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Energy Consumption for Black Liquor Treatment Through A Heat Pump System

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ABSTRACT

The lignin was precipitatedfrom black liquor through an acidification process using sulfuric acid. The remaining acidic filtrate was evaporatedusinga vacuum evaporator to obtain sodium sulfate crystals with a highly concentrated aqueous solution, containing mainly organic compounds. Anenergy consumption model was developed, consistent with the experimental data, for the recovery of lignin and sodium sulfate from black liquor by a heat pump system. Based on the calculations of the model, for a treatment of 1ton black liquor containing 12.0% of dry solid matters, the process generates 28.0kg of lignin, 54.4kg of sodium sulfate and125.6kg ofhighly concentrated solution (containing 40kg of dry organic matter and 25.6kg sodium sulfate). Considering 41.1kWh electric energy and 246.5kg supplementary steam used for the treatment, a total of 713,000kJ of energy is consumed for the treatment process.

Keywords: *Black liquor; Lignin; Sodium sulfate; Energy*

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I. INTRODUCTION

Black liquor is a black and viscous liquid generated in the alkaline pulping process. It usually containsa large amount of sodium, lignin and other organic matter (Haddad et al. (2017). The black liquor is a huge source of pollution, which needs to be effectively treated before discharging (Yan et al. 2008).

In general, black liquor istreated by a combustion-causticization method in the pulp and paper industry. The combustion- causticization process extracts black liquor from the cooking mixture in the following steps:evaporation the extracted black liquor to obtain high concentration of black liquorcombustion of the highly concen trated black liquor by a Thomas boiler to obtain sodium carbonate and recover the combustion heat; and conversion of sodium carbonate to sodium hydroxide by lime (Nong et al. 2016, Luo and Wang2011).Although the treatment removes the pollutants in black liquor completely and recovers the combustion heat and the sodium resource, the disadvantages are: 1) High energy is consumed in the concentration process; 2) The contained lignin, which is a useful nature material, is burned up; 3) A great amount of green-house gases and other harmful gases are released; 4)The process is not suitable for the small scale pulp mills who employ wheat straw as raw materials (Lu et al. 2017, De Blasio et al. 2016, Rastegarfar et al. 2015).

As an alternative to the combustion-causticization treatment, black liquor can be treated by acidification using sulfuric acid, yielding lignin as a product. The remaining acidic filtrate after removal of the lignin can be regarded as pulping wastewater. Itcontains large amounts of salt and organic matter, whichare heavy pollutants. Heat pump evaporation is considered to be an energy-saving way to achieve evaporation, andithas been extensively studied in desalination of sea water (El-Sayed1999, Choi et al. 2005).The acidic filtrate from alkaline pulping, after lignin precipitation, containsab out 4%-12% of sodium sulfate and 2%-6% of organic matter (De and Bhattacharya1996). Due to the similarity of the salt concentration to that of sea water, the filtration may be treated by a heat pump system to obtain additional products of sodium sulfate, in addition toa highly concentrated solution of organic compounds. Therefore, it would be interesting to understand the energy consumption for the heat pump system in order to know its economic effectiveness. The objective of this research was to calculate the masses of products and energy consumption in the black liquor treatment carried out by acidification combined with evaporation.

II. EXPERIMENTAL

2.1. Generating lignin from black liquor

The black liquor sample was obtained from the Jinrong Pulpand Paper Limited Company, Tiandong, Guangxi Province, China. The black liquor contained 12.0% solid matter. One thousand grams of black liquor was acidified with 45.0g sulfuric acid. After the separation by filtering, washing and drying, 28.0g of lignin was obtained with a total of 1,000g acidic filtrate remaining.

2.2. Evaporation of acidic filtrate

The remaininga cidic filtrate contained 4.0% of organic matter and 8.0% sodium sulfate, and it had a COD value of 8600mg/L. Then 1,000g of the acidic filtrate was evaporated usinga vacuum evaporator;54.4g of sodium sulfate crystals, 125.6g of highly concentrated organics in solution (52.0% of dry mass), and 820.0 gof condensate were obtained. The boiling point of acidic filtrate was testedusing a vacuum evaporator under different native pressures.

