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An energy evaluation for thermal conversion of thermoplastic waste to refined oil products using pyrolysis reaction process system

Hossam A. Gabbar*

Faculty of Energy Systems and Nuclear Science,
University of Ontario Institute of Technology,
2000 Simcoe Street North,
Oshawa, L1H 7K4 ON, Canada
Email: hossam.gabbar@uoit.ca
*Corresponding author

Mohamed Aboughaly and Awais Choudhary

Faculty of Engineering and Applied Science,
University of Ontario Institute of Technology,
2000 Simcoe Street North,
Oshawa, L1H 7K4 ON, Canada
Email: mohamed.aboughaly@uoit.net

Abstract: Thermoplastics are converted to hydrocarbon fuels in a chemical reaction called pyrolysis. The work highlights the energy consumption using Aspen HYSYSV8.8 simulations in each process stage starting with granulation, preheating, pyrolysis reactor and condensation as major process steps concluding that the highest required heat duty which is around 61% in the pyrolysis reactor thus having the highest operating cost. Aspen HYSYS is used to calculate energy consumption in each stage. The design is 10 tonne/hour of thermoplastic mixture. Pyrolys is reactor operating temperatures are 550°C at atmospheric pressure. The condensation system shows recovery duty of 3.4 MW of which can be used to heat cold streams. Pinch analysis was also carried out to design a heat exchanger network (HEN) between hot and cold streams in order to reduce energy consumption. Heat recovery from pyrolysis reactor effluent gases shows possible3.439 MW recovery in a 10 tonne per hour pyrolysis plant.

Keywords: sustainable process design; thermoplastics; oil production; Aspen HYSYS simulation V 8.8; mass and energy balance; pyrolysis; hydrocarbon liquids; char; gas; sustainability; cyclohexane; methane; pinch analysis; heat exchange network.

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Biographical notes: Hossam A. Gabbar is the author of more than 160 publications, including books, book chapters, patent, and papers, on the areas of: integrated smart energy grid planning, modelling and simulation; micro grid planning, protection and control; process systems and safety engineering; PHEV planning with hybrid fuel for green and sustainable transportation; risk-based hybrid energy supply and production chains; energy conservation and efficiency management; and process systems and safety engineering.

Mohamed Aboughaly is a Process Engineer with two years Industrial experience in Refinery and Petrochemical Plants including BEng Chemical Engineering from University Technology Petronas. He is a MASc Candidate at UOIT working on Thermoplastic Conversion to Oil Products.

Awais Choudhary is a third year Mechanical Engineer who worked in various fields related to recycling of waste plastics and did extensive analysis of waste recycling methods.

1 Introduction

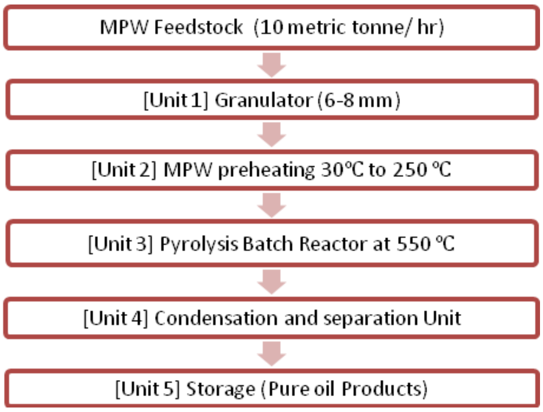
The plastic industry ranks the third among the world industries with nearly 300 million tonnes of plastics produced annually, leaving 275 million tonnes of unrecycled plastic waste deposited globally (Industry, 2010). The usage of plastic materials in daily life has continuously increased by a factor of 60 over the last 30 years (Vasudeo, 2016). However, the technology faced massive challenges due to large energy consumption in thermal cracking process by using energy inefficient gas furnaces or thermal crackers as heat source in pyrolysis reactions. Plastic waste recycling is playing a key role in fuel consumption development since the projected final products are high-end refined products ranging from C_1 to C_{48}. Advancement in pyrolysis helps prevent incineration and landfilling which still accounts for more than 72% of MPW recycling methods according to statistics in the UK (Silvarrey, 2016). And further, plastic to fuel (PTF) technologies offer the potential to manage landfill-bound plastics as an alternative source of fuel production (Sustainability.Inc, 2011).

