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Study of Organic Acid Pollutant Removal Efficient in Treatment of Industrial Wastewater with HDH Process Using ASPEN Modelling

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Abstract: Due to low efficiency and the material choice limitations of traditional evaporation systems to treat acid wastewater, humidification and dehumidification (HDH) as the core process was applied in the treatment and reduction of wastewater with organic acid pollutant concentrations. The forecasting of pH changes and COD reduction is important for the system's design. Therefore, a study of the pollutant removal efficiency with different parameters, such as the reaction temperature, air quantity, and flow rate was conducted with ASPEN modeling. In this article, ASPEN modeling was used to simulate the water and acid material transformation in HDH system. The process was composed of blocks, such as RadFrac, heater and split. The analysis was taken with different air quantities, tower diameters, heat loads and flow rates. The analysis indicated that the pH of the maleic acid wastewater changed from 3.0 to 5.7. The relationship between inlet quantity, air quantity, inlet heat and the clean water yield was also shown in the modeling results. Based on these studies, we determined that the model can help engineers solve the key problems of HDH systems, such as heat balance calculation, equipment selection, and the prediction of incoming and outgoing evaporation materials.

Keywords: humidification and dehumidification; Aspen modeling; acid pollutant remove; evaporation; industry wastewater treatment; pH changing

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1. Introduction

Organic acid pollutant removal is an important topic in industrial wastewater treatment. Neutralization of the acids and the base is the traditional method used to obtain a suitable pH for wastewater treatment plants [1], although this method causes high quantities of additional acid and base [2,3] to be consumed. In this study, it was determined that malic acid should be removed from wastewater before entering the biotreatment system, using an evaporation method.

Due to the low efficiency and material choice limitations of traditional evaporation, HDH technology was chosen as the main process [4]. Before pilot engineering, the pH removal efficiency was observed by simulation modeling.

HDH (humidification and dehumidification) technology is a distillation technology which uses a carrier fluid such as air or water to obtain thermal energy from a heat source [5]. The heat source is then transferred to a humidifier for water evaporation from saline water and then to the dehumidifier for the condensation of the evaporated water to the fresh water [6]. The saturated vapor pressure of water vapor tends to increase with increasing temperatures. Water vapor in the air is very low at room temperature, but near the boiling point of the water, water vapor in the air can be nearly 0%. After that, water or light component organic matter is used in the tower body with different air and water distribution ratios at different temperatures, and is extracted from wastewater or organic matter, so as to realize salt-water separation or material purification and recovery.

The HDH process of evaporation can therefore be employed to remove organic and acid pollutants from wastewater. Due to its efficiency, evaporation can be combined with other novel and traditional processes to realize the recycling and zero discharge of high-salt organic wastewater. In their works, Lawal et al. [7] integrated multi-stage flash (MSF) desalination with humidification and dehumidification (HDH) desalination for brine recovery. Their work concluded that HDH could utilize 67% of the rejected brine by MSF [8,9].

Most recently, humidification and dehumidification (HDH) as a core process has been applied for the treatment and reduction of wastewater with a high salt content and high organic matter concentrations. It has the characteristics of a good separation effect, a high wastewater recovery rate, low investment savings, and low operation costs. It has an advantage over the traditional mainstream evaporation process as it is limited by metal materials, and the large consumption of traditional oil and coal, amongst other factors. Therefore, it has become urgent to develop a new evaporation process with essential breakthroughs in evaporation temperature, energy use and other aspects. Under this market demand, humidification and dehumidification evaporation processes have widely developed. The research scope is expanding globally, and has resulted in several engineering applications with good practical data.

The purpose of this study was to create a model to simulate the flow rate and the energy balance for the HDH process, which is more convenient than setting up experiment equipment. From this model, the researchers can easily obtain all kinds of results and determine energy consumption.

