



# Kinetic Study of Co-pyrolysis of Kelakai (*Stenochlaena palustris*) and Low-rank Coal (Lignite)

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## ABSTRACT

Recently, fossil fuels have still become a main source of energy and chemicals. Biomass conversion has become a promising technology to convert biomass into bio-energy and bio-chemicals. Kelakai, as a potential biomass, is abundant in wetland areas such as Kalimantan (Indonesia) and many other Asian regions, so that low-rank coal (lignite). Co-pyrolysis, the combined pyrolysis of biomass and lignite, is an attractive technique considering to its potential to enhance the efficiency of pyrolysis products. In this study, the thermal decomposition behavior and kinetic of co-pyrolysis of kelakai and lignite at various mass ratio composition (1:0, 3:1, 1:1, 1:3, and 0:1) were investigated. The experiments were performed on a thermogravimetric analyzer (TGA). The TGA result indicated that the kelakai highly decomposed at 257-400°C, while lignite was 286-500°C. Their blends were in between. In addition, thermogravimetric data were subsequently applied to a kinetic analysis based on the Arrhenius equation, with a first-order reaction. The kinetic analysis results, including activation energy and pre-exponential factor, were determined for the kelakai and lignite mixture were found to be in the range of 10.22-10.98 kJ/mol and 0.0651-0.1351 min<sup>-1</sup>, respectively. Knowledge of thermal decomposition characteristics of kelakai and its kinetics is essential for optimizing pyrolysis design. The co-pyrolysis kelakai and lignite resulted in the highest bio-oil yield of 26.86 wt% at the ratio of 0:1 and the lowest yield of 12.51 wt% for the ratio of 1:0, when using mixed ratios of kelakai and lignite, the highest yield was 24.60% (1:3) and the lowest yield was 21.18 wt% (1:1).

**Keywords:** co-pyrolysis, kelakai, kinetics, lignite, yields.

## 1. INTRODUCTION

Energy demand will continue to increase in tandem with population growth and economic expansion. The rising need for energy, particularly fossil fuels, is depleting global oil reserves, making fossil fuels scarcer and significantly driving up their prices [1]. In response, efforts are underway to develop biomass utilization especially, agricultural, forestry, and household waste as a renewable energy source [2,3]. Biomass with high lignin, cellulose, hemicellulose, and carbon is a promising raw material for biofuel production [4–6].

*Stenochlaena palustris*, locally known as kelakai, is a species of fern commonly found in the wetland regions of Kalimantan and Sumatra (Indonesia) and also in many areas of Asian countries. Typically, local

communities only process the young stems and leaves of kelakai, while the older parts of the plant are discarded, leading to waste accumulation [7]. In fact, it contains 35.76, 42.67, and 21.15% of lignin, cellulose and hemicellulose, respectively [7]. Those are benefit for biofuel production.

Lignite, a type of low-rank coal with abundant deposits in South Kalimantan, contains comparatively low energy value and a substantial content of moisture, which make it inefficient for direct combustion to generate energy [8]. Numerous studies have explored optimizing lignite's use as an efficient energy source through processes such as gasification, hydrothermal liquefaction and co-pyrolysis [9–12]. Co-pyrolysis refers to a pyrolysis process involving two raw materials. The co-

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pyrolysis of lignite and biomass shows promise, as biomass can act as a hydrogen donor during pyrolysis, increasing bio-oil yields. In addition, the high content of volatile and high thermochemical reactivity of biomass such as kelakai could promote the co-pyrolysis process [10]. Furthermore, using lignite in co-pyrolysis can reduce its direct combustion for fuel, thereby mitigating environmental pollution [9,13]. Several studies on the co-pyrolysis of lignite with different types of biomass have been conducted, including pine sawdust [10], potato starch [11], and corn cobs [12]. However, based on the current literature, no prior studies have been found on the co-pyrolysis of kelakai waste and lignite. This research will analyze the reaction kinetics of the co-pyrolysis process for kelakai waste and lignite with varying raw material compositions. Understanding the reaction kinetics is essential for designing larger-scale pyrolysis equipment [11,12]. Furthermore, the co-pyrolysis of kelakai and lignite was conducted in a semi batch fixed bed reactor to investigate the effect of pyrolysis temperature on pyrolysis product yield.

