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Trial of Biopellet Prepared by Napier Grass

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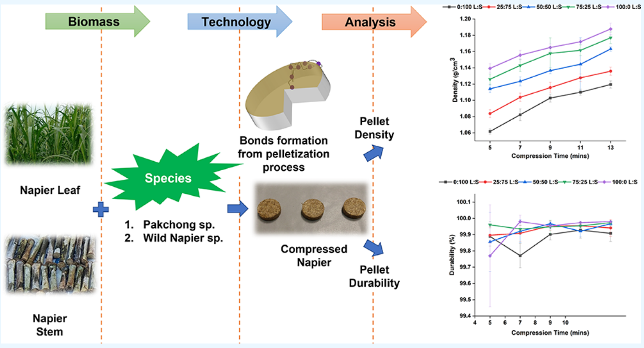
ABSTRACT: Pelletization from nonwoody biomass has gained a lot of attention due to its potential to secure biomass feedstock supply and pricing. Studies have been conducted to produce biopellets from different parts of Napier grass plant. In this study, two different species of Napier grass were used. Two pressure points and five different times were varied accordingly. Proximate, ultimate, and Fourier transform infrared spectroscopy (FTIR) analyses were performed to assess chemical properties. Statistical analysis of the collected data validated and supported the discussion. The produced pellets of Napier grass exhibited good energy density comparable to those in the literature based on gross calorific values. The pellet density demonstrated a significant effect with time, pressure, and feedstock ratios. The pellet durability test showed comparable characteristics to another biomass pellet. Both Pakchong Napier grass (PNG) and wild Napier grass (WNG) biopellet densities obtained were higher than 0.650 g/cm^3, and the durability was higher than 95%. This signifies that the pellet can withstand repeated transfer during handling without breakage.



1. INTRODUCTION

Located in Southeast Asia, Malaysia is blessed with natural resources, which contributes to the development of the biomass industry. As the national green science and technology policy coming into practice, the Tenth Malaysia Plan, the 2010 Economic Transformation Programme, and the 2011 Renewable Energy Act, Malaysia, are tapping the potential of the biomass industry.[1] Among the wide variety of lignocellulosic biomass in Malaysia, Napier grass (*Pennisetum purpureum*) appears to be one of the promising alternative feedstocks that can be utilized for energy conversion, in this case, solid fuels.

Currently, Malaysia is one of the top countries investing in Napier grass cultivation. Napier grass was reported to have a massive potential to produce a high yield of biomass, ~100 boe/ha (barrel of oil equivalent per hectare), and can be harvested every quarter in a year, totaling up to 4 times of harvest in a year.[2,3] Easily grown in tropical conditions, Napier grass can grow to an upward of 4 m in height and has a long lifespan of 7 years.[4] It is also one of the best alternatives in the form of biomass for conversion to solid fuel for industrial purposes due to its high heating value (average of 16 MJ/kg).[5] These advantages summarize the benefits of Napier grass, thus giving it a lot of credit as a great source for use in bioenergy applications (particularly for thermochemical conversion) as compared to other residues.[6]

In Malaysia, many cultivars of Napier grass have been introduced since the 1920s, namely, common Napier, Red Napier, Taiwan Napier, Dwarf Napier, Indian Napier, and so on.[7] However, recently, researchers managed to develop a new hybrid Napier grass in Thailand as the search for a better yield in Napier grass plantation continues. The crossing between *P. purpureum* and *Pennisetum glaucum* resulted in a hybrid Napier called Pakchong Napier (also called Super Napier) with specific characteristics[8] that are superior in terms of dry matter yield and nutritive values as compared to other cultivars. Zailan et al.[9] reported that Common and Red Napier were high-yielding despite the easy degradation and fermentation of Red Napier. Halim et al.[7] investigated nine different species and concluded that the short Napier varieties were leafier and had a higher nutritive quality but had a lower dry matter yield than the taller varieties.

Having a good quality of raw biomass is one thing, but having maximum bulk density for efficient energy production is a must, and it can be achieved through the densification process. Raw biomass materials are densified to obtain high-density products. Among densification processes, pelletization is one of the best methods used for biomass-based direct combustion solid fuel production.[10] The CEN/TS 14588 standard[11] defines biomass pellets as densified biofuel made

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from ground biomass materials with or without additives, typically made in a cylindrical form of diameter 6−10 mm and a random length of typically 5−40 mm. The homogeneity in size, shape, and quality of pellets makes them well-suited for fully automated feed systems.[12] Biomass pellets can be used directly as energy fuel in several applications ranging from residential heating stoves to central industrial heating boilers.[13] They can also be used as fuel sources in large-scale power plants[14] or as part of the process to produce bio-oil.[15] Biomass pellets have been standardized and traded domestically and internationally. The production and consumption of biomass pellets have increased significantly since 2008. According to Matthews,[16] the global biomass pellet production from wood was 28 million tons in 2015. Seventy-five percent of the world's wood pellets are consumed by European countries, followed by North America (14.3%) and Asian countries, mainly South Korea and Japan (7%).[17] Malaysia's energy pellet production is around 168 million tons every year. Many local researchers and universities have shown interest in research and development related to biomass.

Utilizing biomass materials for energy fuels comes with a few challenges. One of the challenges of biomass pelletizing is the logistic feasibility and cost, which require careful management of the agricultural, wood, or even aquaculture residues from nearby areas.[18] Skjeverak and Sopha[19] reported some of the common problems with pellet fuel such as fines produced from pellet during both the handling and the combustion process. In a more recent study, Tan et al.[20] reported that storage can affect dry matter loss, resulting in a decrease in hemicellulose content and bulk density, increase in the ash content, and a significant reduction in energy consumption. Other studies focused on the need for identifying new materials to be used as fuels by mixing well-known, easily available, and good-quality materials, and the characteristics of these materials must be established.[21] To date, there is no information in the existing literature regarding the properties of mixtures of different Napier grass species and parts, as well as scanty information on the characteristics of Napier grass compaction in biopellet production. Napier grass has been actively used as fodder crops by farmers in Africa, Asia, and other tropical/subtropical regions of the world.[22] In conjunction with the entry into force of the EN ISO 17225[23] standard that makes biomass pellets possible for industry and household purposes, evaluation of the best formula for blending of Napier grass species and parts to produce good-quality pellets is investigated.

Biomass pellet is becoming an interesting bioenergy source in Malaysia, and research is needed on the optimization of locally available biomass for production of solid fuels. In this study, the quality of Napier grass was examined from different compositions of two species and the plant parts without any external binders or additives. A single unheated hydraulic press die was used. Availability of the trait properties of Napier grass will create an opportunity for scientists and investors to decide the adoption of the best species to be used as the main feedstock (for example, a species with a higher lignin content will help with the durability of the produced biopellets). This study intends to assess its potential by mixing the different plant parts, varying the pellet-making parameters, and analyzing the performance of the Napier grass pellet. Versatile pelletizing aims at achieving the transformation changes of adapting and adopting feedstock to the pellet technology in order to add value to the feedstock and produce good, clean solid fuels without neglecting the concerns on the sustainability of the raw feedstocks used. At the same time, this will also be able to improve socioeconomics in rural areas of Malaysia.

2. MATERIALS AND METHODS

2.1. Preparation of Raw Materials.

Hybrid Pakchong species Napier grass (PNG) was supplied by a local company, and wild Napier grass (WNG) was collected at Teluk Bakong, Bota, Perak. Collected raw materials were chosen at matured ages of 7−8 weeks (roughly 130 cm tall and higher) and were stored in a cold room at 4 °C before further analysis to minimize infections.

Different species and parts of Napier grass (as shown in Figure 1) were used as raw materials. All feedstocks were air-dried and then oven-dried at 105 °C for further drying. The targeted moisture content (below 10 wt %) was within the global pellet market range.

