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A STUDY ON THE KINETICS AND THERMAL DECOMPOSITION BEHAVIOR DURING THE NON-CATALYTIC CO-PYROLYSIS OF PINEAPPLE CROWN WASTE (PCW) AND POLYETHYLENE (PE)

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ABSTRACT

Unlike plastic, biomass can be converted into high-quality alternative fuel. This study investigated the co-pyrolysis of pineapple crown wastes (PCW) and poly-ethylene plastic (PE). The synergy between these two feedstocks was evaluated using thermogravimetric (TGA) and derivative thermogravimetry (DTG) analyses. The Coats-Redfern method was employed to determine the activation energy during synergy analysis. The results indicated that the activation energy ranged from 17 to 200 KJ/mol, depending on the composition of the co-pyrolysis mixture. The thermal degradation occurred in two stages: PCW degraded at temperatures between 170 and 400°C, while PE degraded at temperatures between 400 and 550°C. The co-pyrolysis of PCW and PE shows potential as an alternative fuel source and offers a solution to community waste problems.

Keywords: pineapple crown waste, polyethylen, TGA, DTG, co-pyrolysis.

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INTRODUCTION

Various researchers worldwide have investigated the use of different types of waste biomass for biofuel production, driven largely by the depletion of petroleum reserves. However, these efforts have often resulted in low fuel yields and suboptimal quality [1], [2]. One way to enhance the quality of bio-oil is through fast pyrolysis, a thermal degradation process conducted at high heating rates [3]. Co-pyrolysis, which involves combining biomass with plastic waste, has been employed to improve both the yield and quality of bio-oil. The efficiency of this process is significantly influenced by parameters such as temperature and the proportion of each feedstock component [4].

Operating the co-pyrolysis process within a temperature range of 400-600 °C has been shown to increase oil yield and reveal potential synergistic interactions between different feedstock types [5], [6], [7]. While much of the existing research focuses on oil yield percentages, fewer studies delve into the synergistic behavior of the mixed feedstocks. Some investigations use thermogravimetric analysis (TGA) to quantify the effects of synergy on oil yield during co-pyrolysis [8]. Synergistic effects are also explored through kinetic modeling, where the activation energy is commonly estimated assuming a first-order reaction based on the Arrhenius equation [9], [10].

The present study aims to evaluate the impact of varying temperatures on the synergistic interaction between pineapple crown waste and polyethylene plastic. The Coats-Redfern method is employed to analyze thermal degradation behavior and determine the extent of synergy, a method previously applied to biomass and coal blend studies [11].

MATERIALS AND METHODS

Pineapple Crown Waste and Polyethylene Feedstock

Pineapple crown waste (PCW) and waste polyethylene (PE) were sourced from various local waste disposal facilities. The collected materials were thoroughly cleaned and subsequently air-dried. For thermogravimetric analysis (TGA), both PCW and PE were finely chopped to an approximate particle size of 1 mm. After preparation, the materials were accurately weighed using an analytical balance. Blends of PCW and PE were then formulated in varying mass ratios (0%, 20%, 40%, 60%, 80%, and 100% PCW). Each mixture was combined in vials and homogenized using a vortex mixer.

TGA Different Mixtures of Pineapple Crown Waste and Polyethylene Plastic

The cleaned and air-dried feed materials were finely ground to approximately 1 mm particle size. Using an analytical balance, blends of pineapple crown waste (PCW) and polyethylene (PE) were prepared in varying mass ratios (0%, 20%, 40%, 60%, 80%, and 100% PCW). Each mixture was placed into labeled vials and homogenized thoroughly using a vortex mixer. The thermal decomposition behavior of the individual feedstocks (PCW and PE) and their blends was evaluated using a PerkinElmer thermogravimetric analyzer. The analysis was conducted under a constant nitrogen purge flow of 20 mL/min, with a controlled heating rate of 10 °C/min over a temperature range of 30 to 900 °C. Both

thermogravimetric (TGA) and derivative thermogravimetric (DTG) data were obtained for each sample. The DTG values were derived as outlined in Equation-1.

$$\frac{dm}{dt} = -\frac{1}{m_o}\left(\frac{dm_t}{dt}\right) \quad (1)$$

The thermal decomposition plots were prepared using OriginLab software.

