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Particle Size and Heating Rate Effects on the Pyrolysis Kinetics of Low-Rank Coal: Kinetics, Thermal Decomposition, and Thermodynamic Studies

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Abstract. This research aims to understand the combustion mechanism of low rank coal by studying the kinetics of the pyrolysis reaction using the thermogravimetric method. This study was conducted by testing coal samples at various pyrolysis temperatures between 0 and 600°C by varying the coal particle sizes of 5, 10, and 15 mesh, as well as different heating rates of 20, 40, and 60 °C/min in a nitrogen atmosphere. The data obtained by TG-DTG are divided into three stages to calculate the mass conversion rate and analyze key parameters, including the reaction rate constant, exponential factor, and activation energy. Thus, the pyrolysis reaction rate and mechanism can be formulated as a function of temperature and time. Mathematical modeling can be compared with the Kissinger, Coats-Redfern, and DAEM models. The findings of this investigation indicate that coal pyrolysis adheres to the first-order kinetics mechanism, exhibiting an average activation energy of 37-75 kJ/mol and a pre-exponential factor of 0.091-1.8 x 10^7 min^{-1}. Thermodynamic parameters, the average enthalpy change (ΔH), entropy change (ΔS), and free energy change (ΔG) associated with coal pyrolysis, are computed to be 2.6-7.3 kJ/mol, –332 kJ/mol/K, and 89-431 kJ/mol, respectively. A pyrolysis, yielding an optimal oil output of 57.04 wt.% at 320°C. This empirical investigation has the potential to enhance the combustion properties of low-rank coal, particularly about ignition efficiency and maximum weight reduction, thereby suggesting the viability of utilizing low-rank coals in co-combustion applications as a fuel source.

Keywords: Coast-Redfern, DAEM, Low-rank Coal, Kinetic, Kissinger, Thermogravimetric

INTRODUCTION

Indonesia possesses significant reserves of coal. Among the regions in Indonesia characterized by extensive coal distribution, the island of Sumatra, particularly Aceh Province, stands out. Raihan (2023) assert that the coal reserves in West Aceh constitute

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approximately 4.7% of the aggregate coal reserves in Indonesia. In a broader context, global coal reserves are classified as low-rank coal (LRC), which encompasses nearly 50% of the total coal reserves worldwide and is regarded as relatively economical, possessing a market value of merely 20-30% in comparison to high-rank coal (Çetinkaya and Bayat, 2020). Notwithstanding its considerable potential, the exploitation of low-rank coal (LRC) within Aceh encounters numerous challenges, including suboptimal utilization technology, diminished combustion efficiency, and the resultant greenhouse gas emissions and air pollution from LRC combustion. Consequently, there exists a pressing need for further investigation aimed at optimizing the utilization of Aceh's coal resources to enhance the quality of the coal produced (Mardhiyah *et al*., 2024).

Conversely, coal constitutes the second most significant energy resource globally, following petroleum and other fuels; its utilization for electricity generation is increasingly critical in light of the escalating energy crisis (Lakhmir *et al*., 2022). LRC can be employed for a myriad of applications, including the synthesis of liquefied natural gas (LNG) through the extraction of volatile components from low-rank coal, thereby supplying feedstock for LNG production and mitigating the presence of impurities (Ahmed *et al*., 2021), as well as in power generation, synthetic fuel production, and the manufacturing of coke (Yudisaputro *et al*., 2021). A pivotal phase in the chemical conversion process of LRC is referred to as pyrolysis.

Pyrolysis constitutes the thermal decomposition of coal within an inert atmospheric environment, such as that provided by nitrogen or argon. When subjected to atmospheric conditions, coal experiences both physical reactions and chemical transformations (Kaur *et al*., 2024). Pyrolysis reaction kinetics of LRC studies the rate and mechanism of LRC decomposition reaction at high temperatures. Understanding the kinetics of the LRC pyrolysis reaction can help in developing accurate combustion models, where this model can be used to predict LRC behavior in the reactor and assist in optimal reactor design (Dwivedi *et al*., 2019). Furthermore, it can find out how to increase combustion efficiency by understanding the LRC combustion mechanism and allowing optimization of the combustion process, thus producing maximum energy with minimal emissions (Zhang *et al*., 2019). Therefore, kinetic information can be used to develop new technologies to utilize LRC more efficiently and sustainably.

