

Research Article

Pyrolytic Oil Yield from Waste Plastic in Quezon City, Philippines: Optimization Using Response Surface Methodology

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Abstract. Plastics play an essential role in packaging materials because of their durability to different environmental conditions. With its importance in the community lies the problem with waste disposal. Plastic is a non-biodegradable material, making it a big problem, especially when thrown in dumpsites. In solving the plastic problem, one efficient way to reduce its volume is through thermal processing such as pyrolysis. This study used the pyrolysis method to recover energy from plastic waste. Liquid oil from plastic was comparable to regular fuel used in powering engines. Before the pyrolysis process, a 3k factorial Box-Behnken Design was used in determining the number of experiments to be used. The output oil yield in each pyrolysis runs was optimized in different parameters, such as temperature, residence time, and particle size using response surface methodology to determine the optimum oil yield. Between polyethylene (PE), mixed plastic, and polystyrene (PS), PS produced its highest oil yield of 90 %. In comparison, mixed plastic produced only its highest oil yield of 45 % in 500 °C temperature, 120 min residence time, and 3 cm particle size. The produced quadratic mathematical models in PE, mixed, and PS plastic were significant in which the p-values were less than 0.05. Using mathematical models, the optimum oil yield for PE (467.68 °C, 120 min residence time, 2 cm particle size), mixed (500 °C, 120 min residence time, 2 cm particle size) were 75.39 %, 46.74 %, and 91.38 %, respectively.

Keywords: Optimization, pyrolysis, response surface methodology, RSM, plastic.

Article History: Received: 17th Sept 2021; Revised: 20th Nov 2021; Accepted: 3rd Dec 2021; Available online: 10th Dec 2021 How to Cite This Article: Olalo, J.A. (2022) Pyrolytic Oil Yield from Waste Plastic in Quezon City, Philippines: Optimization Using Response Surface Methodology. *International Journal of Renewable Energy Development*, 11(1), 325-332. https://doi.org/10.14710/ijred.2022.41457

1. Introduction

The overall global plastic production reached about 359 Mt in 2018 (PlasticsEurope, 2019). With the projected world's urban population rise of 85% by 2030 in both developed and developing countries, urbanization and increasing population will significantly increase plastic waste generation (Lettieri *et al.*, 2009).

The recycling process was one of the methods used to reduce the effects of increasing waste plastic problems. However, better separation of this waste plastic was too complicated and costly, making it labor-intensive (Schaefer 1975). Still, reducing the waste plastic process by recycling was somewhat more dependable since plastics came from petroleum.

Sharuddin *et al.*(2016) stated that studies were conducted to discover a more potent process of energy recovery coming from plastic wastes. In connection, most municipal plastic wastes contain either high-density polyethylene (HDPE), low-density polyethylene (LDPE), polystyrene (PS), polyvinyl chloride (PVC), and polypropylene (PP). These plastics types can be pyrolyzed individually to determine their pyrolytic components (Onwudili *et al.*, 2009).

Through the techniques and methods developed from the energy recovery process, pyrolysis signifies the most beneficial method in the direct recovery of energy from waste plastic compared to numerous methods (Oyedun *et al.*, 2014; Idris *et al.*, 2021). This was because the pyrolysis method thermally degrades waste plastics, thereby reducing the production of serious problems in the regular recycling process treatment (Kaminsky *et al.*, 2004).

Municipal solid waste (MSW) treatment was considered and significantly introduced as the most innovative substitute in waste plastic's energy recovery process and converted it to a more sustainable energy resource (Schaefer 1975). Energy obtained from these waste plastics through the pyrolysis method has lower nitrogen oxides emissions and sulfur oxides. So, a reduction of greenhouse gas emissions can be expected from this processing of municipal solid waste through pyrolysis (Saffarzadeh *et al.*, 2006).

In the economic aspect, the output products for the pyrolysis process have a significantly lower cost than the market price. Lower operational cost and maintenance add up to the advantages of the pyrolysis process in the energy recovery methods (Huijbregts *et al.*, 2008; Ayanoglu and Yumrutas, 2016). The environmental benefits of the pyrolysis process were the possibility of generating carbon credits, making the whole process with no liabilities to the environment. Its fitness in the strategic production of a viable and clean waste-to-energy

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conversion process can help reduce the waste plastic problem (Benvenga *et al.*, 2016; Huijbregts *et al.*, 2008; Li *et al.*, 2016). The integrated pyrolysis method also helps achieve maximum environmental benefits with minimal waste production (Inman 2002). More importantly, the pyrolysis of waste plastics can help reduce fossil fuel use (Olalo 2021; Olalo 2022).

