

Characterisation and quality improvement of binder free bio-pellets from the sugar industry residues and grass jelly food wastes for energy purposes

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Abstract

Torrefied pelletised biomass wastes can be a sustainable and efficient solid fuel; however, the addition of binders is frequently required to improve the consistency, durability and overall quality of the pellets. Such additional processes can increase production costs, reduce stability on exposure to moisture, increase ash content and may reduce the heating value of the fuel. Therefore, the production of durable binder free bio-pellets would create a sustainable and economically viable route for producing these solid fuels. Herein, the binder free pelletisation of waste sugarcane leaves and/or grass jelly leaves/stalks from the agricultural industry was investigated for use as bio-based solid fuels. Importantly, the resulting pellets had a diameter, length, moisture content, unit density, bulk density, calorific value and durability that met the international standards (DIN 51731, SS 187120 and CEN/TS 14961) and Thailand standard (TIS 2772–2560). However, the ash content for grass jelly leaves/stalks pellets was 16%, while the blended pellets of 50:50 sugarcane:glass jelly was 7%, both of which were higher than that required by the Thailand standard. The calorific value of the pellets was improved via a torrefaction process at 200 – 300°C for between 15 – 60 min. Yields of pellets dropped with increasing temperature and residence time, while the calorific value increases as the torrefaction severity increases from 16,630 – 26,334 kJ/kg. Crucially, this is the first reported pelletisation and torrefaction of sugarcane leave, grass jelly leaves/stalks or a 50:50 blend of these wastes to yield bio-based fuels with calorific values comparable to coal-like fuel pellets. Moreover, the mechanical strength of obtained pellets was still maintained without the use of an additional binder, thus reducing additional processing steps and potential cost. The optimal operating conditions for torrefaction were 250°C for 30 min, resulting in the greatest integrity, calorific value, enhancement factor and energy yield.

Keywords: Bio-pellet, Grass jelly, Sugarcane, Torrefaction, Calorific value

1. Introduction

A global increase in energy demand, and current use of non-sustainable solid fossil fuels such as coal which can increase greenhouse gas emission, has resulted in a need for alternative renewable energy sources. Biomass is widely used as a renewable bioenergy resource, however the low bulk density, irregular shape and large particle size distribution, which results in low volumetric energy density and high cost of storage and/or transportation are distinct disadvantages. Mechanical treatment is frequently required to convert the raw biomass into pellets of a uniform size, homogeneity and higher bulk density. Torrefaction can be performed in an inert environment between 200 – 300°C [1] to increase the energy content, reduce hydrophilicity and eliminate the high moisture content, thereby addressing some of the raw biomass problems. The combination of pelletisation and torrefaction improves competitiveness against conventional energy sources including coal. During biomass torrefaction, drying, depolymerization, devolatilization and carbonization processes occurred [2].

In general, there are two different approaches for pelletisation and torrefaction that can be employed: (1) pelletise ground raw biomass and then torrefy to yield torrefied pellets, which can also be described as “pelletisation with subsequent torrefaction”. Approach (2) requires torrefaction of raw biomass, followed by grinding and then pelletised, also referred to as the “torrefaction with subsequent pelletisation”. However, the pelletising of torrefied biomass is a major challenge due to the weakening of cohesive forces between biomass particles and the loss of natural binding characteristics of lignin in the biomass after torrefaction [3]. Hence, the addition of binding agents may be required to produce solid pellets. Torrefaction after pelletisation is the alternative potential pathway to produce

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binder free bio-fuel pellets with no additional processing steps. Therefore, approach (1) “pelletisation with subsequent torrefaction” without binder was selected for investigation in this research.

Most studies utilised binders; scarcely any research omits their use. The addition of a binder during pelletisation can improve the consistency, durability and overall quality of pellets, thereby reducing cracking or breaking during transport and storage as well as to reduce the production of fines during handling [4]. However, binder can be affected pellet density and porosity [5, 6]. Therefore, the common criteria for binder selection are the addition must not significantly affect production cost, durability of the pellet should be improved, should be stable on exposure to moisture, does not increase the amount of ash or unwanted elements and should not reduce the heating value of the fuel [4]. However, the selection of a binder that meet all these criteria is hard to achieve. Kuokkanen et al. [7] found that the addition of 1 wt.% potato flour or 1 wt.% lignosulfonate increases pellet durability compared to binder-free wood pellets from 96.5% to 98% and 96.5% to 97.7%, respectively. Moreover, using 1 wt.% of calcium oxide as an additive can increase durability of pine sawdust pellets by only 0.81% from 91.56% to 92.37% [8], although the pellet durability did not significantly increase. The production of durable pellets with high heating values, low ash content and binder free is challenging but would offer a sustainable and economic route to solid fuel production.

Sugarcane is one of Thailand's top agricultural crops. In 2021, the production of sugarcane in Thailand was around 66.8 million tons, making the country a major global sugar exporter [9]. Sugarcane leaves, bagasse and molasses are residues from sugar production. Bagasse is used in various applications, including paper industries and as feedstock (extracted cellulose) [10], while molasses has been used for ethanol production [11]. Moreover, sugarcane bagasse has also demonstrated potential as feedstock for production of fuel pellet and torrefied pellet [12, 13]. While sugarcane leaves are a true waste, that are typically disposed of through uncontrolled in field burning leading to releases of atmospheric pollutants such as particulate matter and are yet to be investigated as a feedstock to produce torrefied solid fuels.

Furthermore, grass jelly with scientific name of *Mesona chinensis* is part of the mint family (*Lamiaceae*) [14], and is a famous traditional Thai dessert. Grass jelly is made by boiling the leaves and stalks of jelly grass [14]. One of the biggest grass jelly dessert producers in Thailand has a production capacity of around 10 ton per day, and significant quantities of waste leaves and stalks are discarded daily. These solid wastes must be disposed of by landfill, leading to additional waste management issues and costs. The valorisation of this waste to a resource will aid in stimulating new product development, as well as providing a sustainable bio-based fuel source for a local circular economy.

Biomass sources used in the production of fuel pellet include rice straw [15], wheat straw [16], barley straw [16], soybean straw [17], meadow hay [18], pine wood sawdust [17], oil palm empty fruit bunch [19], oil palm frond and corn cob [20]. To the best of our knowledge, there have been no studies carried out to pelletise dry sugarcane leaves or dry grass jelly leaves and stalks, followed by an investigation of the torrefaction behaviours of both materials. The goal of this research is to produce, characterize and evaluate fuel pellets and torrefied pellets from dry sugarcane leaves, dry grass jelly leaves and stalk and/or a blend of both materials. All pellets were produced binder free, which aims to reduce the number of processing steps and produce a cost-effective biofuel.

2. Materials and methods

2.1 Materials

Dry sugarcane leaves or SL (Figure 1 (a)) were received from the Mitr Phol Sugar, Kalasin plant. Dry grass jelly leaves and stalk or GJ (Figure 1 (b)) were collected from the Siam Dessert Co., Ltd. Both raw materials were reduced to smaller sizes using a household blender. The samples were then sieved using a screen size 1 mm. The sieved samples for both raw materials are shown in Figure 1.

