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Multi-Effect Evaporation Coupled with MVR Heat Pump Thermal Integration Distillation for Separating Salt Containing Methanol Wastewater

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Abstract

Due to the high energy consumption for separation of salt containing methanol wastewater, in this work, the multi-effect evaporation coupled with mechanical vapor recompression (MVR) heat pump and thermal integration technologies were raised for the first time. The ELECNRTL thermodynamic model is used to simulate and optimize the evaporation rectification process. Energy consumption and total annual cost (TAC) are taken as objective functions. The results show that multi-effect evaporation coupled with conventional distillation process can save energy consumption and TAC by 44.12% and 39.14%. The multi-effect evaporation coupled with distillation process based on MVR heat pump technology can save energy consumption and TAC by 55.27% and 47.49%, which is super to three-effect evaporation coupled with conventional distillation process. The three-effect evaporation coupled with MVR heat integration process can save energy consumption and TAC by 81.32% and 58.55%, which is more economical than other processes. It can be clearly seen that three-effect evaporation coupled with MVR heat integration process is more competitive to deal with the salt containing methanol wastewater.

Keywords

Salt Containing Methanol Wastewater, MVR Heat Pump, Heat Integration, Energy Saving

1. Introduction

Salt containing organic wastewater is mainly from the chemical industry, such as medicine, pesticide and other industries [1]. The amount of wastewater is very huge and is increasing year by year. As far as we know, the high concentration of soluble inorganic salts and hardly degradable organic compounds in wastewater can cause serious environmental pollution, so the treatment is extremely urgent now. At present, the treatment of wastewater is divided into physicochemical methods and biological methods, specifically focused on the following aspects [2]: Halophilic biology method takes advantage of degradability and salt tolerance of halophilic, but the salt concentration is too sensitive and acclimation time is too long [3]. Membrane technology method utilizes concentration or pressure difference to allow the molecular to pass through selectively, but the metal plate is easy to wear and the operating cost is expensive [4] [5]. Electrochemical method makes use of external electric field to form a current, but the efficiency of removing organic compounds is low [6]. Incineration method can completely remove the organic compounds, but can bring some toxic substances simultaneously [7] [8]. These methods can deal with salt containing organic wastewater to a certain extent, but can't fully recover the organic compounds. So it turns out the waste of resources and irreversibility.

For salt containing methanol wastewater system, methanol and salt can be separated by evaporation process. Meanwhile, methanol and water are separated by distillation process. And thus, the high-purity methanol can be well recovered at the top of column. To the best of our knowledge, methanol is one of the most important cornerstones of chemical industry. It is developed as the raw material for synthesis of chemical derivatives such as formaldehyde, MTBE and acetic acid. It is also applied to adhesives, primers, solvents, detergent and other products [9]. Consequently, the novel process can not only reduce environment pollution, but also fully utilize resources.

Evaporation process requires a large amount of energy to evaporate water, so evaporation can be seen as an energy-intensive operation [10]. Do [11] discovered that evaporation consume 75% of energy, which meant there was a promising prospect for energy conservation. On this basis, the multi-effect evaporation process had been discussed in literatures [12] [13] [14]. Multi-effect evaporator (MEE) needs steam only in first effect; the other effects are heated by secondary steam of previous effect. Therefore, the consumption of steam is greatly saved, the production cost is decreased, and the economic benefit is improved at the same time [15]. For the conventional distillation system, overhead vapor from column is cooled by cooling water. The latent heat of steam is taken away directly, resulting in the waste of energy. The liquid boiling at bottom of the column is provided by utilities, so thermodynamic efficiency of the whole system is lower. If the latent heat of overhead vapor can be used to heat the liquid at bottom of column, it will be more economical and energy-efficient. Therefore, the mechanical vapor recompression (MVR) heat pump distillation process has been noticed more and more widespread attention in recent years.

However, the research about salt (sodium chloride and sodium sulfate) containing methanol wastewater on evaporation coupled with distillation process is not vary comprehensive and the literatures about it are rarely discussed so far. In this work, the innovative MVR heat pump and heat integration technology are further explored.

