

Original Article

# Application of hydrochar as binding material for enhanced fuel properties of torrefied biomass pellets

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Received: 25 July 2023; Revised: 8 November 2023; Accepted: 22 December 2023

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## Abstract

This study aimed to improve the physical and mechanical properties of pellets produced from torrefied sugarcane leaves blended with hydrochar as a binder. The hydrochar was obtained through hydrothermal carbonization of sugarcane leaves. Pellets were produced from blends of torrefied biomass with hydrochar in different ratios. The results showed that adding 10–50% of the binder significantly increased density and compressive strength of the torrefied pellets. Single pellet density ranged from 1,059.2 kg/m<sup>3</sup> to 1,135.5 kg/m<sup>3</sup>, while compressive strength increased by 58–160%. The addition of hydrochar also decreased ash content (3.4–25.2%) and reduced sulfur and nitrogen contents (6–28% and 3–16%, respectively). Optimal conditions for pelletizing were determined at 70/30 proportions of torrefied biomass and binder. The resulting pellets had 10.8% ash content, 9.20% moisture content, 1,111.2 kg/m<sup>3</sup> single pellet density, 6.66 MPa compressive strength, and 18.37 MJ/kg lower heating value. These findings demonstrate the potential of incorporating hydrochar as a binder to improve the properties of torrefied biomass pellets.

**Keywords:** hydrochar, torrefied pellet, compressive strength, torrefaction, binder

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## 1. Introduction

Sugarcane leaves, typically seen as agricultural waste, are produced when harvesting sugarcane. In 2021, Thailand generated 20.19 million tons of sugarcane leaves and tops. Unfortunately, farmers often burn the remaining leaves, causing air pollution. Sugarcane leaves, with an HHV of 17.3 MJ/kg (Menandro *et al.*, 2017), hold energy potential similar to wood (18.54–19.92 MJ/kg) (Domingos *et al.*, 2020). These leaves can be used as an energy crop to reduce waste and air pollution. Unprocessed biomass faces challenges like high moisture absorption, poor storability, and low bulk density. Pretreatment methods, particularly torrefaction and pelletization, are crucial for upgrading biomass for biofuel use.

Torrefaction involves heating biomass without oxygen at temperatures of 225–300 °C. It enhances various properties like energy density, grindability, hydrophobicity, stability, and uniformity. However, torrefied biomass has lower bulk density than the raw biomass. Therefore, pelletization is crucial to increase density and energy content. Binder is an essential component used in the production of torrefied biomass pellets. Natural binders in the biomass itself, such as lignin, protein, starch, fat, and water-soluble carbohydrates (Kaliyan, & Vance Morey, 2010), are partly decomposed and transformed during torrefaction, leading to a reduction in their content. Especially lignin, commonly used in pellet production, strengthens and densifies pellets, forming strong connections between particles (Azargohar *et al.*, 2019). This results in a material that is more brittle and less cohesive compared to raw biomass. Moreover, pelletization is more challenging compared to raw biomass (Butler, Skrivan, & Lotfi, 2023) due to the spring-back effect, which requires

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higher compression force and temperature. Therefore, additional binders may need to be introduced in the pelletization to maintain pellet integrity. Various binders, including tapioca flour, corn starch, vegetable oil, lignin (Abdul Rasid, Elamparithy, Ismail, & Harun, 2021), and molasses (Wang *et al.*, 2019), have been studied. Different binders have shown mixed results; some enhance durability but decrease heating value (Tarasov, Shahi, & Leitch, 2013), while waste engine oil has been found to increase it (Xia *et al.*, 2019). Binders also increased the ash content, requiring consideration for suitable binders that balance both heating value and ash content.

Hydrothermal carbonization (HTC) involves a hydrothermal pretreatment or upgrading in subcritical water, as indicated by the phase diagram of water, within a mild temperature range of 180–250 °C, pressures up to 4.6 MPa, and a residence time of 5–240 minutes (Mlonka-Mędrala *et al.*, 2021). At high temperatures and pressures, water acts as a reactant (Medina-Ramos *et al.*, 2014), breaking down complex organic compounds ultimately reducing volatile matter. In the supercritical state, water serves as a reactant, solvent, and heat transfer medium (Weingartner & Franck, 2005), facilitating biomass conversion to hydrochar.