2.2 Densification and torrefaction

Pellets were produced in a single pellet unit with the hole diameter of 10 mm and 40 mm long without heating system. The sieved sample was added into the hole until full without adding any binder and then manually compressed by hydraulic press machine at 5 ton. The pressure was released for 5 seconds, then the pellet sample was removed from the die. Three types of pellets were produced: (1) 100% sugarcane leaves or SL, (2) 100% grass jelly leaves and stalk or GJ and (3) 50% sugarcane leaves and 50% grass jelly leaves and stalk weight ratio or SL+GJ.

Torrefaction processes were performed using a vertical tubular reactor (length 30 cm and diameter 5 cm) as shown in Figure 2 (a). The reactor was placed in in-house electrical furnace (Figure 2 (b)) and was flushed with nitrogen gas and heated to 200, 250 or 300°C by using heating rate of 10°C/min. Torrefaction temperatures were selected based on previously literature results [1]. Once the desired temperature was reached, the torrefaction temperature was maintained at 15, 30, 45 or 60 min. When the desired torrefaction time was reached, the sample was cooled to room temperature under N₂. The torrefied pellets were retrieved from the reactor after cooling and the mass has been recorded.



Figure 1 As received and milled and sieved of (a) dry sugarcane leaves or SL and (b) dry grass jelly leaves and stalks or GJ.

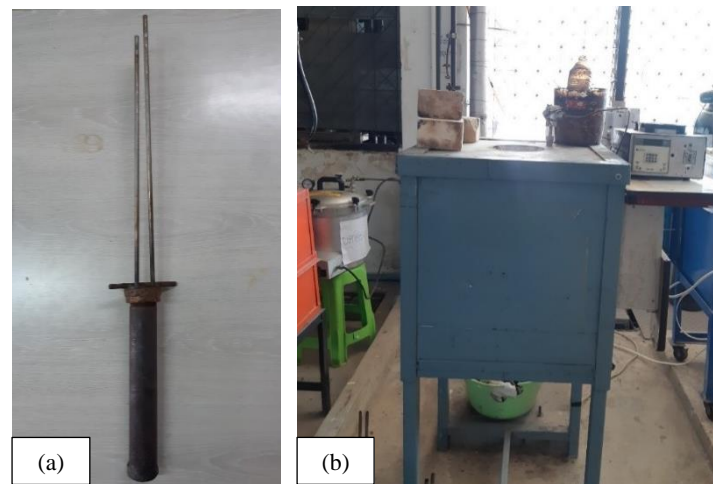


Figure 2 Torrefaction equipment (a) vertical tubular reactor and (b) in-house electrical furnace.

2.3 Characterisation

The quality of the obtained pellets was evaluated considering the physical, chemical, energetic and mechanical properties. The potential uses of pellets were also compared with the recommendations of Germany quality standard DIN 51731 [21], Swedish standard for pellets SS 187120 [22], European standard CEN/TS 14961 [22], EU quality ENplus B [23], and Thailand Standard TIS 2772–2560. The pellets diameter and length which stated in ISO 17225-2 were measured using a vernier calliper for randomly 10 samples and then reported average value. Moisture content, ash content and volatile matter content were determined using standards EN 14774-1 (Solid Biofuels - Determination of moisture content - Oven dry method Part 1: Total moisture-Reference method), EN 14775 (Solid Biofuels - Determination of ash content) and EN 15148 (Solid Biofuels - Determination of volatile matter), respectively. Moisture content was measured by placing the sample in an oven at 105°C until a constant weight is maintained for 24 h. Ash content of a sample was determined as the percentage of residue remaining after combustion at 550°C for 1 h in a muffle furnace (Vulcan Model 3-550A, Neytech) [24]. Volatile matter content was determined by heating the sample at 900°C for 7 min. Fixed carbon content was then calculated from the difference. Furthermore, fixed carbon content and volatile matter can be used to determine the fuel ratio [25]:

$$Fuelratio = \frac{Fixedcarboncontent(\%)}{Volatilematter(\%)} \quad (1)$$

The unit density was calculated from the mass divided by volume of cylindrical pellet and expressed in kg/m³. The bulk density of the samples expressed as the weight of the pellets that fill the volume of a specific container and performed in accordance with the ASTM E873-82 (Standard Test Method for Bulk Density of Densified Particulate Biomass Fuels). Gross calorific value was determined by an oxygen bomb calorimeter (Gallenkamp), following the CEN/TS 14918:2005, with recorded starting and final temperature. The amount of sample was 0.5 – 1 g and the measurements were repeated three times. Two further parameters can be calculated from calorific value, bulk density, and unit density. There are energy density and bulk porosity [26]:

$$Energydensity = Calorificvalue \times Bulkdensity \quad (2)$$

$$Bulkporosity(\%) = \left(1 - \frac{Bulkdensity}{Unitdensity}\right) \times 100 \quad (3)$$

The tests of mechanical durability of pellets were carried out using a sieve shaker by the modified method of Kuranc et al. [27] which was adapted from ISO 17831-1. The prepared samples of 10 pellets were weighted, then they were subjected to vibrations using a laboratory sieve shaker (AS 200 control, Retsch) and a sieve with mesh no. 5 (4 mm). The times of vibrations were 15 and 30 min with amplitude 0.5, 1 and 2. The durability was calculated from the weight of the sample which remained on sieve at each vibration time divided by the original weight.

The compressive strength test was carried out by using Universal Testing Machine or UTM (EZ-LX, Shimadzu). The compression was done on cylindrical pellet in both vertical or axial (Figure 3 (a)) and horizontal or diametral (Figure 3 (b)) directions with rate of 10 mm/min. The compression stopped when the pellet was broken (Figure 3 (c) and (d)). The compressive strength in vertical (σ_v , N/mm²) and horizontal (σ_h , N/mm²) directions were found by using the following formulas, respectively [28]:

$$\sigma_v = \frac{4F_{max}}{\pi D^2} \quad (4)$$

$$\sigma_h = \frac{2F_{max}}{DL} \quad (5)$$

where F_{max} is the maximum force (N), D is the diameter of bio-pellet (mm), and L is the length of pellet (mm).

The performance parameters of the torrefied pellet were represented by the mass yield, enhancement factor, energy yield and degree of torrefaction [25, 29, 30]:

$$\text{Massyield}(\%) = \frac{\text{Mass of torrefied pellets}}{\text{Mass of pellets}} \times 100 \quad (6)$$

$$\text{Enhancement factor} = \frac{\text{Calorific value of torrefied pellets}}{\text{Calorific value of pellets}} \quad (7)$$

$$\text{Energy yield}(\%) = \text{Mass yield}(\%) \times \text{Enhancement factor} \quad (8)$$

$$\text{Degree of torrefaction} = 1 - \frac{\text{Volatile matter of torrefied pellets}(\%)}{\text{Volatile matter of pellets}(\%)} \quad (9)$$

The FTIR analyses of raw materials and torrefied samples were conducted using ALPHA II (Bruker). The spectra were recorded at wavenumbers in the range of 4000 to 550 cm^{-1} and operated with 32 scans at a resolution of 4 cm^{-1} using direct examination of samples without further preparation or attenuated total reflectance (ATR). The thermogravimetric analysis of raw materials and torrefied samples was carried out employing a DTG-60H (Shimadzu). The analysis was conducted from room temperature to 900°C at a constant rate 10°C/min under the flow of air at 60 cm^3/min . The data was acquired in every 20 sec.

