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Valorization of green tea waste from ready-to-drink industry through pelletization and use as solid fuel

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Abstract

The production and characterisation of pellets made from green tea waste from the ready-to-drink industry is investigated in this study. Physical and energetic characterisation of pellets including the length, diameter, proximate analysis (moisture, ash, volatile matter and fixed carbon content), pellet density, bulk density, calorific value, durability, compressive strength and thermogravimetric analysis were carried out to determine the quality of the bio-based fuel pellets. Pellet properties were assessed against comparison with the established Thailand and European standards. Pellet making was conducted with a hydraulic press without binder resulting in pellets with 2 diameters, 7 and 10 mm, and lengths of about 9.5 and 9.8 mm, respectively. The interesting characteristics of these biopellets were the moisture content of 8.78 – 8.82 wt%, the ash of 3.52 – 3.65 wt%, the bulk density of 586 – 603 kg/m^3 and higher heating value of 19.18 – 19.86 MJ/kg. In addition, the pellets produced with both diameters had properties that complied with the standards except for the bulk density of large pellets which were slightly lower. The durability of the 7 mm diameter pellets was 90.2% while 10 mm pellet was 95.6%, which a bit lower than standard value (≥ 96%). The pellet's compressive strengths in vertical direction were much higher than horizontal direction for both diameter pellets. These results suggest that this biomass waste was suitable for alternative fuel applications and has the potential to be developed as a commercial pellet.

Keywords: Biomass, Densification, Energy, Solid waste, Green tea

1. Introduction

Energy consumption is continuously increasing due to activities connected to promoting a more comfortable human life style. Fossil fuels are potential resources but their use generates carbon dioxide and other greenhouse gases emissions that impact on global warming. Therefore, it is necessary to search for new renewable alternative energy sources were their combustion processes have a reduced environmental impact. Biomass is the most attractive energy source because it is abundant, renewable, clean, and low cost. There are several types of biomass such as forestry residues (chip, log, bark and sawdust), agricultural residues, animal residue, industrial waste and municipal solid waste. Non-woody biomass fuels are considered to have a great potential to replace fossil fuels due to their reduced greenhouse gas emissions [1] and aid to maintain the balance of reforestation.

Biomass has several disadvantages when it is directly used as a fuel, including low heating values per unit volume and the low bulk density necessitating large storage facilities and increased transportation costs. A densification process that densifies material into pellet, briquette or cube can improve the biomass feedstocks in terms of increasing the bulk density, improving storability, reduce transportation costs and make the material easier to handle using existing equipment because of the uniform shape and size [2]. Moreover, another advantage of densified material compared to unprocessed biomass is the high energy content per unit volume. Pelletized fuel also has excellent mechanical properties. For example, Wang et al. [3] found that solid fuel pellets produced from municipal solid wastes had high mechanical shock resistance of > 99%. Wheat straw and corn stover pellets had a maximum durability of about 96%, while the lodgepole pine and pinion juniper recorded a maximum of > 96% [4].

Pressure-assisted densification can be done by applied high force to a mass of material within a confined volume to increase density. There is some literature related to the addition of a binder and/or an additive for the improvement of pelletization. For example, Xia et al. used crude glycerin and paraffin as an additive to rice straw pellets [5] Zhai et al. [6] selected molasses and a combination of molasses and lime (CaO) as binder for pelletization of food waste. Moreover, Sukarta et al. [7] applied an adhesive starch and water as a mixture for glue in the manufacture of wood wastes/biosolid sludge pellets. Although a binder and/or additive can improve the quality of pellets from biomass, this needs to be cost-effective and have no negative impact on the environment [5].

The goal of this research is to achieve a non-binder pelletization of green tea waste. Green tea which is a lignocellulosic biomass material used in this study is the waste from the ready-to-drink industry. In 2014 – 2019, the market value of ready-to-drink green tea in Thailand was 376 – 503 million USD. In January – March, 2020, the production of ready-to-drink green tea was 68.4 million litres [8]. So, it is generating more green tea waste. Currently, the only application of green tea waste is as low value animal feed. This study valorises this waste as feedstock to make a more valuable material, which do not use a binder, thereby further reducing the cost of pelletization process.