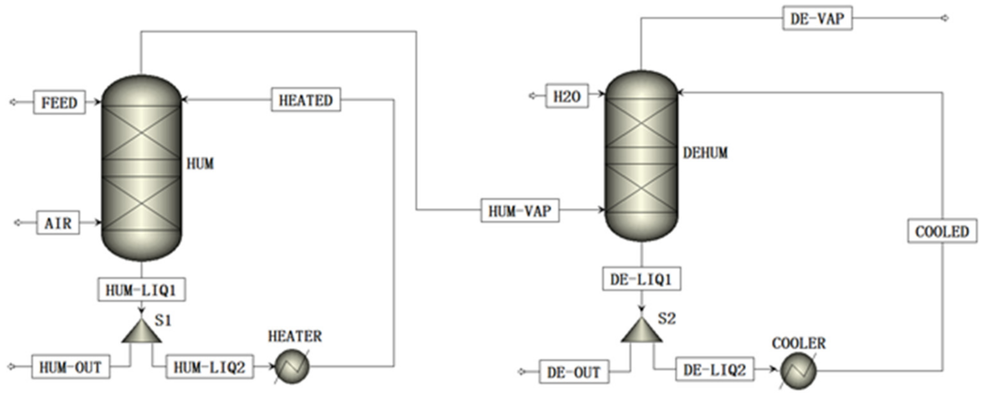
2. Materials and Methods

2.1. ASPEN Simulation

ASPEN Plus (Advanced System for Process Engineering) is the leading chemical process simulator in the world, and allows the user to build a process model and then simulate it using complex calculations (models, equations, math calculations, regressions, etc.).

A.S.Abdullah [10] used ASPEN to simulate the HDH process with different kinds of tower settings to evaluate the effects of solar intensity, temperatures, and relative humidity. Ratnakumar V [11] used ASPEN modeling to simulate the fluidized bed reactor model for chemical looping of synthesis gas. The physical property equations used in these simulation processes are based on NRTL, and the simulation results are consistent with the practical data of gas–liquid reaction processes.

In this study, the HDH system used for humidification and dehumidification (HDH) desalination of maleic anhydride process wastewater consisted of two units, the humidification process and the dehumidification (condensation) process. Figure 1 outlines the overall schematic and model settings [12–14].



(**a**) Humidification schematic in Aspen (**b**) Dehumidification schematic in Aspen

Figure 1. Overall HDH schematic in Aspen Plus.

(a) Humidification process

The RadFrac model was used to replicate the humidifier (HUM) and dehumidifier (DEHUM). The humidification process contained two feed streams, wastewater (FEED) and air (AIR), as showed in Figure 1a. The wastewater flowed in the humidifier from the top and the air flowed in from the bottom. After the two feed streams had a counter-current exchange, the separated gas stream (HUM-VAP) left for the dehumidifier, and the concentrated brine (HUM-LIQ) flowed out and split into two streams: 20% of the brine (HUM-OUT) split out of the system and the remaining brine was fed to a heater (HEATER). The heated stream (HEATED) then returned into the humidifier to create a loop.

(b) Dehumidification process

The dehumidification process contained two feed streams as well, freshwater (H2O) and humid air (HUM-VAP), as showed in Figure 1b. The freshwater flowed in the dehumidifier from the top and the humid air flowed in from the bottom. After the two streams had a counter-current exchange, the separated gas stream (DE-VAP) left the system. The condensed water (DE-LIQ) flowed out and split into two streams. 20% of the condensed water (DE-OUT) was split out of the system and the remaining water was fed to a cooler (COOLER). The cooled stream (COOLED) returned into the dehumidifier to create a loop.

2.2. Thermodynamic Model

When building a chemical process model with stimulation software, the key decision affecting the accuracy of results is a selection of thermodynamic model. In Aspen Plus, the quality of simulation results is determined by the model equations and by their usage in a different system. Using the wrong model or incomplete physical property parameters can lead to great inaccuracies between the simulation results and actual industry data.

In this study, the selection of the thermodynamic model was NRTL (non-random two liquids) activity coefficient equation, and the electrolyte method (ELECNRTL) according to the methods assistant in Aspen Plus. This model is the most frequently used in ideal gas systems, polar liquids systems, and aqueous electrolyte systems. This application is also appropriate for polar substances such as water, alcohols, ketones, ethers, and organic acids [15].