## 2. RESEARCH METHODS

### 2.1. MATERIALS

The kelakai plants were sourced from Handil Bakti, Barito Kuala Regency, South Kalimantan, while lignite was obtained from Rantau, Tapin Regency, South Kalimantan, along with 99% pure nitrogen gas. The kelakai waste and lignite samples were first ground using a crusher and then sieved through an 18-mesh sieve, followed by drying in an oven at 105°C for 2 hours. The blends of kelakai and lignite were prepared by directly mixing at kelakai to lignite mass ratios of 1:0 (KL01), 3:1 (KL31), 1:1 (KL11), 1:3 (KL13), and 0:1 (KL01).

### 2.2. CHARACTERIZATION OF RAW MATERIAL

The kelakai and lignite materials were subjected to ultimate, proximate, and lignocellulosic analysis. Proximate analysis was performed to evaluate the characteristics

of the raw materials (kelakai and lignite) preceding the pyrolysis process. The analysis followed ASTM D3172 standards for moisture and ash content, volatile matter, and fixed carbon. Ultimate analysis was performed to determine the elemental composition of the raw materials (kelakai and lignite), including carbon (C), hydrogen (H), nitrogen (N), sulfur (S), and oxygen (O) using a Perkin-Elmer 2400 CHNS/O elemental analyzer. Lignocellulose analysis was conducted to measure the primary lignocellulosic components in biomass using the Chesson-Data method. Thermogravimetric experiments were performed using a SEIKO TG/DTA 7300 thermogravimetric analyzer. For each test, approximately 2 grams of sample were used, with a high-purity nitrogen flowrate (99.99%) set at 50 ml/min. All samples were tested up to 800°C with a heating rate of 10°C/min.

### 2.3. KINETIC STUDY

Thermogravimetric analysis (TGA) data shows the curve of decomposition of sample correspond to the temperature for the pyrolysis reaction. The TGA data used for kinetic calculations cover the temperature range of 200-550°C. Mass losses due to moisture removal were not considered part of the pyrolysis process [14]. The equation used in this study follows those presented by previous study [9,15,16]. The rate of mass loss is calculated according to Equation (1) as follows:

$$\frac{dW}{dt} = -\frac{1}{W_o} \left( \frac{dW_t}{dt} \right) \quad (1)$$

The mass loss is defined in terms of pyrolysis conversion,  $x$ , and could be calculated using Equation (2), as follows:

$$x = \frac{W_o - W_t}{W_o - W_f} \quad (2)$$

Here  $W_o$  represents the initial sample weight,  $W_t$  is the weight of the sample at a specific time, and  $W_f$  is the mass that remains at the completion of the pyrolysis process. The

integral solution was applied using TGA data to obtain kinetic parameters that involve the activation energy and pre-exponential factor for co-pyrolysis reaction of kelakai waste and lignite. In this study, by using the Arrhenius equation, the reaction kinetics of the co-pyrolysis of kelakai waste and lignite can be described by Equation (3) below:

$$\frac{dx}{dt} = A \exp\left(-\frac{E}{RT}\right) (1-x) \quad (3)$$

Where  $A$  is the pre-exponential factor,  $E$  is the activation energy,  $T$  is the temperature,  $t$  is time, and  $x$  is calculated using Equation (2). For a constant heating rate  $H$  during pyrolysis, defined as  $H=dT/dt$ , Equation (3) is re-arranged and integrated to yield Equation (4), as follows:

$$\ln\left[\frac{-\ln(1-x)}{T^2}\right] = \ln\left[\frac{AR}{HE}\left(1 - \frac{2RT}{E}\right)\right] - \frac{E}{RT} \quad (4)$$

For almost all of the  $E$  values within the pyrolysis temperature interval, the term of  $\ln[AR/HE(1-(2RT/E))]$  in Equation (4) remains fundamentally. Plotting the left side of Equation (4):  $\ln\left[\frac{-\ln(1-x)}{T^2}\right]$  on the y-axis and  $1/T$  on the x-axis result in a linear graph, assuming that the co-pyrolysis reaction follows first-order kinetics. Both the slope and intercept of this straight line will be employed to calculate the activation energy ( $E$ ) and the pre-exponential factor ( $A$ ) [9,13,15].