Herein, this research focusses on the development of bio-fuel pellet that are of sufficient quality to meet the Thailand and European standards of pellet properties for non-woody materials. The Thailand standard defines the quality requirements of non-woody pellets in line with the worldwide European standard, EN 14961-6. The physical properties and thermal characteristics of the pelletized fuels from green tea waste consisting of length, diameter, proximate analysis, pellet density, bulk density, calorific value and thermogravimetric analysis were studied. Crucially this work demonstrates a potential route to waste valorisation through densification.

2. Materials and methods

2.1 Sample and sample preparation

Green tea waste was collected from the ICHITAN Group Public Company Limited, a producer and distributor of ready-to-drink green tea. The waste was sun-dried for 3 days and crushed in a food blender to produce small size samples of 850 μm (mesh no. 20). In Figure 1, photos of raw and crushed materials are exhibited.

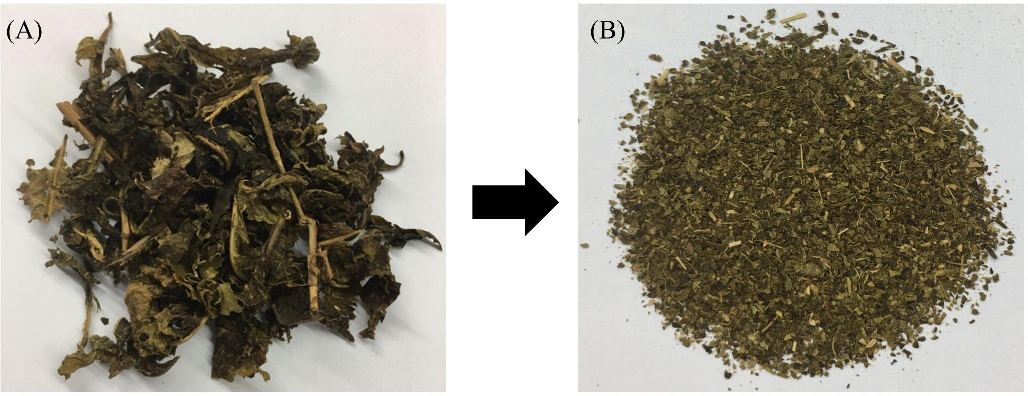

Figure 1 Raw green tea waste material (A) and crushed waste samples (B).

2.2 Pelletizaion of green tea waste

The green tree waste was pelletized using a single pellet hydraulic press (SC AUTO TOOLS). The unit consisted of a cylindrical die and a piston made of steel with the end of the die closed using a backstop. A 7 and 10 mm diameter of channel die was used. Then, the sample was fully filled into the die and a force of 5 tons was manually applied to the press. Subsequent to the compression, the backstop was removed from the die and the pellet was pressed out of the channel. The experimental system of sample pelletizer and the resulting green tea pellet of 7 mm and 10 mm diameters are shown in Figure 2.