The proposed process system uses chemical conversion process called pyrolysis, which is a thermochemical cracking of polymer molecules to oil products at elevated temperatures (400°C to 600°C) in absence of oxygen to convert thermoplastics to oil products (Young, 2010). Pyrolysis can be conducted at a range of temperatures and the chosen operating temperature for the pyrolysis reactor is 550°C. Plastics pyrolysis operating temperatures are defined as low (< 400°C), medium (400–600°C) or high temperature (> 600°C) (Buekens, 2007). The chosen feedstock plastics contains a mass fraction of low density polyethylene (LDPE), high density polyethylene (HDPE), polyethylene terephthalate (PETE), polystyrene (PS) and polypropylene (PP) which account for 90% of total plastic wastes in household and industrial applications (Vasudeo, 2016).

Successful conversion of waste plastics to oil products will promote alternative and renewable energy fuels around the world (Devaraj, 2015). It will also eliminate the tremendous threat caused by waste plastics to the environment due to its non-decomposable properties. Depleting fossil fuel reserves and increasing cost of the petroleum products have promoted millions of dollars to be invested in the search for alternative fuels. Nowadays plastics have become an indispensable part in daily life. Waste to energy is the recent trend in the selection of alternate fuels. Pyrolysis fuels are promising alternative fuels tested in the internal combustion engines (Daniel, 2015).

An illustration of process block diagram of the proposed process system is shown in Figure 1:

Figure 1 Process block diagram of thermoplastic conversion to oil products in a pyrolysis reaction (see online version for colours)



1.1 Process feed characteristics

The following are thermoplastics found in municipal plastic waste account for most of MPW (Silvarrey, 2016).

- LDPE
- HDPE
- PETE
- PP
- PS.

It is to be noted that pyrolysis process systems accepts any thermoplastic mixture composition while the simulation and experimental results are carried mostly on municipal plastic waste statistics in Ontario gathered from Canadian Plastic Industry Association (CPIA) submitted by Kelleher Environmental with the following composition shown in Table 1 (CPIA, 2012). Chemical properties are referred from (Biron, 2007).

Table 1 Polymer mass fraction, chemical formula and molecular weight used in simulation

Polymer element	*Mass fraction (m/m)*	*Chemical formula*	*Thermal conductivity W/(m·K)*	*M_w (g/mol)*
LDPE	0.20	$(C_2H_4)n$	0.30 to 0.34	28.06376
HDPE	0.20	$(C_2H_4)n$	0.46 to 0.52	28.05376
PETE	0.40	$(C_{10}H_8O_4)n$	0.15 to 0.24	192.1711
PP	0.10	$(C_3H_6)n$	0.17 to 0.22	42.08
PS	0.10	$(C_8H_8)n$	0.033	104.1

The Society of the Plastic Industry (SPI) reports that in the USA alone, the plastic industry accounts for 400 billion USD in annual sales (Kutz, 2011).

Due to plastics excellent properties and widespread usage an efficient pyrolysis process system converts a wide range of thermoplastic waste to oil products (Biron, 2007).

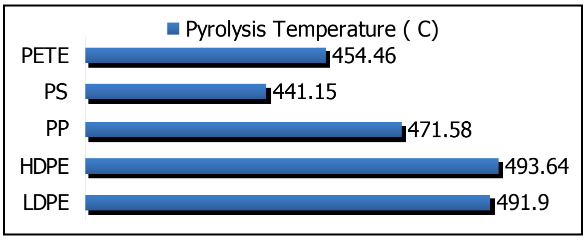
Recent researches have evaluated reaction kinetics, yield, optimum operating temperatures and pressures. Challenges remain in developing reliable reaction kinetics since pyrolysis reactions as well as reducing wax/tar and optimising hydrocarbon liquid production.

Conversions of waste plastics to refined fossil fuels reduce oil production significantly and provide a sustainable solution for plastic waste deposits. It also provides an alternative solution to millions of tonnes of plastic waste deposits in oceans and landfill which harm the environment due to its non-biodegradable characteristics. This mechanism will have a positive impact on plastic industry which encourages more plastic production and provide sustainable solutions to waste deposits that exist as thousands of tonnes as landfills in the environment (North and Halden, 2014).