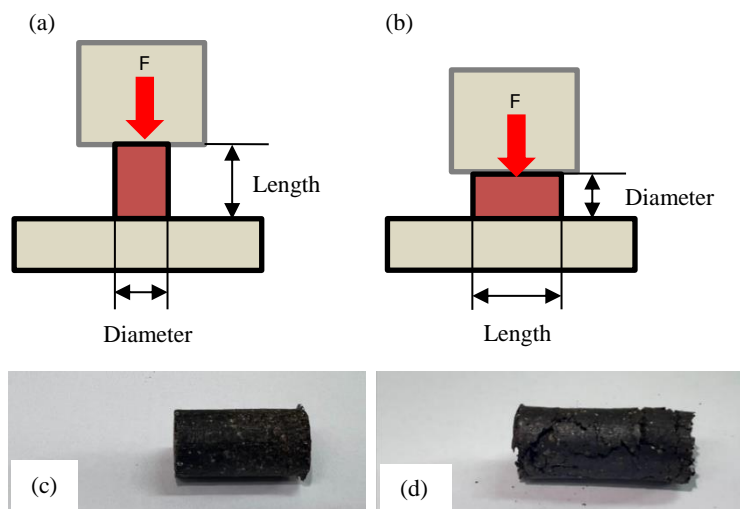


Figure 3 Orientation of compressive strength test (a) vertical and (b) horizontal directions and pellet (c) before and (d) after compression for diametral failure test.

3. Results and discussion

3.1 Physico-chemical properties of raw materials

The result from proximate analysis of the raw residues and the blended sample are shown in Table 1. The moisture content of a fuel has a great influence on the energy value and combustion performance. From the literature it can be observed that the optimum moisture content for pelletised biomass fuels is typically range between 8 – 12% [31]. It was shown that all starting materials had a good moisture content ranging between 11.69 – 12.62%. The amount of fixed carbon and volatile matter directly contribute to the heating value. Greater content of volatile matter in the fuel leads to easy ignited during combustion, while fixed carbon is combusted to heat generator during burning. Therefore, dry sugarcane leaves, and grass jelly waste are suitable for used as an energy source. However, sugarcane leaves, and grass jelly waste had a high ash content. Many studies have also used high ash content biomass as an energy source, for example, pepper plant residue (14.4% dry basis) [32], greenhouse residue (30.2% dry basis) [32], tomato residues (18.0 – 17.8% dry basis) [33], and cucumber residues (25.3 – 33.7% dry basis) [33].

Table 1 Properties of raw materials after milled and sieved.

Parameter	SL	GJ	SL+GJ
Moisture (%)	11.69	12.10	12.62
Ash (% dry basis)	17.74	35.05	26.81
Volatile matter (% dry basis)	69.50	54.80	62.06
Fixed carbon (% dry basis)	12.76	10.15	11.13
Bulk density* (kg/m^3)	60	178	—
Bulk density (kg/m^3)	72	384	143
Calorific value* (kJ/kg)	16,220	15,851	15,921
Energy density* (GJ/m^3)	0.97	2.82	—

*As received form

Bulk density is a parameter which directly affects storage and transportation costs. A higher bulk density promotes product compactness in the container. Bulk densities of as received sugarcane leaves, and grass jelly waste were 60 and 178 kg/m^3 , respectively. After milling, the bulk densities increased to 72 and 384 kg/m^3 , respectively, while the bulk density of the of the mixture was 143 kg/m^3 . This is due to the milling process which is one of the physical treatments to improve the properties of a raw biomass.

Calorific value is a key parameter that determines the quality of solid fuels. The results showed that sugarcane leaves had a higher calorific value (16,220 kJ/kg) than grass jelly leaves and stalks (15,851 kJ/kg), while the mixed sample was between them (15,921 kJ/kg). It was found that the calorific values of both raw materials are competitive when compared to other types of biomass used as solid fuel for combustion, for example, sugarcane bagasse (14,400 kJ/kg) [34], castor stalk (14,627 kJ/kg) [35], rice husks (15,602 kJ/kg) [35], bamboo leaves (15,715 kJ/kg) [35], cotton stalks (16,698 kJ/kg) [35], groundnut shells (16,769 kJ/kg) [35], saw dust (16,853 kJ/kg) [35]. The variation in calorific values may be due to the different composition (like fixed carbon and ash contents), which depends on the type of biomass growing season and even location of planting.

3.2 Pellets production and characterisation

Pellets produced from sugarcane leaves (SL), grass jelly leaves and stalks (GJ) and the mixing of them (SL+GJ) are shown in Figure 4. It can be observed that the physical appearance of all pellets is compacted with a hard texture. Under the same pelletisation conditions all feedstocks produced pellets with the same diameter i.e., 10 mm (Table 2). However, grass jelly leaves and stalk can be produced pellets with the longest length (22.64 mm), followed by mixing of grass jelly and sugarcane leaves (21.83 mm) and for sugarcane leaves (7.35 mm). This is attributed to the low density of sugarcane leaves, when compare with grass jelly leaves and stalks. In terms of pellet diameter and length, all samples satisfy the quality standards as shown in Table 2.

The results for the proximate analysis of all pellets are presented in Table 2. The moisture content of all pellets was in accordance with the requirements of both the international standards and Thailand TIS 2772–2560 standard. The obtained pellets have a high ash content over the threshold requirement, only SL pellet met the requirement of Thailand standard. Yılmaz et al. [36] reported that melon straw pellet had high ash content of 20.73%, greater than those specified by the standards. The high ash content will degrade the pellet quality, reduce the calorific value of the pellet and increase the particle emissions to the atmosphere [22]. The pellet from sugarcane bagasse also had moisture content (8.7%), which was higher than the international standard ($\leq 2\%$) but met the Thailand standard ($< 18\%$) [12].

Fuel ratio is calculated by the ratio of fixed carbon content over volatile matter and indicates the ease of ignition and combustion. Biomass usually has a low fuel ratio due to having high volatile matter, while this ratio in industrial coal is between 0.5 – 1.0 [25]. Bio-pellets produced in this study had a fuel ratio of between 0.11 – 0.17 or around 1/5th that of coal. Therefore, the improvement of bio-pellet by torrefaction to yield a coal-like fuel is need.