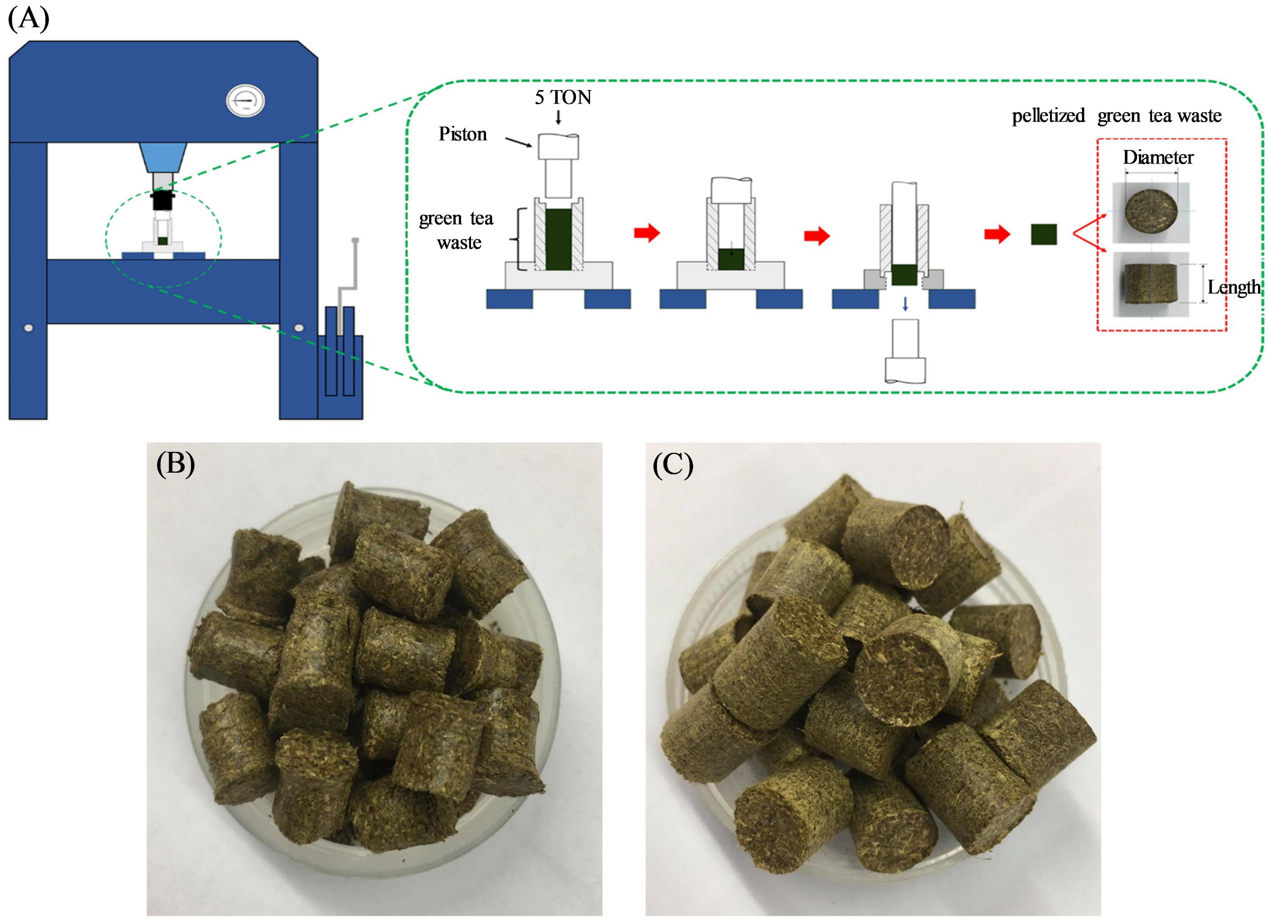

Figure 2 (A) Experimental system of sample pelletizer and photograph of green tea pellets (B) 7 mm diameter and (C) 10 mm diameter.

2.3 Characterisation of raw material and pellet

To determine the biofuel pellet quality, some characteristics were measured including it's diameter, length, proximate analysis (moisture, volatile matter, fixed carbon and ash content), pellet density, bulk density, and calorific value. The detailed characterisation presented in the following.

Pellet size (diameter and length) was measured with a vernier caliper and reported values are the average of 10 measurements for each pellet diameter.

Moisture content, ash content, volatile matter and fixed carbon were determined in proximate analysis. All contents were measured 3 times and the mean value was taken. The moisture content of a sample was determined according to EN 14774-1 Solid biofuels – Determination of moisture content – Oven dry method [9]. A sample of 1 g was put in the crucible and oven dried at 105 °C for 24 h. Finally, the crucible was cooled in a glass desiccator until room temperature was reached. Then, the dried sample and crucible were weighed again and the loss of weight encountered was the moisture of the sample.