2 Methodology

Thermoplastic waste polymers collected from municipal plastic waste with statistical fraction in Ontario is fed to the reaction system, with properties stated in Table 1. Controlled pyrolysis reaction at temperature at 550°C converting thermoplastic feedstock to refined oil products such as PG ($C_1 – C_4$), hydrocarbon liquid ($C_5 – C_{10}$) as desired products and tar/wax (> C_{11}) as an undesired by product. Thermal cracking occurs at atmospheric pressure and chosen reactor temperature is 550°C as our standard operating temperature. After thermal cracking or pyrolysis, tar and wax are removed from lower stream of reactor, gaseous products pass through condensation systems, separated and stored in storage tanks under standard atmospheric pressure and temperature in liquid form for hydrocarbon liquids. In Figure 2 are the pyrolysis temperatures of different types of thermoplastics (Pek, 2015) thus to ensure that all our feedstock of mixture thermoplastics has a complete reaction operating temperature is set to be 550°C.

Figure 2 Pyrolysis temperature of individual plastics (see online version for colours)



Source: Pek (2015)

Waste plastics undergo thermal cracking in a pyrolysis reactor, resulting in the formation of hydrocarbon liquid fuel as the main product and gaseous fuel up to about 20 wt%, as the minor product (Grause, 2011). Our process system involves in several stages of granulation, preheating, pyrolysis (thermal cracking), Condensation and end-product separation as illustrated in Figure 1. Residence time of 60 minutes is sufficient for optimum yield of hydrocarbons and thus chosen as the reactor residence time (López, 2011). The reactor conditions are in Table 2.

Table 2 Pyrolysis reactor operating conditions

Main process design features	
Feed	Thermoplastic waste (LDPE, HDPE, PP, PS, PETE)
Process	Pyrolysis (N_2 environment)
Main equipment	Batch reactor (BR)
Feedstock flowrate	10,000 kg/hr
Main product	Hydrocarbon oil and gas
Operating pressure	1 atm (atmospheric pressure)
Operating temperature	550°C
Reactor classification	BR (Green and Perry, 2008)
Reactor atmosphere	N_2 gas 99.999%
Catalyst	No catalyst added
Reaction residence time τ	60 minutes

Thermal process of converting thermoplastics to oil products has activation energies that explain the three stages of thermal cracking known as random scission, end-chain scission and abstraction of functional substitutes to form small molecules. In many cases these steps occur simultaneously (Vasudeo, 2016).

The products of thermal cracking are categorised as rich petroleum gases ($C_1 - C_4$), non-aromatic liquids ($C_5 - C_{10}$), waxes ($> C_{11}$) and char which are products of thermoplastic pyrolysis (Al-Salem, 2010). Reaction kinetic values used in simulation is stated in Table 3.

Table 3 Activation energy for pyrolysis reaction

Polymer type	*Activation energy, E_o (KJmol^{-1})*	*Reference*
LDPE	259.70	Encinara (2008)
HDPE	147.25	Al-Salem (2010)
PS	217.36	Wang (2013)
PP	202.50	Wang (2013)
PETE	235.7	(Encinara (2008)

The mentioned activation energies is used in our simulations and calculations to estimate energy required for pyrolysis reaction of 60 minutes residence time, τ. In Table 4, yield products according to Williams (2007) are mentioned.

Table 4 Pyrolysis yields of different polymers

Polymer	*(C_1–C_4)*	*(C_5–C_{10})*	*(> C_{11})*
LDPE	0.07	0.93	0
HDPE	0.202	0.132	0.658
PS	0.71	0.02	0.27
PP	95	6	0
PETE	0.32	0.15	0.53

Source: Williams (2007)

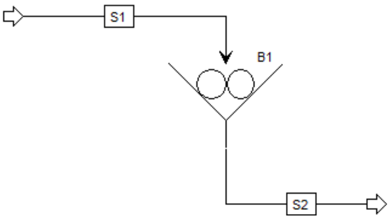
3 Calculations and simulation results

As mentioned in the introduction section, a plastic to oil pyrolysis plant consists of the following process stages which are explained and calculated below:

