Research Article

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Simulation and improvement of the separation process of synthesizing vinyl acetate by acetylene gas-phase method

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Abstract: Vinyl acetate, as an essential organic chemical raw material, can be used to produce polyvinyl acetate, polyester vinyl alcohol, and other products. The existing classical vinyl acetate production process has the problems of low product purity and excessive heat load. In this study, in the classical design of the process, acetylene is separated first, and then acetaldehyde is removed with the formation of an azeotrope between ethylene acetate and water. Meanwhile, considering the solubility of acetaldehyde in water and insolubility of vinyl acetate in water, the process was optimized to separate acetic acid after removing acetylene, so as to avoid the azeotrope formation of vinyl acetate and water. The nonrandom two-liquid-Hayden–O’Connell thermodynamic hybrid model was used to simulate the classical process and improved process (IP). Finally, the reflux ratio and theoretical tray number of the main separation equipment of IP were optimized to get the better parameters. The simulated results show that the purity of vinyl acetate increased from 99.1% to 99.8%, the cooling energy consumption was reduced by 16.83%, and the thermal energy consumption was reduced by 6.18%. At the same time, the equipment investment was also decreased.

Keywords: vinyl acetate, process simulation, separation, azeotrope

1 Introduction

Vinyl acetate, an important organic chemical raw material, can be used in the production of polyvinyl acetate, polyester vinyl alcohol, vinyl acetate-vinyl copolymer emulsion, and other derivatives. It has a wide range of applications in paint, paste, adhesives, vinyl fiber, film, and other aspects [1]. Till date, the industrial production methods of vinyl acetate mainly include acetylene method and ethylene method. The acetylene method is the reaction of acetylene and acetic acid under atmospheric pressure with activated carbon–zinc acetate as a catalyst, and the vinyl acetate products are obtained after separation and purification. The ethylene method is based on ethylene, acetic acid, and oxygen with palladium or gold as the main catalyst and potassium acetate as a cocatalyst for the reaction. The vinyl acetate products are obtained after purification. Due to the energy structure characteristics of “rich coal, less oil, and poor gas” in China, the acetylene resources produced by coal are relatively abundant, and the acetylene method used for the production of vinyl acetate accounts for 63.9% of the total output [2]. As a significant raw material, the purity of vinyl acetate products directly affects the quality of downstream products. Meanwhile, the separation process of vinyl acetate consumes a lot of energy. According to the policy of energy conservation and emission reduction in China, it is very important to reduce the energy consumption and production cost. Therefore, it is very important to improve the acetylene method.

With the development of high-performance computers and the rapid development of computing power, process simulation has been more widely used in process calculation and process optimization. Plesu Popescu et al. [3] simulated the production process of vanillin with sulfate lignin, and several alternative process design schemes with fewer rectifying towers and lower total energy consumption were proposed to make the production process of vanillin cleaner and more energy saving.

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Petrescu et al. [4] extracted phenol with seven entrainers (aromatics, ketones, etc.) based on the process simulation tool to remove phenol from water. By comparing the simulation data, it was concluded that the amount of extractant used in extracting phenol with cyclohexanone was the least, which achieved the purpose of optimizing the process. Kong et al. [5] used the process calculation software to simulate the chemical cycle process, as well as the traditional steam methane conversion and mixed conversion process. The simulated result showed that the chemical cycle process improved the cooling air when compared with the traditional reforming process, and the effective thermal efficiency was also improved. For the process of vinyl acetate synthesis, the research in recent years has mainly focused on the high-performance catalyst and optimized synthesis process. For example, Jiang et al. [6] prepared zinc and acetate/carbon bead catalyst with carbon beads as the carrier, and the test showed that the catalyst had a good catalytic activity, selectivity, and reaction stability. Zhu et al. [7] obtained activated carbon with different oxygen-containing groups through ozone flow heat treatment, indicating that the oxygen-containing groups on the surface of the activated carbon played a key role in the catalytic performance of the vinyl acetate catalyst. Zhang et al. [8] studied and optimized the production of benzene as a by-product in the production process of vinyl acetate with acetylene. It was concluded that the generation of benzene could be inhibited at 453 K under low pressure and in the acetylene/acetic acid molar ratio range of 2–2.6.