2. Simulation Rules and Evaluation Indexes

2.1. Simulation Rules

The treatment capacity of salt containing methanol wastewater was 3000 kg/h. In addition, the content of methanol, water, sodium chloride and sodium sulfate was 10 wt%, 77 wt%, 7 wt% and 6 wt%, respectively. With regard to treatment of the wastewater, the product purity of methanol was specified to 0.995; the COD of salt containing wastewater was less than 500 ppm.

Evaporation processes were operated at pressure-swing, and distillation process was operated at atmospheric pressure. The Heater and Falsh 2 modular were selected to simulate and calculate the evaporators by Aspen Plus software. Radfrac modular was chosen to simulate distillation column and Compr modular was applied to simulate compressors. The distillation column used the type of MELLAPAK as packing column. Since the system related to electrolytes, the thermodynamics data and vapor-liquid equilibrium data were calculated by using the ELECNRTL electrolyte thermodynamics model. Cooling medium with inlet and outlet was the cooling water of 33˚C and 39˚C, respectively. Heating medium was the saturated steam of 130˚C [16].

2.2. Selection of Thermodynamic Model

The correctness of the thermodynamics model is the key for simulation of the process. Therefore, comprehend of phase equilibrium relationship about the salt containing methanol wastewater system is the essential basic research work [17]. What`s more, the solubility of salt (sodium chloride and sodium sulfate) containing methanol wastewater and the change regulations of physicochemical properties of the solution is also necessary.

For the liquid mixture containing salt, usually the vapor-liquid equilibrium point of the system will migrate and the relative volatility will also change. The macroscopic point of view is that the boiling point of the solution will rise. In Aspen Plus software, ELECNRTL thermodynamic model is usually selected to simulate mixed system containing salt, and results turn out that it can generally well simulate the boiling point and data of vapor-liquid equilibrium [18] [19]. In order to investigate the reliability of the thermodynamics model of the electrolyte, the boiling point of methanol-water system containing sodium chloride and sodium sulfate was determined by the experimental method (different ratio of two sodium salts which under atmospheric pressure). Compared with the calculated values of ELECNRTL thermodynamic model, the experimental results were shown in **Figure 1**. It can be seen that the calculated value of ELECNRTL thermodynamics model agree well with the experimental data. That is to say, the ELECNRTL thermodynamics model is reliable to the system.

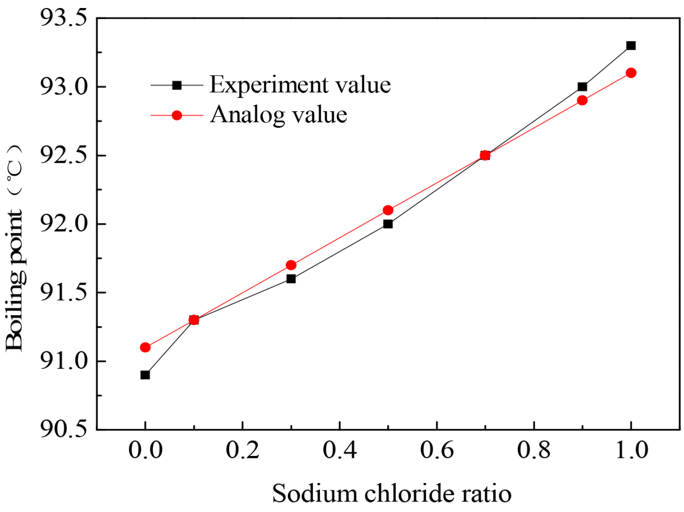

Figure 1. Experimental and calculated values of boiling point.

2.3. Selection of Thermodynamic Model

The total annual cost (TAC) is used as an index to evaluate the economics of all processes. TAC mainly includes capital investment (CI) and operating cost (OC). Capital investment includes distillation column, heat exchanger and compressor costs. Operating cost includes steam costs, cooling water costs, frozen brine costs and electricity costs [20], the formula [21] are as follow:

$$C = 8000 \times \left(\alpha_1 \times q_{c1} + \alpha_2 \times q_{c2} + \beta \times q_r + \gamma \times P\right) \tag{1}$$

