EFFECT OF TORREFACTION PROCESS ON THE COMBUSTION AND PYROLYSIS BEHAVIOUR OF RICE STRAW

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Abstract

Biomass is widely recognized as a renewable energy source that can be used as a substitute for fossil fuels. In order to comprehend and simulate combustion of biomass in large-scale furnaces and to effectively design and operate conversion systems, it is crucial to develop a comprehensive understanding of the pyrolysis characteristics and combustion kinetics. The objective of this study is to conduct a comparative analysis of the thermal properties and kinetic behaviour of raw rice straw and torrefied rice straw under both inert and oxidative atmospheres using thermogravimetric analyser. The results revealed that the lower temperature of initial combustion (T_{ic}) for rice straw was found to be 249 °C, while T_{ic} for torrefied rice straw was 252 °C. T_{ic} serves as a critical parameter for ensuring the safe storage of both raw and torrefied rice straw. The Burnout temperature (T_b) values for both raw and torrefied rice straw fell within the range of 468 °C to 470 °C, indicating that all samples achieved complete combustion at the same temperature. The results showed that the degree of thermal decomposition is higher for the torrefied rice straw, which suggested that the sample was less reactive than raw rice straw. The findings of this study can facilitate the design of combustion reactors and the determination of optimal operating conditions for such reactors.

Keywords: Torrefaction, Rice straw, Pyrolysis, Combustion, Thermogravimetric analysis (TGA)

INTRODUCTION

Global warming has increasingly been paid particular attention to by scholars and policymakers worldwide. The use of fossil fuel is one of the most crucial causes of this issue. Despite the aforementioned environmental concerns, the global energy demand has been gradually increasing every year. About 80% of the energy consumed comes in from of fossil energy such as oil, coal and natural gas. Many efforts are focusing on using renewable energy in order to reduce the use of fossil fuels. Biomass is one of the most common renewable energy used as a substitution of fossil energy. Biomass is defined as matter originating from living plants, including tree stems, branches, leaves as well as residues from agricultural harvesting and processing of seeds or fruits (Wigley, Yip & Pang, 2016). Different types of biomass are used today including lignocellulosic biomass, solid waste, landfill gas and bio gas. Among the different types of biomass, lignocellulosic biomass is widely used to produce energy. The supply of biomass globally shows that primary solid biofuels or lignocellulosic biomass is the most widely used (World Bioenergy Association, 2021).

Thailand, characterized as an agricultural country, dedicates 51% of its total agricultural area to rice cultivation. The production of rice yields a significant agricultural waste known as rice straw, amounting to approximately 35 million tons per year. Unfortunately, some farmers resort to burning rice straw as a means to facilitate plowing and prepare the soil for the subsequent cultivation season. However, this practice not only squanders energy resources but also contributes to air pollution. Therefore, the use of rice straw biomass to produce energy can help reduce the previous mentioned problems. However, biomass still exhibits inferior fuel properties compared to coal (Prins, Ptasinski & Janssen, 2006). One of the major drawbacks is its high moisture content, which gives rise to various negative consequences, including: (1) difficulties during pre-treatment, preparation, and upgrading processes; (2) reduced calorific value and grinding capacity, poor ignition, decreased combustion efficiency, and prolonged residence time in combustion units. Additionally, biomass suffers from a low energy density, characterized by both low bulk density and heating value (Vassilev, Vassileva, & Vassilev, 2015). To gain these advantages, torrefaction is one of the pretreatment processes used to improve fuel properties of biomass. Torrefaction is generally carried out in the temperature range of 200-300°C in an inert atmosphere at slow heating rate. After the process, torrefied biomass has lower moisture content, higher energy density and heating value, good hydrophobicity and is easier to grind than raw biomass (Cahyanti, Doddapaneni & Kikas, 2020; Prins, Ptasinski & Janssen, 2006). The study conducted by Wiangthong et al. (2023) focused on investigating the effects of torrefaction severity on rice straw. They examined three different levels of torrefaction severity, which resulted in mass losses of 10%, 20%, and 30%. The findings of their research indicate that torrefaction has a significant positive impact on the fuel properties of rice straw. Specifically, the researchers observed that rice straw subjected to the most intense torrefaction process (30% mass loss) exhibited the most favorable fuel properties. This included higher levels of fixed carbon and carbon content, as well as a higher high heating value. Additionally, this torrefied rice straw had the lowest humidity content, making it suitable for further energy production.

