

Sensitivity Analysis of Thermal Degradation of Plastic Waste Using Statistically Assumed Exponential Factors and Activation Energies

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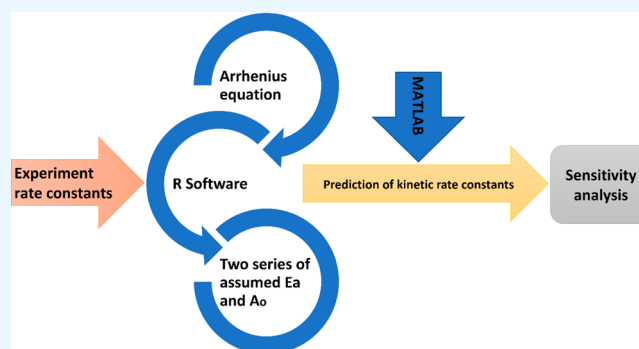
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ABSTRACT: The rise in the production of plastic waste has prompted the exploration of various recovery options instead of landfilling, burning, and other unethical ways of decomposing. The experimentally generated rate constants for the thermal processing of plastic waste do not yield enough liquid fuels and gases for commercial-scale usage. It is imperative to predict kinetic rate constants statistically using an appropriate combination of activation energies (E_a) and frequency factors (A_0) for the optimized thermal valorization of plastic waste. This approach also assists in controlling the selectivity and quantity of the pyrolysis products. A statistical kinetic model was tested to find the best combination of rate constants from different combinations of E_a and A_0 to pyrolyze the high-density polyethylene. Two series of E_a and A_0 were first assumed using R software. These series were then used to predict kinetic rate constants and analyze their sensitivity independently using MATLAB. The rate constants were varied from their originally predicted values during the sensitivity analysis. It was found that the rate constant $k(7)$ dominated the other predicted rate constants where high oil and gas yields were concerned. The gas yield increased from lower to higher extreme positions in the range of 60%–74% with the first series and from 65% to 81% with the second series. The maximum oil content was found around 74% and 65% with the first series and second series, respectively.



1. INTRODUCTION

Plastics have surpassed natural materials as home products due to their diversity, ease of use, and low cost. Roughly, 350–400 million tons of nonbiodegradable plastic are generated each year, and 5–13 million tons of leftover plastic are dumped into the ocean.¹ According to one assessment of the Great Pacific Rubbish Patch, 80 000 tons of plastic garbage have been detected in the Pacific latitudes.² Plastic bags, bottles, containers, and packaging are all made from polyethylene and polypropylene. The environment is impacted by the use of 500 billion to 1 trillion bags globally. The widespread use of high-density polyethylene (HDPE) and low-density polyethylene (LDPE) poses major environmental concerns to terrestrial and marine ecosystems. Additionally, a large number of species from various habitats are on the verge of going extinct due to plastic-based pollution. The use of HDPE in boosting agricultural production has also resulted in a severe societal dilemma. Mulching waste vinyl impedes the soil animals like earthworms as well as the flow of essential elements like air, moisture, and nutrients, as reported by the Chinese Academy of Agricultural Sciences in 2014.³ These factors are causing physiological issues with plant growth,

including germination of seed, root growth, and agricultural output. Therefore, managing polyethylene and other plastic wastes, as well as creating effective technology for plastic breakdown and recycling, is a current necessity.^{4,5}

All recycling activities are classified based on the byproducts and treatment method. Pyrolysis is the use of heat to convert polymers into flammable liquids and gases.^{6,7} Plastic waste is cut into tiny pieces before being heated endothermically.⁸ Depending on the heating method, pyrolysis might be traditional heating or microwave heating of the plastic.⁹ It is critical to understand that decomposition necessitates a great deal of chemistry. To forecast the physicochemical traits of the pyrolysis products, systematic experimental and computational testing is required. A substantial study has been conducted to investigate the degradation of polyethylene (PE) utilizing

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Table 1. Rate Constants Predicted Using Two Series of E_a and A_o

first assumed series			second assumed series		
E_a (J/mol)	A_o (L-mol/s)	predicted rate constant (k_A)	E_a (J/mol)	A_o (L-mol/s)	k_B
80625.019	33.615	$k_{-1} = 0.1086754$	84686.595	33.608	$k_{-1} = 0.08139305$
80625.019	32.664	$k_{-2} = 0.1056029$	81443.176	32.469	$k_{-2} = 0.09903624$
124686.886	42.988	$k_{-3} = 0.006052518$	120686.723	41.349	$k_{-3} = 0.007737653$
122916.918	42.6119	$k_{-4} = 0.006804437$	109352.352	39.533	$k_{-5} = 0.01656573$
113732.624	41.088	$k_{-5} = 0.01260851$	106092.446	40.026	$k_{-6} = 0.02114894$
109446.245	41.256	$k_{-6} = 0.01717254$	111473.947	43.572	$k_{-7} = 0.01570102$
113878.971	44.5344	$k_{-7} = 0.01352471$	82618.8433	31.383	$k_{-8} = 0.08804619$
85148.665	32.268	$k_{-8} = 0.07562118$	130076.545	49.308	$k_{-9} = 0.004731786$
133160.256	50.432	$k_{-9} = 0.003886529$	109352.352	39.533	$k_{-5} = 0.01656573$