Analysis of the Response Variable

In this study, the rate synergy (Ψ) is defined as the deviation between the experimentally measured weight loss rate of the PCW-PE mixture (Rmixture) and the theoretical sum of the individual weight loss rates of PCW (RPCW) and PE (RPE), each weighted by their respective mass fractions (xPCW and xPE) in the blend. This relationship is expressed in Equation-2 [11].

$$\Psi = R_{mixture} - (x_{PCW}R_{PCW}) + (x_{PE}R_{PE}) \quad (2)$$

The values of RPCW and RPE were obtained from the corresponding DTG (derivative thermogravimetric) curves. Rate synergy values (Ψ) were calculated over the full temperature range of each TGA run, starting from 30 °C and ending at 900 °C, for the different blend compositions. Subsequently, Ψ–temperature profiles were generated to evaluate the synergistic behavior across the various PCW-PE mixtures.

Using Values of Activation Energy

One approach to synergy analysis involves evaluating the activation energy (E) associated with the thermal degradation process. In this study, activation energy was determined for the different stages of decomposition using first-order reaction kinetics, commonly referred to as first-order chemical reaction models [12]. The calculations were performed using the models presented in Equations-3 and Equation -4.

$$f(\alpha) = 1-\alpha \quad (3)$$

$$g(\alpha) = -\ln(1-\alpha) \quad (4)$$

In previous studies, pyrolysis and co-pyrolysis were both assumed to involve first order reactions. The kinetic computation in Equation-5 was all based on the equation of Arrhenius [13].

$$\frac{d\alpha}{dt} = Ae^{\left(-\frac{E}{RT}\right)}(1-\alpha) \quad (5)$$

where A is a pre-exponential factor (min-1); E is activation energy (J/mol); R is the ideal gas constant (8.314 J mol-1K-1); T is the absolute temperature (K); t is time; and α is the weight loss fraction or pyrolysis or co-pyrolysis conversion. The latter is defined in Equation-6, where m0 is the initial mass of the mixture, mt is the instantaneous mass (at any time t), and mf is the final mass, i.e., residual mass.

$$\alpha = \frac{m_t - m_o}{m_f - m_o} \quad (6)$$

For a constant heating rate, β = dT/dt, rearranging Equation-3 and integration gives as shown in Equation-7:

$$\ln\left[\frac{-\ln(1-\alpha)}{T^2}\right] = \ln\left[\frac{AR}{\beta E}\left(1-\frac{2RT}{E}\right)\right] - \frac{E}{RT} \quad (7)$$

The first term on the right-hand side of Equation-7, ln[AR/βE(1–2RT/E), is generally considered approximately constant across a broad range of activation energies (E) and within the typical temperature range for co-pyrolysis processes [12]. As a result, plotting the left-hand side of the equation, ln[–ln(1–α)/T2] against 1/T yields a linear relationship. The slope of this line, obtained through linear regression, corresponds to –E/R, allowing for the determination of the activation energy (E). Additionally, the y-intercept, or the value at the median mass (m0+mf)/2, can be used to estimate the pre-exponential factor (A).

Using this method, $\ln[-\ln(1-\alpha)/T^2]$ was plotted against 1/T for each TGA run. The slope obtained through linear regression was then used to calculate the corresponding activation energy value.

For the alternative kinetic models presented in Table-1, a similar approach was applied based on the Coats-Redfern method described in Equation-7. However, in these cases, the functions g(α) and f(α), which represent specific reaction mechanisms, were used according to the respective model. These functions are provided in the two rightmost columns of Table-1. By plotting $\ln[g(\alpha)/T^2]$ versus 1/T and performing linear regression, the activation energy (E) for each model was calculated. The model yielding a regression coefficient (R^2) closest to 1 was selected as the best fit for the experimental data.