2.3. Model development of a heat pump system

The heat pump system model mainly consists of a lignin separation unit,a preheating unit, aheating and evaporating unit and asteam compressor unit, as shown in **Fig. 1**.

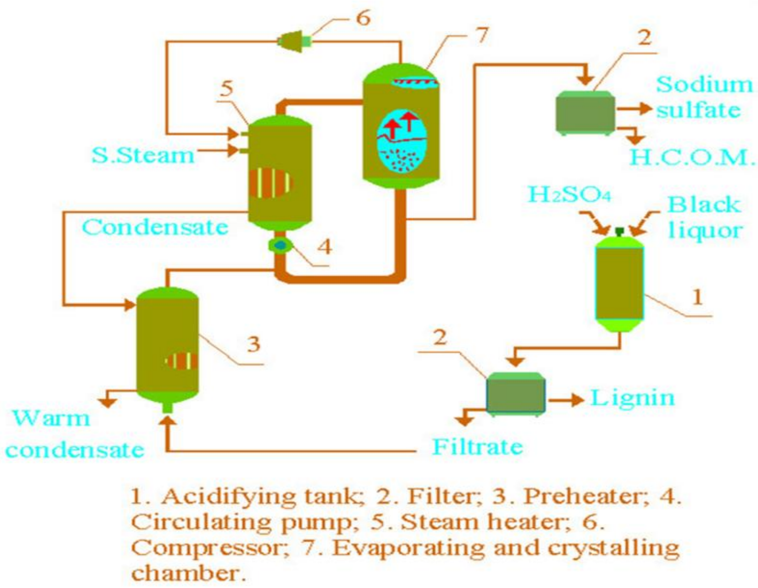

Fig.1 Diagram of the heat pump system

The operation process:

1. Black liquor was fed into a tank and acidified to a pH value of 1-3 using sulfuric acid to precipitate the lignin. The liquid mixture was then separated by a filter to obtainlignin solids and acidic filtrate.
2. The acidic filtrate was fed into a preheating unit, where it was heated from 30°C to 70°Cusingthe heat from the exhaust steam and the hot condensate (100°C) from the heating and evaporating unit.
3. The preheated acidic filtrate was fed into a steam heater and combined with the circulating flow from the evaporating chamber by an axial flow pump. In the steam heater, the acidic filtrate went through the pipes combined with the circulating flow, and the outside of the pipes was heated by steam. The 70°C acidic filtrate and the circulating flow were heated to 90°C by the heating vapor from the compressor and supplemental steam.
4. The 90^{o}C water flow went to the evaporating chamber, where a small part of water evaporated at the 0.06MPa pressure. Thereby, the crystal of sodium sulfate presented because the water flow was concentrated.
5. Using the compressor, the generated vapor was compressed to become the lowpres sure vapor (0.15-0.2MPa) with a raised temperature of 105°C, and was sent to the steam heater to heat the circulating flow, combined with the supplemental steam.
6. To obtain crystal products of sodium sulfate and concentrated organic solution, the concentrated solution combined with the crystal of sodium sulfate was conducted to a filter for separation.

2.4. Mass analysis

(1) Mass of special components in wastewater

Assuming to treat 1,000kg wastewater, the sodium sulfate, total organic matter and water were calculated to be 80kg, 40kg and 880kg, respectively.

(2) Mass of the high concentration of organic components

Assuming the ratio between water and organic matter in the high concentration of organic solution was 60:40, 1,000kg wastewater yielded 125.6kg organic solution in total, including 60kg water, 40kg organic matter and 25.6kg sodium sulfate. The 25.6kg sodium sulfate in there was because it dissolves in the 60kg water at 90°C with the solubility of 42.7g in 100g water.