physicochemical or microbiological techniques or a combination of the two.^{10,11} Thermal, UV, or a mixture of the two is used to shorten polymer chains and produce oxidized groups, including carbonyl, hydroxyl, and carboxyl, on the polymer surface. These treatments alter the original polymer's crystallinity and surface shape, facilitating polymer biodegradation. Polyethylene films have been shown to fragment due to nitric acid oxidation, which is followed by microbial deterioration. Microorganisms or microbial colonies biodegrade PE by altering it and using it as a source of energy. The polymer's physical and chemical characteristics change as a result, including weight loss, structural degradation, and organic matter carbon fixation.¹² On the other hand, PE's high level of hydrophobicity, low specific surface area, and flat surface topography prevent bacteria that break down polymers from forming a biofilm on it.¹³ Additionally, it is yet unknown which specific biochemical processes and enzymes are required for the breakdown of PE.¹⁴

For maximum production and excellent product selectivity, statistical optimization of the process parameters is necessary by applying suitable statistical models. In order to evaluate the effects of modifying operational parameters on process effectiveness, it is critical to create a model that simulates the pyrolysis of polymers. The statistical sensitivity of calculated rate constants for plastic pyrolysis has not been investigated in the published literature. It is possible to examine the efficacy and role of individual rate constants in a pyrolysis process by performing sensitivity analysis. In this study, we conducted a sensitivity analysis by varying values of rate constants from the predicted ones to a level where maximal oil yield is possible. The sensitivity analysis of rate constants has not yet been the subject of past investigations. The A_o and E_a factors can be critical in such analysis to maximize the yield of valuable products. A suitable mix of E_a and A_o is the foundation for forecasting the rate constants that are used to promote oil yield on a commercial basis. Each kinetic rate constant efficiency is solely assessed using sensitivity analysis. This research will show a number of ways for enhancing yields and understanding how temperature affects the kinetic rate constant. What effects do the frequency variables have on the kinetic rate reactions? This study will also provide recommendations for choosing an optimal mix of E_a and A_o to provide an efficient combination of kinetic rate constants. It will be possible to identify which combination of assumed parameters is ideal for increasing gas output while preserving industrial oil production using this model in the R program. Despite the importance of kinetic rate constants in yield efficiency, no research has been performed to examine how they work.

2. METHODOLOGY

Equation 1 is used to suppose two sets of E_a and A_o for predicting the rate constants. Equation 2 is applied to the supposed series factors a_m and b_m in the R software, and rate constants $k(1)$ – $k(9)$ are predicted.¹⁵ In these equations, k is the rate constant, A_o is the frequency factor, E_a is the activation energy, and $(\frac{1}{T_2} - \frac{1}{T_1})$ is the absolute temperature. The rate constant number (1–9) is denoted with M , and supposed values of E_a and A_o are denoted with m . Table 1 lists the values of two series of E_a , A_o , and anticipated rate constants.

$$k = A_o \times e^{-E_a/RT} \quad (1)$$

$$k_{-M} = a_m \times \exp\left(-b_m \times \left(\frac{1}{T_2} - \frac{1}{T_1}\right)\right) \quad (2)$$

The rate constants in Table 1 are then used in MATLAB to analyze the sensitivity of the predicted rate constants by employing eqs 3 and 4. Using sensitivity analysis, we can examine the influence of individual rate constants on the products by varying their values from the predicted values up to a certain level wherever optimum product yield is possible. In this model, eq 3 is utilized in MATLAB to investigate the efficacy of rate constants, while eq 4 indicates a maximum and minimum change in each predicted rate constant. In these equations, t stands for time, B indicates % yield, k refers to the reaction constant, D0 stands for initial conditions, and P shows a change in the original value. Figure 1 gives a graphical representation of the conducted work. The mass of HDPE was taken as 150 kg, the pyrolysis time was varied from 1 to 240 min, and the response of reaction after 60 min of processing time was analyzed. The increasing and decreasing actual values of the projected rate constants imply that each reaction has two extreme positions: the lower extreme and the higher extreme.

$$[t_n, B] = \text{ode23s} (@(t_n, x) \text{ test reaction}(t_n, x, k_n), \text{ time, D0}) \quad (3)$$

$$k_{-P_max} = k(P) + k(P) \quad (4)$$

3. MODEL FRAMEWORK

The proposed equations were solved using the approach described in the literature.¹⁶ Figure 2 depicts a hypothetical workflow for this analysis.¹⁷ It reveals strong dependence of heavy wax (HW), light wax (LW), oil, and gas on $k(1)$, $k(2)$, and $k(3)$ reaction constants, and $k(4)$, respectively. These products are commonly obtained during the pyrolysis of plastic