#### 2.4. CO-PYROLYSIS EXPERIMENT

Co-pyrolysis reaction was conducted in a semi-batch fixed bed tubular reactor with 16 and 25 cm of diameter and height, respectively. The experimental set-up of the pyrolysis reactor is described in our earlier works [8]. The TGA result of kelakai powder was used to determine co-pyrolysis final temperatures (550°C). The co-pyrolysis experiment was observed under the same sample composition and conditions as in TGA experiment (heating rate of 10°C/min, nitrogen flowrate of 200 mL/min, and calculated residence time of 25 min) and the

reaction temperature was maintained for 1 hour since the co-pyrolysis temperature was achieved. About 400 gram of sample was used for each run. Each sample was tested at least three times to ensure repeatability and data accuracy.

### 3. RESULTS AND DISCUSSION

#### 3.1. CHARACTERIZATION OF RAW MATERIAL

In this study, pyrolysis was performed using a mixture of kelakai and lignite. Table 1 shows the results of the proximate and ultimate analysis for the five contents, and Table 2 shows the results of the lignocellulosic composition analysis for kelakai.

**Table 1.** Proximate and ultimate analysis of kelakai (K) and lignite (L).

Sample	K	L
Ultimate Analysis (wt%, dry basis)		
Carbon	42.44	42.09
Hydrogen	5.90	5.56
Oxygen	33.72	36.69
Nitrogen	1.13	0.13
Sulfur	0.20	0.19
Proximate Analysis (wt%, dry basis)		
Water	8.12	8.63
Volatile	76.96	46.80
Ash	8.49	6.71
Fixed carbon	6.43	37.86

It is clear that kelakai has a relatively higher molar ratio of hydrogen than lignite, indicating that the kelakai in the mixture can donate hydrogen during the co-pyrolysis process [12,17], which may subsequently lead to interactions between the two materials. Furthermore, lignite contains a higher fixed carbon content (37.86 wt%) compared to kelakai (6.43 wt%), resulting in slower reaction kinetics [17]. Fixed carbon was formed when the volatile matter is released at the temperature higher than 673 K [18]. In other words, the higher value of fixed carbon relates to lower decomposition rate thus resulted in lower biomass conversion. Kelakai, on the other hand, has a lot of potential to produce more volatile products

(76.96 wt%) than lignite (46.80 wt%) by enhancing the yield of liquid product [9,12,17].

**Table 2.** Lignocellulosic composition of kelakai (wt%, dry basis).

Lignocellulose Analysis (wt%, dry basis)	
Hemicellulosa	13.80
Cellulose	27.93
Lignin	32.11

Based on Table 2, lignin is identified as the dominant macromolecular component found in kelakai waste. Consequently, higher lignin content in kelakai biomass has the potential to enhance the activation energy, as lignin structure was highly compact because of its linkage with a hydroxyl phenolic group [19,20].

### 3.2. THERMOGRAVIMETRIC ANALYSIS

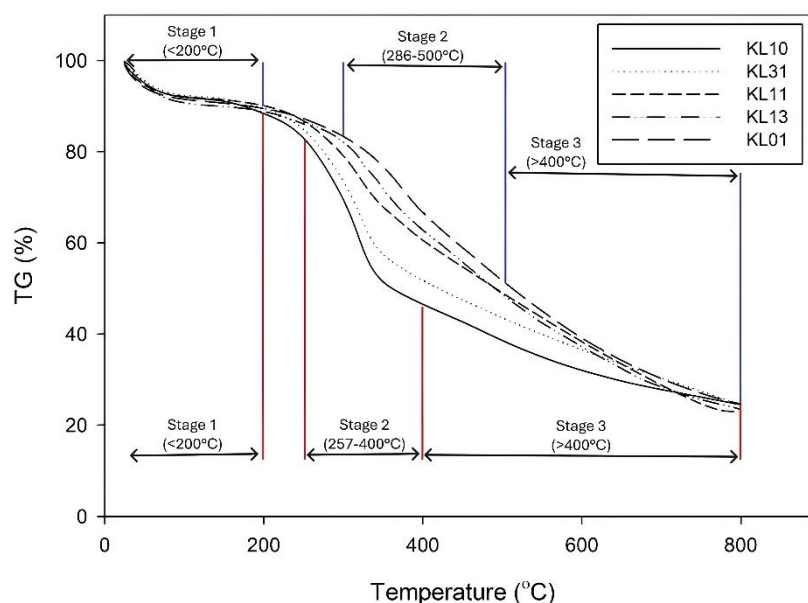
Thermogravimetry analysis (TGA) plots the mass loss of samples as the pyrolysis temperatures increase. Figure 1 presents the TGA curves for kelakai (KL10), pure lignite (KL01), and the mixtures of kelakai and lignite (KL31, KL11, and KL13) with different compositions.