Thermogravimetric analysis (TGA) is a widely employed technique used to rapidly examine and compare the thermal events and kinetics associated with the combustion and pyrolysis of solid raw materials like coal and biomass (Candelier et al., 2016; Ramírez, García-Torrent, & Tascón, 2010; Sedlmayer et al., 2018). It allows for the measurement of mass loss as a function of time and temperature, enabling the tracking of the temperatures at which sample combustion or decomposition reactions initiate. Additionally, TGA curves can be subjected to quantitative analysis to obtain kinetic parameters.

Understanding the pyrolysis characteristics and combustion kinetics is crucial for comprehending and modelling combustion in industrial-scale furnaces, as well as for designing and operating conversion systems. Gaining a solid understanding of biomass decomposition during thermochemical conversion is essential for the development of efficient processing technologies.

The objective of this study is to compare the thermal properties and kinetic behaviour of raw rice straw and torrefied rice straw in both an inert and oxidative atmosphere using a thermogravimetric analyser to investigate the pyrolysis and combustion characteristics of rice straw before and after undergoing torrefaction treatment.

MATERIALS AND METHODS

1. Experimental Apparatus

Thermogravimetric Analyzer (TGA/DSC 1 Mettler-Toledo) is used to measures mass while the temperature of a sample is changed over time. Thermogravimetric Analyzer was used to prepare torrefied biomass and to carry out the study of combustion and pyrolysis behaviour of raw and torrefied biomass.

2. Sample preparation

In this experiment, dry rice straw was chopped into less than 2 cm in length and then was grounded in a ball mill (Retsch PM100) and sieved to obtained particle size less than 100 µm. The sample was dried in the oven at 105 °C for 24 hours to remove the moisture.

The prepared sample was divided into two parts. The first part was kept to study the combustion and pylorosis behavior of raw rice straw. The second portion of rice straw was used to improve fuel properties by torrefaction process. The rice straw was subjected to the torefraction process until the initial weight was reduced by 30%, denoted as Torrefied rice straw in this manuscript, before further studies on its combustion and pylorosis behaviors. Table 1 shows the properties of rice straw before and after treating by torrefaction process. The torrefaction conditions were selected after the study by Wiangthong et al. (2023), it was found that the torrefaction process with the highest intensity, 30% mass loss, demonstrated the best fuel properties for torrefied rice straw. These properties encompassed elevated levels of fixed carbon and carbon content, along with a greater high heating value.

Table 1 Properties of raw rice straw and torrefied rice straw (Wiangthong et al., 2023)

Properties		Raw rice straw	Torrefied rice straw
Proximate analysis	Moisture (%, as received)	8.77±0.06	1.19±0.07
	Volatile matter (%, dry basis)	68.82±0.07	53.18±0.08
	Fixed carbon (%, dry basis)	19.21±0.02	28.48±0.04
	Ash (%, dry basis)	11.97±0.09	18.34±0.11
Ultimate analysis	Nitrogen (%)	1.07±0.02	1.32±0.04
	Carbon (%)	40.23±0.11	46.23±0.11
	Hydrogen (%)	7.32±0.03	4.95±0.07
	Oxygen (%) *by difference	51.38±0.10	47.50±0.14
Higher Heating Value (MJ/kg, dry basis)		16.55	18.88

3. Methodology

The study consisted of 2 parts, the first part was to study the combustion behaviour and the second part was to study the pyrolysis behaviour of raw and torrefied rice straw.