$$CI = C_{cooler} + C_{reboiler} + C_{columm} + C_{com} \tag{2}$$

$$TAC = OC + CI/\theta \tag{3}$$

$$C_{cooler} = C_{reboiler} = 7296 \times A^{0.65} \tag{4}$$

$$C_{column} = 17640 \times D^{1.066} \times L^{0.802} \tag{5}$$

The price of the compressor is taken from the website [22]. A is for the heat area, m^2; D and L are the diameter and height of the distillation column respectively, m; W is for the compressor power, kW. Public works costs: α_1 is for the cooling water price, 0.054 \$·t^{-1}, q_{c1} is for the amount of cooling water, t·h^{-1}; α_2 is for the frozen salt water price, 0.077 \$·t^{-1}, q_{c2} is for the amount of frozen water, t·h^{-1}; β is for the saturated steam price, 30.62 \$·t^{-1}, q_r is for the amount of steam, t·h^{-1}; γ is for the electricity price, 0.12 \$·(kW·h)$^{-1}$, P is for the power consumption, kW·h; θ is for the equipment depreciation period, taking 8 years.

3. Simulation of Evaporation Coupled with Conventional Distillation Process

3.1. Single-Effect Evaporation Coupled with Conventional Distillation Process

As shown in Figure 2, the salt containing methanol wastewater (FD) entered the heater and then fed into evaporator, and the mixed steam of methanol and water

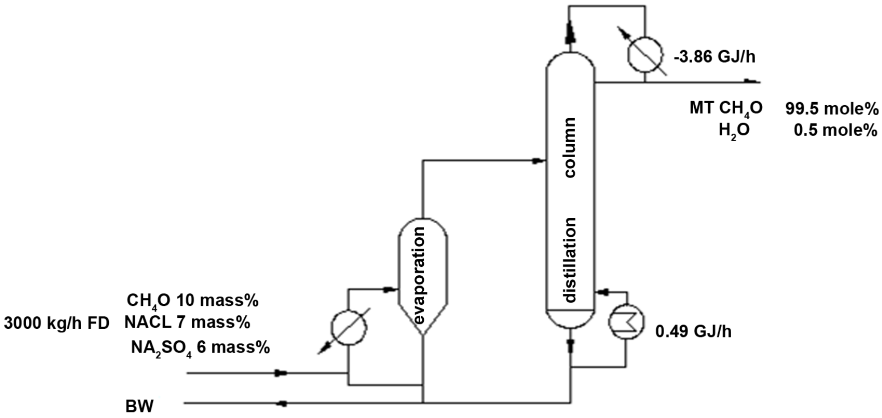

Figure 2. Single-effect evaporation coupled with conventional distillation process.

was directly fed into the distillation column. The top of the column was a qualified methanol product (MT). The bottom of the column was wastewater, and then mixed with the wastewater at the bottom of the evaporator, which turned out as brine waste (BW).

Evaporation and distillation were operated at atmospheric pressure. The feed position of the steam phase was optimized by using the sensitivity analysis modular of Aspen Plus software, and the simulation results were presented in Table 1. The energy consumption data was converted to standard coal data.

3.2. Multi-Effect Evaporation Coupled with Conventional Distillation Process

The energy consumption of single-effect evaporation was higher and reached to 1083.11 t·a^{-1}. To reduce the energy consumption of the evaporation process, the original single-effect process was change into multi-effect evaporation coupled with conventional distillation process. By setting pressure of each effect respectively, the temperature of pre-secondary steam was about 10˚C higher than the boiling point of after-effect concentrate. Due to adoption of multi-effect evaporation, the energy consumption of the evaporation process could be greatly reduced [23] [24].

As the evaporation and distillation were coupled together, the vapor phase entered the distillation column directly, which operated at atmospheric pressure. Therefore, the multi-effect evaporation system had two operating conditions: 1) The final effect evaporation was at atmospheric operation, and the front effects were at pressurized operation; 2) The final effect evaporation was at negative pressure operation, and the front effects were at atmospheric pressure or pressurized operation. For the negative effects operation, the negative pressure steam should through steam compressor to improve pressure first and then injected into the distillation column. The process of three-effect (final effect is at negative pressure) evaporation coupled with the conventional distillation was figured as shown in Figure 3. As the content of methanol between the secondary steam produced by the final effect and the condensate produced by the previous effect

Table 1. Simulation results of single-effect evaporation coupled with conventional distillation.