In recent studies, the study of coal pyrolysis kinetics based on non-isothermal thermogravimetric analysis (TGA) (Mi *et al*., 2023) is widely used to investigate pyrolysis kinetics due to its high sensitivity, repeatability, and reliability for acquiring data in studying the pyrolysis mechanism of low-rank coal (Yan *et al*., 2020). The TGA data obtained is generally accurate and precise, providing comprehensive information. Thermogravimetric analysis has proven instrumental in the evaluation of kinetic parameters associated with such intricate processes. Within these analyses, critical factors including pyrolysis temperature and the rate of heating (whether gradual or rapid) are considered (Kartal and Özveren, 2022). TGA data will produce a curve that shows the change in sample mass against temperature. From this curve, the mass conversion rate, activation energy, and pyrolysis reaction order can be calculated (Wardach-Święcicka and Kardaś, 2023). This method offers several advantages, including being simple and easy to use. For instance, the TGA technique is relatively straightforward and does not require complex equipment.

The methodologies established by DAEM and Kissinger were employed to examine the kinetics associated with the pyrolysis of low rank coal (Fischer *et al.*, 2024). They determined a reaction order that was notably elevated and utilized the Coats-Redfern approach to ascertain a plausible arbitrary reaction order for the analysis of the pyrolytic characteristics of coal (Yang *et al.*, 2023). Given the intricate nature of coal as a material, there exists a deficiency in the literature concerning the consideration of more comprehensive models. According to the author's findings. Although the TGA method has been used to study coal pyrolysis, a comparative study on the effects of coal type, particle size, heating temperature, and combustion time on low-rank coal within the existing literature is lacking. The Coats-Redfern integral approach has been effectively employed to investigate both the activation energy and the pre-exponential factor of complex materials (Huang *et al.*, 2022). The objective of this research is to enhance the understanding of the thermal decomposition behavior, kinetics, and thermodynamic potential of low rank coal through the application of reaction mechanisms utilizing the Kissinger, Coats-Redfern, and DAEM methodologies (Ghorbannezhad *et al.*, 2024). Therefore, further studies are needed on this matter to develop an appropriate method for improving the quality of low-rank coal, maximizing existing coal resources, and optimizing the utilization process of low-rank coal through the development of pyrolysis kinetic models.

MATERIALS AND METHODS

Coal Preparation

Low-rank coal was procured from the Mining Engineering Laboratory of PT. MIFA Bersaudara. A sample weighing 5 kg of low-rank coal was subjected to grinding in an iron mortar, followed by sieving through mesh screens of 5, 10, and 15 sizes and stored in air-tight bags. Proximate analysis was conducted in the MIFA Bersaudara mining laboratory using the ASTM standard (Figure 1) and methodologies detailed in our earlier publication to determine the proportions of moisture, volatile matter, ash, sulfur, and calorific value, as presented in Table 1.

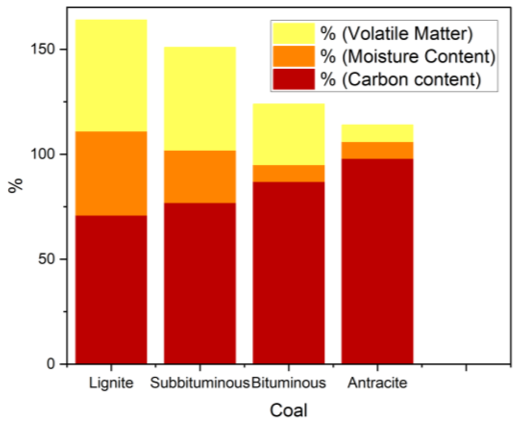

Fig. 1: Classification of coal based on ASTM D388-23 standard

Table 1. Characteristics of low-rank coal

Coal Sample	Moisture Content (%)	Volatile Matter (%)	Ash/ Sulfur (%)	Calorific Value (Kcal/Kg)
Subbituminous	45	39	2/0.2	3400-3200