3.1 Combustion behaviour

About 10 mg of raw rice straw or torrefied rice straw were placed in a 150 µl alumina crucible and heated from room temperature to 800 °C at 5°C.min^{-1} heating rate under air with the flow rate of 50 mL.min^{-1} at STP. The mass of rice straw reduced was recorded to further study the combustion behaviour.

3.2 Pyrolysis behaviour

About 10 mg of raw rice straw or torrefied rice straw were placed in a 150 µl alumina crucible and heated from room temperature to 800 °C at 5 °C.min^{-1} heating rate under nitrogen with the flow rate of 50 mL.min^{-1} at STP. The mass of rice straw reduced was recorded to further study the pyrolysis behaviour.

The tests were repeated three times with similar results, the average of the mass evolution curves was used to determine the kinetic parameter.

4. Kinetic theory

The application of thermogravimetry (TG) analysis is extensive in determining kinetic parameters related to combustion and pyrolysis reactions, as this method is not time-consuming. The combustion and pyrolysis reactions can be described using the Arrhenius formula. Empirical models are commonly employed to calculate the kinetic parameters, which fall into two main categories: the model-free isoconversional method and the model-fitting method. The model-free isoconversional method allows for the calculation of kinetic parameters without assuming a specific reaction model, making it a reliable approach for determining the activation energy of thermally activated reactions (Liu et al., 2021). However, this method necessitates experimental data at various heating rates to access the kinetic parameters. On the other hand, the model-fitting method is primarily utilized for assessing the reaction order and the frequency factor. Unlike the model-free isoconversional method, this approach only requires data from a single heating rate to determine the kinetic parameters. In this study, the model-fitting method was used to determine the kinetic parameters as followed Equation (1):

$$\ln\left[\frac{G(\alpha)}{T^2}\right] = \ln\left[\frac{AR}{\beta E_a}\right] - \frac{E_a}{RT} \qquad (1)$$

with

$$G(\alpha) = -\ln(1-\alpha) \qquad (n=1)$$

$$G(\alpha) = \frac{(1-\alpha)^{1-n} - 1}{n-1} \qquad (n\neq1)$$

$$\alpha = \frac{m_0 - m_t}{m_0 - m_\infty}$$

Where,

α is the conversion from weight loss data

m_0, m_t, m_∞ are the sample mass at initial, at time t and at the end of reaction

T is the reaction temperature (K)

R is the gas constant (J/mol.K)

A is the frequency factor (min^{-1})

E_a is the activation energy (J/mol)

β is the heating rate (β=dT/dt).

To determine the activation energy of reaction (E_a), the curve of the experimental results was plotted, with plot of $\ln((G(\alpha))/T^2)$ versus 1/T. The slope and intercept of the line can be obtained as the activation energy and frequency factor.

5. Method to access Temperature of Initial Combustion (T_{ic}), Temperature of Maximum Weight Loss rate (T_{mwl}) and Burnout Temperature (T_b)

The experimental results were used to determine the temperature of maximum weight loss rate (T_{mwl}), temperature of initial combustion (T_{ic}) and burnout temperature (T_b)

5.1 T_{mwl} indicates the reactivity of the tested sample. This temperature represents the point at which the highest amount of volatile matter is released during the pyrolysis process. As the temperature increases, the reactivity of the sample decreases. The T_{mwl} is identified as the temperature corresponding to the maximum weight loss rate on the DTG curve, as shown in Figure 1.