There are only few studies on the separation process. Li et al. [9,10] simulated the fourth column of vinyl acetate distillation and optimized the operating parameters of the column. Tu et al. [11] calculated the acetaldehyde refining tower and optimized the parameters, thus improving the recovery rate of acetaldehyde. Ye et al. [12,13] simulated and calculated some other processes, such as the coarse separation tower and vinyl acetate refining tower, and then made improvement and parameter optimization to achieve the purpose of improving product purity and reducing total energy consumption. However, these improvements and optimizations were all aimed at a single separation equipment, which had certain limitations. In the current classical process (CP) [14], the mixed solution after the reaction first removes acetylene and then acetaldehyde and finally separates acetic acid and vinyl acetate. The CP has been improved many times by the predecessors, and there are still the problems of low vinyl acetate purity and large energy consumption. We found that vinyl acetate and water can form the lowest azeotrope, which can be used. We try to solve the problems of low product purity

and high energy consumption by adjusting the separation process.

In this study, the whole process of classical vinyl acetate separation was simulated, and the analysis and optimization of the whole process were conducted. Two key process parameters of the reflux ratio and theoretical tray number were compared to obtain lower investment and energy consumption on the basis of vinyl acetate purity requirement.

2 Selection of thermodynamic models

In the whole production process of vinyl acetate, the materials involved include raw materials acetic acid and acetylene, the main product vinyl acetate, and by-products acetaldehyde, ethyl ester, and other polar substances, which belong to a polar nonideal system.

The nonrandom two-liquid (NRTL) equation, which is proposed by Renon and Prausnitz [15], considered that the process of liquid mixture should not proceed randomly and there should be local composition. Therefore, a nonrandom factor a_{ij} was introduced, and the specific nonrandom two-fluid equation was proposed as follows:

$$\frac{g^E}{RT} = x_1 x_2 \left(\frac{\tau_{21} G_{21}}{x_1 + x_2 G_{21}} + \frac{\tau_{12} G_{12}}{x_2 + x_1 G_{12}} \right) \quad (1)$$

$$\ln \gamma_1 = x_2^2 \left[\frac{\tau_{21} G_{21}^2}{(x_1 + x_2 G_{21})^2} + \frac{\tau_{12} G_{12}^2}{(x_2 + x_1 G_{12})^2} \right] \quad (2)$$

$$\ln \gamma_2 = x_1^2 \left[\frac{\tau_{12} G_{12}^2}{(x_2 + x_1 G_{12})^2} + \frac{\tau_{21} G_{21}^2}{(x_1 + x_2 G_{21})^2} \right] \quad (3)$$

$$\tau_{12} = \frac{g_{12} - g_{22}}{RT} \quad (4)$$

$$\tau_{21} = \frac{g_{21} - g_{11}}{RT} \quad (5)$$

$$G_{12} = \exp(-\alpha_{12} \tau_{12}) \quad (6)$$

$$G_{12} = \exp(-\alpha_{12} \tau_{21}) \quad (7)$$

where g_{ij} is the energy parameter representing the interaction between the two components, and α_{12} is related to the nonrandomness of the two components. NRTL equation can be used to study the highly nonideal systems, especially the partial miscible systems. It can be used to express the experimental data well [16].

The Hayden–O'Connell equation [17] of state is used to calculate the thermodynamic properties of the gas

Table 1: Tower equipment operating parameters

Equipment ID	Plate number	Reflux ratio	D/F	Feed plate position	Top pressure (kPa)
5	30				131.33
6	15				111.33
7	10		0.0025	1	161.33
9	20	2.0	0.98	10	161.33
10	25	2.0	0.575	10	151.33
11	25	2.2	0.413	8	161.33
12	22	2.5	0.095	10	191.33
13	20	3.0	0.05	5	161.33
15	30				121.33