Thermogravimetric Analysis

The thermal degradation characteristics of low rank coal during pyrolysis were elucidated via the utilization of a thermogravimetric analyzer (TGA model Simadzu no. 60) within a nitrogen atmosphere. A nitrogen flow rate of 20 ml/min was maintained throughout the experiment while the temperature was programmed to increase from 0 to 600 °C. The initial mass of the low rank coal specimen was measured at 23.8 mg. Three heating rates of 20, 40, and 60 °C/min were employed to assess the mass loss and rate of mass loss during the pyrolysis process. To enhance experimental precision and minimize systematic errors, each experiment was conducted at least three times. The TGA data allowed us to analyze the thermal decomposition of low-rank coal and determine the kinetics and thermodynamics of its pyrolysis.

Kinetic Study

The kinetic evaluation of low-rank coal pyrolysis in Aceh was elucidated through the application of the Arrhenius equation, which elucidated insights regarding the reaction kinetics. The fundamental equation employed for the kinetic assessment of low-rank coal pyrolysis is presented in Eq. (1).

$$\frac{d\alpha}{dt} = k(T).\ f(\alpha) \quad (1)$$

From the research that has been carried out, the combustion characteristics were analyzed using TGA so that the total mass loss value of each sample can be calculated using Eq. (2).

$$\alpha = \frac{(mo - mt)}{(mo - mf)} \quad (2)$$

Coats-Redfern Model

Kinetic analysis using the Coats-Redfern model (Uddin Monir *et al.*, 2024):

$$\beta = \frac{dT}{dt} \quad (3)$$

$$k(T) = A.\ e^{\left(-\frac{E}{RT}\right)} \quad (4)$$

$$f(\alpha) = (1 - \alpha)^n \quad (5)$$

$$\ln\left[\frac{-\ln(1-\alpha)}{T^2}\right] = -\frac{E}{RT} + \ln\frac{AR}{\beta E},\ n = 1 \quad (6)$$

Where β represents the rate of thermal increase (K/min), α denotes the conversion rate associated with the combustion reaction (%), T signifies the instantaneous reaction temperature (K), and R is the universal gas constant (8.314 J/mol·K). The activation energy can be derived by constructing a graph and subsequently calculating the slope from the resulting linear representation. The pre-exponential factor can be determined from the y-intercept of this graphical representation.

Kissinger Model

The rate equation for a solid-state reaction is written in the form of a reaction rate (dα/dt) proportional to some function of the amount of reactant (Ding *et al.*, 2024). If the reaction is assumed to be first order (n = 1) then f(α) = (1 − α),f = −1, ln[−f(α)] = 0 and Eq. (6) is simplified to:

$$\ln\left[\frac{\beta}{Tm^2}\right] = -\frac{E}{RTm} + \ln\frac{AR}{E} \quad (7)$$

Distributed Activation Energy Model (DAEM)

The kinetics of pyrolysis were investigated utilizing the extensively implemented distributed activation energy model (DAEM) (Kristanto *et al.*, 2023). When this model is applied to the pyrolysis of coal, the variation of conversion, denoted as x, about time, represented as t, is articulated by the equation. This methodology has been demonstrated to exhibit simplicity while simultaneously being effective, as it utilizes merely three sets of thermogravimetric analysis (TGA) data obtained during the pyrolysis process at varying heating rates. A streamlined equation was presented in the form of Eq. (8) (Miura, 1995).

$$\ln\left[\frac{\beta}{T^2}\right] = 0.6075 - \frac{E}{RT} + \ln\frac{AR}{E} \quad (8)$$

Where β represents the heating rate and R denotes the value of 8.314 J/mol·K, respectively. By constructing a plot of $\ln(\beta/T^2)$ at predetermined conversion levels (Arrhenius plot), both the activation energy (E) and the pre-exponential factor (A) can be derived from the slope and intercept, respectively.