The ash content of the sample was determined based on EN 14775 Solid biofuels – Determination of ash content [9]. One gram of the remaining sample from the moisture content examination was put in a crucible and then placed inside a muffle furnace (3-550, NEYTECH VULCAN). The temperature of the furnace was increased from room temperature to 550 °C for 1 h. After that, the crucible was removed from the furnace and cooled to room temperature in a glass desiccator. The mass retained in the crucible was expressed as percent ash content. One gram of each of the sample

The content of the volatile matter was found by using EN 15148 Solid biofuels – Determination of the content of volatile matter [10]. One gram of the remaining sample from the moisture content determination was put in a crucible and then heated in a muffle furnace at 700 °C for 7 min. The crucible was cooled down and then weighed again. The volatile matters of the sample were determined by applying the mass of the sample before and after the analysis.

The fixed carbon in percentage was calculated by subtracting 100 with the sum of the moisture, volatile matter and ash content. The equation (1) used to calculate this content as follow:

$$\text{Fixed carbon (\%)} = 100 - [\text{Moisture (\%)} + \text{Volatile matter (\%)} + \text{Ash (\%)}] \tag{1}$$

Pellet density was calculated by dividing the mass by the volume of individual pellets. Its weight estimated by using an analytical balance to three decimal places. The volume of individual pellet is $(\pi/4)\times \text{diameter}^2 \times \text{length}$. Reported values are the average of 5 measurements for each pellet diameter.

Bulk density was determined according to the ASTM E873-82 standard test method for bulk density of densified particulate biomass fuels [11]. Pellets were poured into a 12×12×12 in. container. The weights of the empty container and the combined pellets and container were measured by using an analytical balance to three decimal places. Net weight of the pellets was obtained by subtracting the weight of the empty container from the combined weight of the pellets and container. Bulk density was calculated based on 5 replications as following equation:

$$\text{Bulk density} = \text{Net weight of the pellets/Container volume} \tag{2}$$

The gross calorific value or higher heating value of a sample is obtained by using a bomb calorimeter (GALLENKAMP Autobomb) based on the ASTM E711 standard. About 0.5 g of the sample was placed in a crucible and the bomb was filled with oxygen at a filling pressure of 20 atm. The sample was burnt until complete combustion was reached. The experiments were done three times and the average values were utilized. The difference between the starting and constant final temperatures was used to examine the calorific value. The final experimental results were the average value of three samples. The obtained higher heating value was used to calculate the energy density [12] by multiplying it with the pellet density or

$$\text{Energy density} = \text{Higher heating value} \times \text{Pellet density} \tag{3}$$

The fire time for the complete combustion of the sample was recorded when using the bomb calorimeter. Based on the amount of the sample and fire time, the combustion rate was calculated by using the following equation [13]

$$\text{Combustion rate} = \text{Mass of sample/Fire time} \tag{4}$$

By knowing the gross calorific value and the combustion rate, the heat release rate could be calculated by using the following equation [13]

$$\text{Heat release rate} = \text{Higher heating value} \times \text{Combustion rate} \tag{5}$$

The durability was performed by a modified method of Sette et al. [12]. The pellets of 10 samples were weighed to obtain the initial mass and placed in a vibrating sieve (mesh no. 10 with mesh size of 2.08 mm) for 10 min at 80 rpm. After which, the samples were weighed again and the durability was determined from the ratio between weight before and after being placed on the vibrating sieve. The experiments were conducted in triplicate and the average value was used.

The compressive strength of pellet was determined from the maximum force at break of the pellets, as measured through a universal testing machine or UTM (EZ-LX, Shimadzu). The sample was placed in both vertical and horizontal directions between flat plates, in which the top plate was compressed at the rate of 10 mm/min until the sample reacted breaking point. The maximum force was recorded and the compressive strength in the horizontal direction was calculated according to the Newton equation [14], while the compressive strength in vertical direction was found by using the formula of Gilvari et al. [15]. The experiments were conducted in triplicate for each direction and the average value was used.

Thermal decomposition profiles on raw material and resulting pellets were accomplished using a thermogravimetric analyser (DTG 60H, Shimadzu) under nitrogen gas with constant flow rate of 40 mL/min and a heating rate of 10 °C/min from room temperature to 900 °C. Each sample weighing approximately 10 mg was placed into a platinum pan. Weight loss was recorded throughout the test with respect to the temperature and time and the resulting thermogravimetric (TG) profile was then further processed to obtain the differential thermogravimetric (DTG) profile.