and the washing tower for pickling and washing, and finally the 0105 returns to the reactor. The pickling liquid from the pickling tower refluxes to the degassing tower, and then the degassing tower liquid 0104 enters the crude acetaldehyde separation tower. The mixed liquid phase of acetaldehyde on the top enters the acetaldehyde extraction tower, and the extract enters the acetaldehyde refining tower. The acetaldehyde product 0106 is obtained on the top of the tower. The raffinate in the extraction tower and the kettle liquid of the tower enter into the vinyl acetate crude separation tower. The vinyl acetate crude tower distillate enters into the vinyl acetate refining tower, and the vinyl acetate products 0108 is obtained on the top of the tower. The kettle liquid returns to the coarse separation tower, vinyl acetate crude tower kettle liquid parts backflow pickling after cooling tower, the part into the acetic acid refining tower. The acetic acid 0109 on the top of the tower flows back to the acetic acid evaporator.

After simulation calculation, the tower equipment operating parameters are shown in Table 1, and the main-stream results are shown in Table 2.

As shown in Table 2, the purity of refluxing acetylene is 98.86%, and the content of acetylene in the degassing tower kettle liquid 0104 is almost 0. The purity of vinyl acetate product, by-product acetaldehyde, and refluxing acetic acid reaches 99.1%, 99.9%, and 99.5%, respectively, which meets the design requirements.

4 Simulation of improved process (IP)

4.1 Process improvement analysis

According to the simulation results of the classical separation process, the molar fraction of vinyl acetate in the feed system of the vinyl acetate refining tower is 56.15%, and the water content is 1.03%. Based on the chosen physical property method, the binary phase diagram of vinyl acetate–water system was calculated and analyzed. The analysis and calculation results are shown in Figure 2.

The formation of azeotrope is caused by the deviation from the ideal solution due to the difference in molecular structure among the components in the mixed solution. The lowest azeotrope will be formed when the activity coefficient is greater than 1, and the highest azeotrope will be formed when the activity coefficient is less than 1. Sometimes, when the positive deviation between the real mixed solution and ideal solution is large enough, the mutual solubility decreases, resulting in the incomplete dissolution of lowest azeotropes formed between the components in the liquid phase, that is, forming a heterogeneous azeotrope. Meanwhile, the liquid mixture called homogeneous azeotrope is completely soluble without stratification, at the azeotropic temperature. As shown in

Table 2: Main flow results of CP flow simulation

	0101	0102	0103	0104	0105	0106	0107	0108	0109
Temperature (°C)	20.00	−2.83	13.30	108.08	20.00	38.78	97.99	82.25	133.86
Pressure (kPa)	121.33	311.33	171.33	171.33	301.33	191.33	161.33	151.33	161.33
Mass flow (kg·h ^{−1})	213,928	139,144	74,784	107,055	135,049	1145.78	58,194	38,191	36,142
Mass fraction (%)									
Water	0.06	0.00	0.16	0.12	0.42	0.09	1.03	0.35	0.00
Acetylene	62.41	95.91	0.08	0.00	98.86	0.00	0.00	0.00	0.00
Acetic acid	18.31	0.09	52.22	61.85	0.00	0.00	42.48	0.14	99.54
Vinyl acetate	17.76	3.00	45.22	35.48	0.00	0.00	56.15	99.13	0.00
Acetaldehyde	1.03	1.00	1.11	1.14	0.72	99.91	0.00	0.06	0.00
Crotonaldehyde	0.09	0.00	0.25	0.21	0.00	0.00	0.34	0.32	0.16
The ethyl ester	0.17	0.00	0.48	0.59	0.00	0.00	0.00	0.00	0.00
Anhydride	0.17	0.00	0.48	0.61	0.00	0.00	0.00	0.00	0.31

Figure 2: $T \sim x \sim y$ curve of vinyl acetate in aqueous solution.

Figure 2, when the molar fraction of water is 0.257 and the molar fraction of vinyl acetate is 0.743, the boiling point of the system reaches the lowest of 65.5°C. It can be seen from the gas-phase line in Figure 2 that the boiling point of the system is less than 72.5°C when the molar content of water in the system is between 0 and 0.257. Therefore, in the feed system of the vinyl acetate refining tower, the mixture of vinyl acetate and water has a lower boiling point than the pure vinyl acetate, thereby the products of high purity vinyl acetate cannot be obtained from the top of the tower.