(3) Mass of water evaporating

The mass of water evaporating was calculated based on mass balance. The evaporated water (m_{evap}) was the water in the fed wastewater ($m_{w,ww}$) subtracting the water in the high concentration of organic solution ($m_{w,os}$), as shown in Equation (1). The water in the fed wastewater was 880.0kg, and in the highly concentrated solution (which was rich in organic compounds) was 60kg. The total evaporated water was 820.0kg.

$$m_{W,WW} = m_{w,OS} + m_{evap.} \quad (1)$$

(4) Mass of sodium sulfate crystal

Based on the literature, whenthe temperature was at 0-32.28°C, the sodium sulfate was crystallized in the form of $10H_2O.Na_2SO_4$; while it was above 32.28°C, the sodium sulfate crystallized in the form of small granular anhydrous sodium sulfate (Quist-Jensen et al. 2017). Based on the mass balance, the mass of sodium sulfate crystal was 54.4kg, which was the mass of sodium sulfate in wastewater after subtracting the sodium sulfate in the highly concentrated solution of organics.

2.5. The temperature of wastewater discharged from the preheating unit.

The thermal efficiency of indirect steam heating was in range of 78-83%. Thus, there was about 17-22% of the total supplying heat flow out of the heating and evaporating unit, and passed through the preheating unit. Concurrently, the wastewater was heated by the heat flow out from the unit, which was sufficient to heat the wastewater from 30°Cto 70°C.

2.6. Energy consumption in the compressor unit

(1) The basic equation for the compressor work

The work of the compressor was calculated by Equation (2) (Zhang and Li,2003, Onishi et al. 2017).In the equation, T_e is the absolute temperature of the vapor that enters into the compressor, K; P_c is the pressure of the compressor's exhaust or the heating steam, Pa; P_e is the pressure of the vapor that enters the compressor, Pa; n is the variable index of the compressor, It ranges from 1 to 3, taking 1.2 in this study; R is the gas constant, The R of vapor is 0.4619 kJ/(kg·K).

$$W_c = \frac{m_{evap.} nRT_e}{n-1}\left[\left(p_c/p_e\right)^{(n-1)/n} - 1\right] \quad (2)$$

(2) Electric energy consumption

The electric energy consumption of the compressor is estimated based on the work divided by its energy efficiency, and the result can be expressed as Equation (3),where, ηis the compressor efficiency, η=0.92 for the axial compressors that were employed (Onishi et al. 2017).

$$E = W_c / \eta \quad (3)$$

2.7. Calculation of the supplemental steam

(1) Heat for water evaporation

The heat for water evaporation is the mass of vapor (m_{vapor}) multiplying the latent heat of vaporization, 2293.1kJ/kgat 0.06MPa.

$$H_{evap.} = m_{vapor} \times 2293.1 \quad (4)$$

(2) Heat for heating the wastewater

The heat for heating the wastewater ($H_{r.WW}$) can be obtained as the sum of special components (m_i, including water, Na_2SO_4 and total organic matter) multiplying their heat capacity (C_{pi}) and the difference of temperatures (t_1-t_0, from 70°C to 90°C here).

$$H_{r.WW} = \sum m_i \times C_{pi} \times (t_1 - t_0) \quad (5)$$

(3) Total heat supplying

The thermal efficiency of an indirect steam heater is in the range of 78-83%. Thus, the total heat supply can be calculated by Equation (6). An 80% of thermal efficiency of indirect steam heating was used for the calculation.

$$H_{TS} = (H_{evap.} + H_{r.WW}) / \eta \quad (6)$$

(4) Heat of the supplemental steam

The total supplying heat in the heating and evaporating unit is the sum of the heat of the heating vapor from the compressor and the heat of supplemental steam. Thus, the heat of supplemental steam is the total heat supplied, subtracting the heat of heating vapor, being expressed as Equation (7), where the heat of heating vapor was calculated by Equation (8).

$$H_{SS} = H_{TS} - H_{vapor} \quad (7)$$

$$H_{vapor} = m_{vapor} \times 2293.1 \quad (8)$$

(5) Heat flow out.