3.1 Solid granulator (6–8 mm)

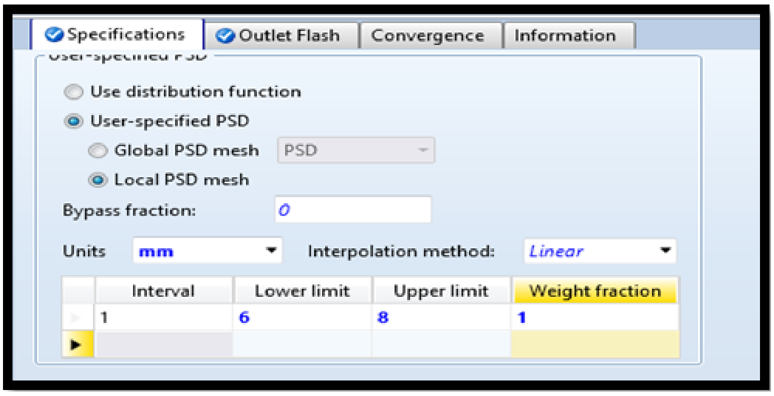
This major equipment involves mechanical granulation of thermoplastic feedstock to granules with particle size diameter (PSD) 6–8 mm based onto increase heat transfer surface area in preheating and pyrolysis stage. It also avoids agglomeration of plastic waste. After plastic solid granulation, the granulated plastic waste is pumped to preheater (unit 2). Figure 3 shows the Aspen HYSYS granulation using model of Rosin-Rammler Sperling Bennet (RRSB) distribution function to calculate power required for feedstock granulation.

Figure 3 Aspen HSYS v 8.8 granulation model



Distribution function for outlet PSD required is (6–8 mm). Granulation is done on dry basis and based on energy consumption at a flow rate of 10,000 kg/hr is 448.36 KW.

Figure 4 Granulator PSD specifications (see online version for colours)



○ Use distribution function
● User-specified PSD
○ Global PSD mesh PSD
● Local PSD mesh

Bypass fraction: 0

Units: mm

Interpolation method: Linear

	Interval	Lower limit	Upper limit	Weight fraction
	1	6	8	1
▶				

3.2 *Feed Preheater (30°C to 250°C)*

This equipment preheats granulated plastic waste to melting temperatures to ensure molten liquid flow to pyrolysis reactor. The feed preheater is a heating agitated tank designed to raise temperature of the granulated plastic mixture to melting Temperature allowing a continuous liquid flow to the pyrolysis reactor. The feed preheater is set to raise temperature to 250°C which is the melting temperature of common thermoplastics as shown in Table 2. The optimum melting temperature chosen is 250°C to ensure that almost all thermoplastic feed is in molten liquid state. The melting temperatures and specific heat capacity of plastic polymers (Biron, 2007) are illustrated in Table 4:

Table 5 Melting temperatures and specific heat capacity of polymers

Polymer	*LDPE*	*HDPE*	*PP*	*PS*	*PETE*
Melting point (°C)	110–120	130	122–124	240	250
Specific heat capacity (KJ/kg. °C)	2.302	2.302	1.9259	1.3398	1.298

Source: Biron (2007)

Simulating an agitated tank preheater as shown in Figure 6, which shows energy required to raise temperature of 10,000 kg/hr with a feedstock mass fraction LDPE: 0.20, HDPE: 0.20, PETE: 0.40, PS: 0.10, PP: 0.10. The feedstock preheater heats granulated thermoplastic mixture from 30°C to 250°C. Heat duty required is 1,406.77 KW at 2 bar as an operating pressure. Inlet stream is granulated solid (6–8 mm) while outlet stream is a molten liquid ready to be pumped to pyrolysis reactor. Variations in specific heat capacity of reactants can be seen in Figure 6 and can be generated from Aspen HYSYS physical property tool.

Aspen HYSYS properties feature simulates accurate specific heat capacity (J/kg.C) of different polymer phases as shown in Figure 6.

Figure 5 Aspen HYSYS V 8.8 simulation for feedstock preheater (see online version for colours)