RESULTS AND DISCUSSION

Thermal Degradation of Low Rank Coal

The pyrolysis of low-rank coal involves a complex process that includes many different reactions (Mi *et al.*, 2023). The TG/DTG curves of Low-Rank Coal samples pyrolyzed between 0-600 °C are depicted in Figure 1, and the characteristic parameters related to the main pyrolysis process are summarized in Table 2. Both TGA and DTG curves show similar trends for the various samples. This phenomenon can be attributed to the degradation of stable configurations present in low-rank coal, such as aromatic rings, which are subjected to extensive fracturing and decomposition; furthermore, active functional groups are swiftly depleted due to the assault of oxygen molecules, resulting in the liberation of carbon monoxide (CO), carbon dioxide (CO_2), and low-molecular-weight organic gases (Fan *et al.*, 2021). The process of low-rank coal pyrolysis generally transpires into three distinct phases: dehydration, devolatilization, and decomposition. These critical processes occur during the thermal treatment, as illustrated in Figure 2 (Yan *et al.*, 2020).

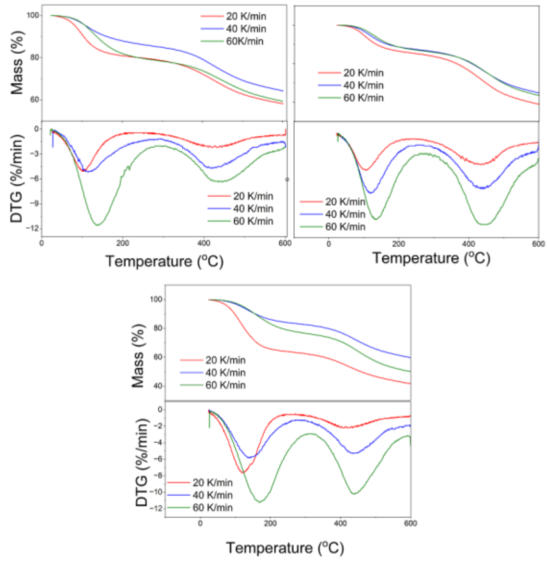

Fig. 2: TG/DTG profile for low rank coal pyrolysis (5, 10, 15 mesh)

As shown in Figure 2, the first stage is dehydration that involves the removal of moisture from materials, which is essential for enhancing their energy content. Of the three graphs, stage 1 usually occurs below 200 °C. The alterations in the slope of the TG curves were comparatively minimal, suggesting that the coals underwent a gradual depolymerization process, which is consistent with the extraction of bound water and the breakdown of carboxylic acid (Angelopoulos *et al.*, 2022). The second stage is devolatilization, which is characterized by the formation of volatile components during 200-400°C (Erić *et al.*, 2022). Thus, at the third stage, the decomposition stage, a gradual decrease in weight loss occurred, the mass loss curves became relatively flat with a slow decomposition until finished at 600°C and producing carbon dioxide and water.

To further illustrate the effect of coal rank on pyrolysis reaction, some characteristic parameters were evaluated: T_i, initial temperature of pyrolysis, T_T, transitional temperature, T_f, pyrolysis end temperature, as shown in Table 2. The degradation temperature of low-rank Aceh lignite is typically inferior to that of higher-rank coal varieties. This phenomenon is attributable to the elevated moisture content and volatile substance content inherent in low-rank coals, which render them more vulnerable to thermal disintegration. The precise degradation temperature may fluctuate contingent upon the particular coal specimen and the pyrolysis parameters, yet it generally resides within a spectrum of 300-500°C (Mergalimova *et al*., 2024). From Table 2, the maximum degradation rate is closely linked to the heating rate, with higher rates leading to increased mass loss (Song *et al*., 2019).

Kinetic Analysis of Low-Rank Coal Pyrolysis

The kinetic analysis of this paper is calculated using three models: the Kissinger method, the Coast-Redfern method, and the DAEM method, as illustrated in Figures 3, 4, 5 and 7. This also provides insights into the reaction mechanisms, activation energies, and pre-exponential factors involved in Table 3. The calculated results show that the kinetic parameters of three different heating rates and particle sizes of three models are different, and the order of activation energy is 5<10<15 mesh. The formulations of mean activation energy, pre-exponential coefficient, and the function governing the reaction mechanism of various coal types are incorporated into each equation.