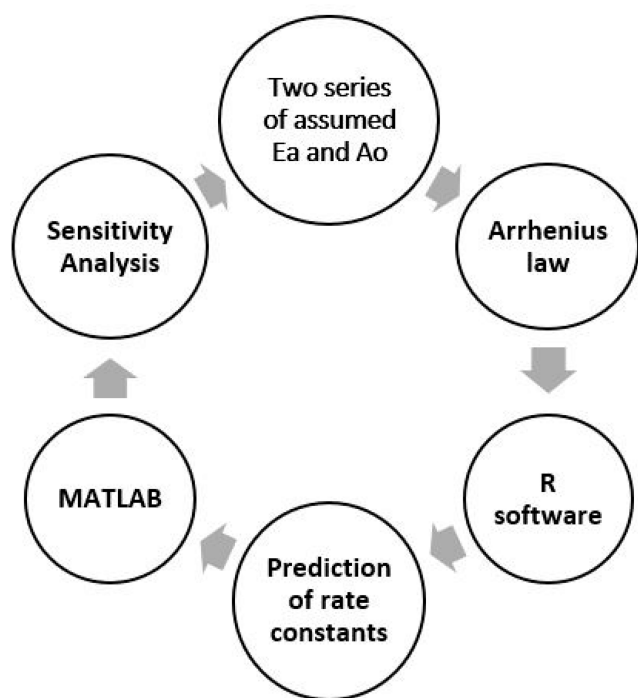


Figure 1. Flowchart of the methodology of the conducted work.

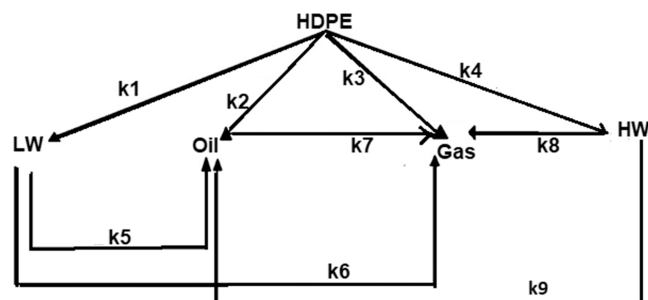


Figure 2. Hypothetical workflow of the thermal pyrolysis of plastic.

under optimum conditions. Free radicals cause the oxidation reactions that change LW into oil using the rate constant $k(5)$, LW into gas using the rate constant $k(6)$, HW into gas using the rate constant $k(8)$, and HW into oil using the rate constant $k(9)$. Using the $k(7)$ reaction constant, some of the oil was instantly transformed into gas. The sensitivity of each rate constant to the yield is checked in a small temperature window of 370–380 °C. The thermal decomposition of PDPE could release gas, oil, and carbon black. Before reaching a steady state, HDPE is broken down into various organic species, primarily LW and HW.¹⁸ The most often used waxes nowadays

are kerosene, paraffin, and aromatics. The high temperature may cause these waxes to break down into smaller molecules, which could eventually result in the formation of carbon black. This material is extremely stable; therefore, it will not undergo further chemical changes.¹⁹ The other reaction parameters, including E_a , A_o , temperature, and catalysts, might also alter the reaction rate and the nature of the products.

4. RESULTS AND DISCUSSION

Table 2 shows initial, lower extreme, and higher extreme values of $k(8)$ and $k(9)$ constants and the corresponding product yield. For the supposed series of E_a and A_o , these rate constants were responsible for the transformation of HW into gas and oil, respectively. Figure 3 shows that for both assumed series, $k(8)$ and $k(9)$ rate constants ensure a constant flow of oil, gas, HW, and LW at a constant temperature. This phenomenon can be influenced by temperature and incomplete conversion of HDPE into HW. As a result, the oil and gas yields remain unchanged after 60 min of response time. For an efficient chemical rate, the reactant molecules must have enough kinetic energy to have collisions among themselves to initiate a proper reaction.¹⁶

The rate responses $k(1)$ and $k(2)$ in both assumed series play a dominant role in the conversion of HDPE into LW and oil, respectively. Table 3 shows the initial, lower, and higher extreme values and their effect on the product yield. Figures 4a and 4b indicate that $k(1)$ behaved a way opposite to that of the $k(2)$ rate constant. At the initial value of $k(1)$, the HW was 10% and 8%, and the gas and oil yields were 32% and 58% for the first assumed series and the second assumed series, respectively. No LW was observed in the case of the first assumed series. However, HW was predicted around 3–12% and oil around 55–59%. The gas yield, on the other hand, decreased from 41% to 28% for series 1. In the second assumed series, HW, oil, and gas remained in the range of 1–7%, 36–27%, and 59–64%, respectively. When the $k(1)$ rate constant is evaluated, one of the constraints of this reaction is the rising amount of HW. The amount of HW reduced significantly with the $k(2)$ rate constant, from 16% to 5% and 10% to 3% for the first series and second series, respectively.