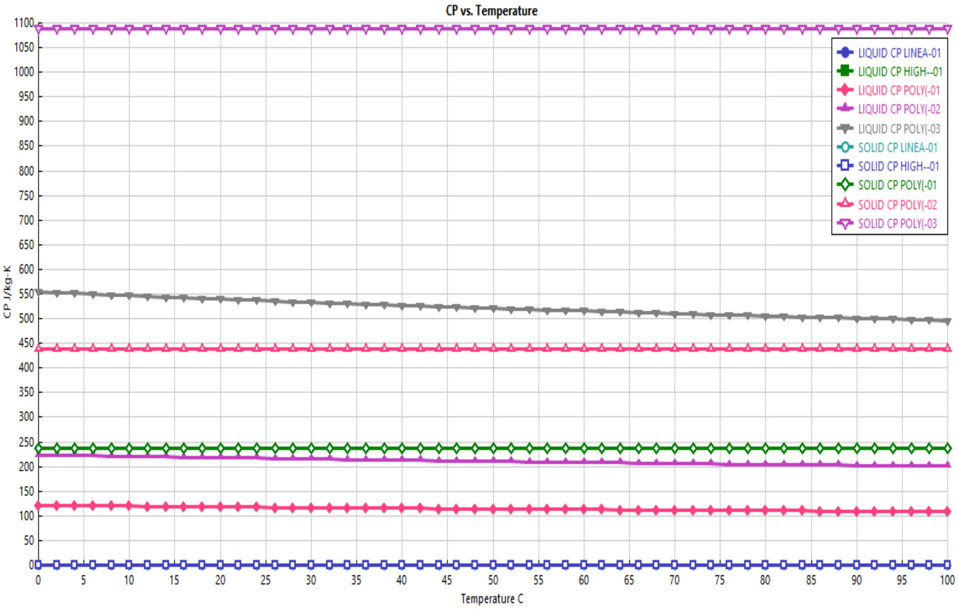
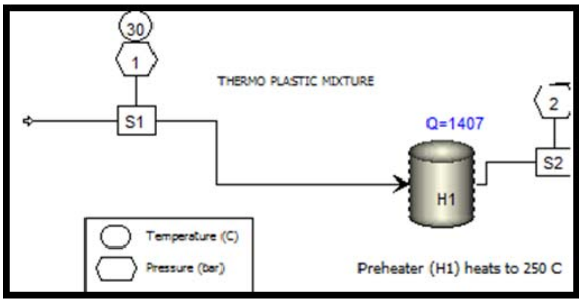


Figure 6 Specific heat capacity simulation according to Peng-Robinson method for plastic polymers in feed preheater H1 (see online version for colours)



3.3 *Pyrolysis batch reactor (550°C, τ = 60 munites)*

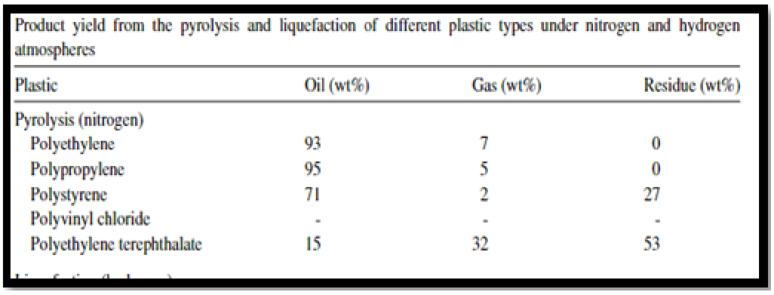
Thermal cracking chosen temperature is 550°C as an optimum temperature than ensures all thermoplastics components are pyrolyzed (Pek, 2015). This temperature is higher than all pyrolysis reactions of all thermoplastic mixtures which are also shown in Figure 2 (Lee, 2007; Papuga, 2016). The pyrolysis reaction of any composition can be expressed as:

$$C_xH_yO_z + Q \rightarrow \text{Char} + \text{Liquid} + \text{Gas} + H_2O \quad \text{(Chen, 2015)} \tag{1}$$

The chosen reactor model in simulation is 'RYield' which is a non-stoichiometric reactor based on known yield distribution. The yields are collected from Figure 7 Williams

(2007). The utility chosen to heat the reactor in simulation is fired heater since it can reach high temperatures required by pyrolysis.

Figure 7 Product yields achieved from pyrolysis above 500°C

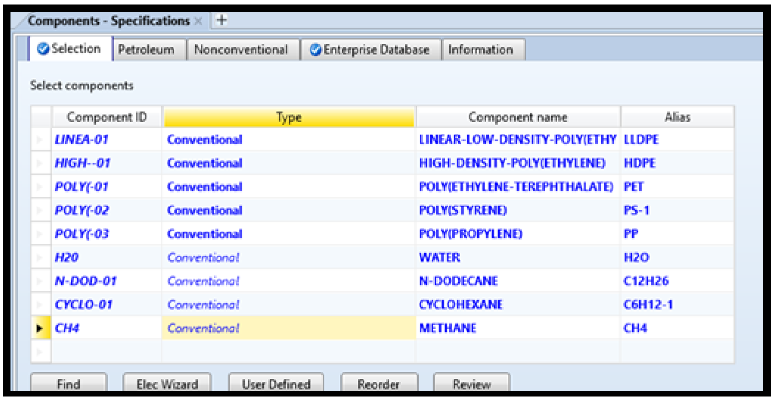


Product yield from the pyrolysis and liquefaction of different plastic types under nitrogen and hydrogen atmospheres