Figure 1. (a) PNG obtained from a local company. (b) WNG obtained near Perak River in Teluk Bakong, Perak.

Napier grass stem and leaf were chopped into small pieces. The cut samples were then crushed using a granulator (AMF SG-21P low-speed granulator) before further grinding in a hammer mill (cutting mill SM 100) using a screen with an opening size of 1 mm. All of the samples were sieved to 300 μm and later stored to reach a uniform moisture content before the pelletizing process and further analyses.[24] All different species and parts of crushed Napier grass were thoroughly mixed by means of blending with different mass mixing ratios, as shown in Table 1. All of the samples were stored in airtight plastic bags for further analysis. Figure 2 shows the detailed process flowchart of this experiment.

2.2. Proximate, Ultimate, and Calorific Value Analyses.

The proximate analysis was carried out to measure the moisture content, volatile matter, and ash content of the raw materials in accordance with BS EN 14774-3 (2009) − Solid biofuels − Determination of moisture content − Oven dry

Table 1. Experimental Design

species	ratio of leaf to stem (L/S) (wt %) leaf	stem	compression pressure (MPa)	compression time (min)
Wild Napier grass	100	0	36.91	5
	75	25		7
	50	50	73.83	9
	25	75		11
	0	100		13
Pakchong Napier grass	100 wt % stem		36.91	5
				7
			73.83	9
				11
				13

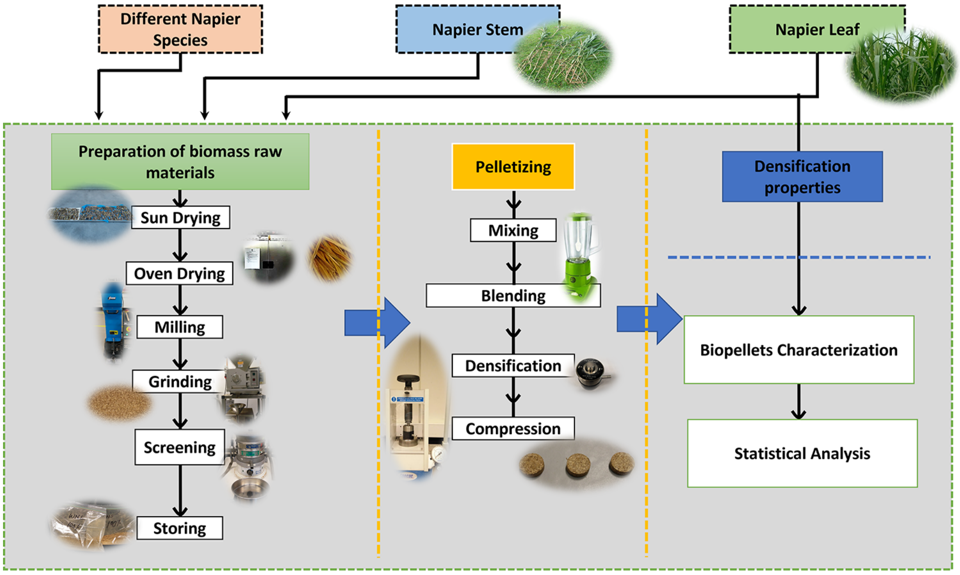

Figure 2. Research flowchart.

method – Part 3: Moisture in general analysis sample;[25] BS EN 15148 (2009) – Solid biofuels – Determination of the content of volatile matter;[26] and BS EN 14775 (2009) – Solid biofuels – Determination of Ash Content,[27] respectively. The fixed carbon content was computed by subtracting the percentage compositions of both volatile matter and ash content. The ultimate analysis was performed to identify the elemental composition of Napier grass in terms of carbon (C), hydrogen (H), nitrogen (N), and sulfur (S) using the CHNS Analyser (Series II CHNS/O Analyzer 2400, Perkin Elmer). The oxygen (O) was obtained by subtracting the sum of C, H, N, S, and ash content. The gross calorific value was determined using a bomb calorimeter (C200 model, IKA Werke) in accordance with the EN 14918 Solid Biofuels Determination of Calorific Value. The heat of combustion with elemental composition was performed based on Boie's equation following Burnham's report.[28] The heat of combustion will reveal how the calorific value, energy content, and elemental compositions are interrelated with each other from the perspective of energy properties of the biopellets.

2.3. FTIR Analysis. The raw feedstocks were also sent for FTIR analysis, where the attenuated total reflection (ATR) mode was used for faster analysis and at the same time to reduce variations in spectral peaks. This is believed to be useful as the results would be more conclusive and easier to interpret in order to improve accuracy in determining the identification of functional groups and bonding properties of the raw materials. The FTIR spectrum was obtained using an FTIR spectrometer (model NICOLET iS10) between the wavelength of 400 and 4000 cm^{-1}.

2.4. Preparation for Pelletization of Napier Grass. The pelleting process was conducted in a single press with a 13 mm diameter die. Sieved Napier grass mixtures (0.5 g) were compressed into pellets at ambient temperature using a laboratory-scale Specac hydraulic press. A cylindrical steel die of dimension 1.3 cm diameter with two pressure points and five different compression times was used, as shown in Table 1. No external binders were used in this experiment. The volume and weight of the compressed biopellets were measured and used to determine the density. The maximum density of the produced biopellets was measured right after pelletizing, while the relaxed density was measured 7 days after pelletizing and in readiness for further analysis.

2.5. Determination of Biopellet Density. The unit density of Napier pellets was determined by measuring the mass using a digital balance and the dimension using a Vernier caliper. In this experiment, maximum and relaxed densities were calculated, where the maximum density is the density of the pellet measured right after being made, while the relaxed density is the density of the pellet measured after 7 days of production to obtain a uniform, true density. The test was carried out 6 times for each sample. Equation 1 shows the equation for determination of biopellet density.

$$\text{biopellet density (g/cm}^3\text{)} = \frac{\text{mass of Napier grass sample}}{\text{volume of compressed Napier grass}} \tag{1}$$

2.6. Determination of Biopellet Durability. The durability of the pellets produced was measured based on a drop test, with reference to a previous study,[29] which involved a free fall of a single pellet from a height of 1 m to a concrete surface. In this case, every variant was tested in triplicate and the mass of each pellet was measured prior to and after the drop, and the weight loss percentage was used as a measure of durability. Equation 2 shows the formula used to obtain biopellet durability.

$$\text{biopellet durability (\%)} = \frac{\text{weight of biopellet after drop}}{\text{weight of biopellet before drop}} \times 100 \tag{2}$$

2.7. Statistical Analysis (by SAS Studio Software). Statistical analysis was conducted using free SAS Studio software, Enterprise Edition, Copyright 2012–2018, SAS Institute Inc. Cary, NC. This analysis helps evaluate the effect of several factors (feedstock ratios, compression time and pressure, as well as species) on the properties of biopellets. The evaluation of effects was carried out through an analysis of variance (ANOVA), with a confidence level of 95%. If the *p*-value is lower than 0.05, a significant statistical effect of the

Table 2. Physicochemical Characteristics of WNG and PNG

properties	biomass					
	WNG (100:0)L/S	WNG (75:25)L/S	WNG (50:50)L/S	WNG (25:75)L/S	WNG (0:100)L/S	PNG (0:100)L/S
proximate analysis						
moisture content (wt %)	7.18	6.38	5.29	4.03	4.79	7.07
volatile matter (wt %)	90.03	91.59	92.4	94.86	93.9	91.29
ash content (wt %)	8.39	6.75	5.39	3.91	2.4	2.64
fixed carbon[a](wt %)	1.58	1.66	2.21	1.23	3.69	6.07
gross calorific value (J/g)	16 587	16 989	17 228	17 509	17 737	17 901
ultimate analysis (wt %)						
carbon	42.44	42.98	44.20	44.94	45.51	45.67
hydrogen	6.02	6.34	6.49	6.33	6.47	6.57
nitrogen	1.72	1.56	1.24	1.08	0.72	0.95
sulfur	0.22	0.20	0.16	0.17	0.18	0.17
oxygen[b]	49.60	48.92	47.91	47.54	47.54	46.64
Boie equation (J/g)	15 374	15 904	16 726	16 859	17 241	17 413

a,b By difference.

experimental factors was identified for the specified response variables.