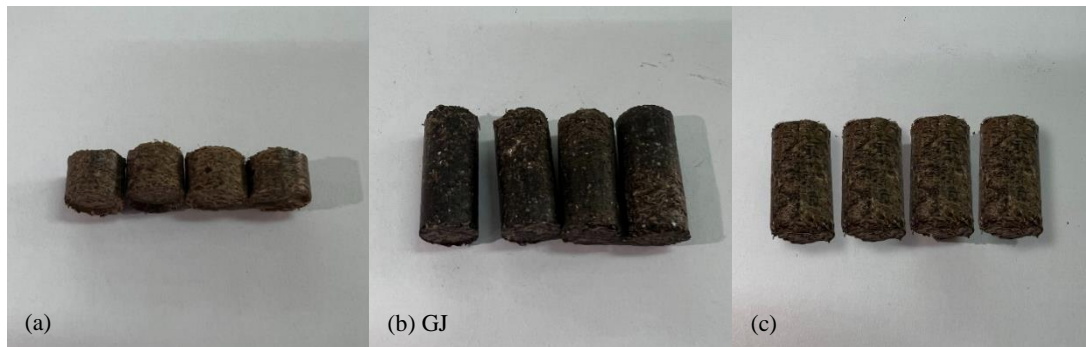


Figure 4 Photographs of the produced pellets from (a) 100% sugarcane leaves (SL), (b) 100% grass jelly leaves and stalks (GJ) and (c) 50% sugarcane leaves and 50% grass jelly leaves and stalk (SL+GJ).

Pellets made from grass jelly leaves and stalk recorded the maximum unit density of 1,334 kg/m³, while pellets made from sugarcane leaves recorded the lowest unit density value of 1,094 kg/m³ as shown in Table 2. According to the Germany quality standard DIN 51731, a single pellet density should be between 1,000 and 1,400 kg/m³. Extremely high density is not good for combustion efficiency because of the difficulty for oxygen to access the tightly packed fuel, which reduces combustion efficiency and can lead to incomplete combustion, that may result in atmospheric pollution [36].

Table 2 Characteristics of obtained pellets.

Parameter	SL pellet	GJ pellet	SL+GJ pellet	International Standard	Thailand Standard
Diameter (mm)	10	10	10	4 – 10*	6 – 25
Length (mm)	7.35	22.64	21.83	< 50*	3.15 – 40
Moisture (%)	10.20	11.04	11.22	< 12*	< 15
Ash (% dry basis)	16.81	34.09	25.76	$\leq 2^{**}$	< 18
Volatile matter (% dry basis)	71.84	56.01	65.05	–	–
Fixed carbon (% dry basis)	11.35	9.90	9.19	–	–
Fuel ratio	0.17	0.13	0.11	–	–
Unit density (kg/m ³)	1,094	1,334	1,150	1,000 – 1,400*	–
Bulk density (kg/m ³)	656	828	764	$\geq 500^{***}$	≥ 600
Bulk porosity (%)	40.04	37.93	33.56	–	–
Calorific value (kJ/kg)	16,387	15,930	16,135	17,500 – 19,500*	$\geq 14,500$
Energy density (GJ/m ³)	10.75	13.19	12.33	–	–
Durability (%)	99.76 – 99.98	99.14 – 99.56	99.41 – 99.74	$\geq 97.5^{****}$	≥ 96.0
Compressive strength (N/mm ²)					
Vertical	49.17	24.35	38.97	–	–
Horizontal	2.28	1.70	2.08	–	–

* DIN 51731, ** ENplus B, *** SS 187120, **** CEN/TS 14961

Bulk density of pellets depends on length, diameter, and single pellet density [37]. The standard value of bulk density is more than 500 and 600 kg/m³ for Swedish standard (SS 187120) and Thailand Standard, respectively. Biomass pelleting can significantly improve the bulk density of milled samples from 72, 384 and 143 kg/m³ to 656, 828 and 764 kg/m³ after pelletising of SL, GJ and SL+GJ, respectively. This confirms that the pelletisation increases the bulk density of biomass. Bulk porosity is the fraction of the volume of void over the total volume and voids spaces in the pellet. Porosity has an influence in transportation and storage, where low porosity of the feedstock indicates less void spaces. Bulk porosity of all pellets is also presented in Table 2. SL+GJ pellet had the lowest bulk porosity (33.56%), follow by GJ pellets (37.93%) and SL pellets had highest (40.04%).

SL, GJ and SL+GJ pellets had calorific value of 16,387, 15,930 and 16,135 kJ/kg, respectively. This was higher than Thailand standard ($\geq 14,500$ kJ/kg) but fell below the international standard (17,500 – 19,500 kJ/kg). Moreover, one of interesting properties of biomass pellets is volumetric energy density. According to the Table 2, the increase in volumetric energy density of bio-pellets rose from 0.97 – 2.82 GJ/m³ to 10.75 – 13.19 GJ/m³ after pelletisation. Pelletisation can improve the bulk density of biomass, consequently giving high energy density. Keeratiisariyakul et al. [38] revealed that energy densities of leucaena and oil palm empty fruit bunch pellets were 9.7 and 10.3 GJ/m³, respectively. Moreover, sorghum-based pellets had value of 11.10 GJ/m³ [39]. Therefore, the results of this research are consistent with energy densities of other biomasses presented in the literature.

Durability is used to describe the physical quality of densified solid biofuels include pellets. It is a measure of their ability to withstand destructive forces such as compression, impact, and shear during storage, handling, and transportation. The production of fines or dust during these activities can lead to an unhealthy working environment. Although there is no specified limit for the production of fines, but it is suggested that up to 5 wt.% would be acceptable level [40]. Moreover, several studies have classified the durability into high (> 80%), medium (70 – 80%) and low (< 70%) with the European standard of greater than 97.5% and Thailand standard of greater than 96.0% [40]. The results of durability tests are generated through vibration at 15 and 30 min with an amplitude of 0.5, 1 and 2 are presented in Figure 5 and Table 2. It was observed that all pellets had high durability and reached the specifications of both international and Thailand standards. The durability of all pellets decreased by only < 1%, with increasing duration time and amplitude (Figure 5).

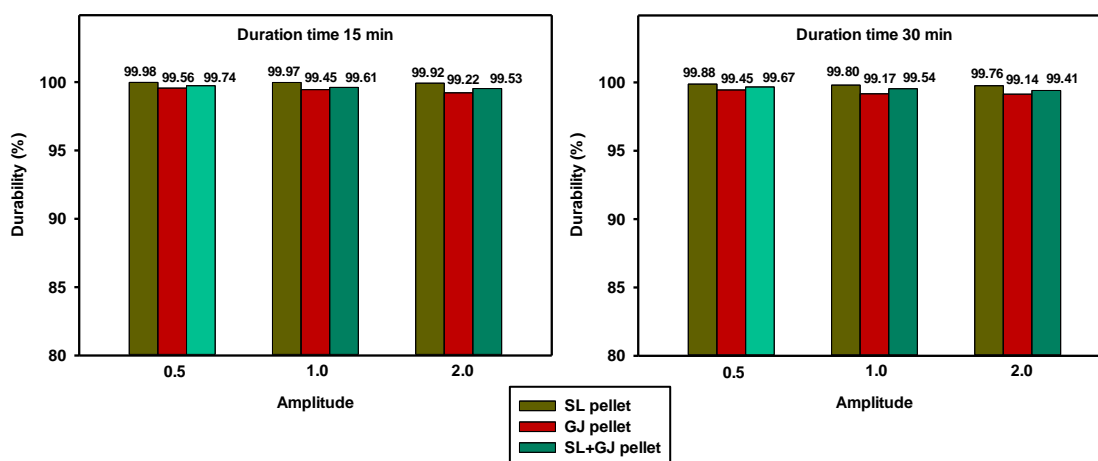


Figure 5 Results of durability of obtained bio-pellets in vibration tests at different duration timing and amplitude.