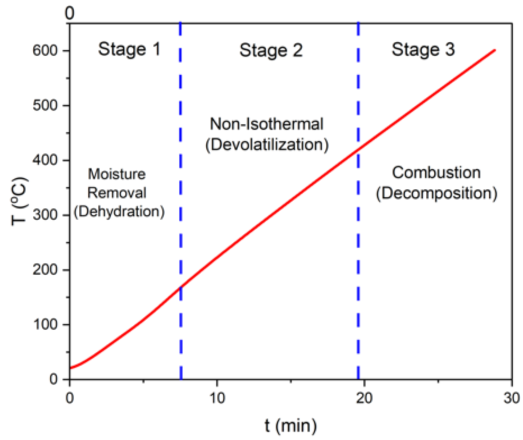

Fig. 3: Heating program for low rank coal pyrolysis in TG experiments

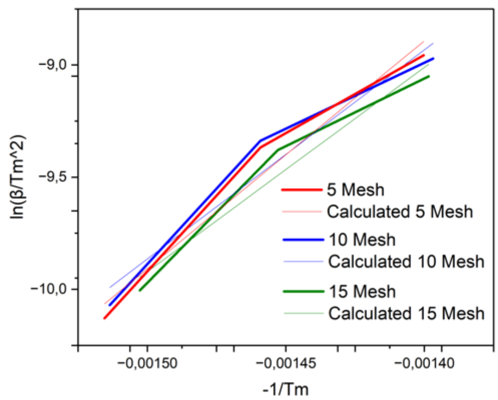

Fig. 4: Kissinger method for low-rank coal from Aceh

Table 2. Characteristics of low rank coal

Heating Rate (K/min)	Particle Size (Mesh)	Temperature (K)			Residue Amount (wt%)	Maximum Degradation Rate (wt%/min)
		T_i	T_T	T_F		
20	5	294.6	524.3	874.5	58.83	3.18
	10	299.4	515.7	874.4	58.10	5.11
	15	297.1	530.5	874.7	41.56	7.66
40	5	299.4	544.7	873.8	64.78	5.18
	10	302.6	521.4	874.2	64.23	5.18
	15	298.9	562.8	874.3	59.57	5.83
30	5	298.9	558	872.9	63.46	7.44
	10	295	569.2	873.4	59.59	11.60
	15	299.1	583.9	872.8	49.6	11.24

Table 3. Calculated kinetic parameters for a low rank coal by three different isoconversional methods

Sample	Coast-Redfern			Kissinger			DAEM		
Particle Size	Ea (kJ. mol^{-1})	A (min^{-1})	R^2	Ea (kJ. mol^{-1})	A (min^{-1})	R2	Ea (kJ. mol^{-1})	A (min^{-1})	R^2
5 Mesh	54.12	1.507×10^2	0.972	52.21	1.2×10^7	0.967	30.59	1.8×10^7	0.982
10 Mesh	37.96	6.6×10^2	0.948	32.32	1.9×10^3	0.948	30.89	9.1×10^3	0.981
15 Mesh	62.85	2.6×10^5	0.994	75.70	3.8×10^5	0.959	63.79	3.6×10^4	0.981

