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Study of Energy Integration in Chlorobenzene Production Process Using Pinch Technology

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Abstract – *In this paper, the industrial application of detailed heat integration in the chlorobenzene production plant has been solved by using the Pinch design method. Designing a Heat Exchanger Network (HEN) through pinch analysis is an effective energy integration technology. In addition, minimizing unit and cost, operability and process control are essential parameters for selecting a suitable HEN design. In this research, the HEN of the existing process has been designed and analyzed using HINT software to minimize energy consumption and cost, thereby achieving maximum energy recovery. The analysis shows that the existing plant has been well integrated, and sound energy-saving effects and minimum energy requirements have been observed through the energy integration between processes. The total energy saving has been 91.80 %, while the energy recovery for heating and cooling have been 86.99 % and 96.23 %, respectively. Based on the economic analysis, the total annual cost of the proposed HEN design has been calculated to be 80912.5 \$.* ***Copyright © 2022 Praise Worthy Prize S.r.l. - All rights reserved.***

***Keywords**: Heat Integration, Pinch Analysis, Heat Exchanger Network, Propylene Glycol Production, Glycerol Conversion*

Nomenclature

ΔT_{LM}	Log mean temperature difference
$\Delta T_{\min}$	Minimum temperature difference
A	Heat exchanger area
a, b, c	Heat exchanger parameters
ACC	Annual Capital Cost
AOC	Annual Operating Cost
$C_{1,2,3}$	Cold stream 1, 2 and 3
CC	Capital Cost
C_u	Utility cost
CI	Cost index
CP	Heat capacity
GCC	Grand Composite Curve
$H_{1,2,3}$	Hot Stream 1, 2 and 3
HEN	Heat Exchanger Network
MER	Maximum Energy Recovery
N_u	Total number of utilities
PTA	Problem Table Algorithm
Q	Heat load
Q_u	Utility heat duty
TAC	Total Annual Cost
T_{in}	Supply temperature
T_{out}	Target temperature
S_i, S_{i+1}	Shifted temperatures
U	Overall heat transfer coefficient

I. Introduction

Chlorobenzene is an aromatic organic compound with the chemical formula C_6H_5Cl that does not occur naturally [1]. The synthetic organic chemical manufacturing industries primarily consume monochlorobenzene as a feedstock. As a result of the simple manufacturing process, monochlorobenzene is often produced in the same plant where it is consumed.

The productions of phenol and o- and p-nitrochlorobenzenes have historically been the two main markets for monochlorobenzene. Monochlorobenzene is also used as a feedstock in the production of diphenyl oxide, dye and herbicide intermediates, and sulfone polymers [2]. The most well-known use of chlorobenzene is in manufacturing the pesticide Dichloro diphenyl trichloro ethane compound. Chlorobenzene is primarily used as an intermediate in manufacturing phenol, chlorobenzene, and other products such as herbicides, dyestuffs, and rubber. Chlorobenzene is also used in producing adhesives, paints, paint removers, polishes, dyes, and medications as a high-boiling solvent [3]. The chemical industry has had to deal with increasing competition in recent decades due to economic growth, rising public concern for the environment, and the actions by national governments in the areas of climate, safety, and health. Continuous optimization by revamp of existing production plants has emerged as a critical strategy in response to these conditions and given the capital-intensive nature of the chemical industry [4]. According to Bhalerao et al. [4], with the aid of the First and Second Laws of

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Thermodynamics, Pinch technology provides a basic technique for systematically studying chemical processes and the surrounding utility structures. The pinch technology has been widely utilized for heat integration analysis [5]. Techno-economic comparison of dual LLE with distillation for manufacturing 10,000 MT of bio-butanol has shown that the bio-butanol production cost from lignocellulose biomass has been much lower than corn and sugarcane due to cheaper feedstock and higher co-product credit [6]. The equation of energy for measuring the changes of enthalpy (ΔH) in streams moving through a heat exchanger is given by the First Law of Thermodynamics. The Second Law of Thermodynamics determines the direction of heat flow.

That is to add, heat energy can only flow from hot to cold. This prevents "temperature crossovers" between the hot and cold stream profiles as they pass through the exchanger unit [7]. A hot stream cannot be cooled lower than the cold stream supply temperature, and a cold stream cannot be heated to a temperature greater than the hot stream supply temperature in a heat exchanger system. The hot stream can only be cooled to the temperature set by the "temperature approach" of the heat exchanger. The minimum allowable temperature difference ($\Delta T_{\min}$) in the stream temperature profiles is the temperature approach for the heat exchanger unit.