The CP is to first remove acetylene, then separate acetaldehyde, and finally separate vinyl acetate in water

and acetic acid. Vinyl acetate and water, as relatively low boiling substances, form an azeotrope that is distilled from the top of the column and is difficult to separate. In the IP, the acetic acid is separated first, so that vinyl acetate as the relatively high boiling substance enters into the distillation tower bottom, thereby avoiding the formation of the lowest azeotrope between vinyl acetate and water and achieving vinyl acetate with high purity. Meanwhile, acetaldehyde, water, and a small amount of vinyl acetate are present at the top of the tower. Acetaldehyde can be extracted with water because vinyl acetate is insoluble in water while acetaldehyde is soluble in water.

4.2 IP simulation

Based on the above analysis, the separation process is optimized and simulated, and the IP simulation process is shown in Figure 3.

After cooling, the syngas 0101 is separated, the separated gas phase and the gas phase of the coarse separation tower enter the second-stage compressor and then undergo cooling and gas-liquid separation. The gas phase 0201 is successively entered into the pickling tower and water washing tower for pickling and water washing. The acetylene gas 0202 is the reflux synthesis section. The liquid phase 0203 and 0204 of the two-stage gas-liquid separator enter the coarse separation tower, and the liquid phase 0205 at the top of the tower enters the vinyl acetate

Figure 3: IP simulation flow chart: 1 – first-stage gas-liquid separator, 2 – compressor, 3 – two-stage heat exchangers, 4 – two-stage gas-liquid separator, 5 – pickling tower, 6 – water washing tower, 7 – acid cooler, 8 – acetic acid refining tower, 9 – acetaldehyde refining tower, 10 – acetaldehyde extraction tower, 11 – vinyl acetate refining tower, and 12 – crude separation tower.

Table 3: Tower equipment operating parameters

Equipment ID	Plate number	Reflux ratio	D/F	Feed plate position	Top pressure (kPa)
5	30				291.33
6	15				111.33
8	20	1.8	0.98	10	161.33
9	22	2.5	0.125	10	181.33
10	20				131.33
11	25	2.2	0.413	8	161.33
12	20	2.5	0.15	10	121.33

refining column, and the liquid phase 0206 of the tower kettle obtains the vinyl acetate product. The liquid phase at the top of the tower enters the acetaldehyde extraction tower, the raffinate reflows to the vinyl acetate refining tower, and the extraction liquid enters the acetaldehyde refining tower. Finally, the acetaldehyde product is obtained by the liquid phase 0207 at the top of the tower. Part 0209 of the crude separation tower liquid goes into the pickling column as the pickling liquid, and the remaining part 0208 returns to the acetic acid evaporator after rectification.

After the simulation calculation, the tower equipment operating parameters are shown in Table 3, and the mainstream results are shown in Table 4.

According to the flow results shown in Table 4, the purity of the reflux acetylene is 98.37%, which meets the reflux requirements. At the same time, the degasser kettle liquid 0209 contains almost no acetylene, and the purity of vinyl acetate and acetaldehyde reaches 99.8% and 99.4%, respectively, which meets the design requirements.

4.3 IP tower equipment parameter optimization

The reflux ratio of the tower and the theoretical plate number both are very important parameters to the tower. Reflux is a necessary condition for mass transfer in vapor–liquid contact. Mass transfer cannot be carried out without gas–liquid contact. At the same time, the difference in volatilization of the component results in favorable phase equilibrium conditions, which makes the rising steam is in direct contact with the self-condensing reflux liquid. The heavy component is transferred to the liquid phase, whereas the light component is transferred to the gas phase. Therefore, the reflux ratio plays a key role in the separation degree of the mixture. Meanwhile, the reflux ratio is also an important factor affecting the heat load of reboiler and condenser. The theoretical plate is a plate in which the gas and liquid phases leave to achieve phase equilibrium. The theoretical plate number is also an important factor in the degree of mixture separation and the cost of tower equipment. Therefore, the theoretical plate number and reflux ratio are set as independent variables, and the other operating conditions of the column remain unchanged to investigate the influence of the theoretical plate number and reflux ratio on the purity and flow rate of the key distillate in the column. After simulation, the optimization curve of coarse separation tower, vinyl acetate refining tower, acetaldehyde refining tower, and acetic acid refining tower is shown in Figures 4–7, respectively.