For a binary solution, the NRTL model for excess Gibbs energy is (Equation (1)) [16]

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

Using the excess Gibbs energy in (Equation (2))

$$ln\gamma_1 = \left[\frac{\partial(nG^E/RT)}{\partial n_i}\right]_{T,p,n_{j[i]}} \quad (2)$$

the activity coefficient can be determined as (Equations (3) and (4))

$$ln\gamma_1 = x_2^2[\frac{\tau_{21}G_{21}^2}{(x_1 + x_2G_{21})^2} + \frac{\tau_{12}G_{12}}{(x_2 + x_1G_{12})^2}] \tag{3}$$

$$ln\gamma_2 = x_1^2[\frac{\tau_{12}G_{12}^2}{(x_2 + x_1G_{12})^2} + \frac{\tau_{21}G_{21}}{(x_1 + x_2G_{21})^2}] \tag{4}$$

where (Equations (5)–(9)):

$$\tau_{12} = (g_{12} - g_{22})/(RT) \tag{5}$$

$$\tau_{21} = (g_{21} - g_{11})/(RT) \tag{6}$$

$$g_{12} = g_{21} \tag{7}$$

$$G_{12} = \exp(-a_{12}\tau_{12}) \tag{8}$$

$$G_{21} = \exp(-a_{12}\tau_{21}) \tag{9}$$

Here, the alpha parameter a_{12} ($a_{12} = a_{21}$) is the binary adjustable parameters estimated from experimental vapor-liquid equilibrium data (varies from 0.2~0.47). The adjustable energy parameters are independent of composition and temperature, but dependent on solution properties [17].

2.3. Input Acid Wastewater

The input flow of organic and acid pollutant was mainly from a maleic anhydride production line, mainly by product and reactor-cleaning water. The modeling was for the design of the process before the real engineering application.

The simulation model of the acid removal HDH system was designed to produce 600 t/day product water under a 20 h operational cycle. Because of this, the feed wastewater was set as 30 t/h (30,000 kg/h). The input streams parameters can be found in Table 1.

Table 1. Inlet streams parameters and conditions.

Stream	Temperature	Pressure	Component	Mass Fraction (%)	Mass Flow (kg/h)
FEED (Wastewater)	25 °C	1 bar	H_2O (H_2O)	98.3	30,000
			Maleic Anhydride (MA)	0.07	
			Acetic Acid (ACETIC)	0.15	
			Acrylic Acid (ACRYLIC)	0.15	
			Dibutyl Phthalate (DBP)	0.07	
			Phthalic Anhydride (PHTHALIC)	0.02	
			Phthalic Acid (PHTHA-AC)	0.25	
			Maleic Acid (MALEI)	0.8	
			Fumaric Acid (FUMAR)	0.08	
			N-butanol (BUTANOL)	0.11	
AIR	25 °C	1 bar	Nitrogen (N_2)	79 (Noble gases included)	15,000
			Oxygen (O_2)	21	
H_2O (Freshwater)	25 °C	1 bar	H_2O (H_2O)	100	30,000

2.4. Units in HDH System

This section describes the units and parameters contained in the HDH system, which are stage number, pressure, packed height and column diameter. Tables 2–4 show the blocks, packing materials and heat exchange parameters in the HDH simulation model [18,19].

Table 2. Block Characteristics.

Humidifier (HUM)	
Number of stages	5
Feed streams convention	FEED: Stage 1 (Above-Stage) HEATED: Stage 1 (Above-Stage) AIR: Stage 5 (On-Stage)
Pressure Packed Height Column Diameter	1 bar 15 m 1.2 m
Dehumidifier (DEHUM)	
Number of stages	10
Feed streams convention	H_2O: Stage 1 (Above-Stage) COOLED: Stage 1 (Above-Stage) HUM-VAP: Stage 10 (On-Stage)
Pressure Packed Height Column Diameter	1 bar 15 m 1.2 m

Table 3. Column Internal Characteristics.

Packing	**Material**	**Dimension**	**Specific Surface Area (m^2/m^3)**
Pall Ring	Plastic	0.625 in/16 mm	364

Table 4. Exchanger Characteristics.

	Temperature	**Pressure**	**Utility Input**	
HEATER	75 °C	1 bar	Medium pressure steam (MS)	4 MPa
COOLER	22 °C	1 bar	Cooling water (CW)	Inlet Temperature 7 °C Outlet Temperature 12 °C 1 bar

3. Results and Discussion

3.1. Stream Acidity (pH) Measurement

The monitoring of system efficiency, product stream composition and concentration during the HDH process is an essential prerequisite for model analysis and process control. The composition of maleic anhydride wastewater is very complicated, and to simplify the calculation, stream acidity (pH) was used to show how many organic acids are remaining in the HDH system. The pH results can be found in Table 5.