Based on energy balance, there was a net positive heat flow out fromthe heating and evaporating unit. The sum of heat flow out (H_{FO}) is the total heat supply (H_{TS}) subtracting the heat for water evaporation (H_{evap}) and the heat for heating the wastewater ($H_{r.WW}$).

$$H_{FO} = H_{TS} - (H_{evap.} + H_{r.WW}) = (1-\eta) H_{TS} \quad (9)$$

(6) Mass of supplemental steam

The mass of supplemental steam was calculated by Equation (10).

$$m_{SS} = H_{SS} / 2293.1 \quad (10)$$

III. RESULTS

3.1. Relationship between the boiling point and the evaporating pressure

Fig. 2 shows the relationship between the evaporating pressure and the boiling point of acid filtrate. It can be seen that the boiling point of acid filtrate increases as the evaporating pressure increasesin the range of 0.04-0.09 MPa. The relationship between the boiling point (B_p, °C) of acid filtrate and evaporating pressure (P, MPa) can be fit as shown in Equation (11). When the evaporating pressure is 0.06MPa, the boiling point of the filtrate is 90°C.

$$B_p = 911.43P + 36.27 \quad (11)$$

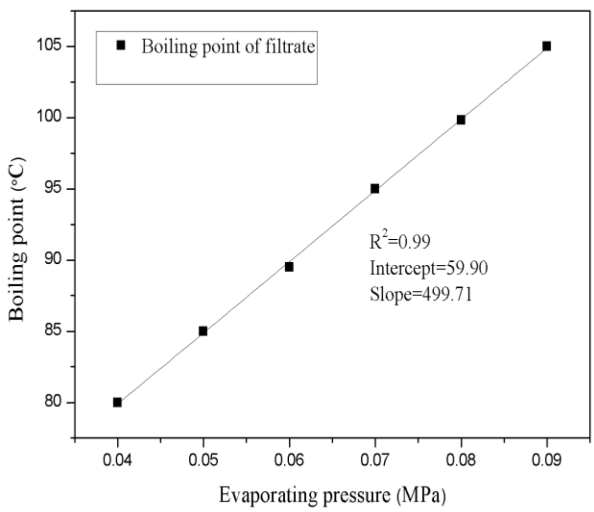

Fig. 2 Relation between pressure and the boiling point

3.2. Products generated from the modeling system

As shown in **Table 1**, the treatment of1000kg of pulping black liquor that contained 12.0% of dry solidsgenerated a total of 95.0kg of wet lignin (wet basis). 28.0kg of dry lignin was obtained after evaporation of 67.0kg water. After removal of lignin and 50.0kg ofwashing water,1,000kg of acidic filtrate remained. The acidic filtrate contained8.0% sodium sulfate and 4.0% total organic matter. The treatment of the filtrate produced54.4kg sodium sulfate, and 125.6kg highly concentrated solution, which contained 40kg organic matter, 60kg water and 25.6kg sodium sulfate.

Table 1 Mass balance for treating the black liquor (1,000kg)

Input	Mass, kg	Output	Mass, kg
Black liquor	1,000	Lign in	28.0
Sulfuric acid	45.0	Sodium sulfate	54.4
Washing water	50.0	Organic solution	125.6
		Water drying up	67.0
		Condensate	820.0
Total	1,095	Total	1,095

