its influence on the integrated distillation-membrane separation process is predicted. The comprehensive solution strategy proposed in this paper can flexibly modify the cost models of various types of membranes and provide assistance for the future industrialization of membranes that are still in the experimental research stage for further modification.

## 2. New GP of distillation-membrane separation integration process

Through the use of GP, a set of efficient distillation-membrane separation integration solutions have been developed. In this paper, GP tree code is used to represent the process relationship. Complex chemical processes are abstracted into tree structures and optimized with computers. A chemical node model library is established, a tree-like code generation rule is established, a suitable fitness calculation method is established, and evolution rules are formulated under the guidance of a chemical engineering specialty.

### 2.1 Establish the cell node model

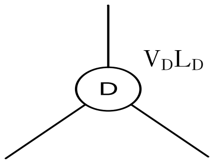
Establishing the node model of chemical unit is the key of this algorithm, which includes the definition of distillation column and membrane separation.

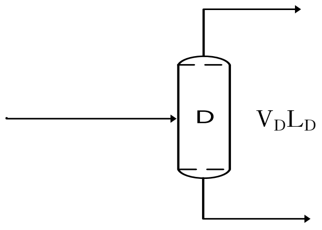
#### 2.1.1 Establishment of distillation column element node D

The distillation column is defined as a simple column with one input and two outputs. The top of the tower is equipped with a full condenser, and the tower kettle is an indirect steam heating reboiler. The separation characteristic values of distillation column nodes are defined as follows:

(i) The theoretical plates is $P_D$.
(ii) The value of reflux ratio divided by minimum reflux ratio is $V_D$.
(iii) The recovery of light components at the top of the tower is $L_D$.

The distillation column node has a feed stream, and the two branches represent the overhead stream and the bottom stream. The column parameters of each structure are imported into the ASPEN software for calculation. The calculation results of material balance and heat balance are returned to VS for subsequent fitness calculations. The distillation column node model and its corresponding distillation column separation structure are shown in Figure 1.




*(a) Distillation node* *(b) Distillation operation*

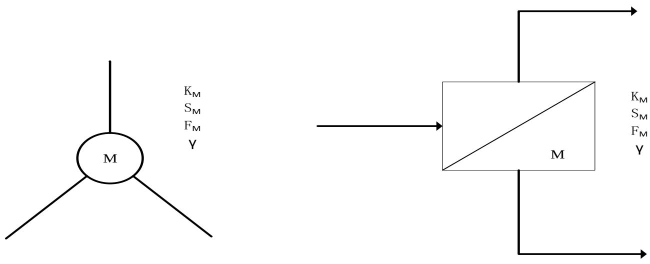
*Figure 1: The distillation column node and corresponding distillation operation*

### 2.1.2 Definition of membrane separation node M

In order to calculate accurately, the research object of the membrane separation node selects the membrane with experimental data support. A membrane separation model was obtained by regression of experimental results. In this paper, a pervaporation model (M) is selected as the membrane separation node model. The membrane separation process is also simplified to one in and two out. In VS, the membrane is abstracted as a mathematical model. The parameters of the membrane separation result are affected by many factors, such as operating temperature, thickness, pressure drop, and crosslinker concentration.
The membrane unit is abstracted into a unique mathematical model with four characteristic parameters:

- (i) The serial number of the membrane is represented by $K_M$.
- (ii) The membrane separation factor is represented by $\gamma$.
- (iii) The membrane separation area unit is $S_M$ ($m^2$).
- (iv) The flux per unit area of the membrane is $F_M$ ($kg \cdot h^{-1} \cdot m^{-2}$).

All membrane units are calculated in VS. Experimental conditions are coded and stored in $K_M$ according to coding rules. The membrane separation node and its corresponding membrane separation operation are shown in Figure 2.




*(a) Membrane separation node (b) Membrane separation operation*

*Figure 2: The membrane separation node and corresponding membrane separation operation*

### 2.2 Algorithm tree graph generation

Tree code generation, inheritance and growth are the foundation of GP. In principle, GP algorithm can select a node randomly as the root in distillation node and membrane separation node. In the application of azeotrope systems, close-boiling systems and membranes studied in this paper, through consideration of the actual situation, the separation results of the distillation nodes can only reach the azeotropic point and cannot cross. There are similar situations in membrane joints. Most of the real membranes have certain requirements for the feed conditions, especially for the feed composition. This paper proposes an intelligent algorithm rule to assist GP in intelligent calculation. In principle, the algorithm can carry out random calculation of distillation node and membrane node at each node. After adding the above practical conditions to improve GP, it can meet the actual production situation without losing the randomness. An example of a tree-like code generation diagram suitable for a distillation membrane integration process is shown in Figure 3. “end” represents the convergence of logistics information. “...” means the tree will continue to grow down.