Finally, the activation energy values (Eblend) for the various PCW-PE blends were compared with theoretical values calculated from the weighted sum of the activation energies of pure PCW and pure PE, as shown in Equation-8, using their respective mass fractions.

$$E_{blend} = (x_{PCW}R_{PCW}) + (x_{PE}R_{PE}) \quad (8)$$

Kinetic Model

Several of the kinetic models listed in Table-1 were applied to the experimental data by fitting the fractional conversion (α) versus temperature (T) using the Coats-Redfern integral method. This approach involves linear regression of the transformed model equations. The thermo-kinetic parameters-activation energy (E) and pre-exponential factor (A)-were determined from the slope and y-intercept of the resulting linear plots, respectively. The quality of fit for each model was assessed based on

the coefficient of determination (R^2), indicating how well the model describes the experimental data.

Table-1. Reaction mechanisms, model names, and their g(α).

Model Names	Code	g(α)
Chemical reaction order 1 (Mampel)	R1	$-\ln(1-\alpha)$
Chemical reaction order 1.5	R1.5	$2[(1-\alpha)^{-3/2}-1]$
Chemical reaction order 2	R2	$(1-\alpha)^{-1}-1$
One-dimensional diffusion	D1	α^2
Two-dimensional diffusion	D2	$\alpha+(1-\alpha)\ln(1-\alpha)$
Jander (three-dimensional diffusion)	D3	$[1-(1-\alpha)^{1/3}]^2$
Ginstling and Brounshtein (three-dimensional diffusion)	D4	$1-(2/3)\alpha-(1-\alpha)^{2/3}$

RESULTS AND DISCUSSIONS

TGA of PCW, PE, and Percentage Compositions

The thermogravimetric (TGA) and derivative thermogravimetric (DTG) profiles are illustrated in Figure-1 for 100% pineapple crown waste (PCW) and in Figure-2 for the various PCW-PE blend ratios. The initial mass loss observed below 150 °C is attributed to the evaporation of moisture and low-boiling-point volatiles present in the PCW. As the proportion of PCW in the blends increased, this early-stage mass loss became more pronounced.

The thermal degradation of pure PCW occurred primarily within the 170-550 °C range. During this phase, the DTG curves exhibited two overlapping peaks: the first peak, occurring between approximately 170 °C and 300 °C, is likely associated with the decomposition of hemicellulose, which typically degrades within 200-260 °C [14] or 220-315 °C [15]. The second peak, observed between 300 °C and 550 °C, corresponds to the degradation of cellulose, commonly reported to occur within the temperature ranges of 320-380 °C [16] or 315-400 °C [15].

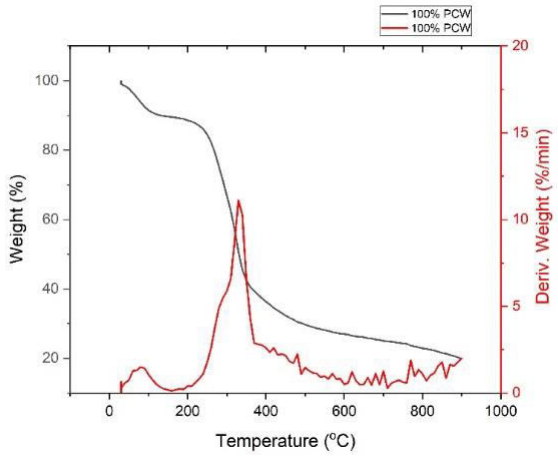
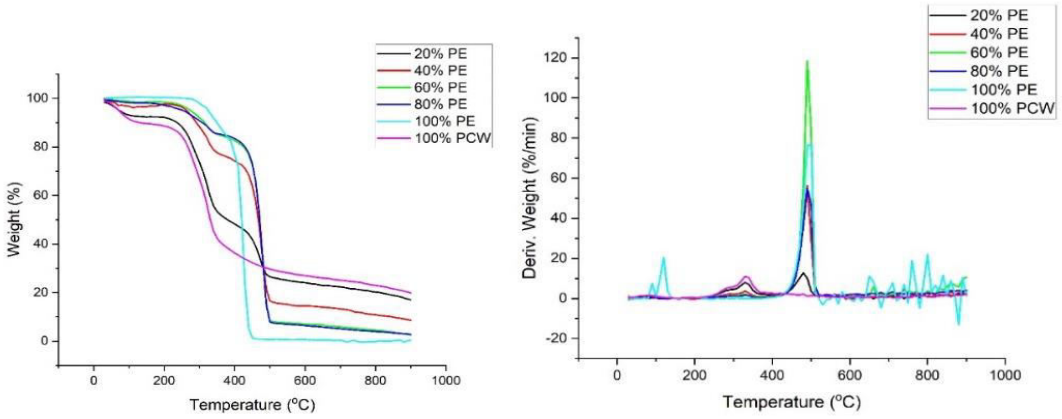