Plastic	Oil (wt%)	Gas (wt%)	Residue (wt%)
Pyrolysis (nitrogen)			
Polyethylene	93	7	0
Polypropylene	95	5	0
Polystyrene	71	2	27
Polyvinyl chloride	-	-	-
Polyethylene terephthalate	15	32	53

The chosen residence time for the reactor is 60 minutes which is recommended residence time τ, for thermoplastic mixture pyrolysis at more than 550°C (Kumar, 2016). The pyrolysis reactor is chosen to be a batch reactor (BR) model with residence time, τ 60 minutes at a constant temperature operating at atmospheric pressure without a catalyst. The reaction kinetic method chosen is by yield due to the complexity and inaccuracy of reaction kinetic models. The yield results achieved according to Williams (2007) in Figure 7. Peng-Robinson thermal model is used to large applicability range in terms of temperature and pressure as well as large interaction parameter database. This is suitable for pyrolysis reactors since it has high operating temperatures and longer residence time.

Figure 8 Input of reactants and products for pyrolysis BR simulation (see online version for colours)



Select components

Component ID	Type	Component name	Alias
LINEA-01	Conventional	LINEAR-LOW-DENSITY-POLY(ETHY	LLDPE
HIGH--01	Conventional	HIGH-DENSITY-POLY(ETHYLENE)	HDPE
POLY(-01	Conventional	POLY(ETHYLENE-TEREPHTHALATE)	PET
POLY(-02	Conventional	POLY(STYRENE)	PS-1
POLY(-03	Conventional	POLY(PROPYLENE)	PP
H2O	Conventional	WATER	H2O
N-DOD-01	Conventional	N-DODECANE	C12H26
CYCLO-01	Conventional	CYCLOHEXANE	C6H12-1
CH4	Conventional	METHANE	CH4

Simulation in Aspen HYSYS V 8.8, the oil, gas, and residue are simulated as the following products as seen in Figure 8:

- methane (CH_4) as any gas product from pyrolysis reactions
- cyclohexane (C_6H_{12})as any hydrocarbon liquid product from pyrolysis reaction
- residue ($C_{12}H_{24}$) as N-dodecane.

Product yield calculations are calculated using the following model:

$$X_n = \frac{Y_o}{W_o - W_\infty} \qquad \text{Equation 2}$$

X_n reactor conversion

Y_o desired product mass (g)

W_o initial reactant mass (g)

W_∞ final reactant mass (g).

The BR chosen in Aspen HYSYS is BR with yield calculations. The yield results used are the same in Figure 8, adding a theoretical energy requirement for pyrolysis to be 1,047.62 KJ/Kg (Gao, 2010).

The heat duty required for a 60 minute pyrolysis using Aspen HYSYS simulation for a preheated 10,000 kg/hr at 200°C is 2,908.3 KW (2.908 MW). The energy required as seen in the simulation is the largest energy duty needed in any process stage.

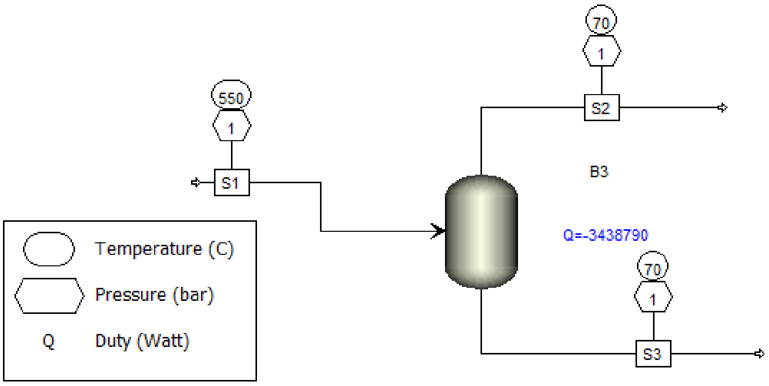
3.4 Condensation system (550°C to 30°C)

The condensation system cools down methane (CH_4), cyclohexane (C_6H_{12}), and N-dodecane ($C_{12}H_{24}$) from 550°C to 30°C for separation and energy optimisation. As cyclohexane and methane are the major products, the condensation system stages are designed with their dew point temperatures that condense the gaseous products to liquid on the condenser trays. A flash separator has an operating setting temperature of 70°C and 1 atm. The simulated energy duty is –3,438.790 KW which can be to heat cold streams.