Parameters	Distillation system	Evaporation system
Pressure/MPa	0.1	0.11
Theoretical stage		-
Feed stage	13	-
Reflux ratio	11.75	-
Column diameter/m	0.85	-
Condenser duty/kW	1072.12	1087.79
Reboiler duty/kW	136.88	
Total heat exchange area/m^2	112.81	135.84
Annual steam cost (10^5\$/y)	4.99	39.24
Annual cooling water cost (10^5\$/y)	5.94	
Total energy cost/t·a^{-1}	1083.11	
TAC (10^5\$/y)	50.76	

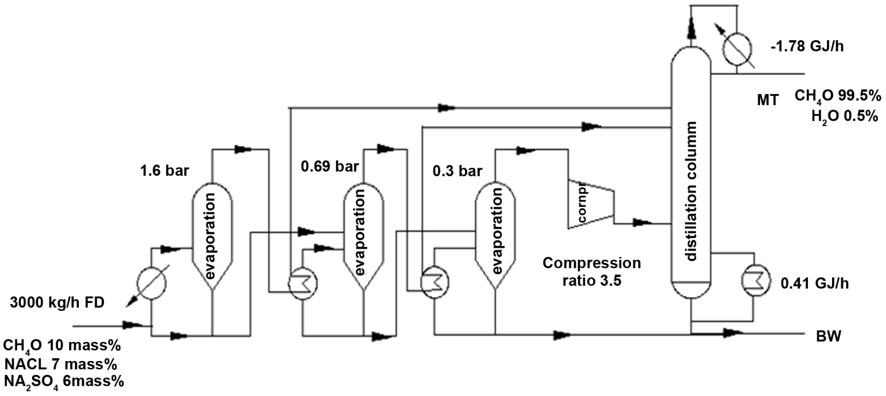

Figure 3. Three-effect evaporation coupled with conventional distillation process.

were different, the feed position of each effect was needed to be optimized. The simulation results of the multi-effect evaporation coupled with conventional distillation process were presented in Table 2 and Table 3.

As seen from Table 2 and Table 3, the multi-effect evaporation coupled with distillation process had economic advantages than the single-effect evaporation coupled with distillation process. Compared with the single-effect evaporation coupled with distillation process, the double-effect process and the three-effect process could reduce energy by 37.6% and 50.53%, and TAC is reduced by 34.22% and 44.05% respectively. Based on the three-effect evaporation coupled with distillation process, the optimization of process was carried out, and the possibility of energy saving was further studied.

4. Simulation of Three-Effect Evaporation Coupled with MVR Heat Distillation Process

It can be seen that either the double-effect or the three-effect evaporation coupled with conventional distillation process, heat load at the top of distillation

Table 2. Summary of simulation results of double-effect evaporation coupled with conventional distillation process.

Parameters	Distillation system	Double-effect system	
		I	II
Pressure/MPa	0.1	0.14	0.063
Theoretical stage	64	-	-
Top temperature/˚C	64.39	100.2	83.4
Bottom temperature/˚C	99.65	108.7	90.1
Condenser duty/kW	628.58	612.95	442.61
Reboiler duty/kW	121.53		
Total heat exchange area/m^2	67.50	80.16	92.78
Annual steam cost (10^5\$/y)	4.43	22.11	
Annual cooling water cost (10^5\$/y)	3.48	-	
Compressor power consumption/kW	-	28.51	
Total energy cost /t·a^{-1}	674.84		
TAC (10^5\$/y)	33.39		

Table 3. Summary of simulation results of three-effect evaporation coupled with conventional distillation process.

Parameters	Distillation system	Three-effect system		
		I	II	III
Pressure/MPa	0.1	0.16	0.069	0.03
Theoretical stage	66	-	-	-
Top temperature/˚C	64.39	101.9	82.7	66.4
Bottom temperature/˚C	99.66	111.0	90.1	72.1
Condenser duty/kW	493.90	441.23	261.90	317.72
Reboiler duty/kW	112.77			
Total heat exchange area/m^2	53.61	55.05	47.69	65.75
Annual steam cost (10^5\$/y)	4.11	92.76		
Annual cooling water cost (10^5\$/y)	2.74	-		
Compressor power consumption/kW	-	51.88		
Total energy cost/t·a^{-1}	535.92			
TAC (10^5\$/y)	28.40			