The "pinch point" is the temperature at which $\Delta T_{\min}$ is detected in the process. The pinch specifies the minimum driving force in the exchanger unit that can be used [8], [12]. Pinch Analysis' basic concepts are as follows. Pinch Analysis is used to determine energy cost and Heat Exchanger Network (HEN) capital cost goals for a process, as well as to recognize the pinch point. Before design, the procedure estimates the minimum requirements for external energy, the unit number, and the network area for a given process at the pinch point. A heat exchanger network design that meets these criteria is developed. Finally, the network is configured by calculating the electricity cost and the network's capital cost to reduce the overall annual cost [8], [10]-[13]. As shown in Figure 1, benzene chlorination has manufactured chlorobenzene in the liquid phase. The process starts with a series of small, externally cooled cast iron or steel vessels containing the catalyst. Ferric chloride is often used as a catalyst, and this can be applied to benzene as a solution. In order to maintain a comprehensive benzene-to-chorine reaction at all points along the reaction stream, chlorine is supplied into each vessel via appropriately placed inlets. The reaction is an exothermic process, and the temperature is kept between 20 and 40 °C in order to avoid the formation of dichlorobezene, which occurs at higher temperatures [14], [15]. Applying pinch technology in the production of chlorobenzene allows process engineers to understand the thermal interactions between chemical processes and the utility systems surrounding them. Before the final full simulation and optimization, such knowledge helps to optimize the overall utility consumption and set-up operations as well as the utility system configuration.

Retrofitting is a term used to describe a task that is performed on continuous processes for the manufacture of bulk products. While other strategies rely on trial and error to improve a design, pinch technology allows the design engineer to set goals and then set out to achieve it.

Calculating the minimum heating and cooling requirements for a heat exchanger network is the starting point for an energy integration analysis using pinch technology [11]-[13]. The focus of this study has been on energy integration and Pinch technology-based retrofit of an existing chlorobenzene facility. Retrofitting an existing flow scheme to debottleneck a plant in order to improve flexibility, reduce energy consumption, and reduce emissions, or modifying the system in order to incorporate a new technology are all examples of retrofit designs. In contrast to a new design, which begins with a blank sheet of paper, a retrofit study begins with a previously optimized procedure [11]-[13]. This study aims to use energy integration analysis to minimize energy consumption in a chlorobenzene production plant with minimal capital investment.

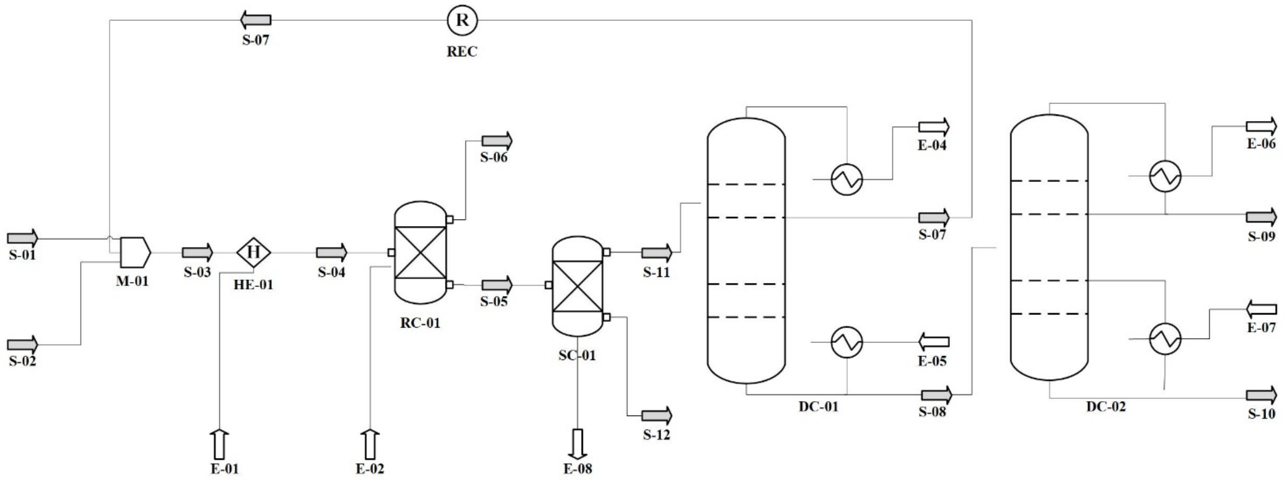

Fig. 1. Process flowsheet diagram of chlorobenzene production in continuous process [9]

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This is achieved by reducing the usage of utilities and the process-to-process heat transfer between existing hot and cold streams.