3.3. Energy conversion in the modeling system

As shown in **Table 2**, at the preheating unit, the exhausted steam and the hot condensate from the heating and evaporating unit contained 737,000 kJ heat in total. That heat heated the 1,000kg of wastewater from 30°C to 70°C, using 152,000 kJ heat. Thus, 584,000 kJ heat flowed to outside environment.The heating and evaporating unit needed 76,100 kJ of heat to heat the wastewater from 70°C to 90°C; noting that this was above its boiling point at a pressure of 0.06MPa. Additionally, 1,880,000kJ of heat was required to evaporate 820kg of water.While the thermal efficiency for indirect steam heater was 80%, it needed total 2,445,000 kJ heat to be supplied, including the heat content of 820kg heating vapor from the compressor and 246.5kg supplemental steam. In the compressor unit, the compressor consumed 148,000kJ electric energy to do 136,000 kJ of work; at the same time, 11,800 kJ of waste heat was exhausted to the outside environment. Based on these values, the utilization of heat pump to evaporate water can be considered as a method for saving energy. It consumes only 37.9% of the heat that would have been required for the conventional evaporation of water.

Table 2 Energy conversion in the modeling system (treatment of 1,000kg of WW)

Energy released	kJ	Energy absorbed	kJ
Preheat unit			
Condensate (100-50°C)	171,380	Filtrate (30-70°C)	152,144.0
Exhaust steam (246.5kg)	565,249.2	Heat flow out	584,485.2
Subtotal	736,629.2		736,629.2
Heating and evap. unit			
Heating vapor (820kg)	1,880,342.0	Waste water (70-90°C)	76,072.0
Sup. Steam (246.5kg)	565,175.5	Water evap. (820kg)	1,880,342.0
		Heat flow out	489,103.5
Subtotal	2,445,517.5		2,445,517.5
Compressor unit			
Electrical energy	147,944.0	Work to vapor	136,108.5
		Heat flow out	11,835.5
Subtotal	147,944.0		147,944.0

3.4. Performance of the modeling system

Based on **Tables 1** and **2**, a total energy of713,000 kJ was consumedto treat 1,000kg of wastewater; including 148,000 kJ (41.1kWh) electric energy and 565,000 kJ heat of supplemental steam (246.5kg). The treatment produced 54.4kg of sodium sulfate and 125.6kg of high concentration organic-containing solution.

3.5. Energy consumption influenced by the concentration of a cidic filtrate

Fig. 3 shows the relationship between the energy consumption and the concentration of acidic filtrate. Under the evaporating pressure of 0.06MPa, the energy consumption was decrease dlinearly with the increase in acid filtrate concentration. The electric energy consumption wasless than the heat of supplementary steam in the range of 6.0%-18.0% of concentration. The maximum electric energy consumption was164,000kJ/ton for 6.0% concentration; while the minimum was132,000kJ/ton for 18.0% concentration.The maximum consumption of supplementary steam was 621,000 kJ/ton for 6.0% concentration; while the minimum was 509,000 kJ/ton for 18.0% concentration. The reason was that the mass of water in the filtrate decreased with the increasein concentration. Therefore, the evaporating water with higher concentration was much less than that with lower concentration, resulting in less electric energy consumption of the compressor and less heat consumption of supplemental steam.

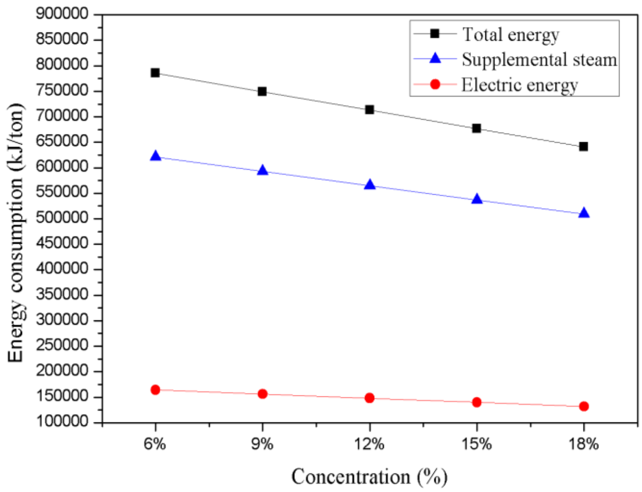

Fig. 3 Relations between energy consumption and concentration of filtrate

3.6. Energy consumption influenced by the evaporating pressure

Fig. 4 shows a relationship between energy consumption and the evaporating pressure. The evaporating pressure stronglyinfluencedthe electric energy consumption and supplementary steam, while it slightly influenced total energy consumption. The maximum electric energy consumption was215,000 kJ/ton at 0.04MPa; while the minimum was 83,000 kJ/ton at 0.09Mpa of evaporating pressure. The maximumconsumption of supplementary steam was 631,000 kJ/ton at 0.09MPa; while the minimum was 523,000kJ/ton at 0.04MPa of evaporating pressure.