3. RESULTS AND DISCUSSION

3.1. Proximate, Ultimate, and Calorific Value Analyses.

Table 2 gives a brief characteristic of the different parts of WNG and PNG supplied by the local industry. In terms of biological appearances, PNG has a larger stem size as compared to WNG, which might affect the physical, chemical, and mechanical properties later throughout the study.

The moisture content varied when going across different parts of the plant. Moisture content is one of the key parameters in biomass pelletizing as it indicates the presence of water within the pellets, which later contributes to the binding properties. Physically, the nature of Napier leaf particles was denser and darker in color compared to Napier stem particles, which were less dense, more porous, and brighter in color. This was the first indicator as it is suspected that the leaf part of Napier grass contains higher moisture compared to the stem. The 100 wt % leaf-to-stem ratio of WNG portrayed a slightly higher moisture content as compared to the other ratios with 7.18 wt %. A decreasing trend was observed with a reduction in the mixing of the leaf-to-stem ratio. This could be due to the Napier leaf containing a higher quantity of water within its overall structure as compared to that of the stem, resulting in a lower moisture content. However, an exception was made for 100 wt % Napier stems from both WNG and PNG. The moisture contents for these two samples were slightly higher, with 4.79 and 7.07 wt %, respectively, even without the addition of the leaf part. This could be due to the nature of the Napier stem particles that is more porous, and without the addition of Napier leaf particles, the spaces between the particles cannot be filled up but instead the absorption of moisture happened during the storing.

Overall, the moisture content of each raw material from Napier grass is in accordance with international normative standards by ISO 17225-6,[23] which suggests that the moisture content should be below 10 wt %. The European Standards CEN/TC 335:M15 criteria[30] also noted that the moisture value for the pelletization process should be lower than 15 wt %. It is also obvious to say that as we mixed the leaf and stem parts, the different ratios affected the moisture content.

Volatile matter is believed to be the key composition in raw materials of biomass regardless of different types. All of the sample compositions showed a high amount of volatile matter, which has a positive influence on the sustainability of the pellet combustion process as reported in previous works.[21,31,32] Generally, the volatile matter for all samples showed no significant differences. However, for the WNG, an increasing trend was observed as the proportion of the Napier stem increased from 90.03 to 94.86 wt % except for the sample ratio with 100 wt % Napier stems from both WNG and PNG, which indicated a slight decrease with 93.9 and 91.29 wt %, respectively. Thus, a higher amount of stem will lead to a higher volatile matter content. The highest value of volatile matter (94.86 wt %) was recorded from the WNG (25:75) L/S ratio. In brief, the high volatile matter makes this Napier feedstock a possible candidate for production of highly reactive fuel with a faster combustion rate during the devolatization phase as compared to conventional fuels such as coal.

Ash content is an underlying property that affects the energy characteristics of biomass. It was previously reported that a higher ash content increases the maintenance cost and reduces the efficiency of heat value as the mineral elements are not involved during the combustion process.[33] In this experiment, the values from all samples tested were lower than 6 wt %, which complied with the ISO 17225-6:2014 standards for Class A solid pellet fuels, except for WNG (100:0) and WNG (75:25). The highest ash content recorded was the WNG (100:0) L/S ratio with a value of 8.39 wt %. The values showed a decreasing trend as the proportion of Napier stem increased with 100 wt % stem for both WNG and PNG species having the lowest values of 2.4 and 2.64 wt %, respectively. This indicates that a higher amount of Napier leaf contributed to the higher ash content. As a short conclusion, the ash content for all samples was low enough to be used to produce Napier grass biopellets.

The portion of biomass samples that remains as residue after the volatile matter distills off, after the sum of moisture and ash content is subtracted, is termed as fixed carbon. It is basically the amount of carbon with minor proportions of hydrogen, oxygen, nitrogen, and sulfur that are not driven off with the gases.[34] From this experiment, the values were reasonable for all pellet materials, with the highest recorded being 6.07 wt % for a 100 wt % stem-to-leaf ratio for both WNG and PNG. An increasing trend was observed as the ratio of leaf to stem was increased, except for the WNG with 75% stem, which decreased to 1.23 wt %. The fixed carbon value corresponds

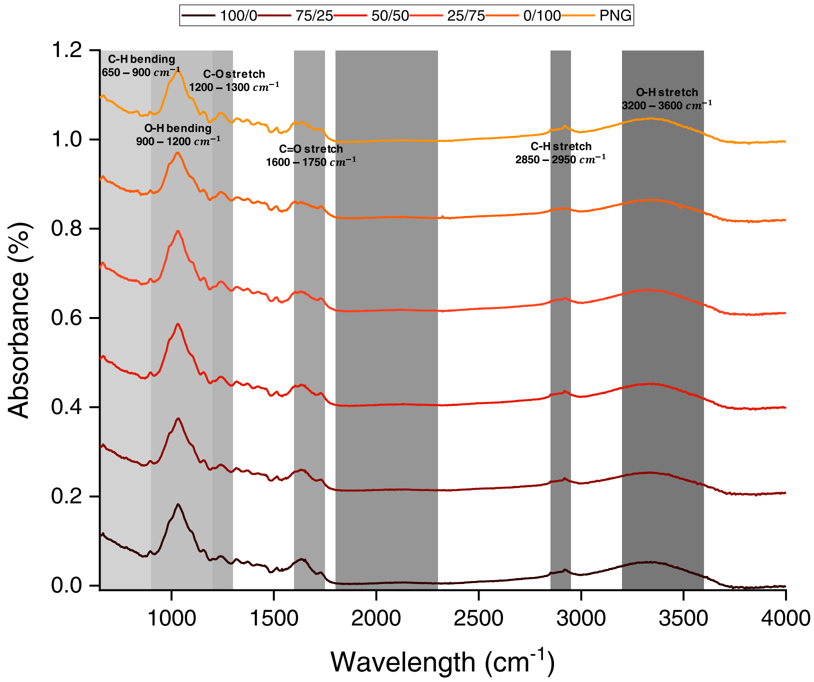

Figure 3. FTIR spectra of Napier grass feedstocks.

to the rise in the calorific value of that biomass, and this has been proven in many biomass-related research.[35] The increment behavior of fixed carbon in this study explained the carbonization mechanism of the Napier grass particles during combustion processes. Besides, this trend is also supported by the rise in the calorific value for all Napier grass samples.