The lignocellulosic components in hydrochar consist of hemicellulose, cellulose, and lignin, as well as a small amount of extractives and ashes. Hydrochar typically contains lower hemicellulose levels than raw biomass, making it more susceptible to hydrolysis. This is due to the amorphous structure of hemicellulose, which facilitates its breakdown (Ahmad & Zakaria, 2019). Research indicates that lignin does not decompose at 200 °C and shows only 15% conversion at 230 °C and 23% at 260 °C (Reza *et al.*, 2014). Notably, lignin itself is not produced during the HTC process; instead, the degradation of hemicellulose and extractives leads to an increase in lignin content.

Hydrochar acted as a filler by filling the void spaces in the torrefied char and binding the torrefied char by solid bridging (through electrostatic attraction, hydrogen bonding, van der Waals forces, mechanical interlocking, and viscous forces) (Islam *et al.*, 2022). The process enhances the durability, strength, and bonding of the pellets without requiring external binders. Lignin decomposition in hydrochar forms solid bridges between particles during cooling, enabling biomass particles to bond (Cao *et al.*, 2020) by heating lignin above its glass transition temperature. Normally, the glass transition temperature of lignin is 100–140 °C. Reza *et al.*, (2014) revealed that hydrochar produced at 260 °C temperature had a higher heating value of up to 26.5 MJ/kg, while maintaining a low ash content of only 0.6%. Importantly, hydrochar has the advantage of a low ash content and an adequate amount of lignin, allowing it to act as a natural binder without the need for external binders.

Numerous studies in the literature have examined the pelletization of both pure and blended biomass samples. Acharya *et al.*, (2012) summarized a comparison of fuel properties and handling characteristics between raw wood, wood pellets, and torrefied wood pellets. Almeida *et al.*, (2017) studied sugarcane bagasse pellets with an average diameter of 9.70 mm, length of 22.70 mm, bulk density of 726.32 kg/m<sup>3</sup>, and 98.2% durability. Proximate analysis showed 8.70% ash, 5.49% moisture, 14.03% fixed carbon, and 77.27% volatile matter. The heating value was 16.0 MJ/kg.

Jarunglumlert *et al.*, (2022) found that untreated pellets had a calorific value of 15.04 MJ/kg, 3.14% ash, 97.36% durability, and a bulk density of 627.5 kg/m<sup>3</sup>. After torrefaction at 280 °C, these had changed to 17.12 MJ/kg, 3.5% ash, 96.76% durability, and bulk density of 620.3 kg/m<sup>3</sup>. Hydrochar pellets from sugarcane bagasse at 180 °C exhibited the lowest ash content (0.97%), highest durability (99.92%), and highest bulk density (637.4 kg/m<sup>3</sup>).

This study aimed to explore the impacts of hydrochar binder on the physicochemical and mechanical properties of torrefied-hydrochar blend pellets. This research addresses a gap in existing knowledge and seeks to produce high-quality pellets from torrefied sugarcane leaves, offering insights for the utilization of biomass from sugarcane and similar sources.

## 2. Materials and Methods

### 2.1 Feedstock preparation

Sugarcane leaves from Kalasin agricultural research center, Thailand, were finely ground and sieved through an 18-mesh screen to ensure a particle size no larger than 1 mm. The samples were then heated at 105 °C for 24 hours, cooled to room temperature, and stored in airtight plastic bags.

### 2.2 Preparation of torrefied biomass

20 g of finely ground sugarcane leaves from Section 2.1 were placed in a stainless-steel reactor and sealed. The reactor was then heated in a high-temperature furnace (Nabertherm LT model, Germany) at a rate of 10 °C/min until reaching 275 °C. Throughout the process, a continuous flow of nitrogen gas (2 L/min) was maintained for 30 minutes. The reactor was then allowed to cool while still under a nitrogen atmosphere. The resulting torrefied product was collected and stored in airtight plastic bags for further analysis.