3. Results and discussions

3.1 Raw material analyses

The results of proximate analyses are listed in Table 1, where the bulk density and the calorific value of the raw material and crushed samples are demonstrated. For comparison purposes, other biomass that was used as raw materials for densification as biofuel pellets was also displayed. Green tea waste exhibited high volatility and low ash content. Ash is a measure of non-combustible inorganics in biomass, higher ash content causes slagging or fouling problems that led to corrosion and reduces the heating value thus leading to a reduction in combustion efficiency [16,17]. Therefore, green tea waste that had a low ash content and a relatively high heating value is suitable for combustion as a fuel. Biomass usually contains a high amount of volatile matter, which can have a positive effect on combustion. Volatile matter consists of combustible gases such as H_2, CO and CH_4 and non-combustible gases like CO_2. The high volatile matter content means that the most of the calorific value would be released as a vapour combustion, which makes the biomass a highly reactive fuel due to its' fast combustion rate [7,16,17].

Table 1 Properties of green tea waste.

Sample	Proximate analysis (wt%)				Bulk density (kg/m^3)	Higher heating value (MJ/kg)
	Moisture	Volatile matter	Fixed carbon	Ash		
Green tea	9.27	70.73	17.84	2.16	97.88	18.17
SD*	1.32	0.56	0.37	0.74	3.27	0.62
Green tea, crushed	9.31	70.04	18.00	2.65	313.56	18.15
SD	1.24	0.70	0.28	0.63	5.29	0.32
Pongamia shell [18]	11.61	70.83	11.86	5.70	146	16.81
Phragmites australis grind [19]	4.06	–	–	8.21	107	17.93
Wheat straw [20]	6.90	70.80	17.00	5.30	–	17.70
Eight species of wood sawdust [17]	11.55 – 41.28	47.30 – 75.16	10.36 – 16.56	0.27 – 1.61	–	13.07 – 20.82

* SD: standard deviation

The moisture content of the green tea waste was 9.27 wt% as illustrated in Table 1. Moisture inversely effects the heating value, where high moisture content lowers the heat liberated during combustion [17]. However, Stelte et al. reported that the stability or integrity of the pellets of wheat straw, beech wood and spruce wood was best in the range of moisture content of raw materials between 5 – 15%. No stable pellets were formed where the moisture content was above 20%. This means that some amount of moisture is needed to improve inter-particular adhesion/attraction and the integrity of the pellets is very poor for completely dry biomass [21]. Green tea waste had a fixed carbon content comparable to other biomass. Fixed carbon content is a measure of the combustible solid in biofuel after the purging of volatile matters. The high value of carbon content means the better quality of biofuel [16].

It is clearly shown that green tea waste is not suitable as a feedstock due to its low bulk density (97.88 kg/m^3). The better option is to pelletize the green tea waste. In addition, some raw materials used to make biofuel pellets also had very low bulk densities such as chopped big bluestem (46.6 kg/m^3), chopped wheat straw (47.7 kg/m^3), chopped corn stover (50.9 kg/m^3) and chopped sorghum stalk (59.3 kg/m^3) [2]. After crushing the raw green tea waste, the bulk density increased to 313.56 kg/m^3. This confirms that crushing is a mechanical method to improve the physical properties of biomass fuel by homogenizing the particle size. Sette et al reported that the bulk density of crushed and milled eucalyptus wood as a raw material for the production of fuel briquettes, was 250 kg/m^3 [9].

The important characteristic of a fuel is its heating value or calorific value which is the amount of heat per kg of sample. The analysis of the calorific value of the raw material showed the value of 18.17 MJ/kg. Calorific value of green tea waste was higher than pongamia shell, phragmites australis and wheat straw. This is due to the larger amount of fixed carbon, leading to the higher calorific value.