According to the lower extreme and higher extreme kinetic rate constants, when the HW dropped, the oil yield climbed from 24% to 37% for the first series and second series, respectively. The gas yield in both assumed series remained nearly identical, as shown in Figure 4b. Since minor differences in E_a and A_o in the assumed series did not affect the reaction rate, these rate constants behaved similarly. The moisture content of the feedstock can only affect gas yield when the temperature is kept lower as in this study. The higher the moisture content, the more gas is produced.²⁰ The wax formation dominates the other products in the first series. In

Table 2. Different Values of $k(8)$ and $k(9)$ Kinetic Rate Constants and Their Effect on the Pyrolysis Products

rate constant	initial value	lower extreme	higher extreme	HDPE	yield (%)				
					oil	gas	HW	LW	
First Assumed Series									
$k(8)$	0.07562118	0	0.15	0	58	32	10	0	
$k(9)$	0.003886529	0	0.0074	0	59	32	7	0	
Second Assumed Series									
$k(8)$	0.08804619	0	0.17	0	61	31	10	0	
$k(9)$	0.004731786	0	0.0097	0	62	30	7	0	

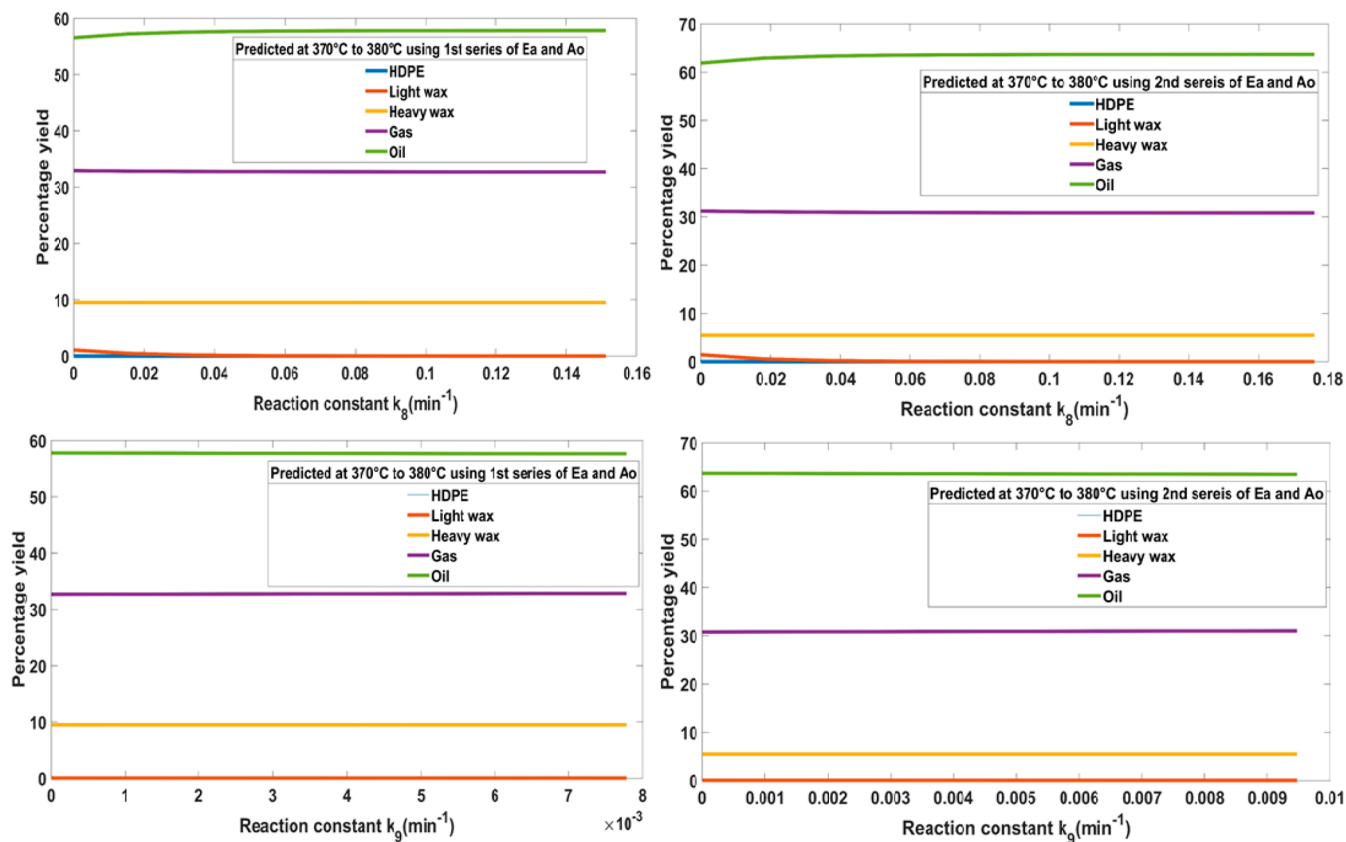


Figure 3. Behavior of $k(8)$ and $k(9)$ kinetic rate constants.

Table 3. Different Values of $k(1)$ and $k(2)$ Kinetic Rate Constants and Their Effect on the Pyrolysis Products

rate constant		oil	gas	HW	LW	
First Series						
$k(1)$	initial	0.108	57	30	11	0
	lower extreme	0	55	41	3	0
	higher extreme	0.217	59	28	12	0
$k(2)$	initial	0.106	57	35	12	0
	lower extreme	0	59	24	5	0
	higher extreme	0.211	56	37	16	0
Second Series						
$k(1)$	initial	0.111	64	30	5	0
	lower extreme	0	63	36	1	0
	higher extreme	0.162	62	27	7	0
$k(2)$	initial	0.133	63	30	5	0
	lower extreme	0	65	23	10	0
	higher extreme	0.198	62	34	3	0

the second series, the wax formation witnessed a decreasing trend while gas and oil formation was more pronounced.²¹