### 2.3 Preparation of hydrochar

The process involved weighing 20 g of finely ground sugarcane leaves and adding them to a hydrothermal reactor with deionized water at a 1:5 ratio (biomass to water by weight) (Volpe *et al.*, 2020). After stirring for 15 minutes, the reactor was heated at a constant heating rate of 6 °C/min to a target temperature of 225 °C and maintained for 30 minutes. Pressure was reduced by cooling the reactor in 10 °C water. The resulting solid, hydrochar, was dried at 105 °C and stored in airtight plastic bags for further analysis and used as a binder in fuel pellet production.

### 2.4 Preparation of blend pellets

In Section 2.2, torrefied solid was mixed with hydrochar binder (obtained from Section 2.3) in the weight ratios 100:0, 90:10, 80:20, 70:30, 50:50, and 0:100. The mixtures were labeled with abbreviations indicating the percentage of hydrochar, namely 0, 10, 20, 30, 50, or 100%. Pelletization was performed using a single pelleting unit apparatus mounted on a universal testing machine (Model No. WDW-100D, Startech, China). The length of a single pellet was controlled by a computer. Weighing 10 g of torrefied

solids and hydrochar, the moisture was adjusted to 17%. Normally, the glass transition temperature of lignin is 100–140 °C (Peng *et al.*, 2015), so the die was heated to 130 °C and held for 30 min. A 1 g sample of the mixture was added and pressed into biomass fuel pellets through an 8 mm in diameter and 10 mm in length die using a rod at a constant speed of 10 mm/min and the pellets were extruded after 1 minute. When cooled to room temperature, the fuel pellets were stored in airtight aluminum bags for further analysis.

## 2.5 Analytical methods

The proximate and ultimate analyses of pellet samples were conducted using thermogravimetric analyzer (TGA 701 model, LECO, USA) and CHNS/O element analyzer (Flash 2000 model, Italy). The higher heating value (HHV) and lower heating value (LHV) were determined in MJ/kg using the empirical equations by Sheng and Azevedo (2005) and Brachi *et al.*, (2016), respectively. The main elements of lignocellulosic biomass were evaluated following the methodology outlined by Basu (2013). The surface morphological features within the pellet were observed by using a scanning electron microscope (JEOL JSM-6460LV). A universal testing machine (LLOYD LR50K model, England) was used to analyze the compressive strength of the pellets. The single pellet density of the pellets was analyzed from the measured mass and volume of an individual pellet (Chew *et al.*, 2018). One-way ANOVA was used to assess the differences between means using SPSS software.

## 3. Results and Discussion

### 3.1 Fracture surface of blend pellets

Figure 1 depicts the fracture surfaces of torrefied, hydrochar, and blend pellets at 10x magnification. In Figure 1(a), the torrefied pellet exhibits numerous cracks, indicating poor cohesion due to the absence of a binder. Conversely, Figure 1(f) shows a smooth, glossy, and relatively uniform surface without significant voids. When hydrochar was used as a binder, the external surface became progressively smoother and shinier, as shown in Figures 1(b) to 1(e), correlating with the binder proportion. It is observed that higher hydrochar content enhances pelletization due to the unique glass transition behavior of hydrochar. This behavior is a distinctive characteristic of natural binders. Lignin and hemicellulose, the two major components displaying natural binding ability in biomass, contribute to this effect (Reza *et al.*, 2014). Feed containing a higher percentage of lignin and hemicellulose exhibits better binding characteristics when compressed under their glass transition temperatures—the point at which they transition from a glassy to a high-elastic state. In accordance with Islam *et al.* (2022), as per the SEM images, the blend pellet compactness increases with the higher doses of hydrochar.

Additionally, Figure 1 shows observable changes in the length of the torrefied pellets. The average length of the torrefied pellets became shorter, suggesting a reduced spring-back effect with an increasing proportion of the binder. The use of hydrochar as a binder improved the cohesion of the torrefied particles during pelletization, owing to its superior binding properties compared to torrefied biomass.



Figure 1. Appearance of torrefied biomass, hydrochar, and blend pellets

Consequently, this reduces the expansion of the pellet after it has been manufactured.

### 3.2 Organic elemental analysis of blend pellets

The carbon, hydrogen, and oxygen contents of torrefied pellets blended with hydrochar derived from sugarcane leaves are presented in Figure 2(a). The torrefied pellet exhibited the highest carbon content (52.29%), while the hydrochar pellet had the lowest carbon content (50.52%). As a result, in the blend pellets, the carbon content decreased from 52.12% to 51.41% with the addition of hydrochar (10–50%), and no significant changes occurred. In contrast, the addition of hydrochar (10–50%) significantly increased the hydrogen and oxygen contents.

