



Determining co-combustion characteristics, kinetics and synergy behaviors of raw and torrefied forms of two distinct types of biomass and their blends with lignite

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Abstract

In this study, the combustion characteristics and kinetics of various mixtures of both raw and torrefied rose pulp and red pine sawdust with each other and with Elbistan lignite were investigated in the context of lignocellulosic biomasses' potential use as fuel. Ignition temperatures, peak temperatures, burnout temperatures, and comprehensive combustion indexes of the fuel mixtures were found to rise with the torrefaction process. This finding indicates that the fuel/combustion performance of the waste biomass can be improved by the torrefaction process. Moreover, the combustion behavior of rose processing waste and pine sawdust has been significantly improved by adding lignite to the samples to be torrefied. Average activation energies of raw pine sawdust, rose processing waste, and Elbistan lignite were found to be 178, 187, and 91 kJ mol⁻¹, respectively. However, the activation energies of both raw samples and their mixtures with lignite as well as each other decreased with the torrefaction process. Furthermore, a synergistic effect was also observed during the combustion of the mixtures of both raw and torrefied biomass with Elbistan lignite.

Keywords Biomass · Co-combustion · Kinetic · Rose pulp · TGA · Torrefaction

Introduction

The lignite reserves of Turkey are estimated to be 16.3 billion tons; while lignite's share in Turkey's total primary energy consumption is about 27% [1]. Imports account for about 75% of Turkey's energy demand [2], making the country heavily dependent on fossil fuel imports. Therefore, it is crucial for Turkey to use domestic lignite reserves to cover a larger portion of its energy needs. Yet, only 1% of lignite reserves in Turkey have a calorific value above 4000 kcal/kg. Against this background, burning these low-quality coals in combination with biomass represents a more stable means of biomass disposal, all the while generating comparatively cleaner energy with existing resources [3]. On the other hand, biomass is usually considered a secondary fuel, due to

a number of reasons such as seasonality and storage issues. When woody biomass is used in combustion systems and is blended with coal, certain economic and technical problems such as low grindability, low energy density, and high transportation cost arise [4, 5]. To overcome these problems, torrefaction—a thermal pre-treatment method—is applied on biomass. Torrefaction is the heat treatment of biomass at certain temperatures (250–350 °C) in order to improve its fuel properties and combustion characteristics [73]. The torrefaction process converts most of the raw biomass into a solid fuel that is relatively odorless and smokeless. Moreover, with torrefaction, biomass turns into a dense, hydrophobic, non-biodegradable and energy-dense fuel that can be stored for longer periods [6–8]. Most crucially, with the help of torrefaction, the share of biomass in co-fired plants can be increased up to 60–70% and thus GHG emissions into atmosphere can be significantly reduced [9].

Turkey is one of the most important production centers of both rose concrete and rose oil worldwide [16]. Approximately 80% of rose oil and rose concrete production in Turkey takes place in Isparta [17]. The amount of distillation waste produced in an average year is approximately 26,000 tons [18]. Uncontrolled disposal of this waste increases the

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pollution burden on surface and groundwater in Isparta and the surrounding provinces, leading to various environmental problems such as odor pollution and aesthetic problems [18]. On the other hand, Turkish red pine (*Pinus brutia* Ten.) is the most wide-spread tree species in Turkey, covering an area of 5,610,215 hectares [19]. One should also note that, in 2019, nearly 850,000 m³ of wood residues from forest products were produced in Turkey [20].

In the light of these figures, it is understood that Turkey has a considerable potential in energy recovery from biomass. Against this background, the combustion kinetics and combustion characteristics of such biomass containing high amounts of volatile organic materials will play a crucial part in designing efficient sustainable co-pyrolysis and co-combustion processes [21]. In this context, thermal decomposition stands out as an important factor during combustion and co-combustion processes, whereas thermogravimetric (TG) analysis is useful for analyzing the kinetics of thermal decomposition of biomass, as well as the combustion/co-combustion characteristics of biomass, lignite, and their blends. TGA is noted as a useful tool for investigating the synergy between biomass and lignite samples during co-combustion processes [21].

Although there are various studies on both combustion and co-combustion kinetics and properties of torrefied lignocellulosic biomass samples including chars made from raw and torrefied willow [10, 11], raw and torrefied duckweed [12], torrefied Norwegian birch and spruce [13, 14], raw and torrefied willow [15], raw and torrefied Canadian yellow poplar, Colombian sugarcane bagasse and their chars [9], and more (corn stalk, olive tree pruning, vine pruning) [5], no studies so far have been found in the literature on the thermal kinetic constants or co-combustion characteristics of both raw and torrefied rose pulp. Furthermore, the majority of the studies in the literature are mostly focused on char production and combustion kinetics, along with combustion characteristics of the chars produced. The studies on the synergistic effect arising with the co-combustion of lignite and torrefied biomass, on the other hand, are rare. Therefore, the

present study aims to evaluate the fuel characteristics of both rose pulp and wood sawdust samples and their blends with low quality Turkish lignite, followed by an investigation of their potential for use as fuel, and how their fuel properties can be improved with the torrefaction process. Another goal is to investigate the synergy between raw/torrefied biomass and lignite in blends thereof, and to determine the activation energy of the samples, as one of the most important pieces of data required for the design of combustion systems.

Material and methods

Material and thermal treatment

In this study, rose pulp and red pine sawdust samples were obtained, respectively, from a local rose oil factory and a sawmill plant in Isparta. Lignite samples to represent local Turkish lignite were taken from Afşin-Elbistan lignite Works. Since both rose pulp (68% ± 5.14%) and Elbistan lignite (55% ± 8.24%) contain substantial amounts of moisture, they were first dried at room temperature and then in an oven at 70 °C. Then, collected samples were crushed using a ball mill (pine sawdust was not extra milled because it was originally obtained in dust form) and sieved in the 300–850 µm range for thermogravimetric analysis (TGA).

The proximate and elemental analysis results of the samples used for the TG analysis are given in Table 1. ASTM D-871–82 and ASTM E-872–82 standards were applied, respectively, for the determination of the moisture and volatile matter (VM) content of the biomass samples [74]. Ash (A) contents of the woody biomass and rose pulp samples were determined using ASTM D-1102–84 and ASTM E-1755–01 standards, respectively [74, 75]. The ASTM D3172-13 standard was also used for the proximate analysis of lignite samples [76]. The fixed carbon (FC) content of both biomass and lignite samples was calculated using Eq. (1) [22].

Table 1 Proximate and elemental analysis results of the pine sawdust, rose pulp, and Elbistan lignite

	Moisture/%	Volatile matter/%	Fixed carbon/%	Ash/%	HHV/kcal/kg
Pine sawdust*	8.2 ± 1.0	85.1 ± 2.9	5.45 ± 2.3	1.20 ± 0.45	4192.4 ± 380
Rose pulp**	6.2 ± 2.1	77.4 ± 4.8	9.56 ± 2.3	6.7 ± 0.46	3924.2 ± 19
Elbistan lignite**	20.9 ± 1.5	43.3 ± 3.2	15.63 ± 2.2	19.7 ± 0.4	5611.1 ± 21
	C %	H %	N %	S %	O* %
Pine sawdust	48.18	6.40	–	–	44.22
Rose pulp	45.58	6.02	4.14	0.29	37.27
Elbistan lignite	35.14	3.82	1.99	2.55	36.80

*As received form, **After dried

$$\text{Fixed carbon (\%)} = 100 - \text{Moisture\%} - \text{Volatile matter\%} - \text{Ash\%} \quad (1)$$

Empirical correlations developed in the literature for raw [Eq. (2)] and torrefied [Eq. (3)] biomass were used to calculate the higher heating values (HHV) of the samples [23].

$$\text{HHV} = -10.81408 + 0.3133(\text{VM} + \text{FC}) \quad (2)$$

$$\text{HHV} = 0.1846\text{VM} + 0.3525\text{FC} \quad (3)$$

where VM and FC, respectively, represent the volatile matter and fixed carbon content of the sample.