Figure 1 clearly demonstrates that the mass loss of each sample increases proportionally to the increase in pyrolysis temperature. Pure kelakai exhibits pyrolysis characteristics similar to those of lignite but within a different temperature range. The pyrolysis process of pure kelakai (KL10) consist of three primary steps. The first stage, which occurs below 200°C, involves the evaporation of sample's moisture. All samples, across various mass ratios of kelakai

and lignite, also exhibit this stage. Mass loss, occurring between 257°C and 400°C, characterizes the second stage, where the decomposition of polysaccharide structures rapidly released volatiles. The third stage entails mass loss above > 400°C, involving the decomposition of residual carbon compounds [11,12,21].

In contrast, the pyrolysis of pure lignite (KL01) also comprises three main stages. The first stage involves mass loss occurring below 200°C, resulting from dehydration and the removal of adsorbed gases. The second stage, the primary devolatilization phase, occurs around 286°C and 500°C, likely involving decarboxylation reactions at a slower rate than that of kelakai. The third stage, which takes place above 500°C, is characterized by slow decomposition due to polycondensation reactions [11,21].

Figure 1 indicates that KL10 begins to decompose at a lower temperature, exhibiting a greater rate of weight loss and more rapid pyrolysis duration relative to KL01. The TG curve of the kelakai and lignite mixture lies between the TG curves of the pure kelakai and lignite samples, with the range of stage 1 almost the same as KL10 and KL01 due to the dehydration reaction. Where as the stage 2 was 259-425°C, 271-480°C, 284-494°C for KL31, KL11, and KL13, respectively. Stage 3 for KL31, KL11, and KL13 took place at 426, 480 and 488°C, respectively. In addition, the residue charcoal yield decreasing as the ratio of kelakai in the mixture increases. This observation is also noted in studies involving lignite and biomass (rice husk, wheat, and bamboo powder) [22].



**Figure 1.** Thermogravimetry curves of kelakai (K) and lignite (L) at various mass ratio composition.

### 3.3. KINETIC STUDY

Equation (4) was employed to calculate the pyrolysis reaction's kinetic parameters, specifically the activation energy and the pre-exponential factor. Plotting the left side of Equation (4) on the y-axis and  $1/T$  on the x-axis result in a linear graph, assuming that the co-pyrolysis reaction follows first-order kinetics. A linear equation model was constructed by plotting  $\ln[-\ln(1-x)/T^2]$  versus  $1/T$  for the pyrolysis of lignite and the mixtures of kelakai and lignite, as shown in Figure 2.