Response parameter like temperature dramatically affects the pyrolytic oil yield. At a higher temperature of 600 °C, pyro-oil from tire pyrolysis promotes the production of liquid hydrocarbons at their highest (Al-Salem 2022). PE, mixed waste, and PS completely degrades at temperatures 467 °C (Marcilla et al., 2005), 400 °C (Donaj et al., 2012; Kaminsky et al., 1996; Ahmad et al., 2014), while pyrolysis temperatures for all types of plastics were used up to 500 °C (Sharuddin et al., 2016). Higher residence time can produce higher oil yield at higher temperatures in cracking of PE (Mastral et al. 2001; Raj et al., 2013). Furthermore, increasing the particle sizes in a pyrolysis process will directly affect the completion time (Oyedun et al., 2012). Certain regularity changes in changing particle sizes are attributed to the important effect of particle sizes in the pyrolysis products. Oil yield increased at higher temperatures in the pyrolysis of Jimsar(China) oil shale (Pan et al., 2021).

This study used a response surface methodology to determine how the different response parameters can affect the possible output of pyrolytic oil yields, such as temperature, residence time, and particle size. By using response surface methodology, a mathematical optimization model can be obtained. Polyethylene, mixed waste plastic, and polystyrene waste will be pyrolyzed based on the developed run numbers using the 3^k factorial Box-Behnken design (BBD).

2. Materials and Methods

2.1 Preparation of materials

Payatas dumpsite in Quezon City, Philippines, was the source of waste plastic used in the study. They were sorted by resin numbers for PE (2,4) for plastic bags, mixed plastic (7) for shampoo sachets, and laminated plastic and polystyrene (6) containers. The plastics were cleaned, cut, and air-dried, shown in Figure 1, before performing pyrolysis in the Department of Science and Technology – Industrial Technology Development Institute (DOST-ITDI) facility. The particle sizes for all feedstocks were 2, 3, and 4 cm in space.

2.2 Response parameters

Table 1 shows the response parameters for temperature, residence time, and particle size for the three types of waste plastics used (PE, mixed waste plastic, and PS). A 3^k factorial Box-Behnken Design was used to efficiently estimate the optimum conditions in determining the maximum possible oil yield output in the pyrolysis process (Annadurai et al., 1999).

2.3 Pyrolysis of PE, mixed waste, and PS

A 5 kW cylindrical continuous pyrolyzer was used in the study, as shown in Figure 2a. It has a pre-heating chamber, a gas condenser, and a water scrubber in which the main furnace was equipped with a rotating paddle driven by a gear motor. The heat source was a liquefied petroleum gas (LPG) burner with a return gas in the condenser. In Figure 2b, the pyrolyzer underwent a preheating process with a temperature of 200 °C (Cepeliogullar and Putun, 2013). When the pre-heat temperature was reached, the feedstock (PE, mixed waste, and PS) was individually pyrolyzed in temperatures 300 °C, 400 °C, and 500 °C as response parameters for temperature.

The residence time in the individual pyrolysis of PE, mixed waste, and PS were from 60 min, 90 min, and 120 min. After completing the individual residence time, the output gas in the reactor will go to the condenser (cold water as a medium) for the production of oil condensate or pyrolytic oil. The non-condensable gas (combustible gas) will be used to fuel the reactor together with the LPG. Then the emission will be scrubbed before releasing into the atmosphere.



(a)

(b)

(c)

Fig. 1 Waste plastic was cut for different particle sizes PE (a), mixed plastic (b), and PS (c).