Table 6 Dew and boiling point of gaseous products

Component	*Dew point (°C)*	*Boiling point (°C)*	*Reference*
CH_4	–40.62°C	–161.5 °C	Green and Perry (2008)
C_6H_{12}	80.85	80.74 °C	Lue (2009)

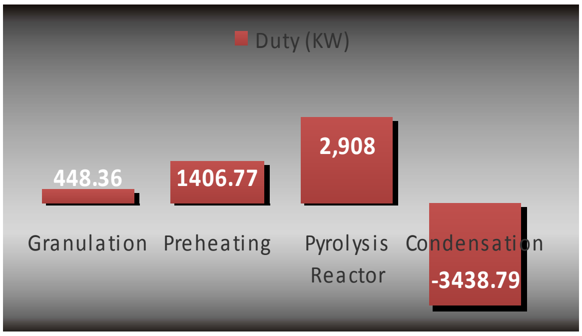
Figure 9 Flash separator heat duty for pyrolysis reactor gaseous products (see online version for colours)



4 Energy analysis for pyrolysis plastic to oil plant

In Figure 10 is the energy profile of major stages in a thermoplastic to oil pyrolysis plant which shows that nearly 61% of heat duty needed is used in the pyrolysis reactor thus showing that this stage requires the largest heat duty due to polymer dissociation energy and residence time required.

Figure 10 Energy consumption for each process stage – thermoplastic to oil chemical plant (see online version for colours)



Also, roughly 3.4 MW can be recovered from condensing the gaseous effluents if our pyrolysis reactor which are mainly cyclohexane and methane. The flash separator used separates cyclohexane (condensate) and methane is the (distillate) at operating temperatures 70°C which is below the boiling point of cyclohexane.

The granulation, preheating and pyrolysis reaction stages requires heat duty while condensation requires a cold duty thus given a negative value.

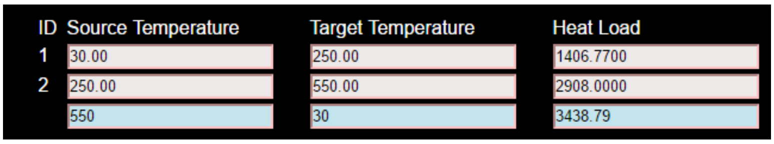
5 Pinch analysis before heat exchanger network (HEN) design

Pinch analysis is a heat exchanger network (HEN) design method to utilise energy exchange for hot and cold streams thus saving energy duty operating costs (Kemp, 2007).

Below are our process design cold and hot streams using online pinch analysis software.

The preheater stream (1) is considered a cold stream with a source temperature of 30°C and a target temperature of 250°C. The pyrolysis reactor equipment stream (2) is a cold stream that is to be heated from 250°C to 550°C. The gaseous effluent of the pyrolysis reactor is a hot stream (3) that need to be cooled from 550°C to 30°C as shown in Figure 11.

Figure 11 Source and target temperatures (see online version for colours)



ID	Source Temperature	Target Temperature	Heat Load
1	30.00	250.00	1406.7700
2	250.00	550.00	2908.0000
	550	30	3438.79

The combined composite curve shows a minimal pinch temperature of ($\Delta T_{\min}$) = 10°C. The pinch analysis laws suggest that no heat transfer is allowed across the pinch, no external cooling above the pinch and no external heating below the pinch. The red arrow in Figure 12 refers to the heat source Q_c while the blue arrow refers to heat sink Q_H. The hot and cold stream curves suggest that these streams can be used in heat exchanging network (Kemp, 2007).