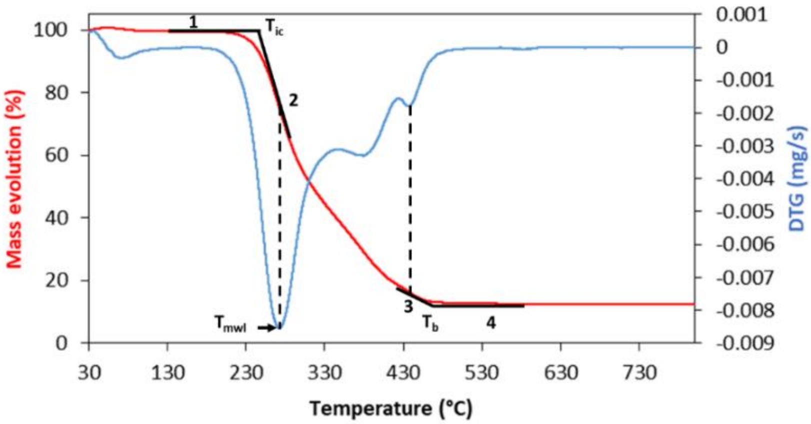

Figure 1 Determination of temperature of initial combustion (T_{ic}), temperature of maximum weight loss (T_{mwl}) and burnout temperature (T_b)

5.2 T_{ic} also referred to as the ignition temperature, signifies the temperature at which the combustion process begins. It provides insight into the material's combustion characteristics, indicating that materials with higher reactivity will initiate combustion at lower temperatures. T_{ic} offers a clear understanding of the combustion behaviour.

5.3 T_b refers to the temperature at which the substance's mass is fully burned and remains constant (Fadhil et al, 2014). Higher burnout temperatures indicate increased difficulty in achieving complete combustion, requiring longer residence times or higher temperatures (Rostam-Abadi, Debarr, & Chen, 1990).

T_{ic} and T_b were determined using the mass evolution and derivative thermogravimetric (DTG) curves under air exposure. The intersection method was used to determine these parameters (Lu & Chen 2015). The T_{ic} is determined by locating the intersection of two tangent lines at point 1 and point 2, as depicted in Figure 1. Point 1 corresponds to the beginning of the devolatilization stage, while point 2 is identified as the intersection formed by a vertical line originating from the first DTG peak and crossing the mass evolution curve. The T_b is determined by locating the intersection of two tangent lines at point 3 and point 4, as illustrated in Figure 1. Point 3 is defined as the intersection formed by a vertical line originating from the last DTG peak and crossing the mass evolution curve. Point 4 is identified as the point where the mass evolution curve stabilizes or reaches a steady state.

RESULTS

1. Pyrolysis behaviour

Figure 2 illustrates the mass evolution and DTG curves. The mass evolution curve indicated that the thermal decomposition follows the pattern observed in biomass pyrolysis. Typically, the mass evolution profile of biomass pyrolysis can be divided into three stages: drying, pyrolysis, and carbonization (Liu et al., 2021). According to Table 2, T_{mwl} was 303°C for raw rice straw and 301°C for torrefied rice straw. At the end of the reaction, the solid residue remaining from raw and torrefied rice straw is approximately 42% and 52%, respectively.

The devolatilization of raw and torrefied rice straw begins at temperatures of 282°C and 288°C, respectively, as determined by T_i. To calculate the kinetic parameters of the pyrolysis stage, it was assumed that the reaction follows a first-order model (n=1) (Liu et al., 2021). The activation energy (E_a) and frequency factor (A) were determined and presented in Table 2. The linear correlation coefficients range from 0.95 to 0.96, indicating that the linear models effectively explain the variability of the data around the mean. The fitting equations for determining the kinetic parameters were also provided in Table 2. The pyrolysis activation energy of raw and torrefied rice straw was found to be 116 $kJ.mol^{-1}$ and 129 $kJ.mol^{-1}$, respectively.