The used values of $k(3)$ and $k(4)$ kinetic rate constants and the corresponding product yields are reported in Table 4. Figure 5 confirms the complete absence of LW in the product while HW was predicted around 9–10% using the first assumed series for both kinetic rate constants. The oil production with $k(3)$ and $k(4)$ constants was almost the same for both assumed series. The oil production ranged from 56% to 65% with a shift in rate constants from lower to higher values.²¹ The gas yield remained at 30–34% with the same rate constants. According to Figure 5, the oil yield increased with processing time for both series due to a decrease in gas and

Table 4. Different Values of $k(3)$ and $k(4)$ Kinetic Rate Constants and Their Effect on the Pyrolysis Products

rate constant		oil	gas	HW	LW	
First Series						
$k(3)$	initial	0.006	57	34	10	0
	lower extreme	0	56	33	10	0
	higher extreme	0.012	59	31	0	0
$k(4)$	initial	0.007	57	32	9	0
	lower extreme	0	56	33	10	0
	higher extreme	0.013	59	31	9	0
Second Series						
$k(3)$	initial	0.008	63	30	7	0
	lower extreme	0	62	31	7	0
	higher extreme	0.015	65	29	7	0
$k(4)$	initial	0.009	63	30	7	0
	lower extreme	0	62	31	7	0
	higher extreme	0.019	65	29	7	0

HW yield.²² The gas and oil yield trend was nearly identical for both the reaction rates and the assumed series.

Table 5 provides information about the initial, lower, and higher extreme values of $k(5)$ and $k(6)$ kinetic rate constants and the product yields obtained using these rate constants. As shown in Figures 6a–6d, the rate constants $k(5)$ and $k(6)$ were responsible for converting LW into oil and gas at a fixed temperature. The maximum oil yield, obtained with lower to higher extreme values of $k(5)$, ranged from 56% to 58% with the first series and 63% to 62% with the second series (Figures 6a and 6c). In the first series, the oil yield slightly increased while a slight decrease was observed in the second series. Figures 6b and 6d demonstrate that the gas yield increased

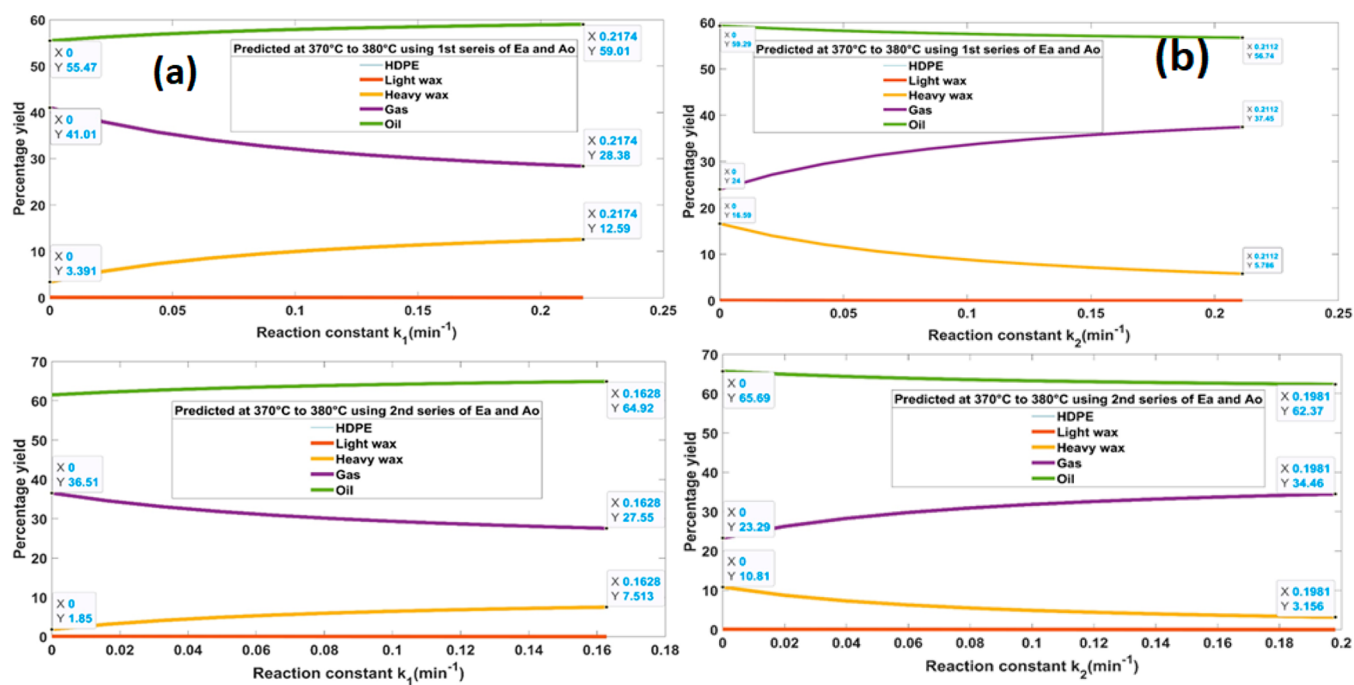


Figure 4. (a) Efficiency of the $k(1)$ kinetic rate constant and (b) the efficiency of the $k(2)$ kinetic rate constant.