Figure 4a is the optimization curve of theoretical plate number, and Figure 4b is the optimization curve of reflux ratio. For Figure 4a, the purity and flow rate of vinyl acetate do not change when the number of theoretical trays is increased to 31. As can be seen from Figure 4b,

Table 4: Main flow results of IP simulation (mass fraction, %)

	0201	0202	0203	0204	0205	0206	0207	0208	0209
Temperature (°C)	−2.87	20.00	−2.87	18.85	53.51	80.81	31.72	126.41	131.12
Pressure (kPa)	311.33	401.33	311.33	201.33	171.33	131.33	181.33	161.33	181.33
Mass flow (kg·h ^{−1})	144,389	140,855	20,421	58,377	39,475	37,930	1,252	40,708	73,624
Mass fraction (%)									
Water	0.00	0.41	0.17	0.16	0.34	0.00	0.04	0.00	0.00
Acetylene	96.00	98.37	0.18	0.04	0.02	0.00	0.50	0.00	0.00
Acetic acid	0.07	0.00	18.40	58.43	0.08	0.08	0.00	89.01	87.91
Vinyl acetate	3.09	0.80	78.14	38.64	96.21	99.81	0.00	8.96	8.72
Acetaldehyde	0.81	0.40	2.23	0.47	3.35	0.00	99.46	0.00	0.00
Crotonaldehyde	0.01	0.02	0.81	1.12	0.10	0.11	0.00	1.78	1.74
Ethyl ester	0.00	0.00	0.01	0.59	0.00	0.00	0.00	0.00	0.83
Anhydride	0.00	0.00	0.07	0.55	0.00	0.00	0.00	0.245	0.80

Figure 4: Optimization curve of coarse separation tower.

Figure 5: Optimization curve of vinyl acetate refining tower.

Figure 6: Optimization curve of acetaldehyde refining tower.

Figure 7: Optimization curve of acetic acid refining tower.

the purity and flow rate of vinyl acetate increase as the reflux ratio continues to increase. The reason is that when the reflux ratio is increased, the liquid–gas ratio in the stripping section and the gas–liquid ratio in the rectifying section are also increased, both of which are beneficial to the mass transfer in the distillation process. However, the increase of reflux ratio is at the cost of energy consumption, so a lower reflux ratio should be chosen. When the reflux ratio is increased to 2.5, the purity and flow rate of vinyl acetate are basically unchanged. Therefore, the optimization results are as follows: the theoretical plate number of coarse separation column is 31, and the reflux ratio is 2.5.

Similarly, it can be seen from Figures 5–7 that the number of theoretical trays of the optimized ethylene acetate refining tower is 14, and the reflux ratio is 2.6. After optimization, the theoretical plate number of acet-aldehydes refining tower is 23, and the reflux ratio is 2.3. After optimization, the theoretical plate number of acetic acid refining tower is 26, and the reflux ratio is 3.1.

5 Comparison of main parameters

5.1 Equipment quantity comparison

By comparing the number of equipment, the specific equipment required for the whole process of CP and IP is shown in Table 5.

For the production of vinyl acetate on the same scale, the CP requires 13 units of equipment in the entire separation process. After the IP, only 11 equipment are needed, which greatly reduces the investment cost of the tower equipment.

Table 5: Number of CP and IP equipment

Project	CP	IP
Total number	13	11
Tower equipment	9	7
Heat exchanger	3	3
Compressor	1	1

5.2 Analysis of energy consumption

The amount of heat required for heating is called heat energy consumption, and the amount of heat removed for cooling is called cold energy consumption. The energy consumption is summarized in Table 6.