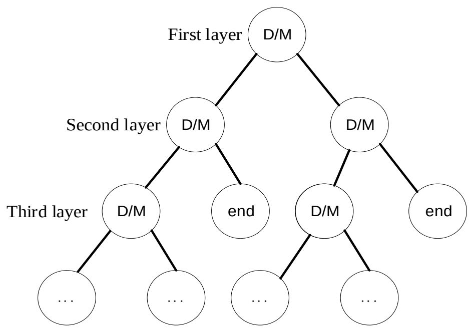



*Figure 3: Flow chart of calculating steps*

### 2.3 Fitness calculations and evolutionary rules

In this paper, fitness is the total annual cost (TAC), which includes equipment costs and operating costs. Such as distillation column equipment costs, membrane equipment costs, hot water electricity costs and cold water electricity costs. This can intuitively reflect the economics of the process. After generating the tree-like code, the fitness is calculated, and the objective functions of the fitness value are discussed separately to solve hot issues such as energy saving and environmental protection, and equipment improvement. This greatly increases the scope of use of GP, which is convenient for its application to different goals or multi-objective optimization. Minimizing the cost of the entire process is the optimization goal of this paper. The lower the TAC of a single process, the higher the fitness of the individual, and the more likely it is that the excellent characteristics can be passed on to the next generation.

The initial population is randomly generated, and subsequent evolution operations are performed on the basis of the initial population. Evolutionary operations include copying, swapping, and mutating to establish an algorithmic evolutionary rule suitable for the chemical industry.

(i) Replication operation: In this paper, competitive selection method is used to select good individuals from the previous generation to new populations, until the optimized replication rate requirements are met.

(ii) Exchange operation: Two different types of nodes cannot be exchanged directly. Only the nodes after the two branches representing the same stream information can perform the exchange operation until the optimized exchange rate requirements are met.

(iii) Mutation operation: A node of an individual is randomly selected, the original content of the node is deleted, and a new node and subsequent branches are randomly generated to generate a new individual to be added to the new population until the optimized mutation rate is met.

### 2.4 Improved GP algorithm

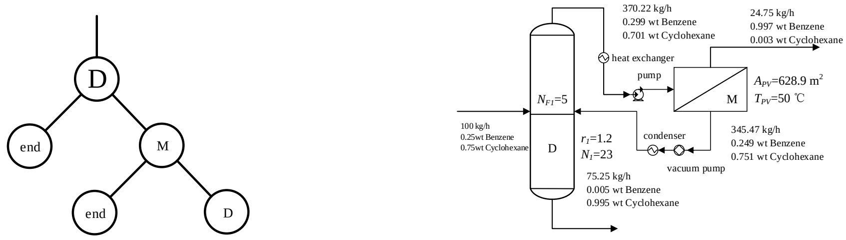
The structure of the solution of the optimization problem is represented by tree structure. The set of initialized solutions is denoted as the initial population. After the initial population is obtained, a series of evolutionary operations are carried out on the population in the next step. The operation object of evolution operation is the population, which is usually called the parent generation by the evolutionary population, and the population obtained after the evolutionary operation is generally called the offspring generation. After the individuals in the parent generation are selected at random for corresponding evolutionary manipulation, they are put back into the population to form progeny with individuals who have not evolved. They are repeated as the parent of the next evolutionary operation. The resulting offspring are repeated as the parent of the next evolutionary operation. There are three types of evolutionary operations: replication, exchange and variation. These three operations correspond to three evolutionary forms of genetic material in nature. The proportions of the three operations are not specified and can be adapted to separate tasks.

The goal of termination criterion design is to stop the evolutionary operation to obtain the optimal solution of the optimization problem when the population evolution reaches a certain degree. An excellent termination criterion should have two performances: stopping the evolution operation when the global optimal solution has been retrieved in the population evolution; Increase the effective evolutionary operation when the population evolution is not globally optimal. In this paper, the maximum permissible evolutionary algebra is used as the termination criterion.