Figure-1. Combined TGA-DTG plots for 100%PCW.



(a) (b)

Figure-2. (a) The TGA curves for pineapple crown waste (PCW) and various PCW-PE blends, (b) The DTG curves for polyethylene (PE) and various PCW-PE blends.

The degradation temperature of cellulose is higher than that of hemicellulose due to its more rigid structure, which results from its highly crystalline polysaccharide arrangement. This structural difference explains why the lower area under the first DTG peak (250-300 °C) compared to the second peak (300-400 °C) indicates that pineapple crown waste contains a smaller proportion of hemicellulose (11-17% [17], 23% [18]) than cellulose (18-21% [17], 33-36% [18]).

Lignin, characterized by a complex structure with various functional groups, degrades over a much broader temperature range, typically between 160 and 900 °C [15]. The TG curve of pineapple crown waste shows that about 26.7% of the mass remains at 500 °C, which can be attributed to lignin. This component continues to degrade slowly beyond 500 °C, with residual mass decreasing to around 18%. The degradation of lignin begins gradually, likely starting near 200 °C, overlapping the degradation ranges of hemicellulose and cellulose. The slow weight loss from 34% at 370 °C to 18% above 900 °C corresponds to this lignin decomposition phase.

In contrast, the thermal degradation of 100% polyethylene (PE) occurs between 400 and 550 °C. Unlike the TG curve for pure PCW, the plastic shows no weight loss below 150 °C, reflecting its lack of moisture or volatile components. The weight loss with increasing temperature is more rapid, and the DTG peak for PE is notably higher than that of PCW.

For the PCW-PE blends, two main degradation stages are observed: the first between 200 and 400 °C, and the second between 400 and 550 °C. The initial stage features overlapping DTG peaks similar to those of pure PCW, representing the decomposition of hemicellulose, cellulose, and lignin components. The second stage corresponds to the degradation of PE, evidenced by a DTG peak akin to that of 100% PE. As the PCW proportion in the blend increases, the first DTG peak becomes more pronounced, while the second peak associated with plastic decomposition decreases.

Interestingly, the second DTG peak, representing plastics degradation, is generally higher than the first peak representing PCW degradation. This phenomenon can be explained by the melting of plastics below 380 °C, which may coat the surfaces of PCW particles and hinder volatile release from the biomass [19].

An exception to this trend is observed in the 80% PCW blend, where the second peak is only slightly higher than the first. This smaller difference is likely due to the lower plastic content (20%) relative to the PCW mass in the mixture. Additionally, the second peak in the 80% PCW blend occurs at a slightly lower temperature compared to other blends. Such peak shifts have been reported in studies involving catalysts and suggest that at higher PCW proportions, catalytic intermediates may accelerate PE degradation at reduced temperatures. At lower PCW-to-PE ratios, the melted PE probably covers more of the PCW surface, limiting the generation and release of catalytic intermediates that would otherwise facilitate plastic decomposition.