It also meets the essential constraint of maximizing the use of usable heat transfer areas with retrofit technology to the greatest extent possible. This article has analyzed data from the process flow diagram, heat exchanger design sheet, chlorobenzene production plant and operation data of chlorobenzene production from benzene and chloride using software HINT-2.2, CHEMCAD version 6 simulation software. The fundamental objective of energy targeting is to evaluate the current network for potential energy and cost savings based on a predetermined minimum temperature. The analysis has demonstrated that HEN design is an effective method for increasing energy recovery and decreasing external utilities.

II. Method

This study has used several data, including the process flow diagram of chlorobenzene production from benzene and chloride as depicted in Fig. 1, heat exchanger design sheet, chlorobenzene production plant operation data, pinch analysis software HINT-2.2, CHEMCAD version 6 simulation software, and pinch analysis excel spreadsheet provided by the Institution of Chemical Engineers (IChemE) [9], [13].

III. Results and Discussion

III.1. Data Extraction

The most important aspect of a process integration analysis is data extraction.

It relates to extracting the data needed for pinch analysis from a given method. The minimal information required is material and energy ratios, physical and chemical properties of materials, and costing data. The stream data needed for calculations in pinch analysis, shown in Table I, have been collected from the plant material and energy balance flow sheets obtained from continuous chlorobenzene production.

These data consist of the streams supply and target temperatures in the integration process. It also includes heat loads in the heat exchangers. P. Kemp, 2010 [17] suggested that it is imperative to extract these stream data during stable plant operation.

III.2. Energy Targeting and Determination of Optimum ΔT_{min}

The primary goal of energy targeting is to examine the current network for possible energy and cost savings based on a selected minimum temperature value.

Thermodynamic profiles of the method have been analyzed by using Composite Curves (CC) and the grand composite curve to assess the hot and cold utility targets, as well as the pinch point location [11], [13].

TABLE I
STREAM'S DATA OF THE CASE STUDY
IN CHLOROBENZENE PRODUCTION PLANT

Streams	Heat Flow (kW)	T_{in} (°C)	T_{out} (°C)
C1	800	230	310
C2	500	230	250
C3	1550	95	250
C4	1125	95	170
H1	1125	320	245
H2	500	260	240
H3	900	180	120
H4	1125	180	105

The selection of the minimum temperature change $\Delta T_{\min}$ for the heat exchanger greatly affects the energy and capital costs in the chemical plant. The cost of energy rises roughly in proportion to $\Delta T_{\min}$, while the cost of capital falls gradually with $\Delta T_{\min}$[14]. Rosna, 2016 [15] did a heat assessment at various minimum temperature differences ($\Delta T_{\min}$) in order to analyze the $\Delta T_{\min}$ reliance on heat consumption or energy consumption, as well as the Equivalent Annualized Operating Costs (EAOC) in a chlorobenzene production plant. Rosna concluded that in this case, for the chlorobenzene production process, varying $\Delta T_{\min}$ did not affect the EAOC and the energy consumption. Hence, $\Delta T_{\min}$ is considered to be 10 °C, which accomplishes the typical value for the petrochemical industries i.e., 10 - 20 °C [12], [18].

III.3. Application of Pinch Analysis Technique

By defining pinch point and estimation of minimum hot and cold utilities and by construction of development of Problem Table Algorithm (PTA), Grand Composite Curve (GCC) and cascade diagram are used to determine minimum energy requirements. This section also presents the design and the retrofitting of the optimum Heat Exchanger Network (HEN) and the determination of the areas and the cost analysis of heat exchanger and its corresponding utilities.

III.4. Problem Table Algorithm and Composite Curve Construction

Maximum Energy Recovery (MER) refers to the minimum number of utilities or heating and cooling loads.