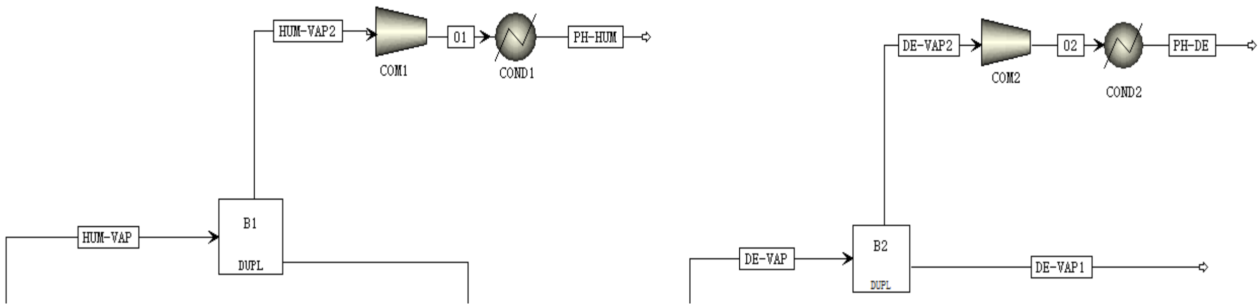
Table 5. pH stimulation results.

Stream	**FEED**	**HUM-VAP**	**DE-VAP**
pH	3.02	4.15	5.74

Liquid stream acidity can be directly measured by setting an ion model with Elec Wizard in Aspen Plus.

Due to the pH can only be measured in the liquid phase, the gas stream liquefaction process is required in stimulation. When the gases are subjected to low temperature and high pressure, the gases begin to liquefy.

The gas stream acidity measure model can be seen in Figure 2. Increased pressure with a compressor brings the gas molecules closer to each other, as in Figure 2a and then feeds to a condenser to convert the gas into the liquid, as in Figure 2b.



(**a**) Humidification model in Aspen (**b**) Dehumidification model in Aspen

Figure 2. Gas stream pH measure model.

3.2. Organic Pollutant (COD) Measurement

Chemical oxygen demand (COD) is an indicative measure of the amount of oxygen that can be consumed by reactions in a measured solution. It is commonly expressed in mass of oxygen consumed over volume of solution which in SI units is milligrams per liter (mg/L). A COD test can be used to easily quantify the amount of organics in water [20].

$$\text{Conversion Factor (COD)} = \frac{(\mathrm{C} * 2 + \mathrm{H} * 0.5 - \mathrm{O}) * 16}{\mathrm{M}}$$

where:

C = Number of carbon atoms
H = Number of hydrogen atoms
O = Number of oxygen atoms
M = Molar Mass

The simulation result is showed in Table 6. The conversion factor is showed in Table 7, the calculation result is showed in Tables 8 and 9.

From these calculations, it is known that the COD has a significand reduction from 19,864 mg/L to 2154.59 mg/L, and the reduction rate is 89.1%.

Table 6. Stream stimulation results.

Stream	FEED (kg/h)	FEED (cum/h)	FEED (kg/cum)	DE-LIQ (kg/h)	DE-LIQ (cum/h)	DE-LIQ (kg/cum)
/	30,000	29.98	1000.70	163,075.17	163.96	994.63

Table 7. COD Conversion Factor.

Component	Chemical Formula	C	H	O	Molar Mass	Conversion Factor
MA	$C_4H_2O_3$	4	2	3	98.06	0.979
ACETIC	CH_3COOH	2	4	2	60.052	1.066
ACRYLIC	$C_3H_4O_2$	3	4	2	72.063	1.332
DBP	$C_{16}H_{22}O_4$	16	22	4	278.344	2.242
PHTHALIC	$C_8H_4O_3$	8	4	3	148.12	1.620
PHTHA-AC	$C_8H_6O_4$	8	6	4	166.13	1.445
MALEI	$C_4H_4O_4$	4	4	4	116.07	0.827
FUMAR	$C_4H_4O_4$	4	4	4	116.07	0.827
BUTANOL	$C_4H_{10}O$	4	10	1	74.14	2.590

Table 8. Calculation of FEED stream COD.