In order to calculate the HHV of the lignite sample, Eq. (4) using the elemental content of Elbistan lignite was applied [24];

$$\text{HHV} = 144.54 * \text{C} + 610.2 * \text{H} + 40.5 * \text{S} - (65.88 - ((30.96 * \text{O}) / (100 - \text{A}))) * \text{O} \quad (4)$$

Previous studies revealed that the sulfur contents of lignite samples are higher than those of other biomass samples. Demirbaş et al. (2008) examined 20 Turkish lignites collected from 20 distinct regions of Turkey and found that the HHV values of the studied samples varied between approximately 5398 kcal/kg and 7022 kcal/kg [25]. Even though Elbistan lignite has almost the lowest calorific value among all Turkish lignites, its calorific value was found to be significantly higher than those of the biomass samples analyzed (Table 1). The total sulfur and ash contents of the Elbistan lignite used in this study were also found to be very close to those of the lignites examined in Demirbaş's (2008) study. Moreover, in the literature, it has been stated that the carbon and nitrogen contents of local Turkish lignites vary from 61 to 72.6%, and from 1.5 to 2.6%, respectively [25]. However, the present study found the carbon (40.51%) and nitrogen (0.68%) contents of Elbistan lignite to be lower than other local Turkish lignites analyzed in the literature [26].

For the torrefaction of the biomass samples, 10 g of samples was placed in 50-mL airtight crucibles. Then, the crucibles were subjected to thermal treatment in an ash furnace. Preliminary experiments to identify optimum torrefaction conditions of rose pulp and sawdust samples were performed in our previous study [27]. During the torrefaction process, biomass begins to decompose at around 150–200 °C [28]. Beyond 300 °C, a significant amount depolymerization is observed [29]. Three different temperatures (250, 270, and 290 °C) and three different holding times (15, 30, and 60 min) were tried. Since the highest energy density was obtained at 270 °C with 60 min holding time for both rose pulp and sawdust samples, [27] these were assumed to be

the optimum torrefaction conditions for the purposes of the present study.

Thermogravimetric analysis and applied kinetic model

In this study, 14 different mixtures were prepared using raw pine sawdust, rose pulp and Elbistan lignite with different rates of combination (Table 2). A further set of 14 blends was prepared using torrefied biomass samples at various rates. TG analyses were performed for each blend, using PerkinElmer-Pyris-1 model TG at four different heating rates, 10 °C min⁻¹, 20 °C min⁻¹, 30 °C min⁻¹, and 40 °C min⁻¹ under 40 mL min⁻¹ dry air flow at a temperature range of 25 °C to 1000 °C in temperature. The repeatability of the TG analysis was also checked in the study, and the process was found to be repeatable at a temperature range of approximately ± 2 °C.

The fundamental rate equation expresses the rate of conversion $-da/dt$ as a function of the reactant concentration (α) and the rate constant (k), at a constant temperature (T):

$$\frac{da}{dt} = kf(\alpha) \quad (5)$$

where

$$\alpha = (m_o - m_t) / (m_o - m_\infty) \quad (6)$$

where m_o is the initial mass of the reactant, m_t is the mass at time t , m_∞ is the final mass. k (s⁻¹) is usually given by the Arrhenius equation as,

Table 2 Mass ratio of blends (mass %)

Blend name*	Pine sawdust	Rose pulp	Lignite
P49R49L2	49	49	2
P48R48L4	48	48	4
P45R45L10	45	45	10
P25R75	25	75	–
P75R25	75	25	–
P50R50	50	50	–
P98L2	98	–	2
P96L4	96	–	4
P90L10	90	–	10
R98L2	–	98	2
R96L4	–	96	4
R90L10	–	90	10
P100	100	–	–
R100	–	100	–
L100	–	–	100

*For torrefied blends; the letter “T” was added in front of the abbreviation

$$k = Ae^{-E_a/RT} \quad (7)$$

where A is the pre-exponential constant (min⁻¹), E_a is the activation energy (kJ mol⁻¹), T is the temperature (K), and R is the universal gas constant (8.314 kJ mol⁻¹). The combination of Eqs. (5) and (7) gives;

$$\frac{da}{dt} = Af(a)\exp(-E_a/RT) \quad (8)$$

For non-isothermal TGA, if linear heating rate is assumed as $\beta = dT/dt$, the reaction rate in Eq. 8 gives;

$$\frac{da}{dt} = \frac{A}{\beta} e^{(-E_a/RT)} f(a) \quad (9)$$

Equations (8) and (9) are used for the calculation of thermal kinetics from thermogravimetric data. In this study, the model-free Flynn–Wall–Ozawa method was used for the calculation of activation energies of the blends [30]. Model-free methods perform an important function in terms of calculating the activation energies of biomass samples [31]. The most important advantages of model-free methods are that they are easy to use and that they prevent problems arising from kinetic model selection. With the Flynn–Wall–Ozawa method, the activation energy was calculated by using temperatures corresponding to specific conversion values at different heating rates for an independent model [32]. This method is defined by Eq. (10) [30],

$$\log \beta = \log \left(\frac{AEa}{Rg(\alpha)} \right) - 2.315 - 0.4567 \left(\frac{Ea}{RT} \right) \quad (10)$$

where β is the heating rate and A is the pre-exponential factor. E_a is the activation energy, $g(\alpha)$ is a function of the conversion, and R is the gas constant. Since $\log(\beta)$ is plotted vs. $1/T$ at any conversion rate for different heating rates, the slope of the straight line will give $0.4567 E_a/R$. Therefore, E_a is calculated based on the slope of this line.

The comprehensive combustion characteristics index (S) was also calculated using the formula below [33],

$$S = \frac{\left(\frac{dW}{dt} \right)_{\max} \left(\frac{dW}{dt} \right)_{\text{ort}}}{T_i^2 T_b} \quad (11)$$

In the equation, $(dW/dt)_{\max}$ and $(dW/dt)_{\text{mean}}$ stand for maximum and average burning rates, respectively. T_i and T_b are the ignition and burnout temperatures of the samples, respectively. High S values indicate high combustion properties for the fuels [34].