Calorific value is the energy per unit mass released upon complete combustion. The gross calorific values for all of the Napier grass feedstocks were above 16 kJ/g, with PNG having the highest value of 17.901 kJ/g. For WNG, the calorific value increased as the proportion of the Napier stem increased in the range of 16.5−17.7 kJ, which corresponds to the amount of fixed carbon as the proportion of stem increased. The higher energy content from the stem part may be attributed to its lower ash content, high carbon content, and lower oxygen level as compared to the samples that contained varying ratios of leaves. All feedstocks met most of the European standards[36,37] for heating value (HHV), particularly in the Italian Standard: CTI-R04/05 under A.2 category, which is for untreated herbaceous biomass: a mixture of these materials.[30] This might be due to the higher fixed carbon content from the stem compared to the leaf as mentioned previously. The difference in compositions of the Napier leaf and stem led to variations of the net calorific value, which is basically affected by fixed carbon, volatile matter, and moisture content. C, H, and O are the main components of biomass materials, and this was proven by Tillman, who found a linear relationship between the net calorific value and carbon contents.[38]

When mentioning the energy content or calorific value, the heat of combustion is another important aspect that can support the energy performance of a biomass pellet. Heat of combustion is also known as the calorific value or the energy value, which can be defined as the amount of heat liberated when a given mass of substance undergoes complete combustion in the presence of oxygen under standard conditions for temperature and pressure. The correlation of heat of combustion with elemental composition was also performed following Boie's equation adapted from Burnham's report.[28] Based on Boie's equation, the derived results were consistent with the measured calorific values obtained from the bomb calorimeter, with the differences ranging from 1200 to 488 J/g across the sample ratios. According to the technical report from Burnham, the factors of oxygen and sulfur coefficients, functional groups, as well as moisture and lignin contents may contribute to the difference in values as compared to the obtained calorific values from the bomb calorimeter.[28] It is worth noting that the biomass has a more diverse set of organic molecules, which may need further improvement or a separate correlation. Nonetheless, for comparison, Boie's equation is qualitatively correct and in agreement with the obtained data. In summary, the energy content of all samples is consistent and predictable from their elemental compositions. The lower the oxygen content, the higher the carbon plus the hydrogen contents, resulting in a more exothermic state once all of the material is oxidized. Boie's equation thus showed a correlation between the heat of combustion with respect to elemental compositions toward the energy content and the calorific value of the biopellets. This shows that they are interrelated with each other.

The biomass underwent heating up, drying, devolatilization, and finally combustion of the volatiles and char during the combustion process. Napier leaves had higher contents of volatile matter than Napier stems, indicating that they could be easily ignited and burned. In addition, pollutant emission is also a factor to be considered when referring to biomass characteristics related to combustion. These emissions are generally affected by the contents of sulfur and nitrogen, resulting in the formation of gaseous pollutants such as SO_2 and NO_2.[3939] Owing to the considerably low ash and sulfur contents, the potential of Napier grass to be one of the promising safe candidates to replace the current energy fuels, along with promoting the go-green movement, is highlighted. Shen and team reported a study on the emission factors of several pollutants from the burning of two types of biomass pellets (pine wood and corn straw) and compared them to that of raw materials' combustion in a traditional cooking stove, and their findings showed that the emission factors of the pellets were significantly lower than those from the combustion of raw materials.[40] This indicates a possible significant reduction of emission factors by means of pelletizing the raw materials. The study on the emission of pollutants from Napier grass biopellets will be discussed in our next publication.

This shows conclusively that Napier grass has good properties as a raw material (Napier grass has high volatile matter and carbon compositions and low ash and sulfur contents). This, combined with a recent technology of pelletizing raw biomass, promises a potentially good-quality solid fuel with less pollutant emissions.

3.2. FTIR Analysis. Figure 3 shows the FTIR spectra for Napier grass feedstocks, while Table 3 summarizes the high-frequency regions, which were between 4000 and 2300 cm^{-1}, and the low-frequency regions, which were between 1670 and 650 cm^{-1}. The analysis will aid in the understanding of the nature of Napier grass as an emerging biopellet fuel for industrial applications. In the range of 650–2300 cm^{-1}, there was the presence of different alkyl, aromatic, alcohol, carbonyl, and ester functional groups within all samples. In the range of 3200–3600 cm^{-1}, the O–H stretching vibration indicated the possibility of the presence of water and hydroxyl groups (alcohol and phenols).[3,41−44] The slightly higher absorption in this range could be because of the presence of moisture content[45] and carboxylic acid[46] in the biomass sample. The alcohol and phenol groups may also be an indication of lignin in the sample.[47] The peak of 2850–2950 cm^{-1} can be associated with the aliphatic saturated C–H stretching bond from lignin components.[48] The fingerprint region 1600–1750 cm^{-1} indicates the C=O stretching band of lignin and could also be derived from the hemicellulose, which has a degree of acetylation presence.[49,50] The absorption in this range was due to the C=O stretching in the conjugated carbonyl of lignin.[45] The lower band observed in this range was possibly caused by adsorbed H_2O,[45] which showed the presence of moisture. The presence of cellulose was also proven based on the peaks in the region 1200–1300 cm^{-1}, which shows C–O stretching, indicating cellulose components from biomass sources. The presence of both cellulose and lignin was further observed at peaks 900–1200 cm^{-1}, as shown by the O–H stretching bond.[46] At peak 650–900 cm^{-1}, the bands may be attributed to the aromatic C–H bending vibration from the lignin in the sample.[50,51]

Table 3. FTIR Bond and Description of Napier Grass Feedstocks

bond	description	wavenumber (cm^{-1})
C–H bending	aromatic compound	650–900
O–H bending	alcohol, phenol (cellulose and lignin)	900–1200
C–O stretch	ester, ether (cellulose from biomass)	1200–1300
C=O stretch	ketone, aldehyde, carboxylic acid, ester (lignin)	1600–1750
C–H stretch	aliphatic/alkyl (alkyl methylene from lignin)	2850–2950
O–H stretch	alcohol, phenol, lignin	3200–3600

Based on WNG feedstocks, which included the combination of both leaf and stem in weight percent, it can be said that there was no new bond formation that took place. Based on FTIR analysis, the amount of C and O–H groups' intensity is depicted by the absorbance (%). The sharpness of the FTIR peak indicates the amount of functional groups' presence in the biopellet feedstocks. The O–H peaks can be associated with the moisture content, while the C–H and C≡C bonds are related to the carbon content. The O–H peak for WNG with a 100:0 leaf-to-stem ratio feedstock indicated the highest moisture content, and this is supported by the proximate analysis shown in Table 2. The carbon content is correlated with HHV, where a higher carbon content will contribute to a higher HHV, which is desired for the formation of biopellets.[52]

Generally, there were no major differences in both species and with different proportions of lead-to-stem ratios. This indicates that a similar chemical structure exists among them. Slight differences were noticeable based on the peaks of specific functional groups, which were mostly supported by proximate and ultimate analyses.

3.3. Biopellet Density and Durability with Statistical Analysis. Pellet density is an essential quality factor to evaluate substantial storage facilities, spaces, and handing systems.[53] Logistics purposes, handling, and storage efficiency are greatly affected by the bulk density of pellets. Higher bulk density leads to greater transport efficiency and lower storage space. Figures 4 and 5 show the density and durability of PNG pellets, respectively, while Figures 6−11 show the density and durability of WNG. Tables 4 and 5 show the ANOVA results for both PNG and WNG biopellets, respectively, with different dependent variables observed. Both WNG and PNG biopellets consisted of two pressure points and at maximum and relaxed densities. Data collections for all Napier biopellet densities were done with six replications, and this was supported by the error bars.

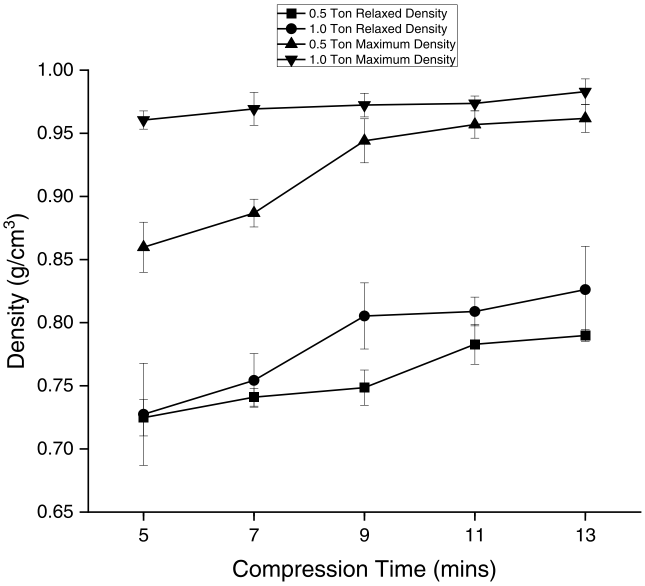

Figure 4. Density of PNG biopellets.