		FEED		
Component	**Mass Flow**	**COD Emission**		
	kg/h	**kg/d**	**t/a**	**mg/L**
MA	21.00	420.00	126.00	686
ACETIC	43.22	864.34	259.30	1536
ACRYLIC	45.00	900.00	270.00	2000
DBP	21.00	420.00	126.00	1570
PHTHALIC	6.00	120.00	36.00	324
PHTHA-AC	75.00	1500.00	450.00	3614
MALEI	240.00	4800.00	1440.00	6621
FUMAR	24.00	480.00	144.00	662
BUTANOL	33.00	660.00	198.00	2851
Total	508.22	1404.34	421.30	19,864.72

Table 9. Calculation of DE-LIQ stream COD.

		DE-LIQ		
Component	**Mass Flow**	**COD Emission**		
	kg/h	**kg/d**	**t/a**	**mg/L**
MA	0.28	5.55	1.66	2
ACETIC	28.00	560.01	168.00	182
ACRYLIC	22.16	443.19	132.96	180
DBP	1.60	31.97	9.59	22
PHTHALIC	N/A	N/A	N/A	N/A
PHTHA-AC	N/A	N/A	N/A	N/A
MALEI	7.59	151.73	45.52	38
FUMAR	N/A	N/A	N/A	N/A
BUTANOL	109.58	2191.50	657.45	1,731
Total	169.20	3383.94	1015.18	2154.59

3.3. Temperature Measurement

From the stimulation, the HEATER temperature was varied from 30 °C to 110 °C to investigate the relationship between product pH and vapor-phase water.

As can be seen in Figures 3 and 4., the vapor-phase water output increases with the heating temperature while the pH decreases.

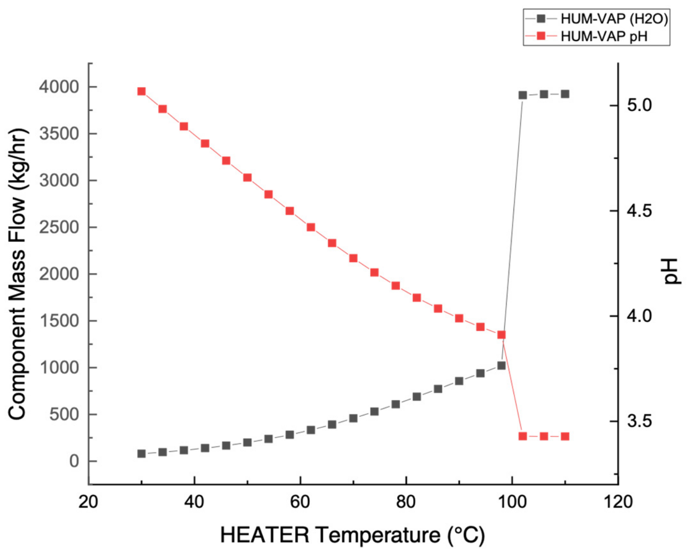


Figure 3. Water vapor mass flow and HUM-VAP stream pH as a function of HEATER temperature.

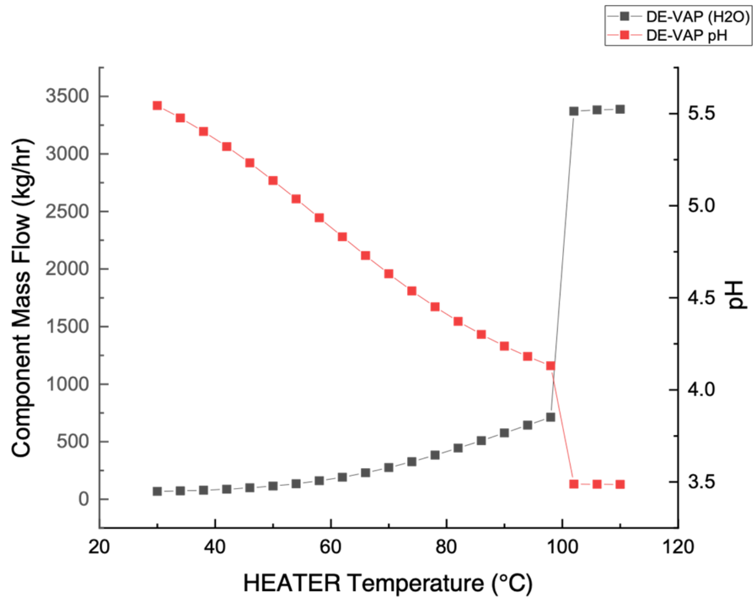


Figure 4. Water vapor mass flow and DE-VAP stream pH as a function of HEATER temperature.