3.2 Biofuel pellet analyses

Table 2 shows the characteristics of green tea waste pellets. All the pellets complied with the Thailand and European standard (Table 3) as regards diameter and length. The geometric dimensions of the pellets, both diameter and length, are crucial factors that might affect the combustion. The thinner pellet allows a uniform combustion, especially for small furnaces. Moreover, pellet dimension might also affect the fuel feed, where shorter pellets can reach continuous flow easier than longer pellets [22]. The results in Table 2 also show that large pellets had lower combustion and heat release rates than small pellets. For large pellets, it required more time consuming and higher heat transfer resistance than small pellets.

Moisture and ash contents of green tea pellets were in the range of 8.78 – 8.82 wt% and 3.52 – 3.65 wt%, respectively. These values meet the requirement of Thailand and European standards. A higher content of small pellets led to a higher combustion rate than large pellets.

As shown in Table 2, the pellet and bulk density of green tea waste pellets was 1,025 – 1,040 and 586 – 603 kg/m^3, respectively. Pellet density values were always higher than bulk density values because the volume of voids was excluded from the calculation. Other studies report the pellet density of 935 kg/m^3 for oil palm frond pellets [23], 991.27 kg/m^3 for para-rubber leaves litter pellets [23], 1,049 kg/m^3 for bagasse pellets [24], 1,119.29 kg/m^3 for para-rubber branch pellets [23], 1,152 kg/m^3 for empty fruit branch palm-oil pellets [24], 1,232 kg/m^3 for wood pellets [25] and 1,247 kg/m^3 for miscanthus pellets [24]. Pellet density is a critical factor affecting the energy density of biofuel pellets, higher pellet density resulting in higher energy density (20.65×10^3 MJ/m^3 for 10 mm diameter and 19.66×10^3 MJ/m^3 for 7 mm diameter). Thailand and European (EN) standards do not have a requirement for the density of fuel pellets. However, the pellet density of green tea pellets which is higher than 1,000 kg/m^3 met the wood pellet standard requirement of Germany (1,000 – 1,400 kg/m^3), DIN 51731.

Pelletization of green tea waste significantly increases the bulk density from 97.88 to 586 – 603 kg/m^3. The higher the bulk density compared to the raw material is helpful to improve its transportation and storage. The bulk density decreases with an increase in the pellet size. This may be due to the obstruction of larger sizes in containers. Compared with the standards in Table 3 ($\geq$ 600 kg/m^3), the bulk density of the resulting small pellets reaches the requirement. Other biomass pellets also had bulk densities which were a bit lower than standard such as sorghum stalk pellets (365 – 479 kg/m^3) [2], wheat straw pellets (469 kg/m^3) [20], miscanthus pellets (481 kg/m^3) [1] and bagasse pellets (590 kg/m^3) [24].

The higher heating values of green tea waste pellets were 19.18 and 19.86 MJ/kg, higher than the raw material (18.17 MJ/kg). This may be due to the fixed carbon content of pellets, which was higher than raw material. All of the pellets had calorific values that met the minimum requirement of the standards in Table 4 ($\geq$ 14.5 MJ/kg). Moreover, the larger size (diameter 10 mm and length 9.8 mm) had a higher calorific value than the smaller sample. This is consistent with the fixed carbon content and pellet density results, with higher fixed carbon and pellet density resulting in higher calorific value. The calorific value of green tea pellets was higher than apple pomace pellets (16.02 MJ/kg) [3], miscanthus pellets (16.2 MJ/kg) [25] and Scots pine pellets (18.37 MJ/kg) [26] and comparable to tea waste pellets (19.52 MJ/kg) [3] and wood pellets (19.57 MJ/kg) [27].

The durability of the pellets was 90.16% and 95.61% for 7 mm and 10 mm diameter, respectively. The standards require these values to be greater than 96% (Table 3). However, many bio-fuel pellets have durability lower than standards such as coffee pulp pellets (76%) [28], big bluestem pellets (86%) [29], groundnut shell pellets (88%) [30] and corn stover pellets (90%) [29]. The compressive strength of pellets in vertical direction were much higher than in horizontal direction. This also in agreement with Gilvari et al. [15] who reported that axial (vertical) compression of miscanthus pellet was higher than diametric (horizontal) compression. However, some literature has reported the compressive strength only in horizontal direction [31,32].