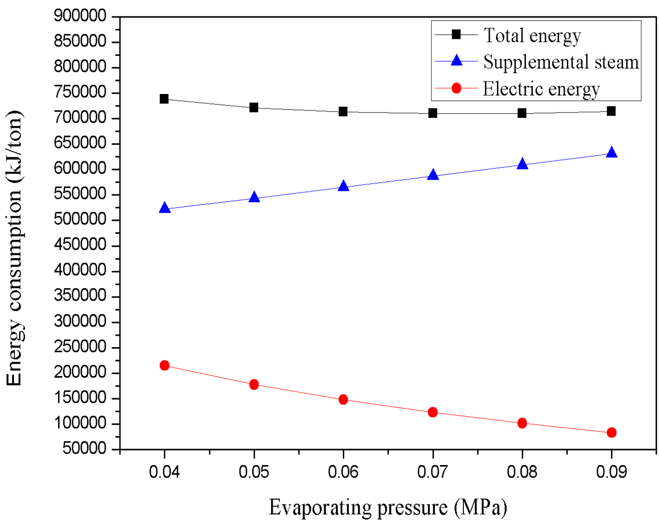

Fig. 4 Relations of between system pressure and energy consumption

3.7. Energy consumption Influenced by efficiency of the heater

Fig. 5 shows the relationship between the energy consumption and efficiency of the heater. The efficiency of the heater strongly influenced the total energy consumption and supplementary steam, while it did not in fluence the electric energy consumption, which was1, 479,000kJ/ton. The maximum consumption of supplementary steam was 628,000 kJ/ton when the efficiency of heater was 78%; while the minimum was

477,000 kJ/ton when the efficiency of heater was 83%.

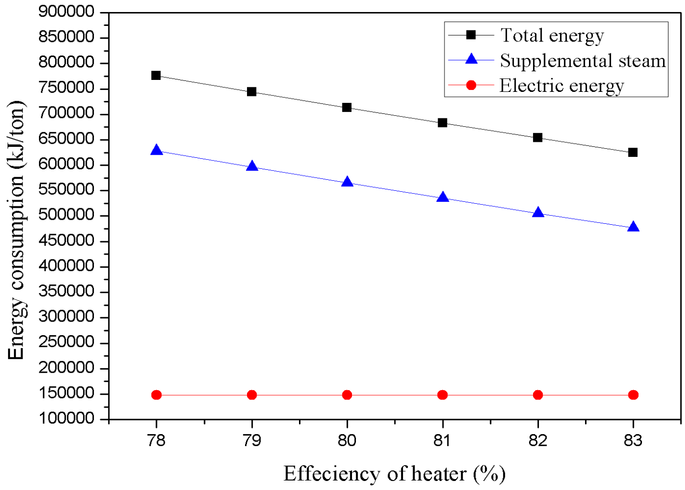

Fig. 5 Relations of between energy consumption and the efficiency of heater

IV. Discussions

As previously mentioned, black liquor is generally treated by the combustion method. This mainly includes the three steps of evaporation, combustion and causticization. A five-stage evaporation system is usually used to evaporate black liquor in pulp mills. The efficiency factor for a five-stage evaporation system is in range of 3.1-3.6, which means that it consumes 1.0kg medium steam to evaporate 3.1-3.6kg of water from black liquor. In this study, 1,000kg 12% black liquor contained 880kg water, in which 800kg of water needed to be removed to obtain 200kg of 60% strong black liquor. The evaporation of 800kg water consumed 222.2-258.0kg of medium steam. Besides the steam, oil was consumed for preheating the Thomas Boiler in the combustion step; and lime was consumed for causticization. An average of 20.0kWh electric energy was consumed in all the three steps for treating 1,000kg of black liquor. However, the method recovered sodium hydroxide and generated steam. The cost of the treatment of 1,000kg12% black liquor can generate profits of 24.4 Y RMB,as shown in **Table 3**.