Fig. 2 Pyrolyzer (a) and the pyrolysis process (b)

While for particle sizes in the individual pyrolysis of PE, mixed waste and PS used 2 cm, 3 cm, and 4 cm. Table 1 shows two kilograms of feed weight for each run to determine the oil yield produced in each specific response parameter. In determining the individual oil yield, Eq (1) was used.

$$\% Yield = \frac{weight of produced oil}{weight of feedstock} x 100\%$$
(1)

2.4 Optimization of produced pyrolytic oil yield

Percentage oil yield produced in each pyrolysis of PE, mixed waste, and PS using Table 2 were optimized to develop an optimum condition for the three types of response parameters used. The overall optimization technique used the Minitab 17 software. The output mathematical model follows Eq. 2 based on the 3k factorial Box-Behnken Design.

$$Y = \alpha_0 + \sum_{i=1}^4 \alpha_i X_i + \sum_{i=1}^4 \alpha_{ii} X_i X_i + \sum_{i=1}^4 \sum_{i< j=2}^4 \left(\alpha_{ij} X_i X_j \right)$$
(2)

where X_i is the input variables, which influence the response variable Y, α_0 the offset term, α_i the ith linear coefficient, α_{ii} the quadratic coefficient, and α_{ij} is the ijth interaction coefficient.

3. Results and Discussion

3.1 Produced pyrolytic oil for the different response parameters

Individual pyrolysis of PE, mixed, and PS plastic in response parameters for temperature, residence time, and particle size produced different percentage oil yields concerning the type of plastic used, as shown in Table 2.

Table 1

Response parameters with corresponding run numbers based on BBD for PE, mixed, and PS

Run No.	Temperature (°C)	Residence Time (min)	Particle Size (cm)
1	300	60	3
2	500	60	3
3	300	120	3
4	500	120	3
5	300	90	2
6	500	90	2
7	300	90	4
8	500	90	4
9	400	60	2
10	400	120	2
11	400	60	4
12	400	120	4
13ª	400	90	3
14^{a}	400	90	3
15^{a}	400	90	3

^a The center point was done 3-times for the overall design variance.

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Table 2

Percentage oil yield for a	different type of plast	c pyrolyzed in differen	t response parameters

		Residence Time	Particle Size	% Experimental			% Predicted		
Run	Temperature				Oil Yield			Oil Yield	
No.	(°C)	(min)	(cm)	\mathbf{PE}	Mixed	\mathbf{PS}	\mathbf{PE}	Mixed	PS
					Plastic			Plastic	
1	300	60	3	35	31	43	36.44	29.69	42.33
2	500	60	3	70	40	87	70.23	40.36	86.15
3	300	120	3	45	30	49	44.67	30.50	49.34
4	500	120	3	75	45	90	73.46	47.17	90.16
5	300	90	2	40	29	47	39.05	30.19	47.08
6	500	90	2	72	42	89	72.35	41.86	88.90
7	300	90	4	42	27	46	41.56	27.99	45.59
8	500	90	4	70	44	89	70.85	43.66	88.41
9	400	60	2	65	25	59	64.33	26.13	58.99
10	400	120	2	69	34	66	70.06	33.42	65.00
11	400	60	4	66	28	58	64.83	29.41	58.49
12	400	120	4	70	30	64	70.56	29.74	63.51
13^{a}	400	90	3	67	32	62	67.11	32.10	61.25
14^{a}	400	90	3	68	33	62.5	67.11	32.10	61.25
15^{a}	400	90	3	66.5	30	60	67.11	32.10	61.25

In PE, mixed plastic and PS oil yield results, an increased oil yields occurred in an increased temperature. Increasing oil yields was also observed while increasing the residence time at a constant temperature. This may be because higher residence time produces high oil yields as also observed by Mastral *et al*, (2001) and Raj *et al*, (2013). In an increased residence time, there was an increase in the conversion of the primary product which yields lighter molecular weight hydrocarbons (Ludlow-Palafox and Chase, 2001). But with regards to particle sizes, it has a weak positive (+) and negative (-) correlation with PE, mixed plastic, and PS oil yields. These results may be explained by the regularity changes in changing particle sizes by Pan *et al*, (2021), in which particle sizes have no direct effect on the pyrolysis product.

It was evident that pyrolysis of PS plastic produced the highest oil yield of 90 %, near to the %wt oil yield of 96.4 % with a temperature of 500 °C (Liu et al., 2000) at response parameters value of 500 °C, 120 min and 3 cm for temperature, residence time and particle respectively. Sharuddin et al. (2016) do not recommend the pyrolysis of polystyrene waste plastic to run above 500 °C to optimize liquid oil production. Demirbas (2004) observed that an increase in temperature to 581 °C reduced the liquid production and an increase in the gaseous product. Sharuddin et al. (2016) stated that the operating temperature in a pyrolysis process deeply relies on the product preference. A higher temperature of more than 500 °C was suggested for gaseous and char products, and a lower temperature in the range of 300 - 500 °C for liquid production. In contrast, the lowest oil yield between the three feedstock used was determined in mixed plastic waste, with the highest percentage oil yield of 45 %, which was lower than the produced oil yield of Ghodke (2021).