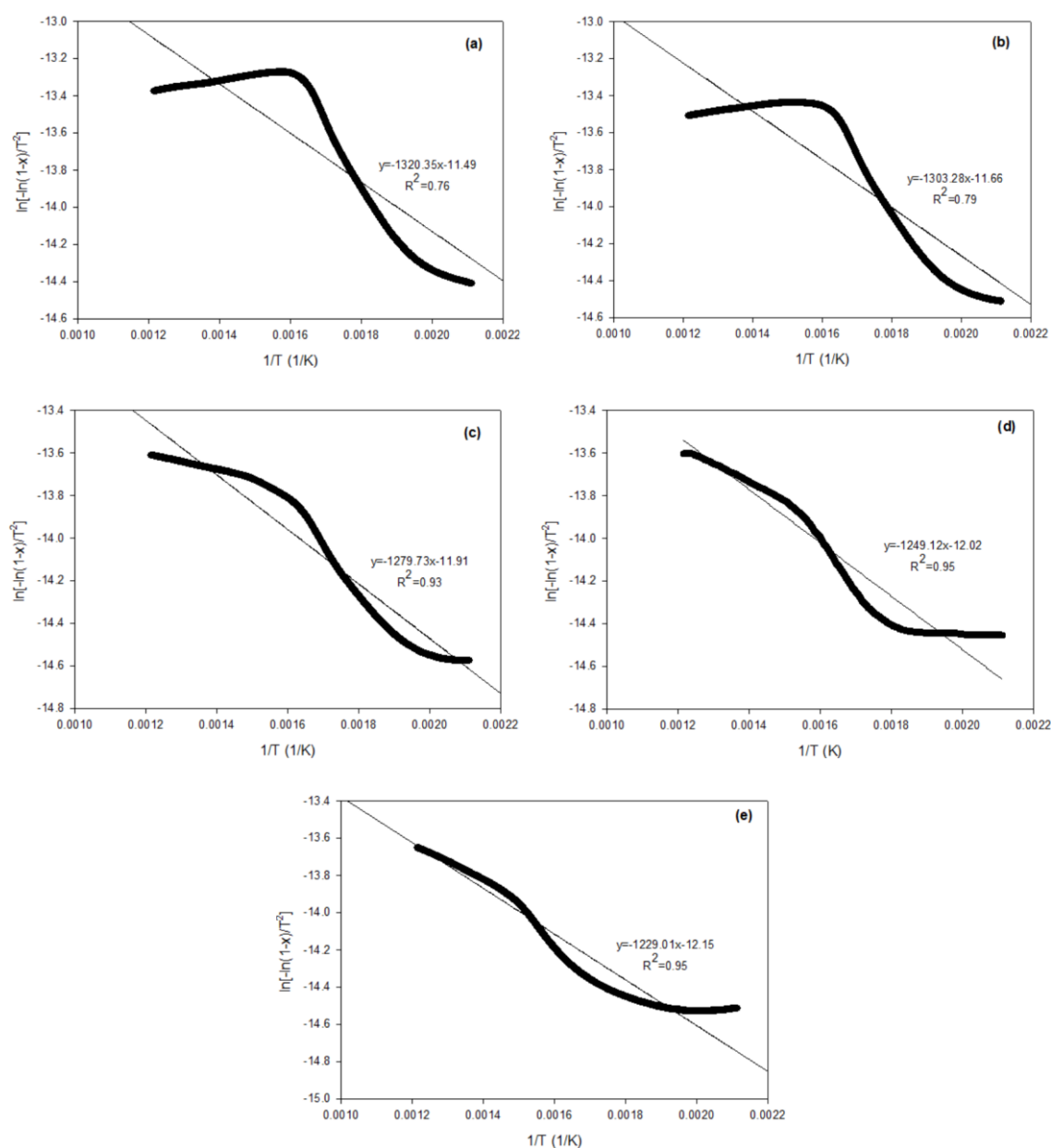
Figure 2 presents the kinetic analysis findings for KL10, KL31, KL11, KL13, and KL01 and they are summarized in Table 3. All of the  $R^2$  values run between 0.76 and 0.95, suggesting a satisfied fit of the experimental data. The variation in the activation energy ( $E$ ) for pure kelakai (KL10) is distinctly different from that of pure lignite (KL01). As the lignite's composition goes up, the  $E$  values decrease for all samples while the pre-exponential factor ( $A$ ) increases. This suggests that the thermal decomposition of the carbonaceous material exhibits multistep kinetics with differing  $E$  values. With the increasing  $A$ , the  $E$  value for pure kelakai (KL10) continues to rise, whereas the  $E$  value for pure lignite (KL01) decreases. This observation is consistent with the findings of

Aulia et al. [9] regarding lignite and polyethylene and Liu et al. [18] with elm sawdust and coal. The interaction between the volatile compounds from kelakai and lignite during co-pyrolysis could significantly accelerate the kelakai pyrolysis, resulting to a lower activation energy.