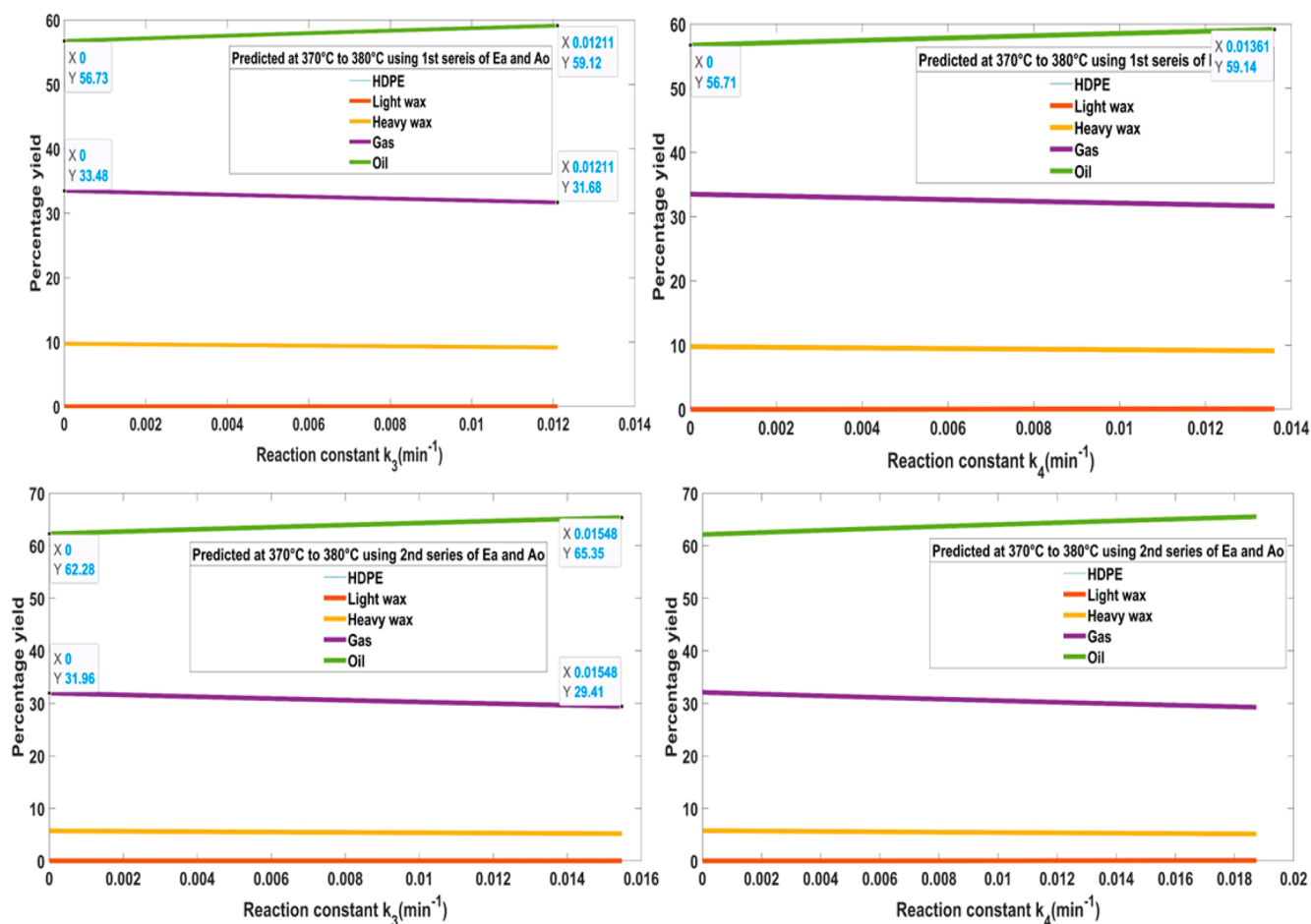


Figure 5. Efficiency of $k(3)$ and $k(4)$ kinetic rate constants.

with both assumed series, which was confirmed by assessing the lower and higher extreme values of the kinetic rate

constants. This increase in gas yield is attributed to a decrease in HW with the processing time.²³ The lower E_a in the second