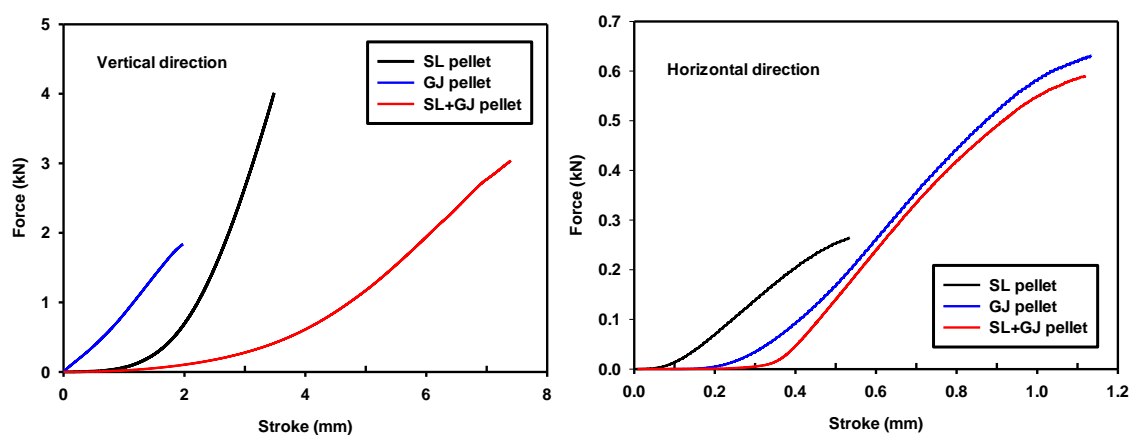


Figure 6 Force versus stroke during compression of pellets.

The compressive strength in vertical direction depends on F_{\max} and D (equation (4)). All pellets had equal values of D , therefore, the compressive strength in vertical direction depends only on F_{\max} . From Figure 6, the maximum force follows the order SL pellet > SL+GJ pellet > GJ pellet, resulting in the compressive strength 49.17, 38.97 and 24.35 N/mm² (Table 2), respectively. For horizontal direction, the compressive strength depends on F_{\max} , D and L (equation (5)). All pellets had the same diameter; therefore, the compressive strength depend on both F_{\max} and L . Although SL pellet showed the lowest maximum force, but the value of length was also lowest, resulting in highest compressive strength of 2.28 N/mm². It was noted that the compressive strength in the vertical direction was higher than in the horizontal direction, due to the pressure applied during the pelletisation process being exerted vertically on the

sample. Therefore, the sample was more tightly packed in this direction. The same trend was found in the vertical direction: SL+GJ pellet (2.08 N/mm²) > GJ pellet (1.70 N/mm²). Tarasov et al. [37] informed that pellet strength depends on pellet size, a longer pellet can be easily broken as compared to a shorter one. Moreover, the results in this current study are consistent with the literature. For example, Caragana pellets had a compressive strength of 13.90 N/mm² in vertical direction [41], while canola straw pellet had compressive strength of 2.21 N/mm² in horizontal direction [42].

3.3 Torrefaction performance and characteristics

Surface morphologies of obtained bio-pellets after torrefaction at different temperatures for 60 min are shown in Figure 7. The change in physical appearance is one indicator that reflects a successful torrefaction process. It was observed that the torrefied products turned a darker colour from the raw pellet colour with an increase in torrefaction temperature. This is due to the process of carbonization and dehydration during torrefaction resulting in the darker colour. According to the physical appearance, it can be said that the torrefaction process succeeded with the use of existing equipment.

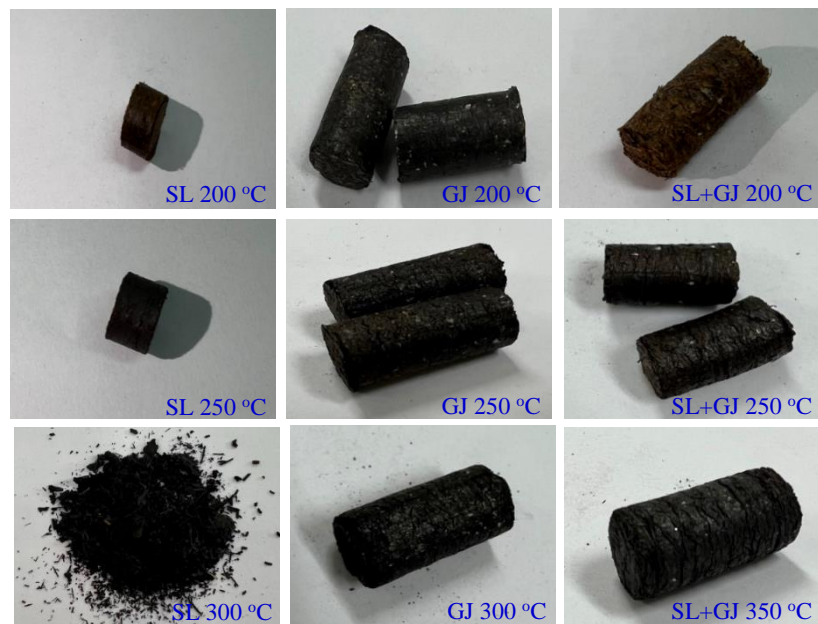


Figure 7 Images of torrefied pellets at different temperatures for a torrefaction time of 60 min.

At a torrefaction temperature of 300°C, cracks appeared on the surface, especially for SL, which still cannot form a pellet shape. This is attributed to the slow dehydration and vaporization of internal moisture of the pellets, which is the major reaction at torrefaction temperatures between 200 – 250°C. At torrefaction temperature in the range of 250 – 300°C, the thermal decomposition of hemicellulose. A large volume of volatile gases accumulated in a short time, creating relatively higher internal pressure within the pellets, causing damage to the surface morphology of the pellets, and thus created cracks. In conclusion, pellets produced at 250°C had a hard texture and was selected for the subsequent study of torrefaction time.