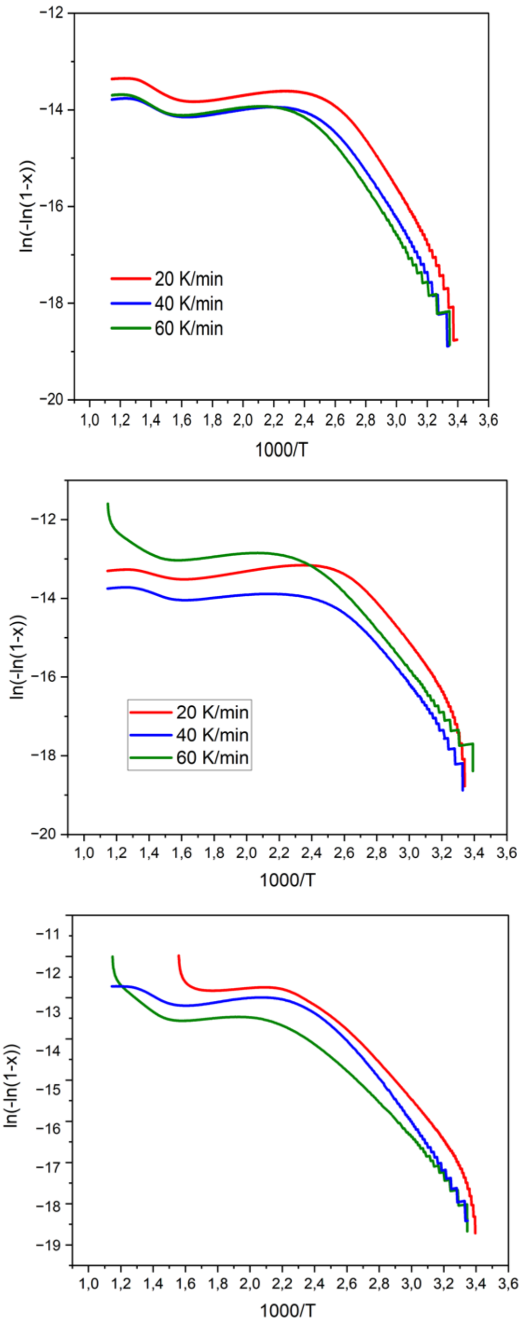

Fig. 5: Coast-Redfern for low-rank coal

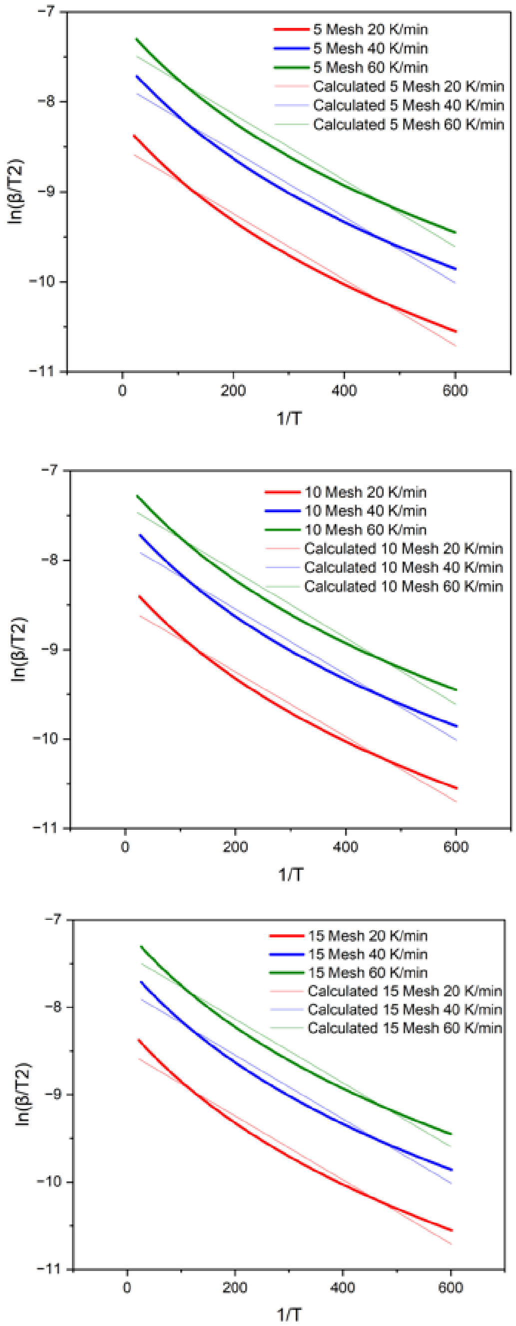

Fig. 6: DAEM method for low-rank coal

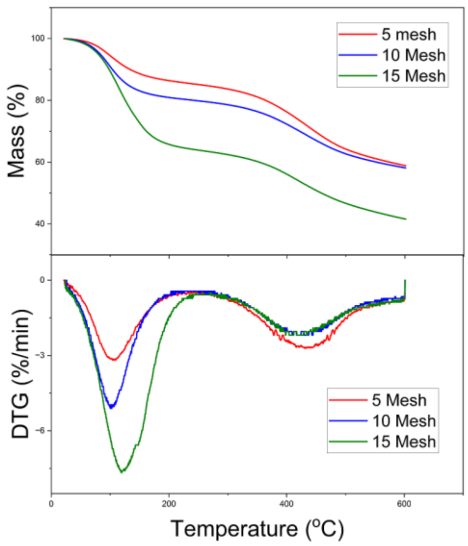

Fig. 7: TG-DTG profiles for three different particle sizes of low-rank coal from Aceh