As can be seen from the results in Table 6, the heat energy consumption of the CP is 107229.76 kW, and the cold energy consumption is 75768.86 kW. The heat and cold energy consumption of the IP are 71084.5 and 89180.25 kW, respectively. The heat energy consumption is reduced by 6.18%, and cold energy consumption is reduced by 16.83%. Therefore, the overall energy consumption of cold and heat is greatly reduced in the IP, which is conducive to energy saving. In this respect, the IP is superior to the CP.

5.3 Purity of vinyl acetate product

From the stream results listed in Tables 1 and 2, it can be seen that the mass fraction of vinyl acetate is only 99.13% without the side line; this is because the boiling point of vinyl acetate and water is the lowest in the CP. The lowest azeotrope formed between vinyl acetate and water is distilled from the top of the tower. This feature is used in the

Table 6: Energy consumption summary

	CP	IP	Decrement	Percentage reduction (%)
Thermal energy consumption (kW)	107229.76	71084.50	4684.36	6.18
Cooling energy consumption (kW)	75768.86	89180.25	18049.51	16.83

Figure 8: Optimization curve of theoretical plate of vinyl acetate refining tower.

IP to first separate acetic acid and vinyl acetate. At the same time, the vinyl acetate is extracted from the vinyl acetate refining tower to obtain a high-purity vinyl acetate product with a purity of 99.81%, realizing the production of high-purity vinyl acetate product. Meanwhile, reducing the side line extraction can simplify the structure of the tower equipment and save costs.

5.4 Optimization comparison of main tower equipment

5.4.1 Theoretical plate number of vinyl acetate refining tower

The number of theoretical plates in the vinyl acetate refining tower of the two processes is optimized, and the optimization results are shown in Figure 8.

Figure 8 shows the optimization curve of the theoretical plate number of the vinyl acetate refiner in the CP and the IP. It can be seen from the figure that the theoretical plate number of the vinyl acetate refining tower in the CP after optimization is 17. The theoretical plate number of the vinyl acetate refining tower in the IP after optimization is 14. Therefore, three theoretical plates can be saved after the improvement.

The heat load of the condenser in the vinyl acetate refining tower in the CP is 11943.7 kW, and the heat load of the reboiler is 11980.6 kW. After the IP, the condenser load of the vinyl acetate refining tower is 2522.99 kW, and the reboiler load is 3095.89 kW. The condenser load can be reduced by 78.9% and the heat load by 74.2%.

Figure 9: Optimal curve of extractant dosage in acetaldehyde extraction tower.

5.4.2 The amount of extraction agent in the extraction column

The extractant dosage of the extraction towers in the two process is optimized, and the optimization curve is shown in Figure 9.

Figure 9a is the extractant optimization curve of the acetaldehyde extraction tower in CP, and Figure 9b is the extractant optimization curve of the acetaldehyde extraction tower in IP. According to the results in the figure, the optimal amount of extractant in the CP is $229 \text{ kmol}\cdot\text{h}^{-1}$, and the optimal amount of extractant in the IP is $180 \text{ kmol}\cdot\text{h}^{-1}$, which is a reduction of 22.7%.

6 Conclusion

In this article, the process simulation software is used to simulate the whole separation process based on the CP of phase synthesis of vinyl acetate from acetylene gas.

(1) According to the properties of the materials involved in the vinyl acetate production, the NRTL-HOC thermodynamic model is used to calculate, and the material balance, heat balance and parameters of each stream are obtained.

(2) Through the analysis of the calculation results in the CP, it is concluded that the main reason for the low purity of the product is the formation of an azeotrope between vinyl acetate and water. It is determined that the improvement of sequence separation in the order of acetylene, acetic acid, vinyl acetate, and acetaldehyde. After simulation, the material balance, heat balance, and the key parameters of the IP are obtained.

(3) When compared with the CP, the IP reduces three equipment for a lower device cost. And for the heat load, the cooling load is reduced by 16.8%, and the heat load is reduced by 6.18%. The purity of vinyl acetate reaches 99.81%. The number of trays in the vinyl acetate refining tower is reduced by 1, the condenser load is reduced by 78.9%, and the reboiler load is reduced by 74.2%.

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