Figure 12 Combined composite curves for hot and cold streams (see online version for colours)

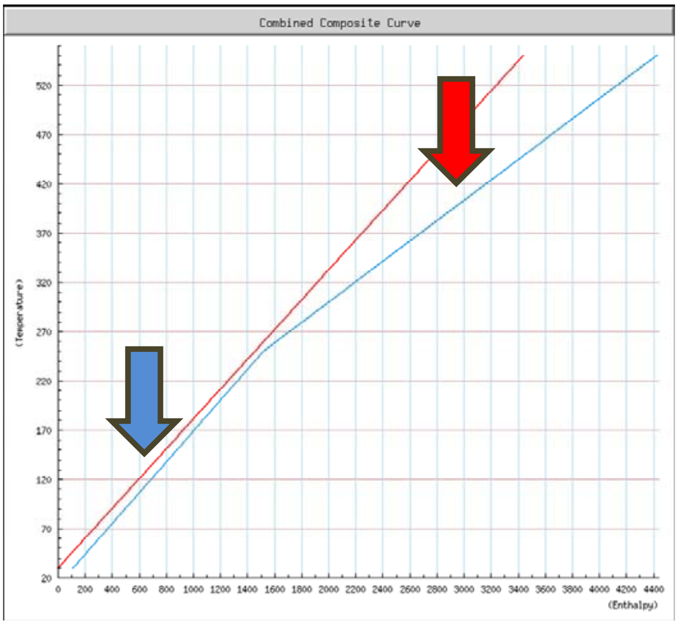
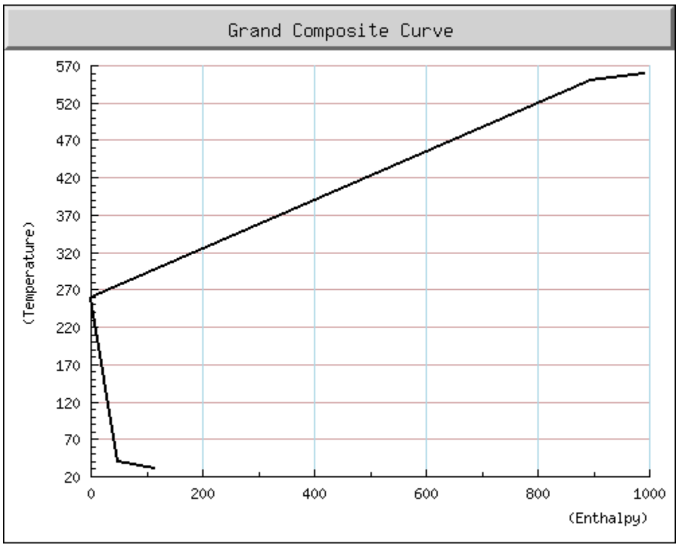


Figure 13 Grand composite curve and pinch temperature (see online version for colours)



Analysing the grand composite curve helps us to identify pockets of heat recovery and decide the appropriate utility for heating and cooling such as LP, MP, HP steam or burning fuels. The pinch temperature is approximately 260°. The curve above the pinch temperature represents the heat sink Q_H while the curve below the pinch temperature represents the heat source Q_C.

6 Conclusions

A 10,000 kg/hr pyrolysis process system has main four process stages which are granulation, preheating, pyrolysis and condensation of gaseous products. The granulation stage consumes around 9.41% of total energy required in form of electric power (0.448 MW). The granulator releases thermoplastic granules with PSD 6–8 mm to the feed preheater tank. The feed preheater converts the thermoplastic mixture to molten plastics at 250°C consuming around 29.53% 1.407 MW. The pyrolysis reactor consumes 61% of heat duty required 2.908 MW for a residence time of 60 minutes. This shows that the pyrolysis reactor consumes the largest power and innovative alternative methods such as plasma arc or renewable energy harvesting techniques can be used to reduce operating cost. The combined composite and grand composite curves are also drawn for the hot and cold streams showing a pinch temperature of the system of 260° and heat sources and sinks that can be used in heat exchanging.

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Nomenclature

BR	batch reactor
HDPE	high density polyethylene
LDPE	low density polyethylene
LP	low pressure steam
PG	petroleum gas
M_w	molecular weight (g/mol)
MP	medium pressure steam
MPW	municipal plastic waste

HP	high pressure steam
PETE	polyethylene terephthalate
PS	polystyrene
PP	Polypropylene
PSD	particle size diameter
W_o	initial mass (g)
X_n	final mass conversion to products
τ	reactor residence time.