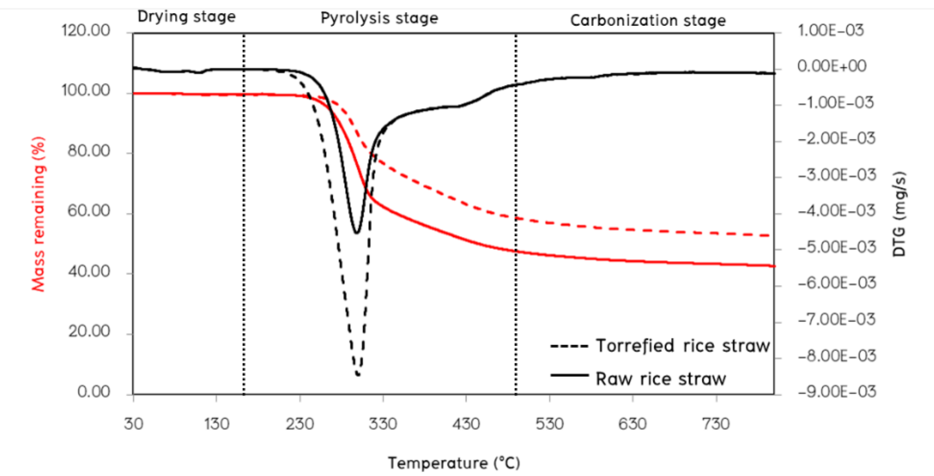

Figure 2 Mass evolution and DTG curves of raw and torrefied rice straw during pyrolysis behaviour study

2. Combustion behaviour

Figure 3 displayed the mass evolution and DTG curves. The observed thermal decomposition in both curves aligns with the pattern seen in biomass combustion. The mass evolution profile can be divided into three stages: drying, devolatilization, and combustion (Liu et al., 2021). Analysis of the DTG curves revealed the presence of three peaks for both raw and torrefied rice straw. Peak 1, which corresponds to T_{mwl}, was identified at 283 °C for raw rice straw and 290 °C for torrefied rice straw. Peak 2 and peak 3 were consistent between the two samples, occurring at 405 °C and 450 °C, respectively. T_{ic} was found at 249 °C for raw rice straw and 252 °C for torrefied rice straw. T_b ranged from 468 °C to 470 °C for raw and torrefied rice straw.

Table 3 presents the determined kinetic parameters for the devolatilization and combustion stages. All the linear correlation coefficients exceeded 0.95, indicating strong linear relationships. The E_a for the devolatilization stage was calculated as 127 $kJ.mol^{-1}$ for raw rice straw and 117 $kJ.mol^{-1}$ for torrefied rice straw. As for the combustion stage, the E_a values were determined as 145 $kJ.mol^{-1}$ for raw rice straw and torrefied rice straw.

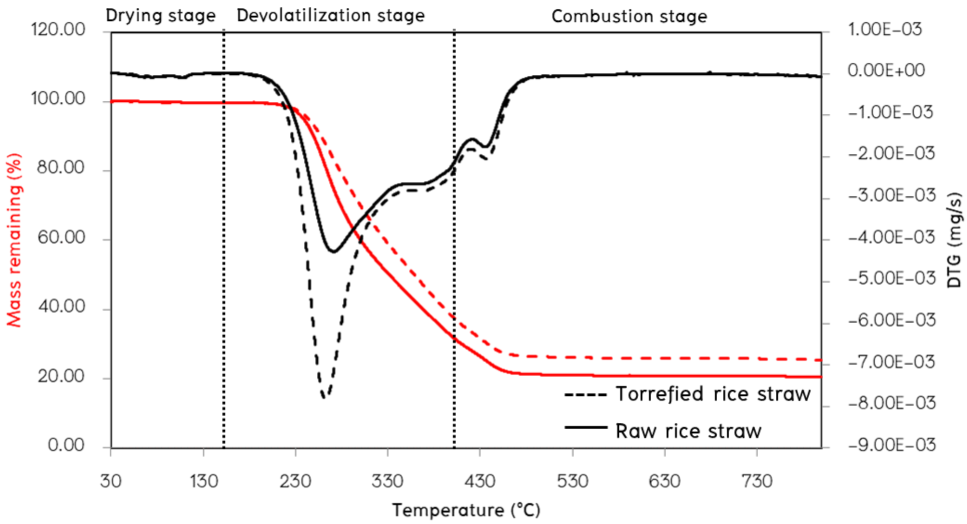

Figure 3 Mass evolution and DTG curves of raw and torrefied rice straw during combustion behaviour study