3.2 Quadratic mathematical model to predict maximum oil yield

The 15 runs for the different response parameters with their output percentage oil yield were analyzed and

produced a quadratic mathematical model for PE Eq. 3, mixed plastic Eq. 4, and PS Eq. 5 using Analysis-of-Variance (ANOVA). It also states the significance of the developed model (Kilic et al., 2014). ANOVA evaluates the effect and interaction of the investigated factors, whether they are significant or not as shown in Table 3. ANOVA results for PE, mixed plastic, and PS showed a significant model with P-value of <0.0001, 0.0021, and <0.0001, respectively. P-values of <0.05 were deemed significant, while higher model F-values indicate the significance of the model. Linear terms of temperature on the three feedstock PE, mixed plastic, and PS had a more significant influence on the oil yield than did the residence time. The interaction term between temperature and residence time in PE and PS had a significant effect on the product oil yield. However, in mixed plastic, the interaction between residence time and particle size had a significant effect on the oil yield, but the value was close enough to the interaction term between temperature and residence time. Quadratic terms for temperature in all feedstock significantly affect the product oil yield.

 $PE \ Oil \ Yield \ (\%) = -208.04 + (1.12 * temp) + (0.204 * residence time) + (4.0 * particle size) - [4.17E - 004 * (temp. residence time)] - [1.0E - 002 * (temp. particle size)] + [0.00 * (residence time. particle size)] - (1.12E - 003 * temp²) + (3.24E - 004 * residence time²) + (0.042 * particle size²)) (3)$

 $\begin{array}{l} \textit{Mixed Plastic Oil Yield (\%) = + 80.58 - (0.45 * temp) + \\ (0.1792 * residence time) + (11.38 * particle size) + [5.0E - \\ 004 * (temp. residence time)] + [1.0E - 002 * \\ (temp. particle size)] - [0.058 * \\ (residence time. particle size)] + (5.542E - 004 * temp^2) - \\ (7.87E - 004 * residence time^2) - (1.71 * particle size^2)) \\ \end{array}$

 $PS \ Oil \ Yield \ (\%) = +57.75 - (0.243 * temp) + (0.242 * residence time) - (3.0 * particle size) - [2.5E - 004 * (temp. residence time)] + [2.5E - 003 * (temp. particle size)] - [8.33E - 003 * (residence time. particle size)] + (5.87E - 004 * temp²) - (1.39E - 004 * residence time²) + (0.375 * particle size²)) (5)$

3.3 Predicted versus actual oil yield

Correlation between predicted and actual values was determined by the correlation coefficient developed in analyzing the effects of the response parameters for temperature, residence time, and particles size on the output percentage oil yield, as shown in Figure 3. The correlation coefficient R² for PE in Figure 3a was 99.57 %. While in Figure 3b for mixed plastic was a 97.28 % correlation coefficient. Finally was PS plastic with the highest correlation coefficient of 99.84 %. The high correlation coefficient of PE, mixed, and PS plastic means that the predicted values were closed to the actual value. These values indicate that the mathematical model developed was consistent with temperature, residence time, and particle size on the output percentage oil yield. When the predicted values were closed to the actual value, as observed by Pinto et al. (2020), the response surface methodology was a good method in predicting the experimental conditions in studying product yields.

Using 2D surface plots, the nature of the response surface and fitness of the model obtained for PE, mixed plastic, and PS were being demonstrated. The 2D contour plots show a classification of a contour shape for various parameters applied. Oil yield increased as residence time and temperature increased (Figure 4a). The highest oil yield occurred between 500 °C and 120 min residence time. The relationship of PS oil yield to residence time and temperature also shows a similar response (Figure 4b). As PS oil yield increased as the residence time and temperature increased. The highest oil yield occurred between 500 °C and 120 min residence time. The relationship of mixed plastic oil yield to residence time and the temperature is shown in Figure 4c. Increased residence time and temperature produced increased mixed plastic oil yield. The 2D contour plot showed different response contour shapes, maybe because of the composition of the mixed plastic. Overall, there was a positive correlation with residence time and temperature concerning mixed plastic oil yield.



Fig. 3 Comparison of the predicted and actual values of oil yield for PE (a), mixed plastic (b), and PS (c).