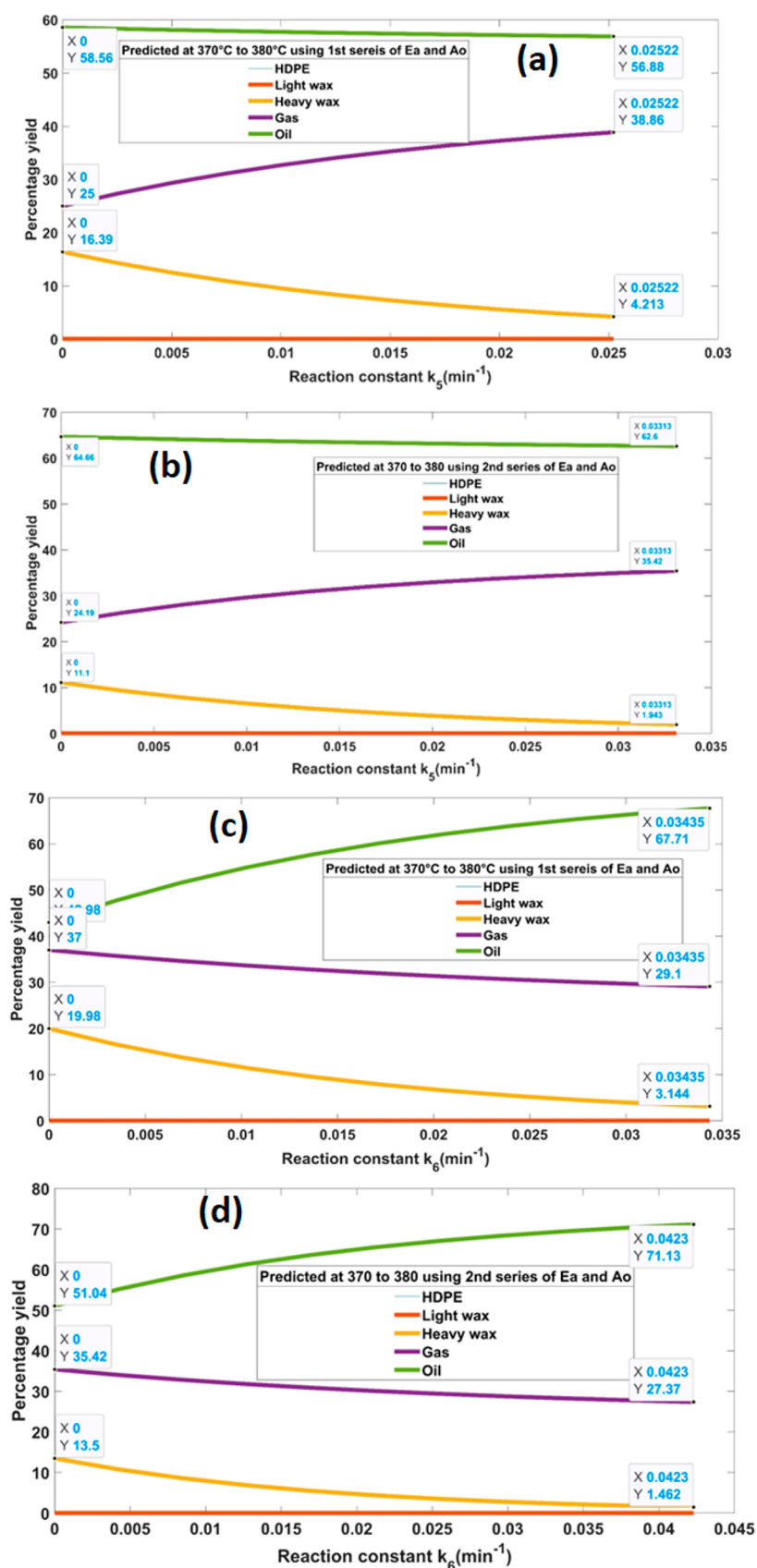


Figure 6. (a) Efficiency of the $k(5)$ rate constant predicted using the first series, (b) efficiency of the $k(5)$ rate constant predicted using the second series, (c) efficiency of the $k(6)$ rate constant predicted using the first series, and (d) efficiency of the $k(6)$ rate constant predicted using the second assumed series.

assumed series compared to the first series increases the reaction rate and consequently the gas yield. The decreasing byproducts result in an increase in primary products at constant temperatures.^{24,25}

Table 5. Different Values of $k(5)$ and $k(6)$ Kinetic Rate Constants and Their Effect on the Pyrolysis Products

rate constant		oil	gas	HW	LW	
First Series						
$k(5)$	initial	0.012	57	32	2	0
	lower extreme	0	58	25	16	0
	higher extreme	0.025	56	38	4	0
$k(6)$	initial	0.017	55	28	19	0
	lower extreme	0	40	37	5	0
	higher extreme	0.034	67	29	3	0
Second Series						
$k(5)$	initial	0.016	63	30	2	0
	lower extreme	0	64	24	11	0
	higher extreme	0.331	62	35	1	0
$k(6)$	initial	0.021	65	29	13	0
	lower extreme	0	51	35	2	0
	higher extreme	0.042	71	27	1	0

Table 6 summarizes the initial, lower, and higher extreme values of the $k(7)$ kinetic rate constant and the corresponding product yield. The LW was completely decomposed for both

Table 6. Different Values of the $k(7)$ Kinetic Rate Constant and Its Effect on the Pyrolysis Products

rate constant		oil	gas	HW	LW	
First Series						
$k(7)$	initial	0.013	60	55	10	0
	lower extreme	0	35	28	10	0
	higher extreme	0.027	74	15	10	0
Second Series						
$k(7)$	initial	0.015	65	56	5	0
	lower extreme	0	37	28	5	0
	higher extreme	81	13	5	0	

assumed series, whereas the HW was in constant flow at 10% and 5% for the first series and second series, respectively. Interestingly, the oil yield increased drastically with a shift in the $k(7)$ value from the lower to the higher extreme. As shown in Figure 7, the oil yield increased from 35% to 74% and 37% to 81% for the first series and second series, respectively. The percentage gas yield decreased from 55% to 15% and from 56% to 13% with the first and second assumed series, respectively. It is also revealed that the oil yield can be increased by adjusting the initial value of $k(7)$. The HW formation decreases with an increase in oil yield. Other kinetic rate constants did not show the same authority on the pyrolysis reaction as the $k(7)$ rate constant, indicating the high commercial significance of this rate constant.