The average values of activation energy from the Coast-Redfern, Kissinger, and DAEM models are 30.59-75.70 kJ/mol. Different heating rates and particle sizes influence the activation energy as shown in Figure 7. At a size of 5 mesh, the lowest activation energy value was obtained at 30.59 kJ/mol due to diminutive particles exhibiting an enhanced surface area-to-volume ratio, which facilitates expedited thermal transfer and accelerated devolatilization (Zhang *et al.*, 2023). This phenomenon may culminate in diminished activation energy as the energy barrier for the reaction is ameliorated (Sun *et al.*, 2024). In contrast, at 15 mesh, larger particles may experience internal temperature gradients, leading to slower heat transfer and a higher activation energy. Additionally, larger particles may have different devolatilization kinetics due to the presence of internal pores and structures (Jingyu *et al.*, 2025). The relationship between heating rates and activation energy is complex, influencing both reaction kinetics and the occurrence of secondary reactions. Accelerated heating rates may result in elevated activation energy due to the swift temperature rise, which consequently restricts the duration available for the reaction to attain equilibrium. This phenomenon may also influence the pre-exponential factor, as the incidence of collisions among reactant molecules is augmented (Smadi *et al.*, 2023). Conversely, slower heating rates may facilitate a more thorough devolatilization process and a diminished activation energy (Enyoh *et al.*, 2024). Nonetheless, this may also precipitate secondary reactions, such as char oxidation, thereby complicating the analytical assessment (Liu *et al.*, 2023). From three different models that showed R^2 (coefficient of determination) > 0.9, this is a statistical measure indicating how well a model fits a given dataset.

Thermodynamic Parameters

The thermodynamic characteristics of distinct phases during the thermogravimetric assessment of Aceh's low rank coal can elucidate insights into the coal's mineralogical composition and its reactions during thermal processing. The thermogravimetric evaluation of Aceh's low rank coal is anticipated to disclose a sequence of mass loss events that correlate with the degradation of multiple organic and inorganic constituents (*Ma et al.*, 2025).

The pyrolysis of low-rank Aceh coal represents a multifaceted phenomenon that is significantly shaped by various thermodynamic parameters, including enthalpy, entropy, and free energy (Wu *et al.*, 2025). The average value of enthalpy during the pyrolysis process is 2.6-7.3 kJ/mol. Enthalpy, which quantifies the overall thermal energy content, is predominantly positive during the pyrolysis process, signifying that thermal energy is absorbed. Meanwhile, the average value of entropy is -332 until -427 kJ/mol, which serves as an indicator of disorder, experiences an increase during pyrolysis as the structural integrity of the coal deteriorates and volatile (Zhang *et al*., 2025) substances are liberated (Wang *et al*., 2024). The entropy change (ΔS) during pyrolysis reflects the increase in disorder as the coal structure breaks down and volatile matter is released (Mishra *et al*., 2024). A positive ΔS is generally expected for pyrolysis based on the other literature (Yao *et al*., 2024).

In addition, the last parameter of thermodynamics is free energy, which is around 89-421 kJ/mol. A negative ΔG indicates a spontaneous process, while a positive ΔG indicates a non-spontaneous process. Free energy, which is the amalgamation of enthalpy and entropy, plays a critical role in ascertaining the spontaneity of the pyrolysis process (Vasudev *et al*., 2020). The pyrolysis of low-rank Aceh coal is typically spontaneous under elevated thermal conditions as a result of the advantageous interplay between changes in enthalpy and entropy.

Based on Table 3, there are different values for various heating rates and particle sizes. Low-rank coal with the lowest particle size (5 mesh) and the lowest heating rate (20 K/min) obtained the lowest value for thermodynamic parameters. Thermodynamic parameters of each phase during thermogravimetric analysis of low rank coal. The rate of heating and the size of the particles can exert a profound impact on the pyrolysis process. Elevated heating rates can expedite the pyrolysis reaction, resulting in varied product distributions and alterations in thermodynamic characteristics (Zhang *et al*., 2022). Reduced particle sizes can facilitate improved thermal transfer and accelerate the pyrolysis reaction, whereas larger particles may exhibit diminished thermal transfer rates and distinct kinetic behaviors (Nawaz *et al*., 2023). A comprehensive understanding of these thermodynamic properties and influencing factors can yield significant insights into the pyrolysis of low-rank Aceh coal, thereby enhancing the optimization of coal-derived product synthesis (Ma *et al*., 2024).