Unlike other studies using pure plastic polymers-where the DTG peak temperature increases with higher plastic, the second peaks for PCW-PE blends containing 20%, 40%, and 60% PCW did not consistently occur at the same temperature, despite a constant heating rate [20].

Finally, the residual mass after pyrolysis increases with higher PCW content in the blends (Figure-2). This could be due to substances produced during plastic degradation that catalyze the breakdown of residual lignin in the pineapple crown waste. Alternatively, products formed from PE degradation may promote lignin decomposition.

Activation Energy Analysis

Plots of $\ln(-\ln(1-x)/T^2)$ vs 1/T for the different PCW-PE blends are shown in Figure-3. The line for 100% PE has the highest slope, i.e., the highest activation energy. The lines for the 100% PCW and the blends have lower slopes, i.e., lower activation energy.

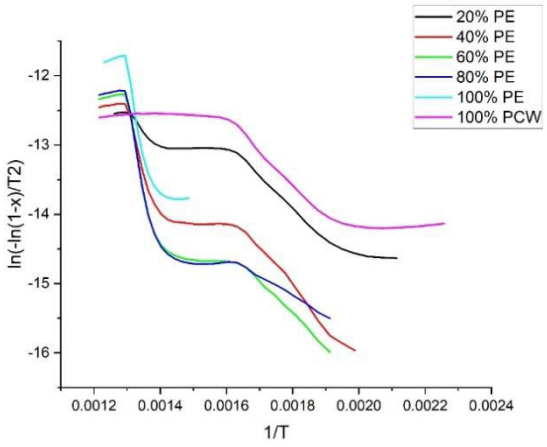


Figure-3. Plot of ln(-ln(1-α)/T2) vs 1/T for the different PCW: PE blends.

Table-2. Activation energy (E) values for the thermal decomposition of Pineapple crown waste (PCW) and polyethylene (PE) according to the first order chemical kinetics. The values were calculated from the slope of the lines in Figure-3, assuming that each curve is described by one slope only.

Feedstock	Temperature range (°C)	E (kJ/mol)	Correlation coefficient (R^2)
100% PE	400-550	86.88	0.87
20% PE	200-520	22.21	0.95
40% PE	230-550	36.41	0.91
60% PE	250-550	42.39	0.83
80% PE	250-550	38.2	0.76
100% PCW	170-400	17.39	0.87

The activation energies for the various PCW-PE feedstock blends are listed in Table-2, under the assumption that the overall thermal decomposition follows a single reaction model. Pure polyethylene (100% PE) exhibited an activation energy of 86.88 kJ/mol, whereas pure pineapple crown waste (100% PCW) had a significantly lower value of 17.39 kJ/mol. The activation energies observed for the blends were considerably lower than the weighted sum of the activation energies of the individual components (100% PE and 100% PCW), suggesting a synergistic interaction during co-pyrolysis. This phenomenon of rate synergy has also been reported in other studies involving biomass (e.g., corn stover, wood) and plastic materials (e.g., LDPE, HDPE) [11, 21].

The activation energy data for each PCW-PE blend are provided in Table-2. Analysis of the plots of $\ln(-\ln(1-\alpha)/T^2)$ versus $1/T$ for both pure PCW and the PCW-PE mixtures reveals two distinct decomposition stages with varying slopes: the first occurring within 200-400°C and the second within 400-550°C. Activation energies for these stages were calculated using different kinetic models, and the values derived from the best-fitting model for each blend are summarized in Table-3. The E values for all PCW-PE mixtures were again found to be lower than the sum of the individual activation energies of PCW and PE, adjusted for their respective mass fractions. This reduction in activation energy across both stages further supports the existence of a synergistic effect during the co-pyrolysis process.

Table-3. Calculated values of activation energy, E, for PCW, PE, and their blends from the best-fit kinetic model using the Coats-Redfern integral method of curve-fitting.