3.4. *Yield Liquid Analysis*

Figures 5 and 6 plots how HEATER temperature affects the liquid stream pH with HEATER temperature varying from 60 °C to 80 °C in increments of 5 °C.

After several simulations, when the ratio of waste liquid, air and fresh water feed was 2:1:2 (FEED:AIR:H2O = 2:1:2), the highest pH (lowest acidity) of vapor product streams was reached.

The overall simulation result is showed in Figure 7.

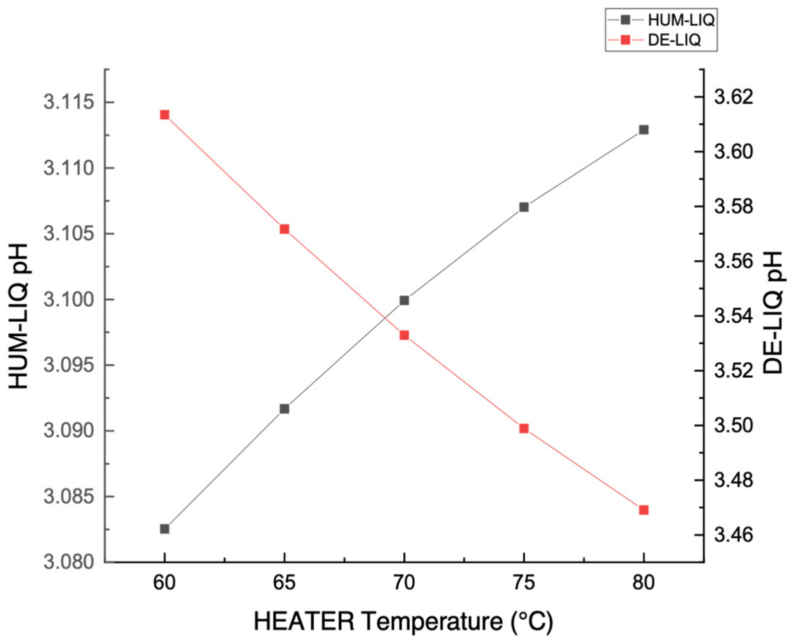


Figure 5. HUM-LIQ pH and DE-LIQ pH as a function of HEATER temperature.

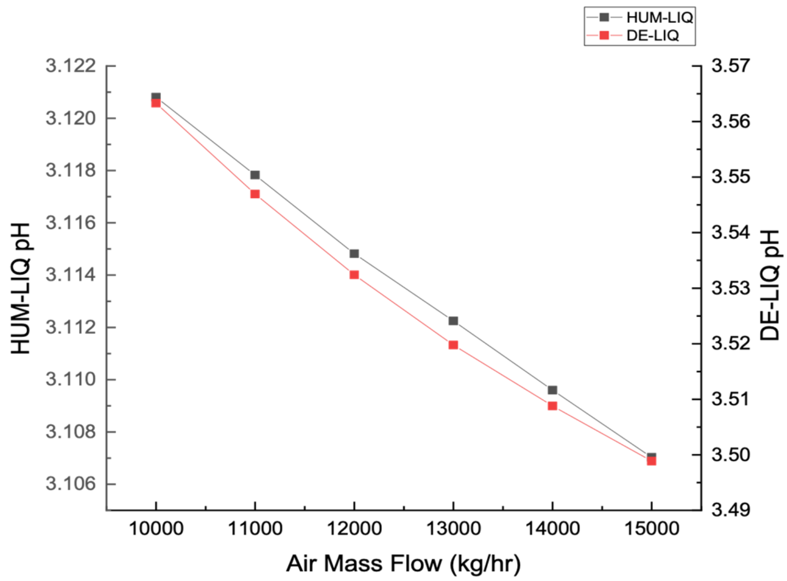


Figure 6. HUM-LIQ pH and DE-LIQ pH as a function of feed air mass flow.