Results and discussion

Analyzing combustion characteristics and activation energies of the raw blends

Combustion characteristics of the raw blends

The TG/DTG curves of the selected blends are presented in Fig. 1. DTG is the first derivative of the TG curve, and it leads to preliminary results based on the oxidative thermal decomposition of the samples. Moreover, the height of the DTG curves also shows the rate of mass decomposition of the sample in the specified temperature ranges [35]. Previous studies noted roughly three regions in lignite-biomass blends [21]. The present study also identified three regions on the TG curves of the blends. These regions were determined based on the approximate starting and ending points of the DTG curves, which showed the thermal decomposition of organic matter/volatile substances or the loss of water in the samples [36]. The first region on the DTG curve is associated with the loss of moisture in the sample. The second region, where the highest mass loss occurred and the main oxidation started, appeared due to the oxidation and removal of volatile substances. Thermal decomposition of hemicellulose and cellulose occurred in this region as hemicellulose decomposes in the 200–300 °C temperature range. Then, decomposition of cellulose occurs between 300 and 450 °C [37]. The last region occurred due to the oxidation of the remaining char after the volatiles were removed from the sample. Lignin breaks down over a broad temperature range (250–500 °C). Therefore, oxidative breakdown of lignin in the samples also occurred in this region [27]. Furthermore, the decomposition of biomass is more prevalent in the blends, compared to the case with lignite. On the other hand, in the last region, the decomposition of lignite in addition to that of the residual biomass is observed as a prominent mechanism [21].

The ignition, peak, burnout temperatures, maximum DTG value and the comprehensive combustion characteristic index (S) were calculated for each blend (Table A.1). Ignition temperature is an important property of fuels as a certain amount of combustion is required in a gasifier to provide the energy required for drying and pyrolysis, and ultimately for the endothermic gasification reaction. Therefore, it is important to know the ignition properties of the fuels. When the temperature reaches a certain value—the ignition temperature—the rate of heat production matches or exceeds the rate of heat loss [38]. Peak temperature (PT) and burnout temperature (BT) are the typical temperatures that can be determined by looking at the DTG curves. The peak temperature on the DTG

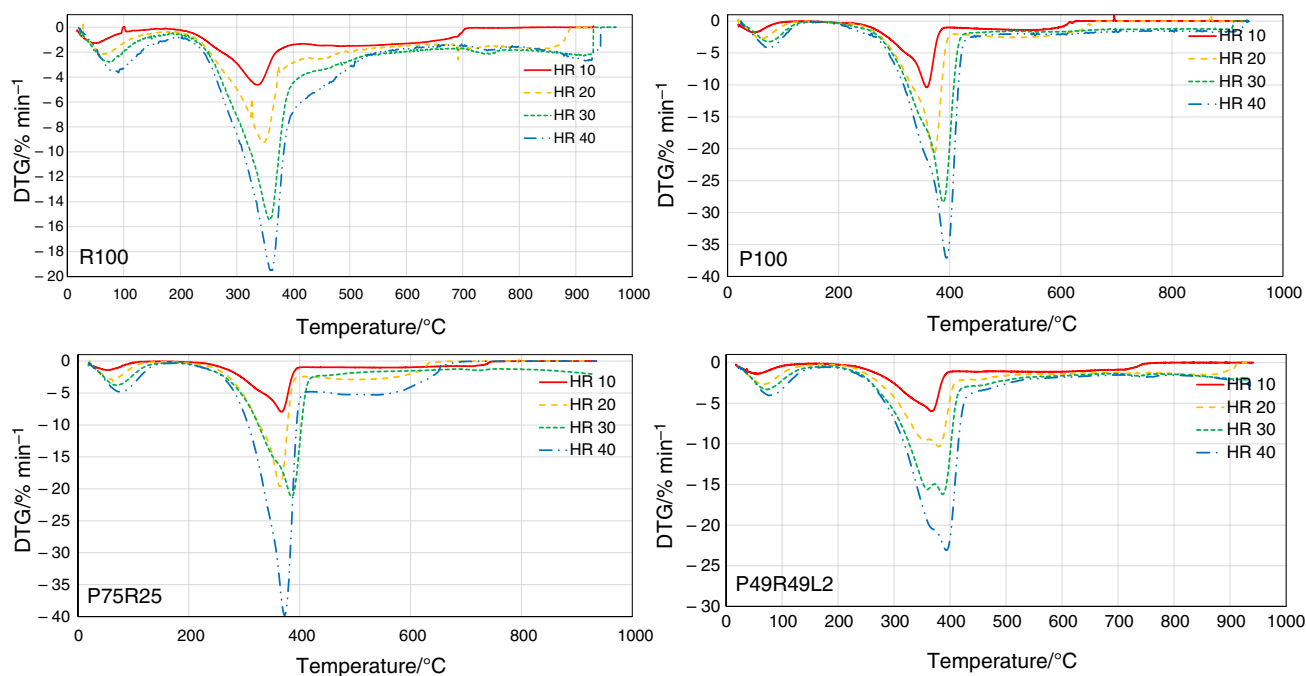


Fig. 1 DTG curves of the selected blends

curves occurred where the rate of mass loss peaked and serves as a shorthand for the flammability of the samples: low PT indicates easier combustion. On the other hand, the burnout temperature indicates the temperature at which the sample oxidation is completed [36].

According to the figures in Table A.1, the ignition temperatures of R100 and P100 were found to be 198 ± 21 °C and 220 ± 13 °C, respectively. Fuels with higher volatile matter content can ignite more easily and complete their burning in a shorter time [39]. In the study, the ignition temperatures of the blends usually increased as the lignite content in the blends increased. The average ignition temperatures of R98L2, R96L4, and R90L10 blends were 210 ± 13 °C, 214 ± 12 °C, and 218 ± 12 °C, respectively. Similarly, the average ignition temperatures of P98L2, P96L4, and P90L10 blends also increased with the addition of lignite and were found to be 225 ± 7 °C, 230 ± 12 °C, and 235 ± 11 °C, respectively. Since the volatile matter content of both pine and rose pulp was significantly higher than that of lignite (Table 1), the ignition temperatures of the blends increased as the lignite content in the blends increased. A similar situation was also with for the biomass and lignite blends, and average ignition temperatures of P49R49L2, P48R48L4, and P45R45L10 samples were recorded, respectively, as 219 ± 14 °C, 220 ± 14 °C, and 223 ± 16 °C. Looking at these figures, one can forcefully argue that both raw pine sawdust and rose pulp samples were easier to ignite compared to their blends with lignite.

The average peak temperatures in the second region where the main decomposition occurred were 374 ± 16 °C and 343 ± 8 °C for P100 and R100 samples, respectively. It is also noteworthy that, in the second region the degradation of hemicellulose and cellulose occurred. The average peak temperatures for R98L2, R96L4, and R90L10 blends were 343 ± 10 °C, 346 ± 13 °C, and 347 ± 11 °C, respectively. On the other hand, the average peak temperatures for P98L2 (377 ± 7 °C), P96L4 (378 ± 12 °C), and P90L10 (379 ± 11 °C) fuel blends showed a significant increase compared to the P100 sample (374 ± 16 °C), but did not exhibit a significant increase overall, parallel to the increase in the percentage of lignite in the blend. A noticeable increasing trend was not observed in the main thermal decomposition regions of the blends, probably due to low percentage of lignite in the blends. The average peak temperatures of the blends consisting of rose and pine were 348 ± 11 °C, 362 ± 8 °C, and 371 ± 7 °C for P25R75, P50R50, and P75R25, respectively. Therefore, it can be concluded that higher thermal decomposition temperatures were obtained as the content of pine increased in the blends.