Table 3 shows the properties of torrefied pellets after torrefaction at different temperatures. With an increase in the torrefaction temperature, the yield of torrefied bio-pellets decreased. The reduced mass yield during the torrefaction process can be attributed to the loss of bound moisture, dehydration and thermal degradation to form volatile products such as CO, CO₂, and other organics [43]. Moreover, the mass yield of GL pellet was higher than SL pellet for the same torrefaction temperature. The effect of the torrefaction temperature on calorific value is also shown in Table 3. The higher the temperature of the torrefaction, the greater the calorific value. The calorific value of torrefied pellets is significantly increased from raw pellets (Table 2), from 15,930 – 16,387 kJ/kg to 16,630 – 26,334 kJ/kg. The calorific values of torrefied pellets were higher than peat ($\leq 14,644$ kJ/kg), lignite (14,644 – 19,292 kJ/kg) and sub-bituminous (19,292 – 24,405 kJ/kg) and comparable to bituminous (24,405 – 32,539 kJ/kg) [44]. Dehydration and the loss of volatile substances during the torrefaction, leads to an increase the carbon and subsequent increase in calorific value. The enhancement factor was used to define energy output of torrefied pellets relative to non-torrefied pellets. The results demonstrated that the enhancement factor also increased with greater torrefaction temperature. Bampenrat et al. [45] highlighted that an enhancement factor of around 1.3 – 1.4 is attained under optimal torrefaction conditions for biomass. In the current study, the factor lies between 1.04 – 1.63, which is higher than other biomass reported in the literatures, for example torrefied oil palm fibre pellets have a factor of 1.07 – 1.24 [46], 1.05 – 1.27 is observed fortorrefied soybean straw pellets [17], while torrefied pine wood pellets have an enhancement of 1.01 – 1.29 [17].

Table 3 Effect of torrefaction temperature on properties of torrefied pellets.

Parameter	SL pellet			GJ pellet		SL+GJ pellet		
	200°C	250°C	200°C	250°C	300°C	200°C	250°C	300°C
Mass yield (%)	85.11	80.42	90.07	86.35	74.91	89.52	85.65	68.16
Calorific value (kJ/kg)	21,701	23,848	16,630	20,108	23,108	19,949	22,150	26,334
Enhancement factor	1.32	1.46	1.04	1.26	1.45	1.24	1.37	1.63
Energy yield (%)	112.34	117.41	93.67	108.80	108.62	111.00	117.34	111.10

Energy yield is a parameter that is influenced by the calorific value and mass yield. It is a measure of the balance between mass loss and calorific value enhancement. If the value of energy yield is above 100%, the torrefaction process has been able to increase the energy of the pellets [47]. Table 3 also shows that the energy yield of all torrefied bio-pellets exceeded 100%, except for grass jelly leaves and stalk pellets torrefied at 200°C (93.67%). Pahla et al. [30] reported that an energy yield below 70% is not desirable from an energy efficiency point of view. Therefore, the values reported in this current study are still acceptable.

Figure 8 shows the torrefied pellets obtained using a torrefaction temperature of 250°C for between 15 to 60 min. All torrefied pellets maintained their integrity after torrefaction and became darker with increased residence time. It is important to note that testing demonstrated that densified torrefied biomass powder could not be made into a stable pellet form without the use of a binder, adding additional process steps and cost. This confirms the weakening of cohesive forces between biomass particles and the loss of natural binding characteristics after torrefaction. Therefore, torrefaction after pelletisation is an attractive alternative route for torrefied pellet production.



Figure 8 Images of pellets torrefied at 250°C with different times.

Table 4 shows the mass yield and fuel properties of torrefied pellets at various torrefaction time. It is observed that increased residence time decreased the mass yield. The greatest mass yield was obtained after 15 min, in contrast the smallest mass yield was obtained after 60 min for all three samples. When torrefied at the same temperature but for longer durations (from 15 to 30 min) the calorific value, enhancement factor and energy yield significantly increase. After this time these values demonstrated only a limited increase, therefore the pellets torrefied for 30 minutes were selected for additional characterisation. It is important to note that this treatment time is shorter than optimum torrefaction time for many types of biomass, such as peanut shell (60 min) [48], sugarcane bagasse (90 min) [45], palm kernel shell (90 min) [45], and cocopeat (90 min) [49].

Data from proximate analysis of torrefied SL pellet, GJ pellet and SL+GJ pellet are given in Table 5. The moisture content decreased after torrefaction at mild temperatures, resulting from the evaporation of water and some reduction in the moisture/extractives content. This suggests that the hydrophobicity of fuel pellets is improved. The ash content of torrefied pellets has met the Thailand standard of < 18%, with GJ pellets being the exception. Iryani et al. [50] also demonstrated that the ash content decreased after torrefied sugarcane bagasse, Napier grass, palm empty fruit bunches and cassava stems. Torrefaction increased the fixed carbon content, while decreased the volatile matter when compared with original pellets. The SL, GJ and SL+GJ torrefied pellets had fixed carbon content

increased from 11.35 to 21.33%, 9.90 to 20.55% and 9.19 to 20.20%, respectively. The volatile matter of pellets decreases from 71.84 to 68.75%, 56.01 to 53.30% and 65.05 to 63.16% for SL, GJ and SL+GJ sample respectively, after torrefaction. These trends were related to the decomposition of hemicelluloses during the torrefaction process and the partial depolymerization of cellulose and lignin. The fuel ratio of torrefied pellets (0.28, 0.34 and 0.19 for SL, GJ and SL+GJ) increased from raw pellets (0.17, 0.13 and 0.11 for SL, GJ and SL+GJ), due to the increase of fixed carbon and the reduced volatile content. When the torrefied pellets have a greater fuel ratio, leading to a more efficient combustion with fewer emissions. This demonstrates the superiority of torrefaction as a means of providing energy densification and enhancing the properties if the resulting solid fuel as compared to the raw biomass. The degree of torrefaction was found to be 0.10, 0.17 and 0.07 for torrefied SL, GJ and SL+GJ pellets, respectively. Sarker et al. [25] reported the degree of torrefaction between 0.1 – 0.4 for torrefaction of canola residue. The higher degree of torrefaction means a larger loss of volatiles during torrefaction process.

Table 4 Effect of torrefaction time on properties of torrefied pellets.

Parameter	SL pellet				GJ pellet				SL+GJ pellet			
	15 min	30 min	45 min	60 min	15 min	30 min	45 min	60 min	15 min	30 min	45 min	60 min
Mass yield (%)	86.20	82.47	80.85	80.42	91.21	89.10	88.62	86.35	90.34	86.98	86.65	85.65
Calorific value (kJ/kg)	20,327	22,049	22,932	23,848	18,404	19,007	19,293	20,108	19,213	21,706	21,937	22,150
Enhancement factor	1.24	1.34	1.40	1.46	1.16	1.19	1.21	1.26	1.18	1.33	1.34	1.37
Energy yield (%)	106.89	110.51	113.19	117.41	105.80	106.03	107.23	108.80	106.60	115.68	116.11	117.34

Table 5 Physical and fuel properties of torrefied pellets at 250°C for 30 min.