Sample	β (°C/min)	Stage 1	T (°C)	Ea (KJ/mol)	Model: The functions g(α) are in table 1.	R^2 %
100% PE	10	2	400-550	126	Jander (three-dimensional diffusion) D3	88.88
20% PCW	10	1	200-400	43.67	Jander (three-dimensional diffusion) D3	97.45
20% PCW	10	2	400-550	175.06	Jander (three-dimensional diffusion) D3	91.25
40% PCW	10	1	200-400	65.73	Jander (three-dimensional diffusion) D3	96.75
40%PCW	10	2	400-550	168.9	Jander (three-dimensional diffusion) D3	90.9
60%PCW	10	1	200-400	71.31	Jander (three-dimensional diffusion) D3	95.27
60%PCW	10	2	400-550	123.12	Jander (three-dimensional diffusion) D3	90.94
80%PCW	10	1	250-400	44.91	Chemical reaction order 1.5	96.36
80%PCW	10	2	400-500	30.997	Two-dimensional diffusion) D2	88.31
100%PCW	10	1	210-390	37	Chemical reaction order 1.5	94.15

Kinetics of PCW and PE co-Pyrolysis

As presented in Table-3, the kinetic model that best describes Stage 1 of the pyrolysis of pineapple crown waste (PCW) is the reaction order model R1.5, with a correlation coefficient (R^2) of 94.15%. Other top-performing models for PCW include R2, D3, and D4, which showed R^2 values of 93.28%, 90.27%, and 88.6%, respectively. In the case of 100% polyethylene (PE), the D3 model (Jander's three-dimensional diffusion) demonstrated the highest correlation, with an R^2 of 88.28%.

At both low and high PCW proportions, the dominant reaction mechanism transitioned to the Jander diffusion model (D3). Meanwhile, for intermediate PCW compositions-specifically 40% and 60% PCW-the D3 model remained the best fit, with correlation coefficients ranging from 90.9% to 96.75%. These results indicate that the D3 model generally yields high R^2 values across both reaction order and diffusion-based models. Thus, it is evident that no single kinetic model can fully describe the entire pyrolysis process. The reaction rate, influenced by the remaining mass of the material, varies across stages, making it challenging to determine a universal model that explains the thermal degradation mechanism for co-pyrolysis systems such as wood or corn stover with different plastics [11].

In Stage 2, the activation energy was observed to increase with higher PE content. Pure PE exhibited the highest activation energy of 126 kJ/mol using the D3 model. As the proportion of PCW increased, the activation energy declined, reaching as low as 31 kJ/mol at 80% PCW. For pure PCW, the highest activation energy recorded was 37 kJ/mol using the R1.5 model. This trend aligns with findings from other co-pyrolysis studies involving biomass (e.g., corn stover) and plastics, where the reduction in activation energy with increased biomass content is strongly associated with synergistic interactions [22].

CONCLUSIONS

This research explored the feasibility of using co-pyrolysis to convert waste polyethylene (PE) and pineapple crown waste (PCW) into liquid fuel. It provided insights into the behavior of these materials during thermal decomposition and identified key factors influencing the co-pyrolysis process. The study led to the following key findings.

At a constant heating rate of 20°C/min, PE undergoes thermal degradation between 400°C and 550°C. In contrast, PCW decomposes at lower temperatures, within the range of 170°C to 400°C, occurring in two distinct stages: the first, between 170°C and 300°C, is attributed to hemicellulose decomposition, while the second stage (300°C to 400°C) corresponds to cellulose breakdown. For the PCW-PE blends, thermal degradation also follows a two-stage process, with the initial stage corresponding to PCW components (hemicellulose and cellulose), and the second stage involving the decomposition of PE, remaining PCW fractions, and lignin.

Evidence of synergistic interaction between PCW and PE during co-pyrolysis was observed through a notable reduction in activation energy. This synergy enhanced the degradation of PCW and resulted in a lower amount of solid residues.

The findings from this study offer valuable insights for the practical design and optimization of co-pyrolysis systems and can guide future efforts to enhance the efficiency and effectiveness of converting biomass and plastic waste into fuel.

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