The average burnout temperatures of the P100 and R100 samples were found to be 692 ± 76 °C and 866 ± 109 °C, respectively (see Table A.1). With the addition of lignite, the burnout temperatures of the blends containing pine increased significantly and the average burnout temperatures of the P98L2, P96L4, and P90L10 blends reached 804 ± 121 °C, 834 ± 112 °C, and 824 ± 101 °C, respectively. Even though the burnout temperatures of blends increased with addition

of the lignite, no systematic trend was observed. Similar findings were also observed during the co-combustion of biomass/coal blends. Similarly, higher or lower burnout temperatures were reported in the literature, depending on the fuel properties and blending ratios [77, 78].

Maximum DTG values were found to be $-23 \pm 11\%$ min^{-1} and $-12 \pm 7\%$ min^{-1} for the P100 and R100 samples, respectively. The maximum DTG levels decreased with the addition of lignite into the fuel blends. In other words, in this study, raw pine samples were found to be the most reactive and the easiest to burn woody fuel. The increased lignite content of the blends could cause less volatile matter formation in the oxidative environment. Accordingly, average maximum DTG values for R98L2, R96L4, and R90L10 blends were found to be $-11 \pm 5\%$ min^{-1} , $-11 \pm 6\%$ min^{-1} , and $-9 \pm 5\%$ min^{-1} , respectively. Average maximum DTG values for P98L2, P96L4, and P90L10 blends were found to be $-22 \pm 10\%$ min^{-1} , $-22 \pm 11\%$ min^{-1} , and $-20 \pm 9\%$ min^{-1} , respectively. The addition of 2% and 4% lignite by mass produced did not lead to significantly altered maximum DTG values. Although the maximum DTG is also considered to be directly related to the reactivity of the sample, the temperature at which the maximum DTG is achieved is generally considered inversely proportional to the reactivity and combustion of the sample [40]. The average maximum DTGs of the fuel blends containing both pine and rose (P25R75 ($-11 \pm 6\%$ min^{-1}), P50R50 ($-21 \pm 14\%$ min^{-1}), and P75R25 ($-19 \pm 15\%$ min^{-1})) were calculated, leading to the conclusion that the reactivity of the blends with equal amounts of rose and pine wastes was higher than those of the other two blends.

S represents the reactivity of coal combustion during the oxidation reaction. The fuels with higher S values exhibit better performance during combustion [22], as higher S values are associated with lower ignition and burnout temperatures and higher mass loss rates [41]. Therefore, coal with the highest S value is easier to ignite [22]. In this study, average S values for R98L2, R96L4, and R90L10 blends were found to be $7.41\text{E-}07 \pm 5.22\text{E-}07$, $6.47\text{E-}07 \pm 4.76\text{E-}07$, and $6.2\text{E-}07 \pm 4.58\text{E-}07$, respectively. Average S values for P98L2, P96L4, and P90L10 fuel blends were found to be $1.51\text{E-}06 \pm 1.07\text{E-}06$, $1.42\text{E-}06 \pm 1.07\text{E-}06$, and $1.15\text{E-}06 \pm 7.53\text{E-}07$, respectively. It was observed that S values decreased as more lignite was added to both pine and rose pulp blends. As the volatile content of the samples was reduced, lower S values were obtained for the samples containing lignite, compared to the raw samples. All blends containing lignite exhibited a better combustion performance compared to lignite itself. The average S values of P25R75, P50R50, and P75R25 samples containing both rose and pine were $8.14\text{E-}07 \pm 6.16\text{E-}07$, $2.39\text{E-}06 \pm 2.43\text{E-}06$, and $1.7\text{E-}06 \pm 1.89\text{E-}06$, respectively. A glance at the bigger picture

involving the S values of all blends reveals that the best combustion performance was obtained with P75R25 blend.

The flammability of the samples increased significantly with increasing heating rates. Table A.1 shows that ignition, peak, and burnout temperatures are also shifting to higher ranges as the heating rate increases, given the higher efficiency of transfer occurs at lower heating rates [42]. In the light of the results presented so far, one can forcefully argue that higher thermal decomposition temperatures are observed for all samples, with increases in heating rates due to heat transfer limitations.

Activation energies of the raw blends

In the study, the Flynn–Wall–Ozawa method was used to calculate the activation energies of raw blends. The average activation energies of the blends are presented in Table 3. The activation energies of the blends varied from 101 kJ mol^{-1} (R90L10) to 187 kJ mol^{-1} (R100). According to data presented in Table 3, the activation energies of both raw pine and raw rose samples and blends thereof decreased with the addition of lignite. The highest activation energies were recorded with raw rose and pine samples. No study examining the thermal kinetic constants and combustion characteristics of rose pulp was found in the literature so far. However, the E_a values obtained for the biomass blends in this study were found to be similar to the values noted in the literature for other lignocellulose biomasses and their blends with coal, including wood ($96\text{--}147\text{ kJ mol}^{-1}$) [43], pine wood ($91.8\text{ to }175.8\text{ kJ mol}^{-1}$) [44], wood chips, nutshells ($139\text{--}155\text{ kJ mol}^{-1}$) [45], Turkish Beypazari Lignite

Table 3 Calculated activation energies of the blends

Raw blend/mass %	Activation energy/kJ mol^{-1}
R100	187
R98L2	164
R96L4	144
R90L10	101
P100	178
P98L2	174
P96L4	157
P90L10	133
P25R75	175
P50R50	172
P75R25	158
P49R49L2	183
P48R48L4	174
P45R45L10	147
L100	91

and their blends ($90.9\text{--}215.3\text{ kJ mol}^{-1}$) [46], branched millet and beet root ($43.6\text{--}133.3\text{ kJ mol}^{-1}$) [47], pine sawdust, coal, and their blends ($143.1\text{--}167.7\text{ kJ mol}^{-1}$) [34].

Analysis of combustion characteristics and activation energies of the torrefied blends

Combustion characteristics of the torrefied blends

The TG/DTG curves of the selected torrefied blends are given in Fig. 2. In the study, the average ignition temperatures of the torrefied TP100 and TR100 samples were found to be $228 \pm 7\text{ }^\circ\text{C}$ and $230 \pm 8\text{ }^\circ\text{C}$, respectively (Table A2). Therefore, the ignition temperatures of the torrefied pine and rose samples increased about $8\text{ }^\circ\text{C}$ and $32\text{ }^\circ\text{C}$, respectively, when compared against their raw form. Because of the degradation of organic matter (e.g., hemicellulose) in the biomass, the ratio of the lignin content rich in carbon increases during torrefaction [48]. Agricultural residues have relatively higher hemicellulose content than that of woody biomass [49, 50]. Such higher hemicellulose content of the rose pulp sample compared to the pine-based sample is most probably the reason why the carbon content of the rose pulp sample increased further after the torrefaction, and higher ignition temperatures were observed for rose pulp samples compared to pine samples. A similar trend was also observed with the addition of lignite into the torrefied rose pulp samples (see Table A2). Accordingly, higher ignition temperatures were observed for TR98L2 ($244 \pm 11\text{ }^\circ\text{C}$), TR96L4 ($254 \pm 6\text{ }^\circ\text{C}$), and TR90L10 ($258 \pm 5\text{ }^\circ\text{C}$) blends compared to the case