## 3. Industrial calculation examples

In this section, in order to verify the feasibility of the comprehensive solution of the integrated distillation-membrane separation process in the near-azeotrope system, the benzene-cyclohexane azeotrope system is selected as the research object. The benzene-cyclohexane system forms a near-azeotrope when the benzene content is 45 wt%. At this time, the normal distillation operation is difficult to perform effective separation. The research uses three typical feed flows (100 kg/h, 300 kg/h, 500 kg/h) to search for the optimal integration process under multiple feed combinations. Taking polyurethane membrane as the research object of integration process optimization, the impact of possible modification of membrane in the future on the profit is explored. In this paper, the theoretical extension of membrane separation factor and flux is studied. The basic conditions for a polyurethane membrane are: operating temperature is 50 °C, vacuum degree downstream of the membrane is 0.1009 MPa, and membrane thickness is 0.12 mm. (Yao et al., 2016)

When the feeding situation is 100 kg/h and the benzene content is 25 wt%, the optimal individual tree structure calculated by the comprehensive solution strategy is shown in Figure 4.




*(a) GP tree structure* *(b) Corresponding flow chart*

*Figure 4: GP tree structure and corresponding flow chart*

### 3.1 Optimization results and analysis of real polyurethane membrane

Table 1 lists the optimal integration processes searched when the benzene content of other feed flows is 25 wt% and their corresponding process parameters. All processes are distillation-membrane (D-M) circular logistics with the benzene content of 25 wt% in the back-flow at the residual side of membrane infiltration. When the membrane separation factor is low, the comprehensive solution strategy will search out the D-M cycle structure at a lower feed flow.

*Table 1: Summary table of integrated distillation-membrane separation processes for optimized benzene-cyclohexane system*

| Feed flow (kg·h$^{-1}$) | Theoretical forcast price ceiling of per unit membrane area ($·m$^{-2}$·y$^{-1}$) | $N_1$ | $r_1$ | $A_{PV}$ (m$^2$) | $O_{cost}$ ($·y$^{-1}$) | $S_{cost}$ ($·y$^{-1}$) | $M_{cost}$ ($·y$^{-1}$) | $T_{cost}$ ($·y$^{-1}$) | TAC ($·y$^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|
| 100 | 44 | 23 | 1.2 | 629 | 1,594 | 36,594 | 27,670 | 36,112 | 65,858 |
| 300 | 39 | 29 | 1.5 | 1,870 | 2,617 | 50,700 | 72,939 | 59,239 | 134,519 |
| 500 | 41 | 36 | 2.1 | 3,053 | 3,487 | 54,075 | 125,209 | 63,562 | 192,688 |

$N_1$ is tray number; $r_1$ is repreflux ratio; $A_{PV}$ is membrane area; $O_{cost}$ is operating cost; $S_{cost}$ is equipment cost; is $M_{cost}$ is membrane cost; $T_{cost}$ is column cost; TAC is total annual cost.

The data in the table show that with the increase of the feed flow, the membrane area used per unit feed amount gradually decreases, but the relative decrease of the membrane area does not change the characteristics of the low scale effect of membrane separation, resulting in the overall decrease of the virtual membrane cost, which is consistent with the characteristics of the distillation-membrane integration process.

### 3.2 The influence of polyurethane membrane performance prediction on the integration process

In this section, different theoretical predictions of polyurethane membranes are performed in two directions: separation factor modification and flux modification. The effects of different modification directions and modification ranges on the virtualization cost and the distillation-membrane integration process are discussed. In the direction of separation factor modification, two different cases of 20 % increase in separation factor and 10 % decrease in separation factor are discussed. These two cases represent two common situations in the optimization direction of the separation factor, namely increasing the separation factor without affecting the flux and increasing the separation factor and appropriately reducing the flux. The first case is bound to have a higher theoretical forecast price ceiling of per unit membrane area. The second case has an improvement that requires actual calculation and discussion. These two similar cases are also discussed in the flux modification direction. The optimal process retrieved by all membranes at the highest virtualization cost is the D-M cycle process. Table 2 lists the optimal integration process parameters for different modification cases.

*Table 2: The optimal integration process parameters for different modification cases*

| Separation factor increase | Total flux increase | Theoretical forcast price ceiling of per unit membrane area ($·m$^{-2}$·y$^{-1}$) | $N_1$ | $r_1$ | $A_{PV}$ (m$^2$) | $O_{cost}$ ($·y$^{-1}$) | $S_{cost}$ ($·y$^{-1}$) | $M_{cost}$ ($·y$^{-1}$) | $T_{cost}$ ($·y$^{-1}$) | TAC ($·y$^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|---|
| 0 | 0 | 44 | 23 | 1.2 | 629 | 1,594 | 36,594 | 27,670 | 36,112 | 65,858 |
| +20 % | 0 | 50 | 21 | 0.9 | 587 | 1,467 | 35,025 | 29,356 | 33,969 | 65,848 |
| +20 % | -10 % | 42 | 22 | 1.2 | 677 | 1,770 | 35,783 | 28,422 | 35,433 | 65,974 |
| 0 | +20 % | 61 | 20 | 1.2 | 511 | 526 | 34,356 | 31,139 | 31,339 | 66,021 |
| -10 % | +20 % | 54 | 24 | 1.7 | 477 | 1,686 | 37,235 | 26,728 | 37,227 | 65,649 |