Table 2 Characteristic and kinetic parameters of pyrolysis behaviour for raw and torrefied rice straw

Samples	Fitting equation	Correlation coefficient R^2	E_a (kJ/mol)	A (s^{-1})	T_i (°C)	T_{mwl} (°C)
Raw rice straw	y = −14.003x + 10.766	0.9529	116	3.32E+06	282	303
Torrefied rice straw	y = −15.594x + 12.5	0.9668	129	2.09E+07	288	301

Table 3 Characteristic and kinetic parameters of combustion behaviour for raw and torrefied rice straw

Samples	Stages	Fitting equation	Correlation coefficient R^2	E_a (kJ/mol)	A (s^{-1})	T_i (°C)	T_{mwl} (°C)	T_b (°C)
Raw rice straw	Devolatilization	y = −15.247x + 13.694	0.9704	127	6.72E+07	249	283	470
	Combustion	y = −17.449x + 11.463	0.9575	145	8.30E+06			
Torrefied rice straw	Devolatilization	y = −14.074x + 11.434	0.9633	117	6.50E+06	252	290	468
	Combustion	y = −17.388x + 11.331	0.9559	145	7.25E+06			

DISCUSSIONS

1. Pyrolysis behaviour

In Figure 2, the mass evolution and derivative thermogravimetric (DTG) curves are presented. The mass evolution profile of biomass pyrolysis generally consists of three stages: drying, pyrolysis, and carbonization. In the drying stage, which occurs between 30 °C and 150 °C, moisture is eliminated from the sample. The subsequent stage is pyrolysis, taking place within the temperature range of 150 °C to 500 °C. This stage involves the removal of volatile matter, resulting in a decrease in mass. During pyrolysis, a prominent peak is observed in the DTG curves, which corresponds to cellulose pyrolysis. T_{mwl} is determined as 303 °C for raw rice straw and 301 °C for torrefied rice straw. These temperatures align with the temperature range of cellulose decomposition, typically between 315 °C and 375 °C (Di Blasi, 2008).

The third stage is carbonization, occurring from 500 °C to 800 °C. During this stage, the primary reaction is the decomposition of lignin. A slight decrease in mass is observed, and the DTG curve tends to flatten. This observation is consistent with the findings of Liu et al. (2021); Gani & Naruse (2007).

The temperature at which devolatilization initiates (T_i) is found to be 282 °C for raw rice straw and 288 °C for torrefied rice straw. This indicates that torrefied rice straw undergoes a higher degree of thermal decomposition compared to raw rice straw. It can be concluded that the torrefaction process improves the thermal properties of biomass, resulting in higher thermal stability due to a reduction in volatile matter (Agar & Wihersaari, 2012).

The pyrolysis E_a is determined as 116 kJ.mol^{-1} for raw rice straw and 129 kJ.mol^{-1} for torrefied rice straw. Activation energy represents the minimum energy required to initiate a reaction, and higher values indicate greater difficulty for reactants to undergo the reaction (Kaur et al., 2018). It is evident that the activation energy increases after the torrefaction process.

2. Combustion behaviour

The DTG curves indicate that the thermal decomposition follows the pattern observed in biomass combustion. The mass evolution profile can be divided into three stages: drying, devolatilization, and combustion. In the drying stage, which occurs between 30 °C and 150 °C, moisture is removed from the sample. The subsequent devolatilization stage takes place within the temperature range of 150 °C to 400 °C. In this stage, hemicellulose and cellulose undergo pyrolysis, leading to the production of volatiles and a significant decrease in mass. The third stage is combustion, which occurs between 400 °C and 800 °C. During this stage, the pyrolysis of lignin is the primary reaction, resulting in the formation of coke, followed by its combustion.