with torrefied pine and lignite blends –TP98L2 ($227 \pm 8\text{ }^\circ\text{C}$), TP96L4 ($235 \pm 9\text{ }^\circ\text{C}$), and TP90L10 ($238 \pm 3\text{ }^\circ\text{C}$). Therefore, the highest ignition temperature was that of TR90L10, followed by lignite ($259 \pm 14\text{ }^\circ\text{C}$). Again, a similar trend was observed for TP49R49L2, TP48R48L4, and TP45R45L10 samples, the ignition temperatures of which were recorded as $235 \pm 8\text{ }^\circ\text{C}$, $239 \pm 7\text{ }^\circ\text{C}$, and $243 \pm 8\text{ }^\circ\text{C}$, respectively. On the other hand, the average ignition temperature of fuels consisting of both pine and rose decreased with the increase in the content of pine in the blends, with ignition temperatures of the TP25R75, TP50R50, and TP75R25 blends being $236 \pm 4\text{ }^\circ\text{C}$, $236 \pm 92\text{ }^\circ\text{C}$, and $231 \pm 47\text{ }^\circ\text{C}$, respectively. Against this background, it can be concluded that it is easier to ignite the raw blends compared to their torrefied forms, because fuels with high volatile matter content are easier to ignite and take a shorter time to burn [39]. As seen in Tables A1 and A2, since lignite was poorer in terms of volatile matter compared to both raw pine and rose pulp, the ignition temperatures have increased in the blends of both raw and torrefied biomass with lignite.

In this study, the peak temperatures of torrefied blends were also found to be slightly higher than those of their raw forms, probably due to the carbonization of pine and rose pulp with torrefaction [4]. The mean peak temperatures in the second region were recorded as $377 \pm 9\text{ }^\circ\text{C}$ and $344 \pm 7\text{ }^\circ\text{C}$ for the TP100 and TR100 samples, respectively. The average peak temperatures of TR98L2, TR96L4, and TR90L10 blends, in turn, were, respectively, $344 \pm 10\text{ }^\circ\text{C}$, $347 \pm 8\text{ }^\circ\text{C}$, and $351 \pm 11\text{ }^\circ\text{C}$. The average peak temperatures of TP98L2, TP96L4, and TP90L10 blends were recorded

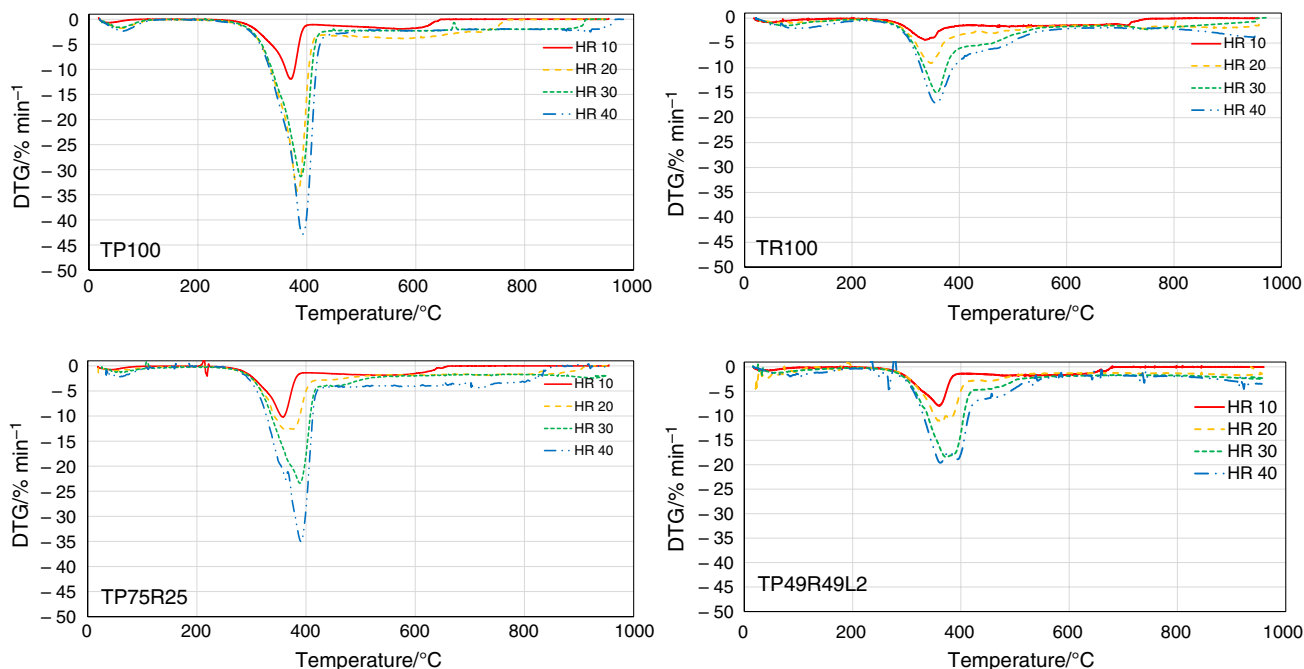


Fig. 2 DTG curves of the selected torrefied blends

as 378 ± 15 °C, 382 ± 13 °C, and 383 ± 14 °C, respectively. Therefore, like the case with raw blends, higher peak temperatures were observed with the addition of lignite into the torrefied biomass. Table A2 shows that the peak temperatures in the third region where char combustion occurred were generally more affected by the torrefaction process compared to measured peak temperatures in the second region. The result of the torrefaction process for this region was higher peak temperatures.

Average burnout temperatures were found to be 805 ± 134 °C and 882 ± 98 °C for TP100 and TR100, respectively. The burnout temperatures increase with the addition of lignite to the torrefied pine and the average values for TP98L2, TP96L4, and TP90L10 samples reached 806 ± 83 °C, 825 ± 154 °C, and 839 ± 68 °C, respectively. The burnout temperatures of torrefied blends containing rose pulp also increased with the addition of lignite, with average values being measured as 872 ± 8 °C, 890 ± 54 °C, and 893 ± 90 °C for TR98L2, TR96L4, and TR90L10 blends, respectively. It has been observed that the burnout temperatures of the torrefied samples have shifted to higher temperatures with the torrefaction process, compared to their raw forms. Similar findings were also reached for both biomass and lignite-biomass blends. Consequently, due to devolatilization and depolymerization [23], higher ignition and burnout temperatures were observed with torrefaction of blends, compared to their raw counterparts.