If Q_{hu} is the heat supplied by the hot utility and Q_{cu} is the heat extracted by the cold utility, calculating energy goals entails deciding the minimum values of Q_{hu} and Q_{cu} that should be met under thermodynamic constraints [13], [18]. Pinch analysis using two techniques (composite curve and problem table) is used to estimate minimum hot and cold utilities as a first step in HEN design. All hot streams from the T-H diagram are converted to a single curve based on the sum of streams of the same load interval. This curve is known as the hot composite curve. A cold composite curve can be obtained by doing again the same step with cold streams, as shown in Figure 2.

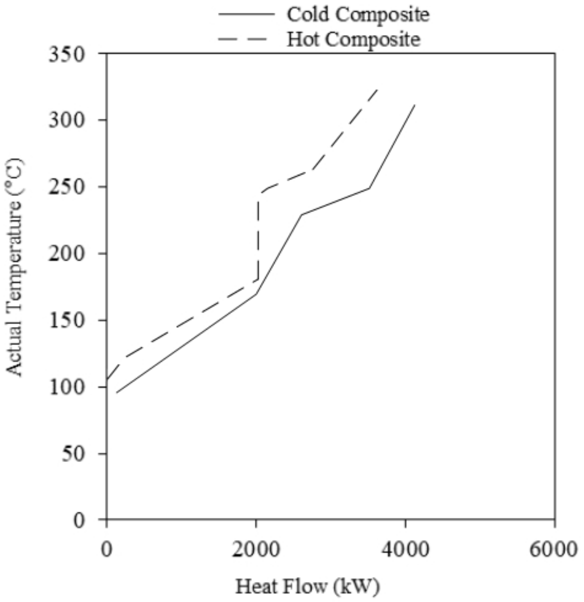


Fig. 2. Hot and cold composite curves

The energy recovery limits are in the overlap area, while the necessary heating and cooling are in the non-overlap region. The temperature difference can be adjusted by moving composite curves horizontally, which affects utility loads and heat transfer field.

Maximum energy recovery and minimum utilities are realized when detecting $\Delta T_{\min}$ value. Linnhoff and Flower created the problem table in 1978, which displays the related process stream data to the analysis of energy in an ordered table format. These data are used to calculate the energy balance needed when using Pinch technology [18]. The identification of pinch temperatures and minimum energy targets necessitates the creation of PTA. The requirements of the minimum hot and cold utility have been determined by using the $\Delta T_{\min}$ =10 °C assumption. The steps of constructing problem table are summarized as follows [13], [18]:

a. The process is divided into intervals;
b. The intervals limits are shifted for both hot and cold streams. ½ $\Delta T_{\min}$ is subtracted from the temperatures of the hot streams and ½ $\Delta T_{\min}$ is added to the temperatures of the cold streams as shown in Table II. All the temperatures are arranged in a descending order, excluding temperatures that are common to both hot and cold streams and serve as the upper and lower limits for different temperature intervals. This phase confirms that there is enough $\Delta T_{\min}$ driving force between the cold and hot streams for heat transfer to occur throughout each interval;
c. Balances of enthalpy of each interval restricted by shifted temperatures (S_i and S_{i+1}), can be easily determined as follows:

$$\Delta H_i = (S_i - S_{i+1})(\sum CP_{Hot} - \sum CP_{Cold})_i \quad (1)$$

d. The positive magnitude of the enthalpy balancing equation is called “surplus”, and the negative is called “deficit”, as shown in Table III. In order to develop a feasible network design based on the concept that all “surplus” intervals discarded heat to cold utility and all “deficit” intervals received heat from hot utility;
e. Starting with zero external heating and building up interval loads to define the largest deficit benefit, which represents the smallest hot utility (Q_h);
f. By cascading the phase interval with (Q_h) and accumulating interval loads, the minimum cold utility (Q_c), which is the load of the last interval, is reached.

The cascade diagram included in Table III represents the feasible and infeasible regions. They display heat flow data from various temperature intervals, which is useful for determining the minimum energy requirement and pinch field. In each temperature interval, the total heating and cooling requirement implies that excess heat is transferred to a cold utility and heat is supplied from a hot utility. The minimum energy demand for hot and cold utilities is illustrated in the cascade of Table III. The minimum energy demand for hot and cold utility has been evaluated to be 475 kW and 150 kW, respectively.