Parameter	SL	GJ	SL+GJ
Moisture (%)	5.88	6.50	7.11
Ash (% dry basis)	9.92	26.15	16.64
Volatile matter (% dry basis)	68.75	53.30	63.16
Fixed carbon (% dry basis)	21.33	20.55	20.20
Fuel ratio	0.28	0.34	0.19
Degree of torrefaction	0.10	0.17	0.07
Unit density (kg/m ³)	913	1,019	976
Bulk density (kg/m ³)	539	665	570
Bulk porosity (%)	40.96	34.74	41.60
Energy density (GJ/m ³)	11.88	12.64	12.37
Compressive strength (N/mm ²)			
Vertical	46.17	21.87	5.57
Horizontal	1.91	1.52	1.41

The density decreased after torrefaction due to the formation of void spaces between particles in the pellets arising from the weakening of the inter-particle hydrogen bonding leading to biomass degradation [3]. It was noted that the pellet shape and integrity was maintained after torrefaction. The bulk density of torrefied pellets decreased from the original pellet i.e., 656 to 539 kg/m³ for SL pellet, 828 to 665 kg/m³ for GJ pellets and 764 to 570 kg/m³ for SL+GJ pellets. This was due to the decrease in mass of the pellets during torrefaction, while almost maintaining the original volume (retain length and diameter). The bulk density of all torrefied pellets met the SS 187120 standard of ≥ 500 kg/m³ (Table 2) but was lower than coal (700 – 900 kg/m³) [51].

The energy density of torrefied SL pellets increased from raw SL pellet (10.75 to 11.88 GJ/m³). For SL+GJ pellets the energy density value remains constant, while GJ pellets decreased compared to the raw from 13.19 to 12.64 GJ/m³. Although torrefaction can improve the calorific value, it can also reduce the bulk density. The values of energy density found in this study were in the range of previously reported results, for example, torrefied leucaena pellets (10.3 GJ/m³) and torrefied oil palm empty fruit bunch pellets (11.1 GJ/m³) [38]. Upon torrefaction treatment, the compressive strength of the pellets slightly decreased in both directions. The decrease in the compressive strength of torrefied pellets at this temperature was due to the reduced lignin content in pellets, weakening bonding between the particles.

Combustion performance is crucial to evaluate the potential of materials as heating fuel. The TGA and DTG curves of raw materials and torrefied samples are shown in Figure 9. The combustion process could be divided into three stages. The first stage with a small peak between 50 – 150°C represented the removal of water. The weight loss of the torrefied sample was lower than the untreated sample and the obtained values were also in agreement with proximate analysis results (Table 2 and Table 5). The second stage was revealed in the temperature range 200 – 500°C, which was ascribed to the decomposition of hemicellulose, cellulose, and lignin. Hemicellulose has low thermal stability because of the amorphous polymeric structure, which decomposes at a low temperature range (220 – 320°C) [41]. Cellulose decomposes at higher temperature (280 – 400°C) due to this component composed of a long chain of glucose without branches [42]. Lignin possesses better thermal stability due to its complex structure, thereby decomposing over a broad temperature range of 200 – 900°C [52]. Two clearly visible peaks in DTG curves can be observed in this stage. The first peak represents the decomposition of hemicellulose, and the second peak represents cellulose and lignin decomposition. TGA and DTG profiles of both raw materials significantly different. Sugarcane leaf waste was unprocessed biomass, while grass jelly leaves and stalks were previously boiled with water to extract pectin, and other polysaccharides [14]. Therefore, the GJ sample showed less pronounced mass losses compared to SL. The third stage that occurred above 500°C corresponded to decomposition via dehydration and decarboxylation reactions. Moreover, the peak of torrefied samples shifted to lower temperature. This mean that both SL and GL need lower temperatures for combustion after torrefaction. The residues of torrefied samples after TGA were less than that of raw samples, indicating that torrefied samples resulted in lower ash formation. This was consistent with proximate analysis (Table 2 and Table 5). Lower TGA profile and DTG peaks at higher temperature for the torrefied samples when compared with original materials agree with the work of Kajina et al. [53].

Two combustion characteristic parameters i.e., ignition and burnout temperatures are also crucial for fuels. The ignition temperature (T_i) is defined as the minimum temperature at which a fuel ignites spontaneously in an environment without external source of ignition.

This parameter related to its safety in storage and delivery [54]. The higher the ignition temperature, the safer the fuel is to store and deliver. The burnout temperature (T_b) refers to the temperature at which the fuel is almost completely consumed. The lower burnout temperature was, the less time it took for fuel to burn out. Both temperatures are key parameters to evaluate fuel selection, consumption, and combustor design [54]. In this present study, both parameters can be determined by using the intersection method as described by Lu and Chen [54]. It can be seen in Table 6 that the ignition temperatures of sugarcane leaves, and grass jelly leaves and stalks are 265 and 260°C, respectively, which is a normal ignition temperature for biomass such as sugarcane bagasse (250 – 263°C) [55], bamboo (241 – 265°C) [54], or pine sawdust (210 – 290°C) [56]. The burnout temperatures of both raw materials are also consistent with other biomass [54].