Table 2 Results analyses of pellets from green tea waste.

Property	Green tea waste pellet	
	Ø 7 mm	Ø 10 mm
Pellet density (kg/m^3)	1,025	1,040
SD	91	102
Bulk density (kg/m^3)	603	586
SD	21	13
Length (mm)	9.5	9.8
SD*	2.4	1.3
Proximate analysis (wt%)		
Moisture content (SD)	8.78 (0.87)	8.82 (1.3)
Volatile matte (SD)	68.51 (1.86)	65.52 (2.23)
Fixed carbon (SD)	19.19 (3.10)	22.01 (3.04)
Ash content (SD)	3.52 (1.1)	3.65 (1.5)
Higher heating value (MJ/kg)	19.18	19.86
SD	0.27	0.36
Energy density (MJ/m^3)	19.66×10^3	20.65×10^3
Combustion rate (g/min)	0.18	0.12
Heat release rate (J/min)	3,452	2,478
Durability (%)	90.2	95.6
SD	1.30	0.18
Compressive strength (N/cm^2)		
Vertical (SD)	1,443 (19)	2,116 (25)
Horizontal (SD)	16 (6.8)	44 (7.5)

* SD: standard deviation

Table 3 Quality requirements for solid biofuel pellet (non-woody).

Property	Unit	Thailand Standard TIS 2772-2560 (2017)		European Standard EN 14961-6:2012
		Class I	Class II	
Diameter	mm	6 – 25	6 – 25	6 – 25
Length	mm			
diameter 6 – 10 mm		3.15 – 40	3.15 – 40	3.15 – 40
diameter 12 – 25 mm		3.15 – 50	3.15 – 40	3.15 – 50
Bulk density	kg/m^3	≥ 600	≥ 600	≥ 600
Moisture content	wt%	≤ 15	≤ 15	≤ 10
Ash content	wt%	≤ 10	≤ 18	≤ 6
Calorific value	MJ/kg	≥ 14.5	≥ 14.5	≥ 14.5
Durability	%	≥ 96	≥ 96	≥ 96

In this study, TG and DTG curves of the raw material and the pellet samples were accomplished as shown in Figure 3. Initial weight loss of about 8 – 10 wt% at room temperature to 100 °C is attributed to the loss of adsorbed moisture related to the moisture content in the samples. These values are consistent with the values analysed from proximate analysis. The second step of weight loss at 200 – 350 °C for raw material and 200 – 300 °C for the crushed sample and pellet samples is attributed to the degradation of hemicellulose and cellulose [22]. The third degradation region at 350 – 520 °C for raw material and 300 – 520 °C for the crushed sample and pellet samples could be attributed to the cellulose and lignin degradation [22]. The DTG curves show two significant peaks for all samples. However, the highest mass loss for raw material was the second peak (high temperature of about 500 °C) whereas the highest mass loss for the crushed sample and pellet samples was first peak (lower temperature of about 300 °C). This mean that after physical treatment, crushing and densification with pressure, samples can easier to decompose or made the samples more reactive. The final mass about 3 wt% of all samples showed total decomposition of organic matter.