Table 3. Thermodynamic parameters of each phase during thermogravimetric analysis of low rank coal

Sample		Phase	Temperature	ΔH	ΔS	ΔG
Particle size	**Heating rate**		**(K)**	**kJ.mol^{-1}**	**kJ.mol^{-1}. K^{-1}**	**kJ.mol^{-1}**
5 Mesh	20 K/min	1	339.07	2.76	-332.46	89.14
		2	438.65	3.63	-438.95	177.89
		3	874.45	7.27	-427.37	421.76
	40 K/min	1	345.87	2.82	-332.46	102.93
		2	465.18	3.85	-438.95	189.72
		3	873.75	7.26	-427.37	420.58
	60 K/min	1	345.5	2.83	-332.46	113.08
		2	520.32	4.31	-438.95	216.79
		3	872.88	7.25	-427.37	409.14
10 Mesh	20 K/min	1	324.50	2.62	-332.46	62.79
		2	417.36	3.44	-438.95	155.00
		3	874.39	7.27	-427.37	-

Sample		Phase	Temperature	ΔH	ΔS	ΔG
Particle size	Heating rate		(K)	$kJ.mol^{-1}$	$kJ.mol^{-1}. K^{-1}$	$kJ.mol^{-1}$
	40 K/min	1	339.34	2.77	-332.46	99.32
		2	558.27	4.63	-438.95	246.63
		3	874.19	7.26	-427.37	412.92
	60 K/min	1	412.41	3.38	-332.46	131.90
		2	648.38	5.38	-438.95	280.25
		3	873.35	7.24	-427.37	373.63
15 Mesh	20 K/min	1	327.76	2.66	-332.46	83.42
		2	460.90	3.82	-438.95	190.61
		3	874.45	7.22	-427.37	315.05
	40 K/min	1	377.39	3.13	-332.46	167.97
		2	571.22	4.74	-438.95	260.03
		3	874.28	7.23	-427.37	318.78
	60 K/min	1	347.04	2.84	-332.46	112.54
		2	638.02	5.29	-438.95	274.77
		3	872.83	7.23	-427.37	365.79

CONCLUSIONS

The pyrolysis process of low-rank coals from Aceh can be divided into three stages: the dehydration stage, the devolatilization stage, and the decomposition stage. As the heating rate increases, the temperature at which the most rapid release of volatile matter occurs also rises, resulting in a greater overall amount of volatile matter being released from the coal. The activation energies of Aceh's low-rank coals are estimated to range between 37–75 kJ/mol, with a pre-exponential factor of 0.091–1.8 × 10^7 min^{-1}. Thermodynamic parameters, including the average enthalpy change (ΔH), entropy change (ΔS), and free energy change (ΔG) associated with coal pyrolysis, have been calculated at 2.6–7.3 kJ/mol, –332 kJ/mol/s, and 89–431 kJ/mol, respectively. Based on three models—Kissinger, Coats-Redfern, and DAEM. It can be concluded that each method provides reliable results, with average R^2 values greater than 0.9. This research highlights the significance of particle dimensions and heating rates in elucidating the pyrolysis kinetics of Aceh low-rank coal. A comprehensive understanding of these parameters is crucial for improving thermal conversion processes that transform coal into economically valuable products. Future investigations may further examine the influence of coal composition and mineralogical content on the pyrolytic behavior of Aceh coal.

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NOMENCLATURE

α	:	conversion rate of the combustion reaction
t	:	time [s]
$f(\alpha)$	:	kinetic equation
$k\,(T)$	:	rate constant

A	:	pre-exponential factor [min^{-1}]
Ea	:	activation energy [kJ/mol]
R	:	universal gas constant [8.314 J/mol/K]
T	:	reaction temperature [K]
β	:	heating rate
Tm	:	Peak temperature [K]

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