Since the sample for PNG was only from the stem, there were no leaf-to-stem-ratio parameter results. Figure 4 shows the trends for both maximum and relaxed density of biopellets produced from a 100% stem of PNG. Both maximum and

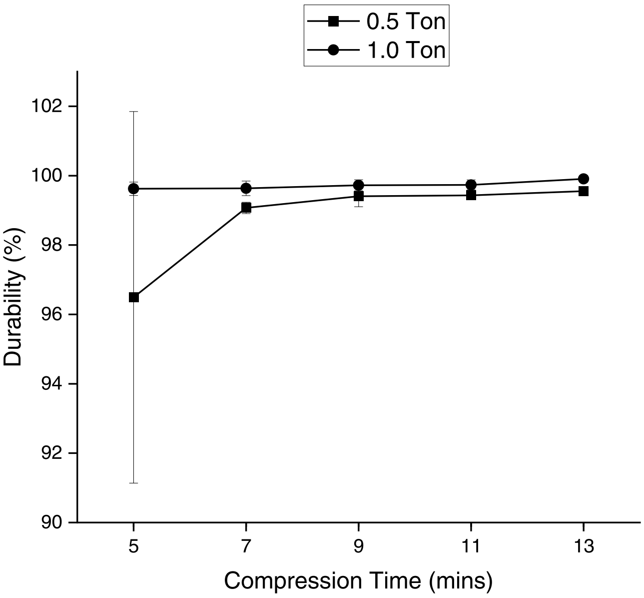

Figure 5. Durability of PNG biopellets.

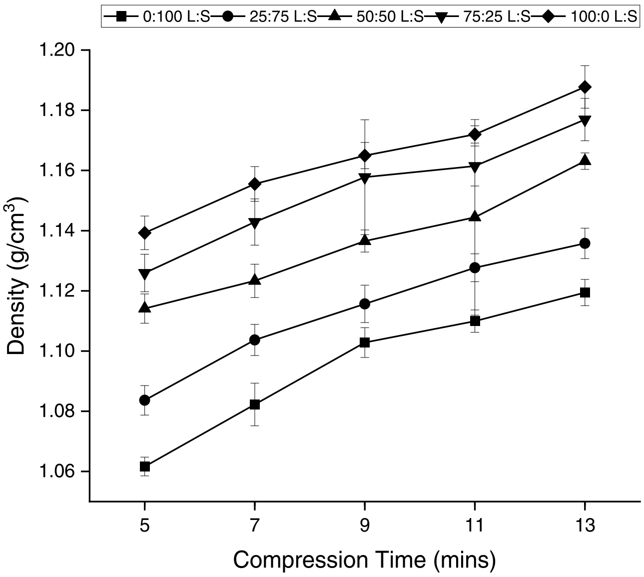

Figure 6. Maximum density of WNG biopellets at 73.83 MPa pressure point.

relaxed densities are directly proportional to the compression time. This trend was supported by the statistical analysis shown in Table 4, where compression time (T), compression pressure (P), and their interactions at various orders significantly affected both maximum and relaxed densities of PNG biopellets (p-values below 0.0001), indicating there was a significant interaction between compression time and pressure toward PNG biopellet density.

When higher compression times of 5−13 min were used, the maximum density increased. However, at a higher pressure, the increment of the maximum density was not significant, especially the changes from 7 to 9 min and from 9 to 11 min. Both resulted in p-values of 1, as shown on the least-square means (LSMs) for the effect of both compression time and pressure, which indicated insignificant increment. For the maximum density at 36.91 MPa, for PNG biopellets, the same increasing trend was observed when higher compression times were used. The increasing trend observed was significant when the compression times change from 5 to 7 min and from 7 to 9 min. The p-values obtained were 0.0122 and <0.0001, which indicated the significant effect of having a higher compression time at a lower pressure point. When comparing the pressure points between maximum densities, the differences were significant when applying a higher pressure at lower compression times, especially at 5 and 7 min. The differences when using a higher compression pressure were significant with p-values lower than 0.0001.

For relaxed density, at 73.83 MPa compression pressure, a similar increment trend was observed as higher compression times were used, as shown in Figure 4. The LSM showed a significant increment when going from 7 to 9 min compression time with a p-value of 0.0215; meanwhile, on going from 9 to 11 min, there was an insignificant effect with a p-value of 1. At a lower pressure of 36.91 MPa, the relaxed density obtained showed the same increasing trend as the compression time was increased. The biggest significant effect however was recorded when the time changed from 9 to 11 min, supported by a p-value of 0.1681, while the least recorded was for 7−9 min compression time with a p-value closer to 1 (0.9998). When comparing the relaxed densities from both pressure points, it is obvious to say that the effect of applying a higher compression pressure was minimal. The recorded relaxed densities from both pressure points with the same compression times used were almost similar to one another with minimal significant effects. This trend was later supported by the ANOVA test based on Table 4, where the interaction between time and

Table 4. ANOVA Results for PNG Biopellets

dependent variable	variables	sum of squares	DF	mean square	F value	P value
maximum density	time (T)	0.03387357	4	0.00846839	56.32	<0.0001
	pressure (P)	0.03735216	1	0.03735216	248.4	<0.0001
	$T \times P$	0.01813573	4	0.00453393	30.15	<0.0001
	error	0.00751865	50	0.00015037		
relaxed density	time (T)	0.04775516	4	0.01193879	26.24	<0.0001
	pressure (P)	0.01140654	1	0.01140654	25.07	<0.0001
	$T \times P$	0.00405131	4	0.00101283	2.23	0.079
	error	0.02274651	50	0.00045493		
durability	time (T)	60.01559793	4	15.0038995	0.98	0.441
	pressure (P)	22.78361373	1	22.7836137	1.49	0.237
	$T \times P$	55.41864806	4	13.854662	0.9	0.4801
	error	306.3775817	20	15.3188791		

Table 5. ANOVA Results for WNG Biopellets

dependent variables	variables	sum of squares	DF	mean square	F value	P value
maximum density	ratio (R)	0.24643124	4	0.6160781	903	<0.0001
	time (T)	0.13001611	4	0.03250403	476.4	<0.0001
	pressure (P)	0.6091185	1	0.6091185	8928	<0.0001
	$R \times T$	0.00603768	16	0.00037735	5.53	<0.0001
	$R \times P$	0.00453127	4	0.00113282	16.6	<0.0001
	$T \times P$	0.00232548	4	0.00058137	8.52	<0.0001
	$R \times T \times P$	0.00287579	16	0.00017974	2.63	0.0008
	error	0.0170567	250	0.00006823		
relaxed density	ratio (R)	0.52338976	4	0.13084744	1582	<0.0001
	time (T)	0.06156274	4	0.01539068	186.1	<0.0001
	pressure (P)	0.61427792	1	0.61427792	7426	<0.0001
	$R \times T$	0.0086741	16	0.00054213	6.55	<0.0001
	$R \times P$	0.02945765	4	0.00736441	89.03	<0.0001
	$T \times P$	0.000718	4	0.0001795	2.17	0.0729
	$R \times T \times P$	0.00589275	16	0.0003683	4.45	<0.0001
	error	0.02068002	250	0.00008272		
durability	ratio (R)	0.8309242	4	0.20773105	13.89	<0.0001
	time (T)	0.33796177	4	0.08449044	5.65	0.0004
	pressure (P)	1.70377633	1	1.70377633	113.9	<0.0001
	$R \times T$	0.35508102	16	0.02219256	1.48	0.1206
	$R \times P$	0.42788881	4	0.1069722	7.15	<0.0001
	$T \times P$	0.04964703	4	0.01241176	0.83	0.5091
	$R \times T \times P$	0.47042226	16	0.02940139	1.97	0.0226
	error	1.49528484	100	0.01495285		

pressure showed an insignificant trend toward the relaxed density with a p-value of 0.079. This indicated that the interaction between these two factors did not manage to significantly affect the relaxed density even at the onset, and the single interaction for each compression time and pressure did affect the relaxed density significantly. It is obvious from this that high compression time will not be profitable for industrial applications from an energetic and economic point of view. The cost that may be incurred from a higher compression time does not result in significant densification.