The maximum DTG value of the Elbistan lignite ($2.62 \pm 1\%$ min⁻¹), which is suitable for pulverized combustion and which has a low calorific value, was found to be the lowest DTG value when compared with other blends. In the study, torrefaction led to an increase in the maximum DTG values of the blends in general, and the maximum DTG values of the torrefied TP100 and TR100 samples were found to be $-29.81 \pm 13\%$ min⁻¹ and $-11.80 \pm 6\%$ min⁻¹, respectively. Furthermore, the reactivity of the torrefied pine samples was found to be higher than that of rose pulp samples. This point is also supported by the lower ignition, peak and burnout temperatures of torrefied pine samples compared to those of the torrefied rose pulp samples [51]. Average maximum DTG values for TR98L2, TR96L4, and TR90L10 blends were $-12.40 \pm 7\%$ min⁻¹, $-12.72 \pm 7\%$ min⁻¹, and $-11.79 \pm 6\%$ min⁻¹, respectively. The average maximum DTG values for P98L2, P96L4, and P90L10 blends were found to be $-26.57 \pm 13\%$ min⁻¹, $-26.84 \pm 13\%$ min⁻¹, and $-24.90 \pm 910\%$ min⁻¹, respectively. Thus, the addition of 2% and 4% lignite by mass led to DTG values of the torrefied blends as well as raw blends approaching maximum. Moreover, the pine-lignite blends were more reactive compared to rose pulp and lignite blends and that the highest reactivity was observed with the blends with the highest pine content.

Table 4 Calculated activation energies of the torrefied blends

Torrefied blend/mass %	Activation energy/kJ mol ⁻¹
TR100	148
TR98L2	136
TR96L4	129
TR90L10	120
TP100	172
TP98L2	149
TP96L4	124
TP90L10	118
TP25R75	103
TP50R50	135
TP75R25	123
TP49R49L2	128
TP48R48L4	124
TP45R45L10	121
L100	91

The lowest S value measured in the study was that of lignite ($1.01\text{E-}07$). When the S values of the torrefied blends were examined, it was seen that the comprehensive combustion index has improved with the torrefaction process. Moreover, S values were found to decrease with the addition of lignite into the torrefied biomass, with mean S values for TR98L2, TR96L4, and TR90L10 blends being $7.85\text{E-}07 \pm 7.40\text{E-}07$, $7.05\text{E-}07 \pm 6.46\text{E-}07$, and $6.99\text{E-}07 \pm 6.23\text{E-}07$, respectively. Mean S values for TP98L2, TP96L4, and TP90L10 blends, again, were found to be $1.52\text{E-}06 \pm 1.51\text{E-}06$, $1.45\text{E-}06 \pm 1.41\text{E-}06$, and $1.21\text{E-}06 \pm 1.15\text{E-}07$, respectively. Therefore, inverse correlation exists between S values and the amount of lignite added into torrefied blends. Moreover, all torrefied blends with lignite had a better combustion performance compared to lignite itself. The respective S values for TP25R75, TP50R50, and TP75R25 samples were also found to be $8.16\text{E-}07 \pm 6.83\text{E-}07$, $1.7\text{E-}06 \pm 2.41\text{E-}06$, and $2.17\text{E-}06 \pm 2.85\text{E-}06$. Accordingly, it can be concluded that higher S values were obtained with increase of torrefied pine content in the blends.

A review of Table A2 reveals that the peak, ignition, burnout temperatures and maximum DTGs were increased by the torrefaction process, following a trend comparable to the one observed in previous studies [33, 52]. The fuel performance of the blends during combustion has been improved as a result of the torrefaction process. At the same time, adding lignite to the torrefied samples significantly improved the burning behavior of rose pulp and pine, compared to their raw forms.

Activation energies of the torrefied blends

The activation energies of the torrefied materials were also calculated and are presented in Table 4. The average activation energy of Elbistan lignite (91 kJ mol^{-1}) was found to be close to the calculated activation energies of other low quality Turkish lignites, namely Orhaneli ($84.74 \text{ kJ mol}^{-1}$), Tunçbilek (34.6 kJ mol^{-1}), Elbistan ($108.6 \text{ kJ mol}^{-1}$), and Soma (25.3 kJ mol^{-1}) [53]. The average activation energies of the blends, on the other hand, were found to be between 172 kJ mol^{-1} (TP100) and 103 kJ mol^{-1} (TP25R75). As shown in the table, all thermally treated blends with the exception of TR90L10 saw a significant decrease in their activation energies compared to the activation energy levels of their raw forms. Moreover, the addition of lignite into both raw and torrefied blends led to an observed decrease in the activation energies of the blends. Similar results are reported in the literature [40, 42, 54]. The fall in the activation energies of the blends containing torrefied biomass is due to dehydration and decarboxylation of the biomass during the torrefaction process [55].

In the literature, synergy and additivity were observed between various biomass and lignite samples during the co-combustion studies [32, 34, 39, 56–68]. However, the studies on synergy in the co-combustion of coal/lignite and torrefied biomass are still in their infancy. Although a number of studies suggest a synergy between torrefied biomass and lignite during co-combustion [69, 70], Mi et al. (2016) found that there was no synergy between torrefied bamboo, torrefied wood and their blends under the oxidative atmosphere [41].

Rizkiana et al. (2020) also found no interaction between coal and biomass on solid yield during co-torrefaction [71]. Therefore, more studies should be performed to understand the effect of synergy on thermal decomposition of various torrefied biomass and lignite/coal samples during the co-combustion process, to successfully design and operate thermo-chemical processes.

In the present study, to understand the combustion of raw/torrefied biomass with the addition of lignite or other biomass, first of all, DTG curves of the blends were calculated using the formula [69];

$$(dw/dt)_{\text{calculated}} = x_1(dw/dt)_1 + x_2(dw/dt)_2 \quad (12)$$

where x_1 and x_2 represent mass fractions of the pine/rose pulp/lignite in the blend and $(dw/dt)_1$ and $(dw/dt)_2$ represent the corresponding mass loss rates ($\% \text{ min}^{-1}$). Experimental and calculated DTG curves for both selected raw and torrefied blends are given in Fig. 3. As indicated in the figure, the experimental and calculated mass loss figures did not match, indicating synergy between raw/torrefied biomass and lignite.

Secondly, the average activation energies of the blends were also plotted against mass% of the biomass in the blends [27]. A synergistic effect was observed for both raw rose pulp/lignite and torrefied rose pulp/lignite blends (Fig. 4). Furthermore, the synergistic effect was found to be stronger for raw rose pulp/lignite blends ($R^2 = 0.488$) compared to torrefied rose pulp/lignite blends ($R^2 = 0.8371$). As in the case with rose pulp samples, the synergistic effect was also observed for raw pine/lignite and torrefied pine/lignite