This table also shows the pinch temperature distribution, which is located between the fourth and the fifth temperature intervals. This pinch temperature serves as a guide for decomposing the design problem. Above the pinch, only heat is supplied, while below the pinch, heat is rejected to a cold utility. Plotting the shitted temperatures against the net heat flow presented in Table III yields the Grand Composite Curve (GCC) displayed in Fig. 3. It illustrates temperature variation as a feature of heat flow. The same figure shows that as the temperature rises, the enthalpy of the heat source rises, and the enthalpy of the heat sink falls. Fig. 3 also shows that at a temperature of 175 °C, heat interactions seen between heat source and the heat sink approach zero.

Table III shows that the there is a single pinch problem in this case that has occurred at temperature of 175 °C.

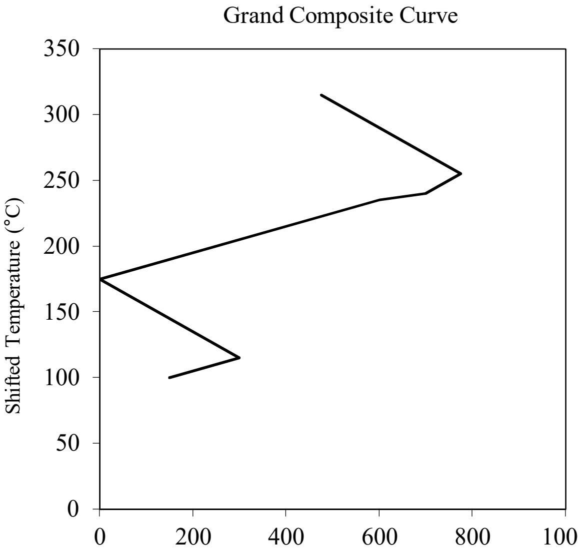


Fig. 3. Grand composite curve

III.5. Heat Exchanger Network Design and Energy Targeting

The streams that are in Table I have been inserted into HINT software, as illustrated by Fig. 4, in order to study the possible matches and to design the proposed heat exchanger network. HEN can be used to describe the proper installation between heat flow and cold flow in order to obtain utmost and efficient energy recovery with the lowest number of units. Making HEN or installing between one stream and another cannot be arbitrary and should follow the existing rules as follows [11], [12]:

a. The problem should always be solved from the most constricted region – the Pinch point;
b. No heat transfer across the pinch;
c. There is no hot utility below the pinch and no cold utility above the pinch;
d. Pinch matches should follow the following rule and the procedure illustrated in Figs. 5:
    1. Above pinch $H_{hot} \leq H_{cold}$
       $Cp_{hot} \leq Cp_{cold}$
    2. Below pinch $H_{hot} \geq H_{cold}$
       $Cp_{hot} \geq Cp_{cold}$

It can be shown that the network above the utility pinch comprises two hot streams and three cold streams based on the methodology used in the HEN design (Fig. 4). On the other hand, there are two hot and cold streams in the network below the pinch. HINT software is used to apply the match methodology described in Figures 5 on the grid diagram for process-to-process Heat Exchangers (HE) that should be installed to meet the minimum utility targets. In the existing procedure, Figure 6 shows the results of combination and matching, which yields 8 heat exchangers. Five process-to-process heat exchangers, one cooler (C), and two heaters are included (H).

III.6. Economic Analysis of HEN Design

After HEN design has been introduced, it is major step to check the candidate design by testing the economic evaluation in order to determine the return on investment.

The economic analysis usually involves evaluating the capital and the operating costs of the process. The overall economic cost or the Total Annual Cost (TAC) should be computed for new design of HEN.