Table 3 Cost of treating 1,000kg 12% black liquor by combustion (RMB)

Contains	Price Y.	Number	Income, Y	Expenditure, Y
Steam produced	0.12 Y/kg	450kg	54.0	
NaOH recovery	3.0 Y/kg	30kg	90.0	
Oil	4.0 Y/kg	1.5kg		6.0
Lime	0.8 Y/kg	50.0kg		40.0
Steam consumed	0.12 Y/kg	222.2kg		26.7
Electric energy	0.6Y/kWh	20kWh		12.0
Address white mud	0.1Y/kg	120kg		12.0
Depreciation	1.0% income	144Y		1.4
Maintenance	1.0% income	144Y		1.4
Salary	20Y/ton	1ton		20.0
Profit				24.4
Total			144.0	144.0

In this study, the treatment of 1,000kg of 12% black liquor by a heat pump system consumed 50.0kg sulfuric acid, 246.5kg steam and a total of41.1kWh of electricenergy. The treatment generated 28.0kg lignin, 54.4kg sodium sulfate and 125.6kg of solution with ahigh concentration of organic matter. Therefore, the treatment of 1,000kg 12% black liquor can yield 43.7YRMB of profit, as shown in **Table 4**.

The utilization of a heat pump to evaporate water has often been considered as a method for saving energy. Based on the present analysis it consumes only 37.9% of the heat that would have been required for water evaporation. That energy efficiency of water evaporation was higher than the energy efficiency of 38.0%-46% for a three-stage evaporation system; but was lower than the energy efficiency of 27.8%-32.3% required for a five-stage evaporation system. Additionally, the cost of steam compressor is high. And it might be corroded easily by the sewage vapor,it might be difficult to be employed ina large-scale mill, in comparison to a multiple-stage evaporation system.However, the treatment of black liquor by a heat pump system can achieve higher profits, avoid discharging white mud and reduce the greenhouse gas emission when used asan alternative methodfor the treatment of black liquor.

Table 4 Budget of treating 1,000kg of 12% black liquor by a heat pump system (RMB)

Contains	Price Y.	number	Income, Y	Expenditure, Y
Lignin	4.0 Y/kg	28.0kg	112.0	
Na_2SO_4	0.5 Y/kg	54.4kg	27.2	
Steam	0.12 Y/kg	246.5kg		29.6
Ele. Energy for compr.	0.6Y/kWh	41.1kWh		24.7
Ele. Energy for others	0.6Y/kWh	10.0kWh		6.0
Address HCOM	0.1Y/kg	125.6kg		12.6
Depreciation	1.0% income	139.2Y		1.4
Maintenance	1.0% income	139.2Y		1.4
Salary	20Y/ton	1ton		20.0
Profit				43.5
Total			139.2	139.2

V. Conclusions

A heat pump system model was used to simulate the process of the filtrate treatment and energy consumption. A treatment of1,000kg 12% black liquor generated 28.0 kg lignin, 54.4kg sodium sulfate and 125.6kgof solution having a high concentration of organic matter. The process consumed 246.5kg of supplementary steam (565,000 5kJ heat),counting 41.1 kWh electric energy (148,000 kJ). The consumption accounted for a net 37.9% of the heat of water evaporation. The treatment of black liquor by a heat pump system might beprofitable, avoid discharging white mud, and reduce the greenhouse gas emissions, while provid in gan alternative method for the black liquor treatment.

Acknowledgments

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