Based on the membrane before the theoretical prediction, when the separation factor increases by 20 % and the flux decreases by 10 %, theoretical forecast price ceiling of per unit membrane area (TFP) decreases. When the separation factor increases by 20 %, TFP increases by only 13.6 %. That means, when the separation factor is increased by 20 %, if TFP increases by 13.6 % more, economic benefits cannot be obtained through this modification method. There is a huge change when the flux changes. When the flux increased by 20 %, TFP increased by 38.6 %. That is, when the flux is increased by 20 %, if the TFP increases by 38.6 % lower, such a membrane modification method can bring economic benefits. For polyurethane membranes, the impact of flux on membrane separation effect is much higher than the separation factor. Modification of polyurethane membranes in the direction of flux enhancement can obtain greater modification benefits. This conclusion provides a basis for the comprehensive solution strategy applied to guide the modification direction of the membrane. The comprehensive solution strategy can calculate the modification income in advance to achieve the purpose of suggesting the membrane modification.

## 4. Conclusions

There are many integration possibilities in the distillation-membrane integration process, such as the complete coupling between the membrane and the distillation unit and the incomplete coupling. Under the background of multiple integration possibilities, the calculation and optimization of each logistics parameter should be done. It is urgent to develop a general algorithm that can quickly and flexibly solve the matching optimal distillation-membrane separation process. GP algorithm is proposed to solve these problems. The distillation-membrane integration process can be widely used in the study of different azeotropic or near-azeotropic systems, providing a basis for the industrial development and modification of different types of membranes in the future. Taking the benzene cyclohexane near-azeotropic system and polyurethane membrane as examples, the optimal distillation-membrane integration process with various feed flows is optimized. The reliability of the algorithm is verified by the in-depth analysis of the optimization results. The GP algorithm proposed in this paper is universal and can match more suitable membrane types and materials for different systems. The same applies to lab-scale membranes. By comparing the optimal process cost with the cost of the industrial process being used, the industrial application scale of the integrated process with the greatest economic benefit under the current membrane cost. It can provide suggestions for membrane modification, better promote the development of membrane and integrated technology, and have far-reaching significance for energy conservation and environmental protection in the chemical industry.

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## References

Ahadi H., Karimi-Sabet J., Shariaty-Niassar M., Matsuurac T., 2018, Experimental and numerical evaluation of membrane distillation module for oxygen-18 separation, Chemical Engineering Research and Design, 132, 492-504.

Khalid A., Aslam M., Qyyum M. A., Faisal A., 2019, Membrane separation process for dehydration of bioethanol from ferm entation broths: Recent developments, challenges, and prospects, Renewable and Sustainable Energy Reviews, 105, 427-443.

Muhammad A.Z., Muhammad F.I.Z., Munawar Z.S., Kamarul A.I., Mohd K.A.H., 2019, Economic, feasibility, and sustainability analysis of energy efficient distillation based separation processes, Chemical Engineering Transactions, 72, 109-114.

Roth T., Kreis P., Górak A., 2013, Process analysis and optimisation of hybrid processes for dehydration of ethanol, Chemical Engineering Research and Design, 91, 171-1185.

Schwantes R., Chavan K., Winter D., Felsmannb C., Pfafferottc J., 2018, Techno-economic comparison of membrane distillation and MVC in a zero liquid discharge application, Desalination, 428, 50-68.

Sun H.Y., Jiang L.F., 2011, Research progress of hybrid-distillation-pervaporation process, Chemical Industry Times, 25(12), 48-52.

Wang X.H., Li Y.G., 2010, Stochastic GP intergration of heat integrated nonsharp distillation sequences, Chemical Engineering Research and Design, 88, 45-54.

Wang X.H., Hu Y.D., Li Y.G., 2008, Synthesis of nonsharp distillation sequences via genetic programming, Korean Journal of Chemical Engineering, 25(3), 402-408.

Yao L.L., Ye H., Song Y., Cui P., 2016, Conditions and process of pervaporation benzene/cyclohexane mixture by waterborne polyurethane membranes, CIESC Journal, 67(S1), 289-295.