Based on the DTG curves, three peaks are observed for both samples. The combustion process begins with pyrolysis, where hemicellulose and cellulose pyrolyze to produce volatiles, while lignin pyrolysis results in char formation (Worasuwannarak, Sonobe & Tanthapanichakoon, 2007). Peak 1, 2, and 3 correspond to the decomposition of hemicellulose, cellulose, and lignin, respectively. Hemicellulose decomposes within the temperature range of 225 °C to 325 °C, cellulose at 375 °C, and lignin at higher temperatures between 250 °C and 500 °C (Prins, Ptasinski & Janssen, 2006). Peak 1 for raw and torrefied rice straw is found at 283 °C and 290 °C, respectively, which align with the temperature range of hemicellulose decomposition. Peak 2 and peak 3 for both raw and torrefied rice straw are similar, occurring at 405 °C and 450 °C, respectively, representing the temperature range where cellulose and lignin decomposition take place (250 °C to 500 °C).

The T_{mwl} was higher for torrefied rice straw compared to raw rice straw, with values of 290 °C and 283 °C, respectively. These findings suggest that torrefied biomass exhibits greater reactivity. These results align with a study by Broström et al. (2012), which reported T_{mwl} for torrefied biomass to be around 300 °C.

A lower temperature of T_{ic} indicates a more reactive material, as it signifies the ease with which the material reacts in the presence of air. In the case of this study, T_{ic} was found to be 249 °C for raw rice straw and 252 °C for torrefied rice straw. The higher T_{ic} for torrefied rice straw indicates its higher reactivity compared to raw rice straw under dry air conditions. The increased T_{ic} can be attributed to the reduction in combustible volatile matters during the torrefaction process. These results are consistent with the findings of Cruz Ceballos, Hawboldt & Hellleur, (2015), who observed an increase in T_{ic} after the torrefaction process due to a decrease in hemicellulose and cellulose content.

The T_b represents the difficulty of the sample to burn and indicates the residence time or temperature required to achieve complete combustion. The T_b values for both raw and torrefied rice straw were found to be in the range of 468 °C to 470 °C, indicating that all samples achieved complete combustion at around the same time.

The difference in activation energy (E_a) for the devolatilization stage highlights that torrefied rice straw requires more energy to initiate devolatilization compared to raw rice straw.

In this stage, the main reaction involves the pyrolysis of hemicellulose and cellulose. The torrefaction process leads to the decomposition of these components in torrefied rice straw, resulting in a lower content of hemicellulose and cellulose compared to raw rice straw. As a result, raw rice straw contains a higher amount of hemicellulose and cellulose, requiring a lower E_a for their decomposition. On the other hand, the E_a for the combustion stage, where the pyrolysis of lignin is the primary reaction, is the same for both samples. Since lignin is not decomposed during the torrefaction process and the amount of lignin might be similar in raw and torrefied rice straw, the samples exhibit similar E_a values for this stage.

CONCLUSION

TGA (Thermogravimetric Analysis) was conducted on raw rice straw and torrefied rice straw in order to investigate their pyrolysis and combustion behavior using nitrogen and air as carrier gases. The aim was to examine the impact of torrefaction on the characteristics of pyrolysis and combustion. The pyrolysis process of these biomass samples was observed to comprise three stages: drying, pyrolysis, and carbonization. Similarly, the combustion process consisted of three stages: drying, devolatilization, and combustion. By analyzing the TGA data, we gained a deeper understanding of the pyrolysis and combustion characteristics of raw and torrefied. Furthermore, we examined their ignition performance, burnout performance, and overall combustion performance, which serve as essential factors for guiding fire prevention. Additionally, the ignition temperature (T_{ic}) was measured, serving as a critical value for ensuring the safe storage of raw rice straw and torrefied rice straw. We also concluded that the degree of thermal decomposition is higher for the torrefied rice straw, which suggested that the sample was less reactive than raw rice straw. The lower activation energy of the sample resulted in the more easiness to decompose and high risk of self-heating and auto-ignition. The findings of this study can facilitate the design of combustion reactors and the determination of optimal operating conditions for such reactors. Moreover, these findings also assist in preventing self-heating and spontaneous combustion that can occur during the storage of biomass and torrefied biomass.

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