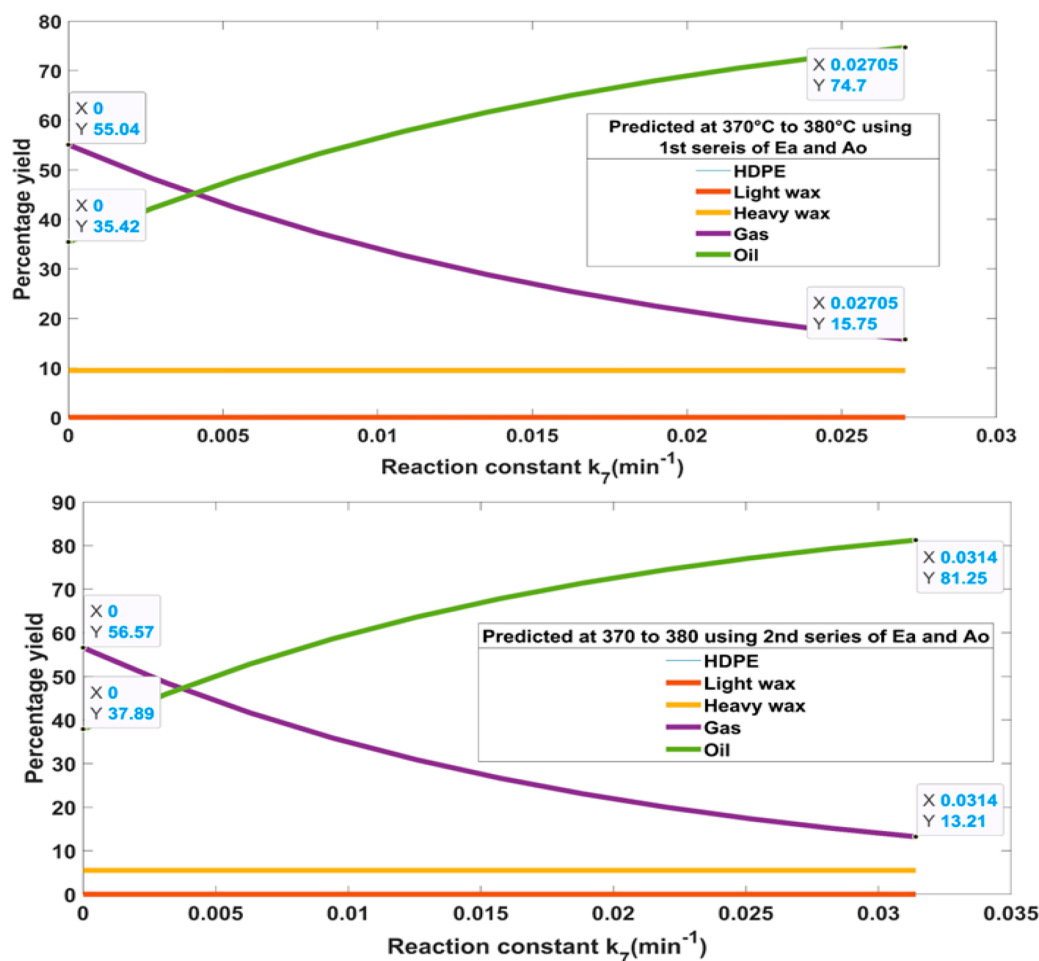


Figure 7. Efficiency of the $k(7)$ kinetic rate constant.

5. CONCLUSIONS

This work is focused on extracting experimental rate constants from pyrolysis experiments that do not produce enough liquid fuels and gases for commercial application. Using information from experimental rate constants, we forecasted statistical rate constants and analyzed their sensitivity in the thermal pyrolysis of plastic waste. Only sensitivity analysis of kinetic rate constants can determine their efficiency during a pyrolysis reaction. This study advances our understanding of how temperature affects the kinetic rate constant and what effect frequency factor changes have on the rate of chemical reactions. Two sets of E_a and A_0 at 370–380 °C in the R software were assumed with the Arrhenius equation to predict rate constants and analyze the sensitivity of each kinetic rate constant. The statistically predicted $k(7)$ rate constant outperformed the other rate constants for high oil and gas yield. Using this rate constant, the gas amount decreased from 55% to 15% and from 56% to 13% with first series and second series, respectively. It is also revealed that the oil yield can be increased by adjusting the initial value of $k(7)$. These results confirm the high significance of the conducted analysis for commercial-scale pyrolysis of plastic waste.

■ ASSOCIATED CONTENT

Data Availability Statement

The reported data is available from the corresponding authors upon valid request.

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Notes

The authors declare no competing financial interest.

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