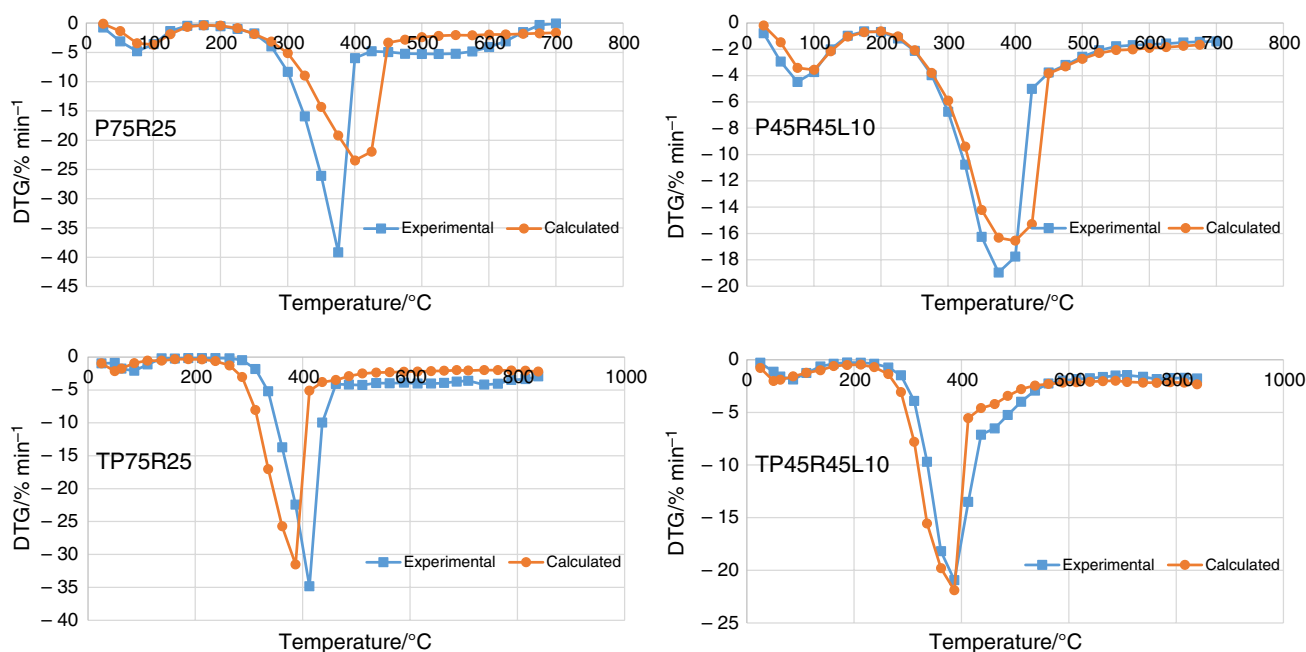
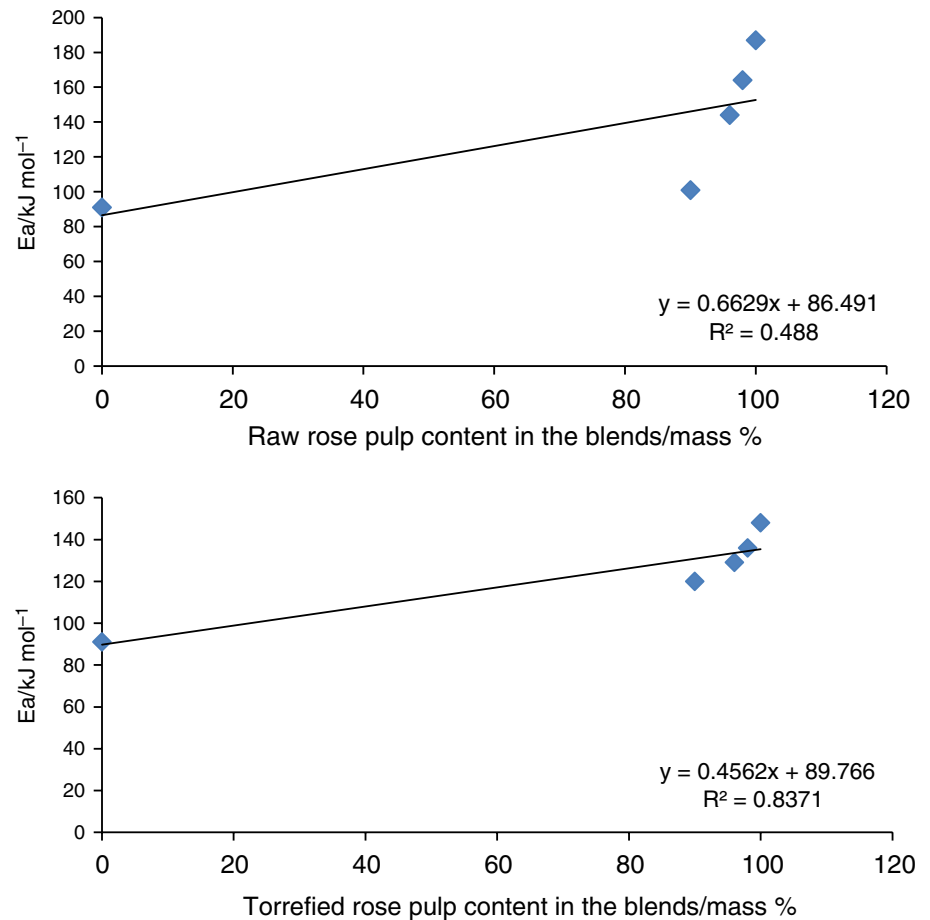


Fig. 3 Experimental and calculated DTG curves of selected raw and torrefied blends at a heating rate of $40 \text{ }^\circ\text{C min}^{-1}$

Fig. 4 Average activation energies of the blends vs mass % of the raw and torrefied rose pulp in the blends



blends. However, in this case the effect was stronger with the torrefied pine/lignite blends ($R^2=0.5916$) compared to raw pine/lignite blends ($R^2=0.8242$) (Fig. 5). Decomposition of agricultural residues is much faster compared to that of woody biomass, because of the higher hemicellulose content of the former [49, 50]. Different synergistic effects were observed during the decomposition of agricultural versus woody residues, probably due to different proportions of hemicellulose, cellulose and lignin content thereof [72].

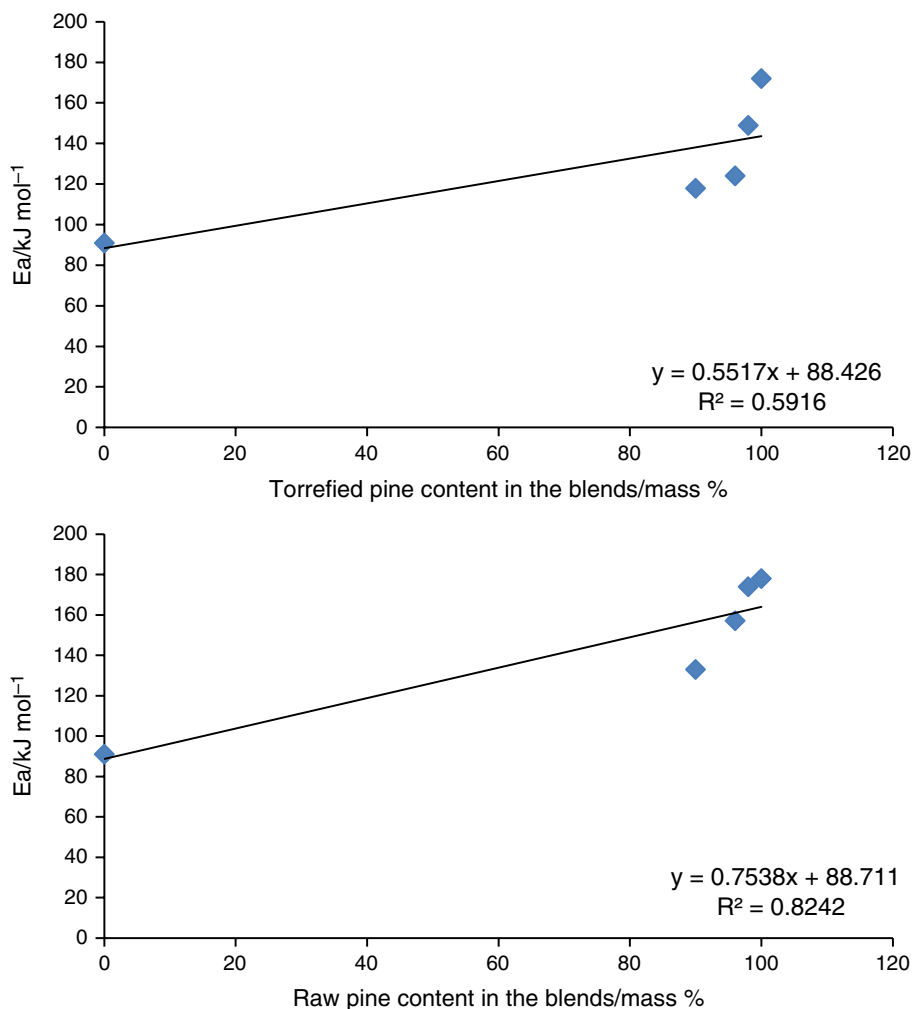
And finally, to investigate the effect of blending on the co-combustion process for all blends, the average activation