In view of the above, even though the relaxed densities of PNG biopellets produced followed similar trends to that of maximum densities, the values were much lower. This was due to a phenomenon called "spring back effect," where the pellets underwent reduction in density due to expansion caused by high residual stress within the pellets.[24,54] Maximum density measurement produced the highest density values for PNG biopellets as compared to relaxed density. This was expected as the maximum density was calculated right after the pellet was made. In general, a high-quality pellet should have a specific density of at least 1 g/cm^3, which was not achieved here (this is the effect of spring back, which may be smaller when the temperature factor is added). It is suspected that the compacted biomass remains intact, which contributed to a higher density as compared to that of a relaxed density. In terms of pressure points however, for maximum density, opting for a higher pressure had a significant effect only at lower compression times. Taking this into consideration, even the maximum density produced the highest density for the PNG feedstock, and the relaxed density remained the true density for logistic purposes. This true density affects the durability of PNG biopellets.

As for the durability of PNG biopellets, Figure 5 shows the durability at different compression times and two different pressure points. The durability test was performed using the drop-type method with three replications. Generally, an increasing trend was observed at the 36.91 MPa pressure point. For 73.83 MPa compression pressure, the increasing trend was minimal. The durability of PNG biopellets was directly proportional to the compression time. Looking at the ANOVA data from Table 4, both single and multiple interactions of time and pressure had no significant effects on the durability of PNG biopellets with higher p-values of 0.441 and 0.237 for single interactions, respectively, while for cross-multiple interactions, the p-value was 0.48. This indicated that opting for a higher compression time at an already high-enough pressure results in minimal durability values. Regardless of all of these insignificant effects from the interaction of both compression time and pressure, all PNG biopellets produced managed to satisfy the European Standard for Pellet Durability Index by having more than 95% score. It is worthy of note that a typical DU test has a different measurement procedure than the one in this work (ISO 17225-2:2014).

For WNG biopellets, Table 5 shows the ANOVA of the factor affecting the WNG biopellet maximum density. One extra independent variable, which is the ratio of leaf to stem (L/S) feedstocks (R), was varied, and each ratio system was tested for both maximum and relaxed densities. Figure 6 shows the maximum density of WNG biopellets at different leaf-to-stem ratios and at 73.83 MPa pressure point. Based on the figure, at different pressure points with different leaf-to-stem ratios, the graph showed an increasing trend when higher compression times were used, with WNG biopellets having a 100 L/S ratio resulting in the highest maximum density of 1.18 g/cm^3. For the maximum density at 36.91 MPa pressure point of WNG biopellets as shown in Figure 7, all sample ratios exhibited the same increment trend when applying longer compression times, with the highest maximum density recorded at 13 min compression time with a 100:0 leaf-to-

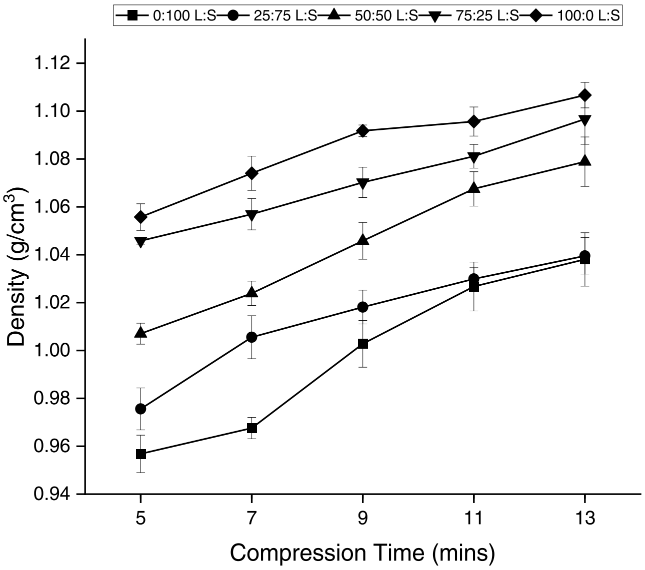

Figure 7. Maximum density of WNG biopellets at 36.91 MPa pressure point.

stem ratio. It can be deduced that the maximum densities of WNG biopellets at both pressure points are directly proportional to that of compression time. The same increasing trend was observed for the feedstock ratio, indicating a significant effect as the ratio of leaf to stem increased. In other words, the higher the proportion of leaf used, the higher the maximum density produced. Based on Table 5, multivariate analysis of maximum density with different factors (R, T, and P) and their interactions showed highly significant effects for all single factorials and for all two interactions with p-values lower than 0.0001. For the full factorial (interaction among all three independent variables R, T, and P) however, a slightly lower p-value with 0.0008 was observed. Nevertheless, this value is still way lower than 0.05, indicating the high significant effect toward WNG biopellets' maximum density.

From both maximum densities' graph from both pressure points, 100:0 L/S resulted in the highest densities as compared to the other ratios. In terms of compression time, 13 min remained the best compression time to be used to produce pellets with a high density. Opting for higher compression pressure promises the best maximum density for WNG biopellets, noting a very significant effect with less than 0.001 p-values.

As for the case of the relaxed density of WNG biopellets, Figures 8 and 9 show the relaxed densities for WNG obtained at 73.83 and 36.91 MPa pressure points, respectively. The exact same increasing trend was observed when opting for longer compression times and when going for higher leaf-to-stem ratios, with the best relaxed density turning out to be at the 100:0 L/S ratio and 13 min compression time. Looking specifically at the interaction between both compression time and pressure however showed insignificant effects toward the relaxed density of WNG biopellets. The overall interactions from both independent variables gave a p-value of 0.0729 (Table 5), which is slightly higher than 0.05, indicating a less significant effect toward the relaxed density of WNG biopellets. An in-depth LSM analysis on the interaction effects from both T and P, for instance, at the 73.83 MPa pressure point showed that there were insignificant effects when jumping from 9 to 11 min and from 11 to 13 min compression time, noting slightly higher p-values of 0.0855 and 0.0646, respectively. As for the full factorial analysis of all three independent variables however, a p-value less than 0.0001 was obtained, speculating a highly significant effect toward the relaxed density of WNG biopellets. In short, it can be summarized that the interaction between T and P did not influence the WNG relaxed density significantly, which contrasts with that of the single interaction from both variables, where they independently showed highly significant effects toward WNG relaxed densities. The full factorial analysis showed that the inclusion of the ratio of leaf to stem (R) did help minimize the effect between T and P, thus lowering the p-values below 0.05, indicating the R variable plays a significant role in increasing the pellet density as compared to other variables.

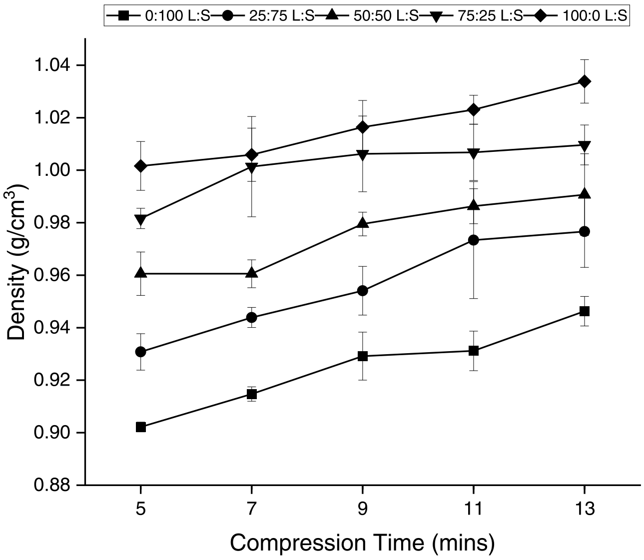

Figure 8. Relaxed density of WNG biopellets at 73.83 MPa pressure point.