was higher than the bottom of the column, as shown in Table 2 and Table 3. This is because the feeds are vapor phase, resulting the condensation heat load at top of the column become larger. Simultaneously, the latent heat of top steam is taken away directly by cooling water, which led to a large amount waste and irreversibility of energy. If the latent heat of the top steam (methanol steam) can be fully utilized, the energy consumption of the whole process can be greatly reduced. Consequently, to supply the heat for evaporators or reboiler of distillation column, the top steam of distillation column was compressed by compressor to increase its temperature and pressure. For this purpose, the following two MVR heat integration energy-saving process were put forward.

4.1. Three-Effect Evaporation Coupled with MVR Heat Pump Distillation Process

The methanol steam at the top of distillation column was compressed by compressor and then used for heating of the bottom of column (MVR heat pump distillation process) [25] [26]. Three-effect evaporation coupled with MVR heat pump distillation process was showed in **Figure 4**. The three-effect evaporation process was unchanged, that is to say, the condensate of first effect and second effect and the steam of third effect entered the different parts of the distillation column respectively. For the normal operation of the methanol-water separation system, the bottom temperature of column is about 100˚C. If the heat transfer temperature difference of the reboiler was specified as 10˚C, the methanol vapor need to be compressed to 0.5 MPa, and then the saturation temperature of methanol vapor is 111.3˚C. For three-effect evaporation coupled with MVR heat pump distillation process, the process parameters of the three-effect evaporation system were invariable, so only the simulation and optimization of MVR heat pump distillation system were needed, and the simulation results were shown in **Table 4**.

For the three-effect evaporation coupled with MVR heat pump distillation process, the total energy consumption was the sum of energy consumption of evaporation system and power consumption of compressors in distillation system, and the total value was 55.27 $t \cdot a^{-1}$. For distillation system, the energy consumption was reduced by 5.46% when the MVR heat pump process was adopted. For the whole process of three-effect evaporation coupled with MVR heat pump distillation, the energy consumption was reduced by 9.59%, and TAC reduced by 6.14%. Obviously, whether the energy or TAC, the decline was not large. It was because the feeding of distillation column was steam phase, then the

Table 4. Simulation results of MVR heat pump distillation system.

Parameters	value
Pressure/MPa	0.1
Top temperature/˚C	64.39
Saturated temperature of compressed steam /˚C	111.3
Bottom temperature/˚C	99.66
Compression ratio	4.5
Compressor power consumption/kW	106.44
Cooler heat duty/kW	487.58
Annual cooling water cost (10^5\$/y)	2.70
Total energy cost/$t \cdot a^{-1}$	55.27
TAC (10^5\$/y)	26.66

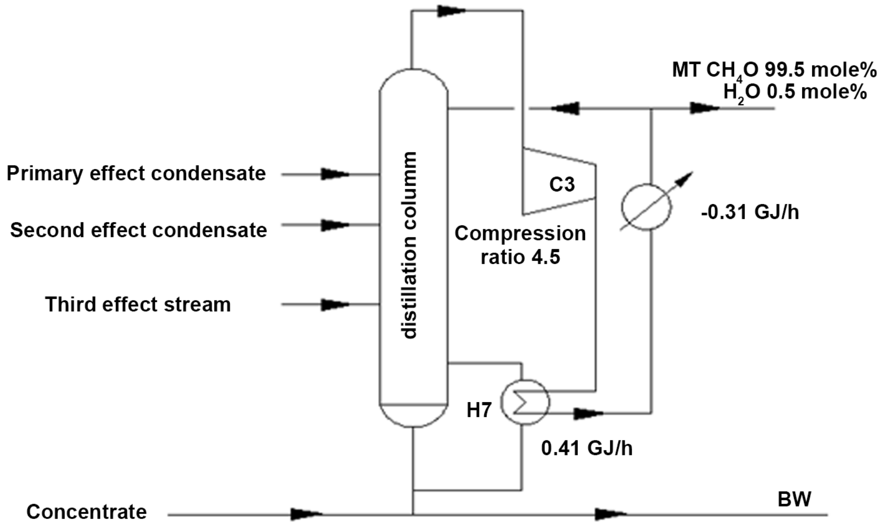

Figure 4. Three-effect evaporative coupled with MVR heat pump distillation process.

heat load for bottom of column was not large. So that latent heat of the top steam was excess and most of the latent heat was still taken away by cooling water. Besides, although the operating costs were reduced, but due to the larger investment in the compressor, the decline in TAC was not so obvious.