energies of both raw and torrefied blends were calculated using the formula [69];

$$E_{\text{exp}} = m_b \times E_b + m_l \times E_c \quad (13)$$

where m_b and m_l refer to the mass% of the biomass and lignite in the blends, respectively. E_b and E_c represent the activation energies (kJ mol⁻¹) of the biomass and lignite. Nearly all of the experimental activation energies of raw and torrefied blends (Fig. 6) were found to be higher than the corresponding calculated activation energies. Therefore,

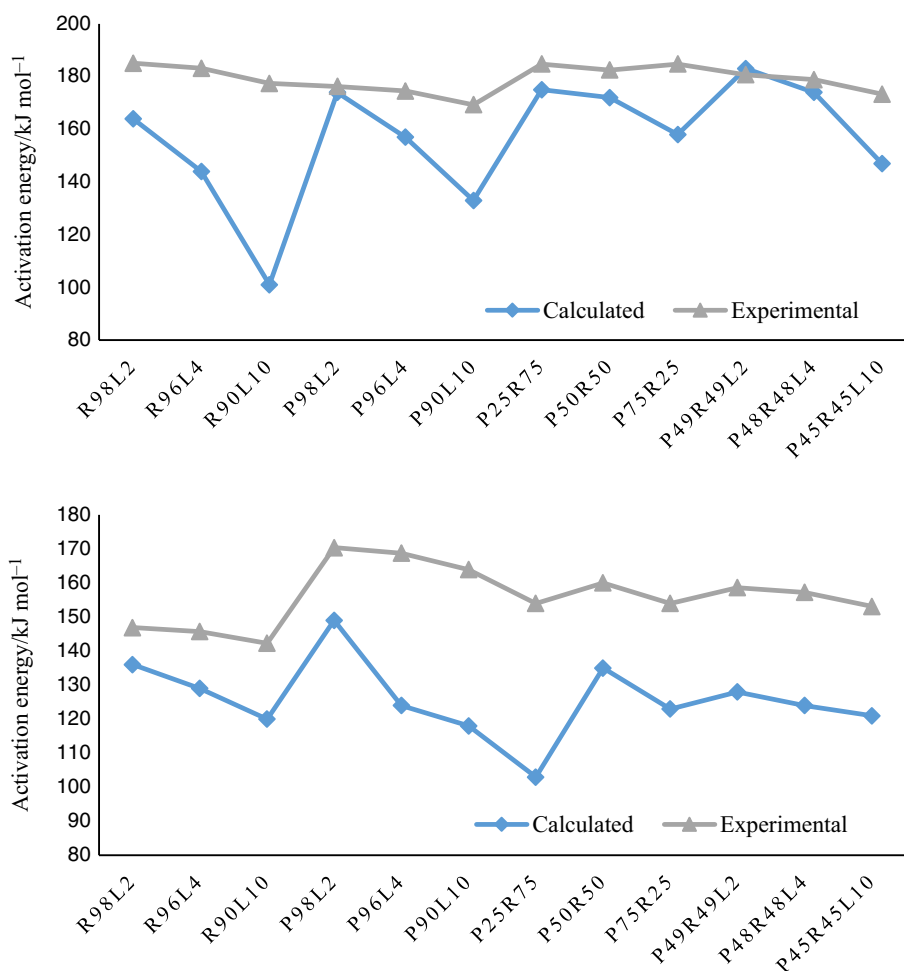
Fig. 5 Average activation energies of the blends vs mass % of the raw and torrefied pine in the blends



a synergy was observed for both raw biomass/lignite and torrefied biomass/lignite blends in the co-combustion process [69]. Moreover, with increasing lignite content in the blends, synergistic effects became stronger for both raw

and torrefied blends. Besides, in addition to biomass/lignite blends, the synergistic effect was also observed for raw and torrefied rose pulp/pine blends during the co-combustion process.

Fig. 6 Average activation energies of experimental and calculated both raw and torrefied blends



Conclusions

In the study, raw pine and rose pulp blends were found to have the lowest ignition and burnout temperatures among all. Therefore, the raw blends were identified as the most reactive fuel blends. The peak temperatures, ignition temperatures, final combustion temperatures and comprehensive combustion performances of the blends examined also increased with torrefaction. As a result, the fuel/combustion performance of raw biomass was observed to be improved by the torrefaction process. At the same time, by adding lignite to the torrefied biomass, the burning behaviors of both rose pulp and pine were significantly improved compared to their raw forms. Therefore, the results of this study can be useful in the development of thermochemical systems utilizing raw/torrefied wastes of pine and rose pulp. Moreover, the findings of the study also emphasize the potential of blending waste biomass with low quality local lignite during the disposal of waste, in the context of environmentally friendly energy production.

The average activation energies of raw pine, rose pulp, and Elbistan lignite were calculated to be 178, 187, and

91 kJ mol⁻¹, respectively. With torrefaction, average activation energies of pine and rose pulp samples decreased to 172 and 148 kJ mol⁻¹, respectively. The activation energies of the raw pine/lignite and torrefied pine/lignite blends varied between 133–174 kJ mol⁻¹ and 118–149 kJ mol⁻¹, respectively. The activation energies of the raw rose pulp/lignite and torrefied rose pulp/lignite blends varied between 101–164 kJ mol⁻¹ and 120–136 kJ mol⁻¹, respectively. Moreover, the activation energies of pine/rose pulp blends also decreased as the amount of rose pulp in the blend increased. It has also been observed that there was a synergistic effect during the co-combustion of both raw and torrefied biomass with lignite. Since the kinetic parameters of raw/torrefied biomass and lignite or their blends are important for the proper design and operation of thermochemical systems, it is recommended to design co-combustion systems taking such synergetic effects between raw/torrefied biomass and lignite into account. Against this background, further studies are required to examine the effects of synergy during the thermal conversion of various biomass/lignite blends, especially with reference

to the design and operation parameters of co-processing systems.

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Authors' contributions SA was involved in the methodology, formal analysis, and software. MC contributed to writing—reviewing and editing. SY helped in the conceptualization, project administration, supervision, data curation, and writing—original draft preparation.

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Declarations

Conflict of interest The authors have no conflicts of interest to declare that are relevant to the content of this article.

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