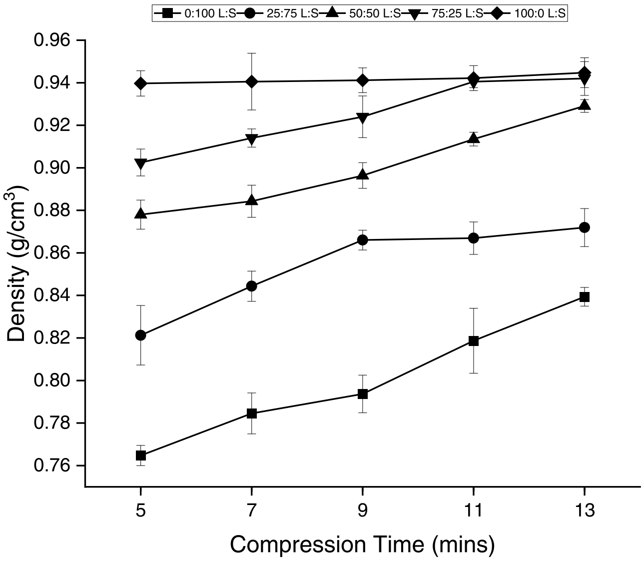

Figure 9. Relaxed density of WNG biopellets at 36.91 MPa pressure point.

Figures 10 and 11 show the durability for WNG biopellets at 73.83 and 36.91 MPa pressure points, respectively, while Table 5 shows the ANOVA data of the factor affecting the WNG biopellet's durability. At 73.83 MPa, there was an increment

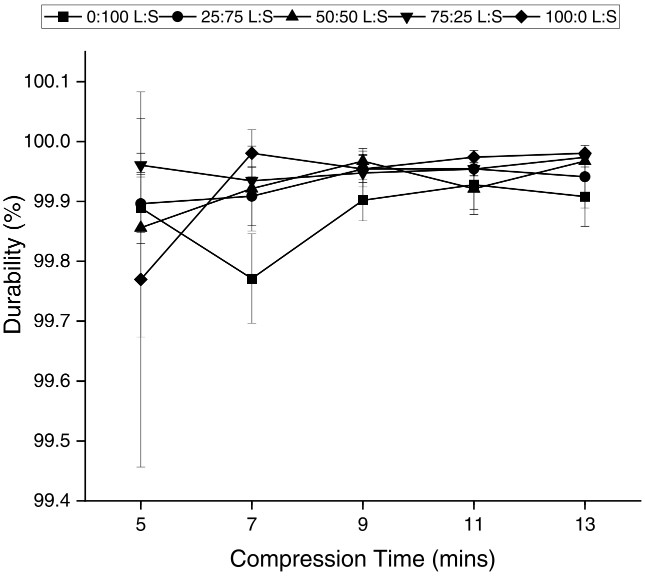

Figure 10. Durability of WNG biopellets at 73.83 MPa pressure point.

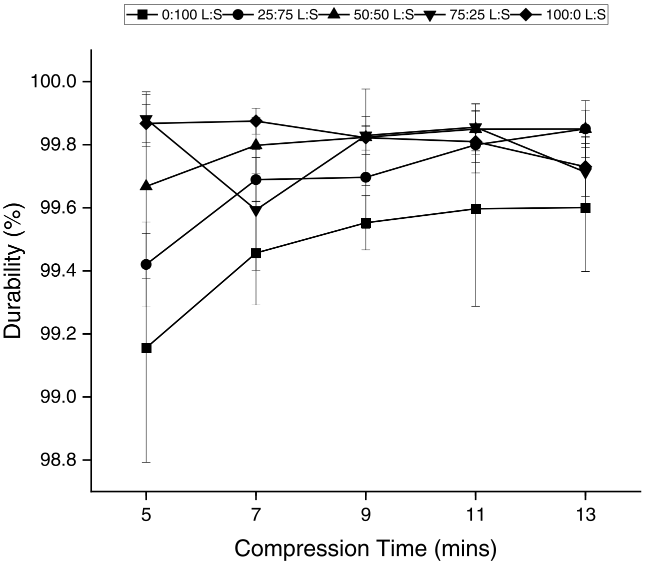

Figure 11. Durability of WNG biopellets at 36.91 MPa pressure point.

trend when using a higher proportion of leaf; however, it was not always the case. For example, at 73.83 MPa pressure point and 5 min compression time, the durability showed a decreasing trend when the proportion of leaf increases with the exception of the 75:25 L/S ratio. At 7 min and above however, an increasing trend was observed for the durability of WNG as the leaf proportion increased. Furthermore, increasing compression times seemed to help increase the WNG durability for most ratios except for the 5 min compression time.

More inconsistent data were observed for the durability of WNG at the 36.91 MPa pressure point, as shown in Figure 9. Increasing the compression times resulted in an increasing trend for the durability of WNG biopellets for the first three compression times (5, 7, and 9 min), with a single drop at 7 min for the 75:25 L/S ratio. At 11 min, the durability increased as the ratio increased; however, a slight drop was observed at the 100% L/S ratio. Meanwhile, at 13 min, the only increment was observed at the 25:100 L/S ratio, while the rest of the ratios seemed to drop in durability. The inconsistency was supported by a statistical analysis. All three independent variables achieved p-values less than 0.05 for single factorial analysis, which indicated there were significant effects toward the durability of WNG biopellets. However, for the cross-interactions ($R \times T$ and $T \times P$), p-values were more than 0.05 (0.1206 and 0.5091, respectively), which showed disagreement, noting insignificant effects toward the WNG durability. Results for the full factorial, however, gave a significant effect toward WNG durability with a p-value of 0.0226. Even with the inconsistent data, all WNG biopellets, like PNG biopellets, managed to satisfy the European Standard for Pellet Durability Index by having values higher than 95%.

A function of moisture content against durability was plotted, as shown in Figure 12. The intention was to study and report the effect of moisture content on the strength of the biopellets. The moisture content values were taken from Table 2, and the set of durability data was taken at the 73.83 MPa pressure point. Based on Figure 12, in general, the effect of moisture content on the durability of biopellets showed a consistent trend except for 7.07 wt % moisture. The decreasing trend was observed when going across each moisture content, which supported the previous statement, noting that the higher leaf-to-stem ratio resulted in a higher durability. The exception could be due to the different species or ratio used. At 7.07 moisture content, the ratio was 100% stem of PNG Napier grass. The fibrous nature and structure of the Napier stem, which has the tendency to take up and trap the moisture from the surrounding, could be the reasons why the moisture content was slightly higher. This resulted in a slightly lower durability when compared to that of other ratios with leaf proportions. This indicates the importance of having the optimum amount of lignin and moisture for them to synergically function as binding agents to improve the durability of the biopellets.

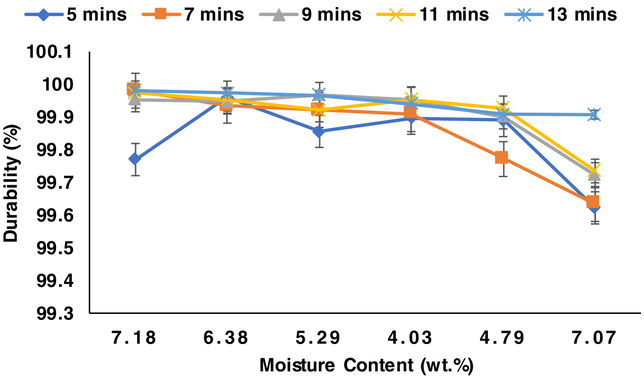

Figure 12. Durability of the Napier grass biopellets at 73.83 MPa against the moisture content of the raw Napier grass.