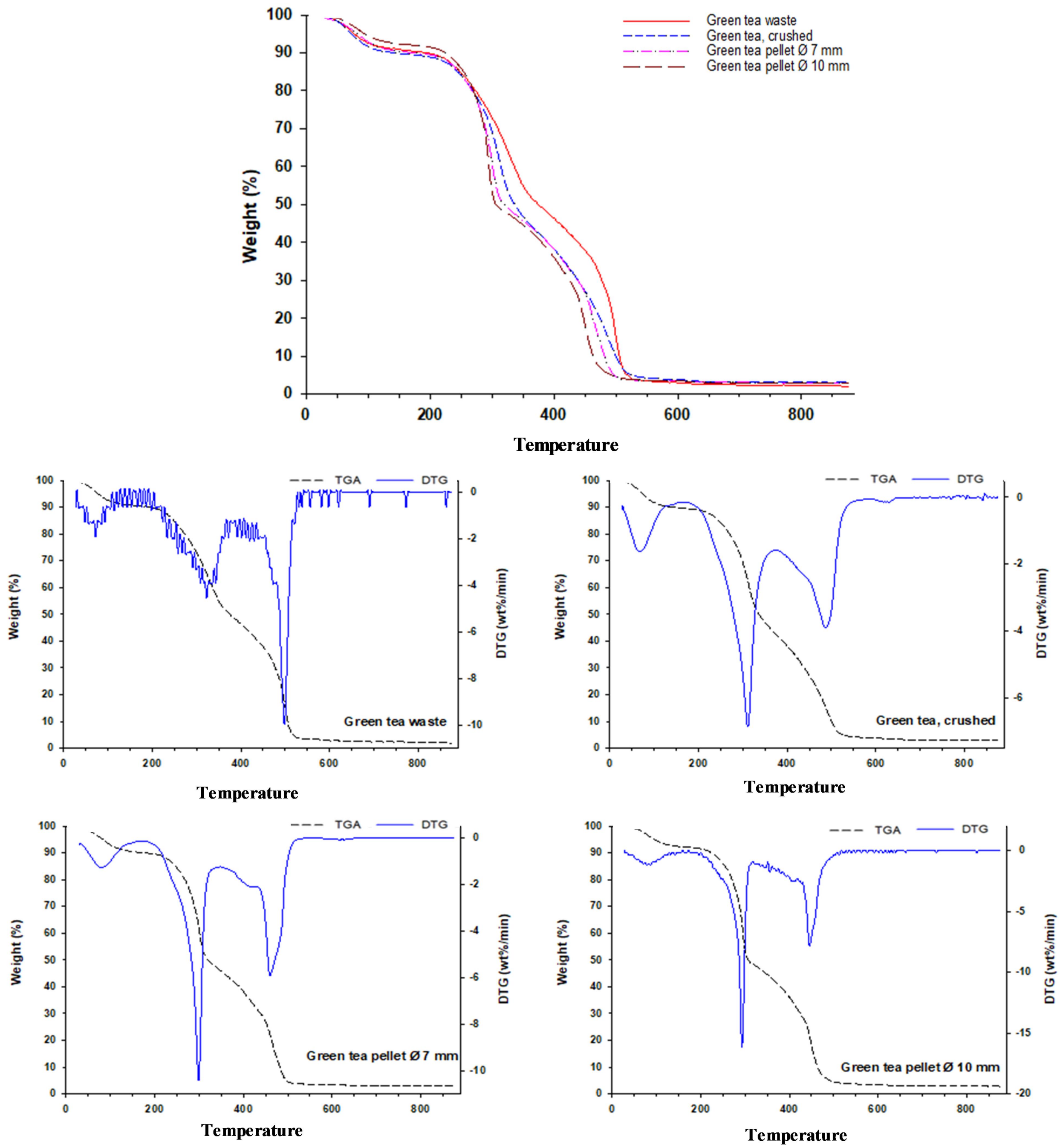

Figure 3 Thermogravimetric analysis of raw green tea waste and its pellets.

4. Conclusion

Pelletization is one of the most widely method used to densify biomass feedstocks to form biofuel with high bulk density. In Thailand, the production of ready-to-drink green tea quite high due to high market value. Therefore, it generates solid tea waste in large amount. In this study biofuel pellets from green tea waste were successfully developed without the additional binders. To make bio-pellets, a hydraulic press was used. The bio-pellet quality reference standard refers to the Thailand and European standards. The six properties of pellets met the requirements of the standards: diameter 6 – 25 mm, length 3.15 – 40 mm, bulk density ≥ 600 kg/m^3, moisture content ≤ 10 wt%, ash content ≤ 6 wt% and calorific value ≥ 14.5 MJ/kg with a bit lower durability than the standards (≥ 96%). This indicated that biofuel pellets made from green tea waste exhibited good properties for use as a quality bio-based fuel. Thus, this study demonstrates a route for the valorisation of biomass waste through the production of sustainable biofuel pellets from green tea waste.

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