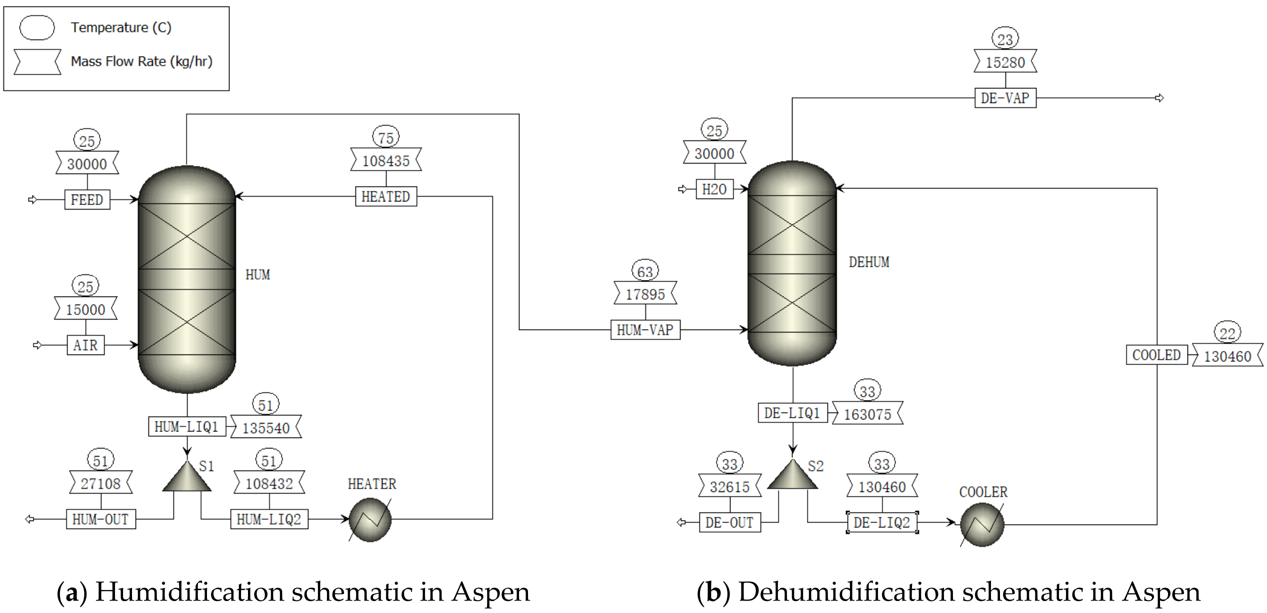


Figure 7. Overall HDH schematic with simulation result.

4. Conclusions

After the ASPEN simulation, it was observed that the COD reduction is significant, from 19,864 mg/L to 2154.59 mg/L, with a reduction rate of 89.1%, the pH change of the maleic acid wastewater was from 3.0 to 5.7, and the ratio of waste liquid, air, and fresh water feed was 2:1:2 (FEED:AIR:H2O = 2:1:2). From the process of the organic acid matter migration simulation, we can conclude that the proportion of acid matter production formation at different boiling points is different based on varying heating conditions.

It can be concluded that, the HDH process can achieve a good organic acid pollutant removal rate for industry wastewater [21], and the cleanliness of the water production is very high, which is suitable for biochemistry [22].

Although HDH is widely studied these days in the world, studies on real operation systems are seldom found, which is why it is important to develop a modeling system to analyze the output material from HDH system and to find suitable parameters for these reaction equipment settings.

In this study, HDH process was successfully simulated in the application of industry wastewater. With the ASPEN simulation, sensitivity analyses of the air volume, tower diameter, tower height, and heat addition can be conducted, and better parameters can be selected. Therefore, the use of this model is of great significance for the selection and calculation of pilot test equipment. In practical applications, the model can help engineers solve the key problems of HDH systems, such as heat balance calculations, equipment selection, and the prediction of incoming and outgoing evaporation materials [23].

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Data Availability Statement: The authors confirm that the data supporting the findings of this study are available within the article [and/or] its supplementary materials.

Conflicts of Interest: The authors declare that they have no known competing financial interest or personal relationship that could have appeared to influence the work reported in this article. The authors declare no conflict of interest.

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