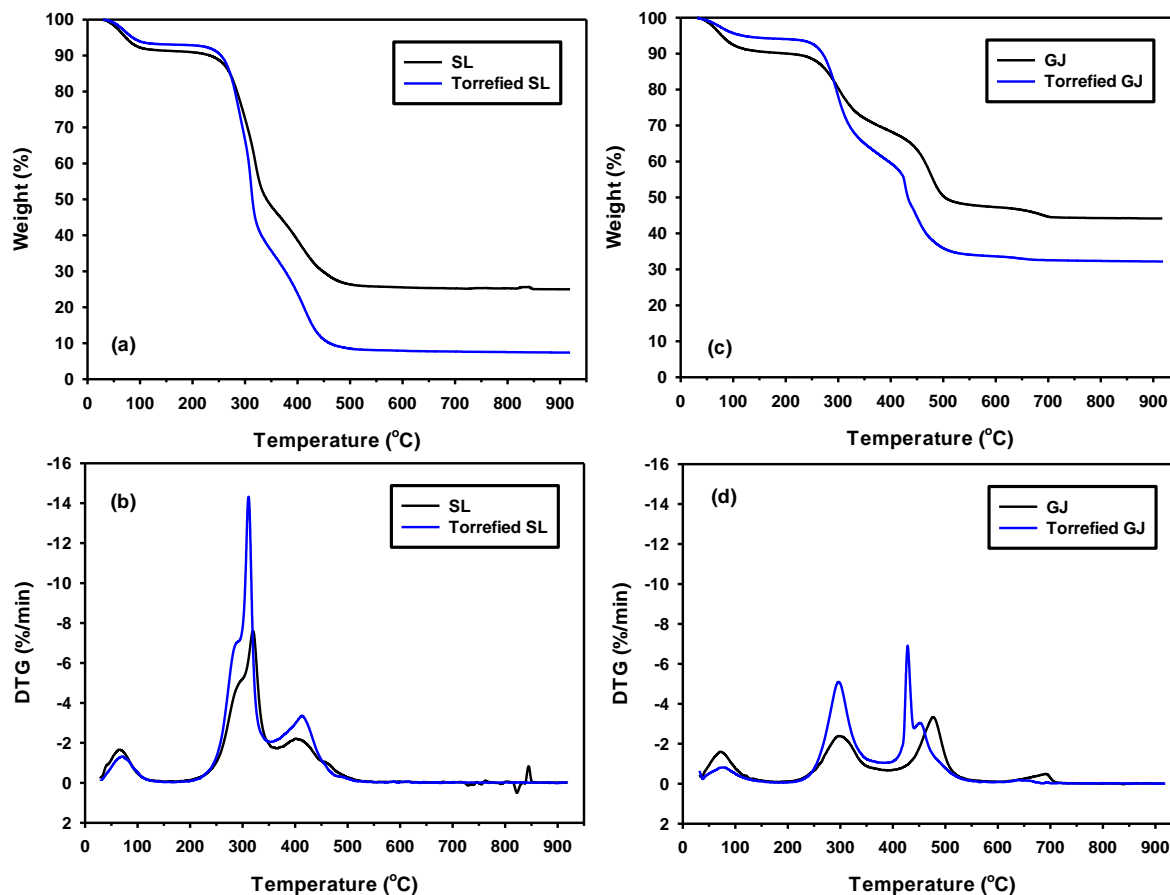


Figure 9 (a) TGA and (b) DTG curves for raw materials and (c) TGA and (d) DTG curves for torrefied samples at 250°C for 30 min.

After torrefaction, the ignition temperatures increased for both materials, thus making the pellets less reactive than untreated biomass. The ignition temperature depends on the release of volatiles, with the greater number of volatiles lower T_i . The volatile matter decomposed during the torrefaction process, therefore, the raw materials contained higher volatile matter than that of the torrefied samples (Table 2 and Table 5). Similarly, higher burnout temperatures were obtained for the torrefied samples compared to their original biomass. This was also due to less water/volatiles and more fixed carbon contained in torrefied samples. Aich et al. [57] reported the ignition and burnout temperatures for sixteen coal samples as 296 – 343°C and 467 – 488°C, respectively. This means that the torrefied samples in this current work had slightly lower ignition temperature than coal (280°C for torrefied SL and 265°C for torrefied GJ) and lower burnout temperature for torrefied SL (460°C), indicate this fuel is burn quicker, while torrefied GJ (518°C) needs longer time.

Table 6 Combustion characteristic parameters of raw materials and torrefied samples.

Sample	Ignition temperature (T_i , °C)	Burnout temperature T_b , °C)
SL	265	450
Torrefied SL	280	460
GJ	260	478
Torrefied GJ	265	518

The FTIR spectra of the raw materials and torrefied samples at 250°C for 30 min are shown in Figure 10. The raw materials and torrefied samples demonstrated similar FTIR spectra but with different intensities, indicating common surface components. Raw sugarcane leaves and grass jelly leaves and stalks had a wide band at a wavenumber of 3321 cm^{-1} attributed to the vibration in the O–H bonds [29], while the intensity of this band decreased on torrefaction. This might be related to the dehydration of the biomass waste during the torrefaction process. The loss of –OH groups signify enhancement of hydrophobicity of torrefied pellets from a hydrophilic raw biomass. The band around 2900 cm^{-1} has been ascribed to C–H stretching vibrations normally associated with aliphatic hydrocarbon [58]. The peaks in the range of 1500–1100 cm^{-1} correspond to the stretching bending peaks of C–O–C, C–H and C–OH

[17]. The spectra of torrefied samples showed no obvious differences compared with the spectra of the raw biomasses. The pronounced band at 1032 cm^{-1} for SL series and 1028 cm^{-1} for GJ series has been attributed to multiple C–O, C–C and C–C–O stretching in lignin, cellulose, and hemicellulose [58]. These bands also decreased upon torrefaction.

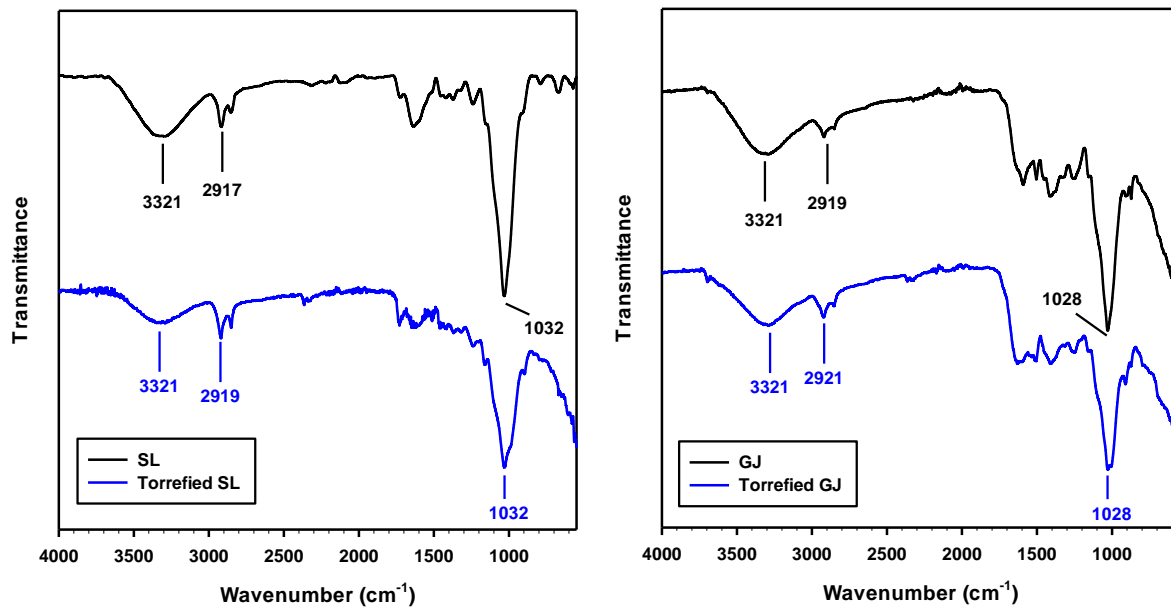


Figure 10 FTIR spectra of the raw materials and torrefied samples at 250°C for 30 min.

4. Conclusions

In summary, biomass wastes from sugar and grass jelly were used to produce bio-pellet fuel without an additional binder. The physical properties of the fuel pellets complied with the standards and increased the bulk density from $60 - 178\text{ kg/m}^3$ to $656 - 828\text{ kg/m}^3$. The unit density and durability of the fuel pellets were in the range of $1,094 - 1,334\text{ kg/m}^3$ and $99.14 - 99.98\%$, respectively. Pellets were further upgraded by the torrefaction at 250°C and 30 min to yield bio-based fuel pellets that demonstrated a calorific value ranging between $19,007 - 22,049\text{ kJ/kg}$. This study indicates that waste sugarcane leaves, grass jelly wastes, have the potential as a sustainable alternative to fossil fuels for households, power plants and factories, although, ash management must be considered. Future work should focus on the evaluation of combustion and exhaust gas emissions.

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