Factors	Su	m of Squa	ıre		df		Me	ean squar	е		F-value			P-value	
	PE	Mixed	\mathbf{PS}	PE	Mixed	\mathbf{PS}	PE	Mixed	\mathbf{PS}	PE	Mixed	\mathbf{PS}	PE	Mixed	\mathbf{PS}
Model	2501.3	551.9	3806.4	6	6	6	277.9	61.3	422.9	127.3	19.9	352.4	< 0.0001	0.0021	< 0.0001
Tempera-ture	1953.1	364.5	3612.5	1	1	-	1953.1	364.5	3612.5	894.6	118.2	3010.4	< 0.0001	0.0001	< 0.0001
Residence Time	66.12	28.1	60.5	1	1	1	66.1	28.1	60.5	30.3	9.1	50.4	0.00	0.0294	0.0009
Particle Size	0.5	0.1	61	1	1	1	0.5	0.1	2.0	0.2	0.04	1.7	0.65	0.8484	0.2532
Tempera- ture* Rosidence Time	6.25	6	2.3	1	1	1	6.3	9.0	2.3	2.9	2.9	1.9	0.15	0.1482	0.2292
Tempera-ture* Particle Size	4	4	0.3	1	1	1	4.0	4.0	0.3	1.8	1.3	0.2	0.23	0.3063	0.6672
Residence Time* Particle Size	0	12.3	0.3	П	1	1	0.0	12.3	0.3	0.0	3.97	0.2	1.00	0.1028	0.6672
${ m Tempera-ture}^2$	463.9	113.4	127.4	1	1	1	463.9	113.4	127.4	212.5	36.8	106.2	< 0.0001	0.0018	0.0001
${ m Residence}$	0.3	1.9	0.1	1	1	1	0.3	1.9	0.1	0.1	0.6	0.05	0.72	0.4733	0.8351
Particle Size ²	0.01	10.8	0.5	1	1	1	0.01	10.8	0.5	0.0	3.5	0.4	0.96	0.1205	0.5397
Residual	10.9	15.4	9	2	ũ	2	2.2	3.1	1.2						
Lack of fit	9.8	10.8	2.5	က	က	ŝ	3.3	3.6	0.8	5.6	1.5	0.5	0.16	0.4177	0.7310
Pure error	1.2	4.7	3.5	5	2	61	0.6	2.3	1.8						

Table 3 ANOVA for response surface quadratic model of oil yield for PE, mixed and PS

14

14

14

3812.4

567.3

2512.2

Total



Fig. 4 Two-dimensional contour plot of oil yield against residence time and temperature for PE (a), PS (b), and mixed plastic (c).

Table 4Optimum condit	tion for optimum o	il yield			
Dlastie Trme	Temperature	Residence Time	Particle Size	Oil Yield	
Plastic Type	(°C)	(min)	(cm)	%	
PE	467.68	120	2	75.39	
Mixed	500	120	2.75	46.74	
PS	500	120	2	91.38	

3.4 Optimum value

Table 4 presents the optimum percentage pyrolytic oil yield for response parameters of temperature, residence time, and particle size for PS, mixed, and PS plastic. The optimum value for the three feedstock regarding residence time was 120 min, while 460 °C – 500 °C were the optimum temperatures. In comparison, the highest oil yield occurred in the smaller particle sizes between 2 cm and 3 cm. This was comparable to the optimization results of liquid fuel from pyrolysis of waste polyethylene by Pan *et al.* (2021), which produced 83.63 % with a maximum operating temperature of 488 °C. Sharuddin *et al.* (2016) also concluded that to produce liquid oil, the temperatures should be below 500 °C. The study produced an R-squared value of 99 % from its experimental liquid fuel production of 83.50 %.

4. Conclusion

Between PE, mixed, and PS plastic, PS possesses the most significant possible percentage oil yield, and the lowest is mixed plastic. Using ANOVA, the correlation coefficient of the three feedstock indicates that the mathematical model developed was significant in predicting the oil yield based on the three response parameters for temperature, residence time, and particle size. The optimum conditions for temperature for the three feedstock should be as high as 500 °C. The higher residence time of 120 min and a smaller particle size between 2 cm and 3 cm will produce a higher percentage oil yield.

Acknowledgments

It was great gratitude by the author to the Department of Science and Technology – Industrial Technology Development Institute (DOST-ITDI) for lending their pyrolyzer machine.

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