**Table 3.** Kinetics parameters of co-pyrolysis kelakai and lignite.

Sample	$E$ (kJ/mol)	$A$ ( $\text{min}^{-1}$ )	$R^2$
KL10	10.98	0.1351	0.76
KL31	10.83	0.1125	0.79
KL11	10.63	0.0858	0.93
KL13	10.38	0.0750	0.95
KL01	10.22	0.0651	0.95

The  $E$  values indicate that kelakai needs a higher energy to decompose in comparison to lignite. The calculated  $E$  values were decreased in order: 10.98, 10.83, 10.63, 10.38, and 10.22 kJ/mol for KL10, KL31, KL11, KL13, and KL01, respectively. As more lignite was added in the mixing, the reaction could be easier and faster occurred. This result also might be associated with kelakai's higher ash content compared to lignite that leads to thermal and mass transfer limits [21,23].



**Figure 2.** Curves of  $\ln(-\ln(1-x)/T^2)$  vs  $1/T$  for kelakai and lignite with various mass ratio: (a) KL10, (b) KL31, (c) KL11, (d) KL13, and (e) KL01.

### 3.4. THE EFFECT OF KELAKAI AND LIGNITE MIXTURE ON CO-PYROLYSIS PRODUCT YIELDS

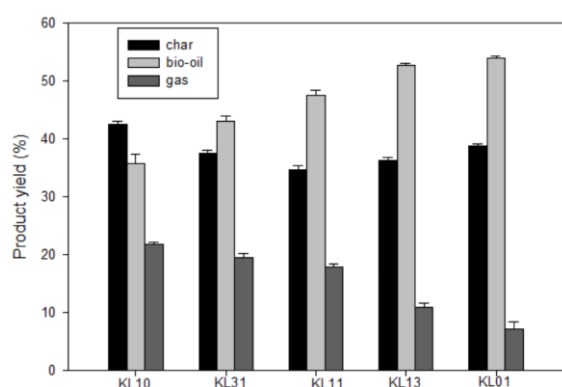
Figure 3 shows that there is a significant difference in the yield of liquid product (bio-oil) obtained with the addition of lignite in the pyrolysis process of kelakai.

Adding lignite increases the liquid yield compared to the pyrolysis of lignite alone. This trend was also observed by Onay [24]. The highest liquid yield was obtained from KL01, with a value of 53.96 wt%, while the lowest was from the KL10 variation at 35.6

wt%. Meng et al. reported that the synergetic effect of co-pyrolysis of potato starch (biomass) and lignite was beneficial to the bio-oil yield [11]. Furthermore, He et al. confirmed the existence of synergetic effect between the coal and biomass from the yields of pyrolysis products are higher than their calculated values [4]. Kelakai without the addition of lignite showed the highest char yield, amounting to 42.56 wt%, while the lowest char was produced by KL11 at 34.58 wt%. As for the gas yield, the highest was produced by KL10 at 21.76 wt%, while the



lowest gas yield was from KL01 at 7.20 wt%. From Table 1, the H/C atomic ratio of kelakai was greater than that of lignite. As a result, during co-pyrolysis, a large number of active radicals are released, while the secondary reaction is reduced [24-25]. The addition of lignite enhances the degradation of kelakai, which decomposes into larger quantities of gas and bio-oil compared to pyrolysis without lignite. This is consistent with the findings of Guo et al. [10] and Wu et al. [26], who reported that lignite addition increases bio-oil yield while reducing gas production.



**Figure 3.** Effect of kelakai and lignite mixture on co-pyrolysis product yields.

#### 4. CONCLUSION

Thermogravimetric analysis (TGA) was performed to investigate the thermal degradation and kinetic of co-pyrolysis of kelakai and lignite at various mass ratio composition. The kinetic analysis revealed that the first-order satisfied the model of co-pyrolysis of kelakai and lignite. In terms of both activation ( $E$ ) and pre-exponential factor ( $A$ ), adding lignite in pyrolysis of kelakai is beneficial. Co-pyrolysis of kelakai and lignite significantly reduce activation energy compared to pyrolysis of kelakai itself. The co-pyrolysis kelakai and lignite resulted in the highest bio-oil yield of 26.86 wt% (KL01) and the lowest yield of 12.51 wt% (KL10), when using mixed ratios of kelakai and lignite, the highest yield were 24.60 wt% (KL13) and the lowest yield were 21.18 wt% (KL11). Thus, the optimum mass ratio for co-pyrolysis kelakai and lignite was 1:3 (KL13).

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