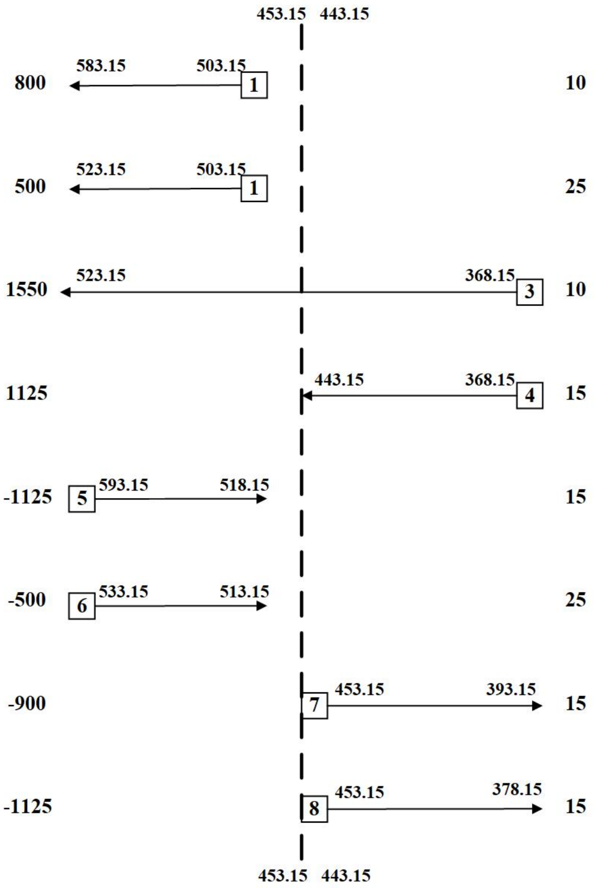


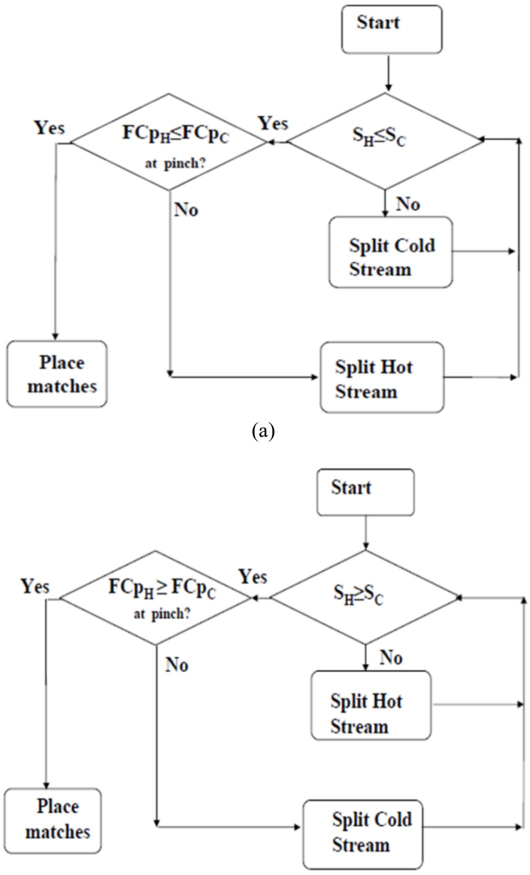
Fig. 4. Grid diagram of the streams

TABLE II
SHIFTED STREAMS

Interval	Stream	C1	C2	C3	C4	H1	H2	H3	H4
	mCp (kW/K)	10.0	25.0	10.0	15.0	15.0	25.0	15.0	15.0
	Heat Flow (kW)	800.0	500.0	1550.0	1125.0	1125.0	500.0	900.0	1125.0
Interval	Shifted Temp (°C)	COLD	COLD	COLD	COLD	HOT	HOT	HOT	HOT
	315	▲				●			
1		│				│			
	255	│	▲	▲		│	●		
2		│	│	│		│	│		
	240	│	│	│		▼	│		
3		│	│	│			│		
	235	●	●	│			▼		
4				│					
	175			│	▲			●	●
5				│	│			│	│
	115			│	│			▼	│
6				│	│				│
	100			●	●				▼

TABLE III
PROBLEM TABLE AND CASCADE

Shift Temperature	Interval	$T_{(i+1)}$-T_i	mCp_{net}	dH		Infeasible Cascade		Feasible Cascade	
°C		°C	kW/K	kW					
315						▼	0	▼	475
	1	60	5.0	300.0	surplus	300		300	
255						▼	300	▼	775
	2	15	-5.0	-75.0	deficit	-75		-75	
240						▼	225	▼	700
	3	5	-20.0	-100.0	deficit	-100		-100	
235						▼	125	▼	600
	4	60	-10.0	-600.0	deficit	-600		-600	
175					**PINCH**	▼	-475	▼	0
	5	60	5.0	300.0	surplus	300		300	
115						▼	-175	▼	300
	6	15	-10.0	-150.0	deficit	-150		-150	
100						▼	-325	▼	150



(a)

(b)

Figs. 5. Sequences for designing analysis of HEN above pinch (a) and below pinch (b) [9]

Cost estimation equations for HEN design are listed below [17]-[23]:

$$TAC = ACC + AOC \tag{2}$$

$$\begin{aligned} ACC &= CC \times \text{Module factor} = \\ &= (a + bAC) \times \text{Module factor} \end{aligned} \tag{3}$$

$$A = \frac{Q}{U \times \Delta T_{LM}} \tag{4}$$

$$AOC = \sum_{u=1}^{N_u} Q_u \times C_u \tag{5}$$

Table IV illustrates the heat exchangers in the process with its corresponding areas and costs. In order to calculate the cost targets and annualize them, the following data have been applied:

a. Heating cost: \$50/kJ;
b. Cooling cost: \$50/kJ;
c. Pay-back time: 10 years;
d. Interest rate: 5%;
e. Module factor: 3.4.