4.2. Three-Effect Evaporation Coupled with MVR Heat Integration Process

In three-effect evaporation coupled with MVR heat pump distillation process, energy consumption mainly lies in the first-effect evaporator and compressor of the distillation system. So that most part of the latent heat released by the compressed steam could be used to heat the first-effect evaporator, and the excess part could be utilized for bottom reboiler. In order to meet the energy balance of distillation system, an auxiliary reboiler was needed, SQ was supplementary energy from outside, as shown in Figure 5. In addition, to make full use of sensible heat of the compressed steam condensate, a feed preheater H1 was added to preheat the feed. Since the temperature of first-effect evaporation and bottom of the distillation column was different, in order to meet the heat transfer temperature difference of H3 and H7 at 10˚C , the top vapor of column were compressed respectively, namely, two compressors (C1 and C3) with different compression ratios. The condensate from the heat exchangers H3 and H7 were mixed and went through preheater H1 and the cooler H2 respectively. In the end, a part of them returned to column, while others were extracted as methanol product.

As shown in Figure 5, energy consumption was mainly the sum of power consumption of three compressors C1, C2 and C3. Compressor power consumption depended mainly on the amount of compression and compression ratio, because top steam of distillation column was a certain amount, finally the compressor power consumption was determined by compression ratio only. Since

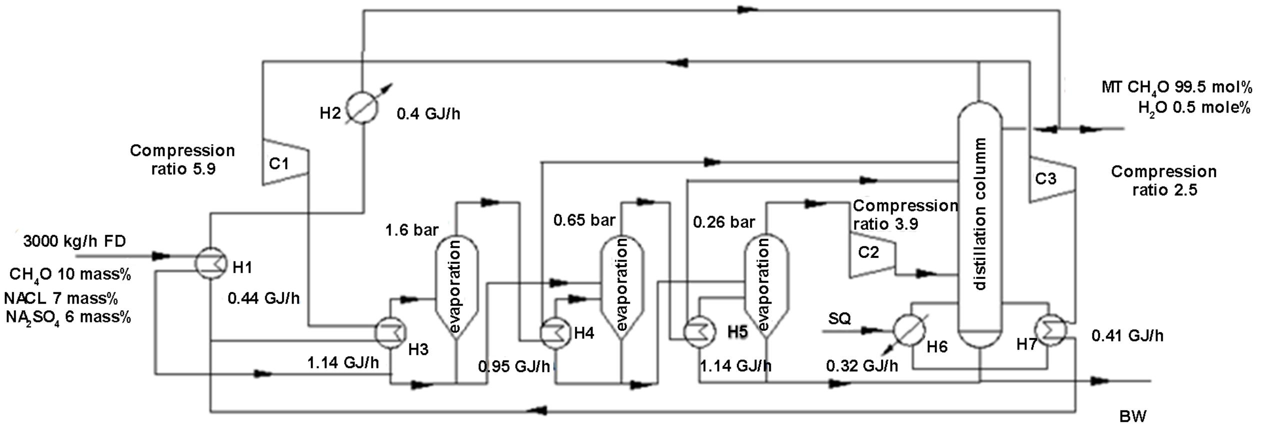

Figure 5. Three-effect evaporation coupled with MVR heat integration distillation process.

distillation column was operated at atmospheric pressure, the compression ratio of the compressor C3 was constant. Because the heat transfer temperature difference (10˚C) of heat exchanger H1 was specified, the compression ratio of the compressor C1 depended on operating pressure of first-effect. At the same time, the first-effect operating pressure in turn affected the third-effect operating pressure, which resulted in the changes to compression ratio of compressor C2. Therefore, the first-effect operating pressure would influence operation parameters and energy consumption of the whole process. To investigate the energy consumption and influences on process parameters of whole system, Aspen Plus software was used to specify the different operating pressure of the first-effect. And the optimized simulation results were shown in Table 5.