Comparing the results between two different species is important since the type of species is also one of the major factors that contribute to the pellet properties. While there were two different densities studied, the focus was more on the relaxed density since it is also defined as the true density for the pellet market. As for the relaxed density, both species were able to produce densified biomass pellets, achieving more than 0.650 g/cm^3, a criterion set by the European Standard for

Pellet Production. As for the durability comparison, both satisfied the European Standard for Pellet Durability Index, even with the inconsistent results. The difference was however noticeable when a comparison between different compression times was made. The WNG, with the inclusion of the leaf part, exhibited a higher density and durability as compared to the PNG, especially at a higher pressure point and ratio of leaf to stem used. This was suspected earlier as the leaf part of Napier grass contains higher moisture and lignin contents.

The explanations on density and durability have been elaborated in previous studies. With regard to the effect of applied compression force, it was reported that in the first stage of the densification mechanism, rearrangement of particles happened, where the major pore volume decreased significantly, thus increasing the particle packing. This would cause an increase in the pellet density due to the reduction of the empty spaces between the particles.[55] At a low pressure point, the particles will rearrange and maintain their original physical properties, provided the compression time is fixed. At a higher pressure point, elastic and plastic deformations occur, which cause the particles to flow into the void spaces and increase the contact area between the particles as well as with the walls of the die before entering the second stage.[56] In the second stage, the applied pressure forces the particles to collide with each other and with the walls of the die, leading to interparticle contact. Some papers reported that the capillary forces in the movable liquid phase, adhesion and cohesion forces in the nonmovable phase, natural binders, formation of solid bridges, and mechanical interlocking are some factors that contribute to pelletization formation.

The moisture content of both feedstock species was already in the optimum range. The lower initial moisture content of biomass evidences the occurrence of the natural evaporation of water and plasticization. It is believed that the lower the amount of moisture content, the lower the heat required for the evaporation. In any case, even with a much lower moisture content, the presence of this minimum quantity of moisture is necessary for the gelatinization of the starch in the biomass and it is needed for capillary pressure and interfacial forces to develop, which later promote the binding of particles.[57] In a more recent study, the use of different binders was investigated as reported by Rajput et al.,[58] where aqueous-based binders showed a positive impact on the pellet properties as compared to oil-based binders. In another study, increasing the moisture content to more than 20% reduced the durability and hardness of the pellets due to the leachate squeezing out of the materials during pelletization.[59] It can be deduced that moisture content plays a major role in producing durable biopellets.

The effect of lignin content is obvious especially when referring to how the pelletization mechanism happened. Typically, lignocellulosic biomass contains chemical components, for example, lignin, proteins, and starch. As mentioned earlier, the inclusion of the leaf part of WNG endows WNG with better density and durability due to the higher lignin content as compared to the other ratio, which consists of a combination with the stem part of Napier grass. In general, the presence of lignin helped enhance the binding characteristics of the densified pellets due to its low melting point (140 °C),[60,61] which commonly melts during the pelletization process, especially with the introduction of higher temperature as one of the parameters studied. However, in this study, no temperature factor was studied and it was impossible to check for the rise of temperature during the pelletization process. It was suspected that the rise of temperature happened as a result of the frictional forces formed between the particles and the walls of the die, which could potentially soften and melt the lignin from the sample to a certain degree. The external surfaces of the biopellets were assumed to have an increase in the temperature due to the friction forces against the die wall. The increase in temperature was suspected to be in the range of the plasticization temperature where the lignin was believed to softened and eventually solidifed, acting as binder for the particles to bind to each other and form the biopellets. An almost similar study by Tilay et al. reported, in the range of 70 °C with the applied temperature of 90 °C, that the temperature was sufficient for the crude protein present in the sample to denature and melt the lignin and hemicellulose to form the pellet.[62]

Besides lignin, protein denaturation will also improve the solid bridge formation and attraction forces between the particles, hence producing a more compact pellet structure.[63] Xie et al. reported the presence of protein content in Napier grass up to 5%,[64] and this was further supported by a study by Rusdy et al., stating that the range of the protein content of Napier grass varies from 4.4 to 20.4%.[65] Formation of a new bond was expected to happen during protein denaturation with other available molecules, where the functional groups of the unfolding protein like hydroxyl, carboxyl, and amine are available to form new hydrogen bonds with other materials and water molecules present as moisture.[66] The reassociation of the protein and chemical bonding upon cooling also influences the hardness and durability of the pellets.[66] It is this cooling phenomenon that necessitates the need to study the effect of compression time. The effect of compression time was critical in this case as it gives the meaning of holding the compression force at specific time. It is believed that a longer compression time will help in both the melting and cooling of the chemical components from the biomass sample. In other words, the particles will have a higher heat energy generated from the frictional forces among themselves as well as with the die walls. The same mechanism is suspected to help in cooling the melted components associated with the bindings and bonding of the particles. This is true in some cases as the maximum energy generated occurs right after the force is applied on the biomass sample. It is suspected that the speed of the die used to compress the sample will affect and contribute to the amount of heat generated through collisions. However, the speed of the die press was not investigated in this study.

Each of the mechanisms has different effects on the pelletization process. The solid bridge is responsible for the determination of the final strength of the pellets, where it usually forms by crystallization of dissolved substances, hardening of natural binders, and some from the effect of chemical reactions. Different biomass composition approaches also affect the pelletization mechanism, where most of the major ingredients such as starch, proteins, lipids, fibers, and water contribute to different types of binding and respond differently to the different temperatures and pressures.

4. CONCLUSIONS

This study mimics a simplified model of a common industrial pelletization practice in which Napier grass is used since it has rarely been discussed in the literature and because this species is considered versatile. Certain significant physical and mechanical parameters were considered to understand and study the density and durability of Napier grass biopellets.

Some conclusions were deduced based on the specific compression parameters, for example, the compression time and pressure together with the pelletization materials and method used. The moisture contents of all materials used were below 15%, which meet the minimum requirements under European Standards CEN/TC 335:M15 criteria.[30] The high volatile matter and fixed carbon of Napier feedstock made it a good candidate for energy pellet production. This was supported by its high gross calorific values. The ash content values were negligible, with less than 10%, as they passed most standards for pellet solid fuels. The FTIR spectrum was in line with proximate and ultimate analyses, showing similar chemical structures across different parts and species of Napier grass. The Pakchong species biopellet recorded the highest durability of 99.95% at 73.82 MPa pressure and 13 min compression time. However, at high pressure, the effect of compression time was minimal. For wild Napier species, using a higher pressure resulted in the highest density with values of more than 1.0 g/cm^3 for maximum density and around 0.9 g/cm^3 for relaxed density. The highest recorded durability for the WNG biopellet was 99.98% for a 100:0 L/S ratio at 13 min compression time and at a 73.82 MPa pressure point. Even with all of the inconsistent data, both PNG and WNG biopellet densities obtained were higher than 0.650 g/cm^3. All in all, the overall data indicated that high-quality biopellet fuel was produced, thus demonstrating the sustainability of utilizing Napier grass for energy applications.

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The following research activities were performed by specific authors: conceptualization, A.S.I., N.B.O., D.O.P.; data curation, A.S.I., N.B.O.; formal analysis, A.S.I., N.B.O.; funding acquisition, N.B.O.; investigation, A.S.I., D.O.P.; methodology, A.S.I., N.B.O., D.O.P.; project administration, N.B.O.; resources, A.S.I., N.B.O.; validation, N.B.O.; visualization, A.S.I.; writing—original draft, A.S.I.; writing—review and editing, N.B.O., D.O.P.

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Notes

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