When calculating total area of the heat exchangers and its economic analysis, the following results have been obtained:

a. Minimum number of heat exchangers: 8;
b. Area target: 182.037 m^2;
c. Annual Operating Cost (AOC): \$31,250/year;
d. Capital Cost (CC): \$496,625;
e. Annual Capital Cost (ACC): \$49,662.5;
f. Total Annual Cost (TAC): \$80,912.5.

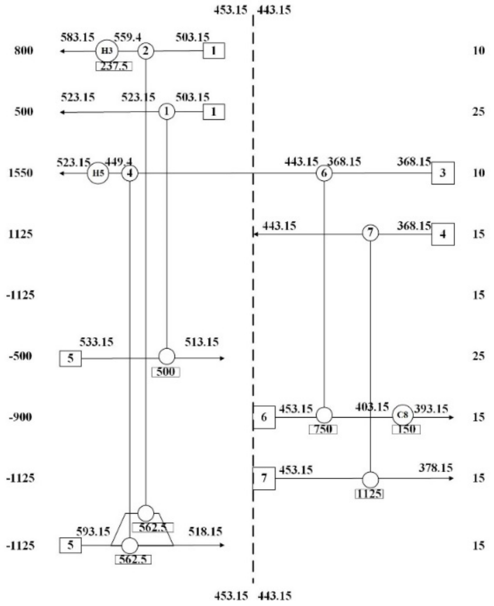


Fig. 6. The Heat Exchangers Network (HEN)

TABLE IV
HEAT EXCHANGERS IN THIS STUDY

H.E.	Heat Load (kW)	Hot Str	T1C (K)	T2C (K)	Cold Str	T1F (K)	T2F (K)	Area (m^2)	Cost ($)
1	500.	5	533.15	513.15	2	503.15	523.15	50.	80506.8
2	562.5	10	593.15	518.15	1	503.15	559.4	24.3279	56412.6
3	237.5	SG	-	-	1	559.4	583.15	9.49081	38682.
4	562.5	9	593.15	518.15	3	443.15	499.4	6.69431	34560.9
6	750.	6	453.15	403.15	3	368.15	443.15	37.5829	69487.7
7	1125.	7	453.15	378.15	4	368.15	443.15	112.5	126992
8	150.	6	403.15	393.15	SG	-	-	4.31523	30620.9

From the entire method of performing pinch analysis using HINT software, MER (maximum energy recovery rate) can be calculated. MER is the quantity or maximum energy load during heating or cooling, which can be reduced after integration with pinch analysis. The MER value is the difference between the heat and cooling loads before and after the integral analysis. Following are the MER for the studied case and its percent energy savings [21], [22]:

a. MER: 3500 kW/year;
b. Energy recovery for heating: 86.99%;
c. Energy recovery for cooling: 96.23%;
d. Energy saving: 91.80%.

IV. Conclusion

This article discusses the application of the retrofit design and pinch techniques to the integration system application in the chlorobenzene production plant. When the heat interactions between the heat source and the heat sink approach have been interacted, a single pinch problem has been revealed in this study case.

Furthermore, the $\Delta T_{\min}$ has been chosen to be 10 ºC based on the assumption and the outcomes of the previous studies. The results of the study have been calculated by using HINT software. According to the results, it has been proved that designing HEN is an efficient method for boosting energy recovery and so on, reducing external utilities. The energy saving for the study case has been high at 91.80 %, and the percentages of utilities' savings have reached 86.99 % and 96.23 % for both hot and cold utilities, respectively. Economic analysis results indicate that the costs are slightly high since there are only 8 heat exchangers in the process.

However, these prices are economically applicable since the target areas of the total heat exchangers are also high, 182.037 m^2. The HEN design has been successfully developed using the principles of pinch technology. The new HEN design can suggest energy targeting, cost analysis, and energy savings. It can be applied to industry in order to achieve energy saving.

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