Table 5 showed that when the operating pressure of first-effect was 0.15 MPa, energy consumption of three-effect evaporation coupled with MVR heat integration process was 58.25% less than three-effect evaporation coupled with MVR heat pump distillation process and the TAC was decreased by 21.11%. As it can be demonstrated, three-effect evaporation coupled with MVR heat integration process had obvious economic advantages. Because three-effect evaporation coupled with MVR heat integration process greatly reduced the amount of cooling water, the process is especially suitable for areas with severe water shortage.

5. Comparison and Analysis of the Results

To comparison and analysis easier, Table 6 summarized the main data about the above four kinds of evaporation coupled with distillation processes. It could be observed that compared with single-effect evaporation coupled with conventional distillation process, multi-effect evaporation coupled with conventional distillation process could reduce the energy consumption and TAC by 44.12% and 39.14% respectively. Three-effect process was more energy-efficient than the single-effect process. Multi-effect evaporation coupled with distillation process based on MVR heat pump technology was more economical than the multi-effect evaporation coupled with conventional distillation process, among them

Table 5. Summary of simulation results of three-effect evaporation coupled with MVR heat integration process.

Parameters	Distillation system	Three-effect system		
		I	II	III
Pressure/MPa	0.1	0.15	0.065	0.026
Theoretical stage	71	-	-	-
Feed stage	9 10 17	-	-	-
Top temperature/˚C	64.5	100.0	81.1	63.2
Bottom temperature/˚C	99.67	109.1	88.6	68.8
Top duty/kW	497.76	315.34	262.71	317.53
Bottom duty/kW	112.55			
Total heat exchange area/m^2	200.49			
SQ/kW	35.55			
Compression ratio	C1:5.9, C2:3.9, C3:2.5			
Annual steam cost(10^5\$/y)	1.30			
Annual cooling water cost(10^5\$/y)	0.44			
Total compressor power consumption/kW	192.86			
Total energy cost /$t \cdot a^{-1}$	202.28			
TAC (10^5\$/y)	21.03			

Table 6. Summary of main data of evaporation coupled with distillation process.

Parameters	Single-effect evaporation coupled with distillation process	Double-effect evaporation coupled with distillation process	Three-effect evaporation coupled with distillation process	Three-effect evaporation coupled with MVR heat pump distillation process	Three-effect evaporation coupled with MVR heat integration process
Annual operating cost (10^5\$/y)	50.17	32.24	27.37	24.13	18.74
Equipment depreciation cost (10^5\$/y)	0.59	0.85	1.03	2.53	2.29
TAC (10^5\$/y)	50.76	33.39	28.40	26.66	21.03
energy cost/$t \cdot a^{-1}$	1083.11	674.84	535.92	484.52	202.28
Save energy/%	-	37.70	50.53	55.27	81.32
Save TAC/%	-	34.22	44.05	47.49	58.55

three-effect evaporation coupled with MVR heat integration process was the optimal. The reason was that the process took full advantages of the total latent heat and most sensible heat of the top compressed steam of distillation column. Meanwhile, not only the amount of cooling water was reduced, but also the amount of external steam was greatly reduced. The energy data in Table 6 was converted to standard coal data.

6. Conclusions

Taking the separation of salt containing methanol wastewater as the research object, the simulation and optimization of evaporation processes coupled with distillation processes were carried out by using Aspen Plus software, and conclusions were drawn as follows:

1) For the methanol-water system containing sodium chloride and sodium sulfide, through the validation of the experimental data, it was appropriate to use the ELECNRTL thermodynamics model in Aspen Plus software.

2) Compared with single-effect evaporation coupled with conventional distillation process, double-effect and three-effect evaporation coupled with conventional distillation process could save energy by 37.70% and 50.23%, and reduce TAC by 34.22 % and 44.05 %, respectively.

3) The three-effect evaporation coupled with distillation process based on MVR heat pump technology could save energy consumption and TAC by 55.27% and 47.49%, which is super to three-effect evaporation coupled with conventional distillation process.

4) The three-effect evaporation coupled with MVR heat integration process could reduce energy consumption and TAC by 81.32% and 58.55% respectively, which had economic advantages than three-effect evaporation coupled with MVR heat pump distillation process. Therefore, it was the optimal process for treating similar salt containing organic wastewater.

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