Figure 2(b) displays the nitrogen and sulfur contents in pellets of torrefied biomass blended with hydrochar. Torrefied biomass pellets had the highest nitrogen (0.67%) and sulfur contents (0.18%), while hydrochar pellets had the least with nitrogen at 0.45% and sulfur at 0.07%. The addition of hydrochar (10–50%) decreased nitrogen and sulfur contents in the blend pellets by 2.98% to 16.41% and 5.56% to 27.78%, respectively, due to these elements being dissolved in gaseous and liquid components during hydrothermal carbonization. This is beneficial for reducing NO<sub>x</sub> and SO<sub>x</sub> emissions during combustion or cofiring (Kongto *et al.*, 2021).

### 3.3 Proximate analysis of blend pellets

Table 1 shows the proximate analysis of raw leaves. The volatile matter, moisture content, fixed carbon, and ash of raw biomass were 74.9, 7.0, 15.3, and 9.8%, respectively. Figure 3 presents the proximate analysis of sugarcane leaf-derived torrefied biomass, hydrochar, and blend pellets. The moisture content ranged from 7.85% to 9.01%, complying with the ISO/TS 17225-8:2016 standard in not exceeding 10%. Torrefied pellets had the lowest VM (58.9%), and the highest FC (21.6%) and ash (12.7%). Hydrochar pellets

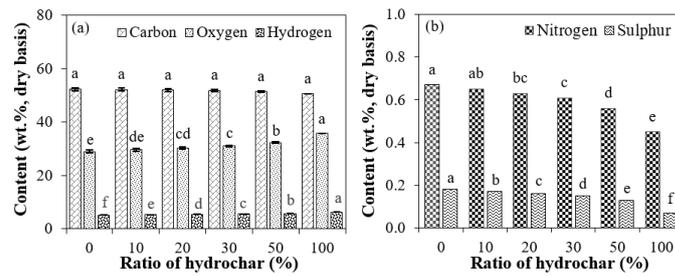


Figure 2. Chemical composition of torrefied biomass, hydrochar, and blend pellets (Significant differences ( $p \leq 0.05$ ) between samples are indicated by different letters.)

Table 1. Properties of raw biomass, hydrochar, and biochar derived from sugarcane leaves

Parameter	Raw	hydrochar	biochar
Mass yield (%)	100	48.4	68.2
Fiber analysis (wt.%, dry basis)			
Hemicellulose	36.7	1.9	4.0
Cellulose	41.4	57.1	46.1
Lignin	6.4	17.3	36.5
Other	15.5	23.7	13.4
Ultimate analysis (wt.%, dry basis)			
C	44.5	50.0	51.0
H	6.1	6.2	5.1
N	0.7	0.4	0.7
O <sup>a</sup>	38.8	36.3	29.4
S	0.2	0.1	0.2
Higher heating value (MJ/kg)	17.7	20.0	19.4
Lower heating value (MJ/kg)	16.9	18.6	18.3
Proximate analysis (wt.%, dry basis)			
Fixed carbon	15.3	18.0	22.71
Volatile matter	74.9	75.0	63.6
Ash	9.8	7.0	13.6
Moisture (wt.%, as received)	7.0	1.1	1.4

<sup>a</sup> Calculated by difference (C + H + N + O + S + Ash = 100%).

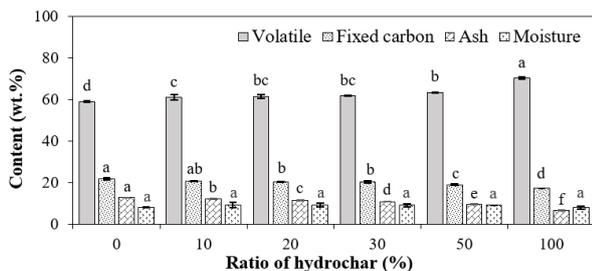


Figure 3. Proximate analysis of torrefied biomass, hydrochar, and blend pellets (Significant differences ( $p \leq 0.05$ ) between samples are indicated by different letters.)

showed the highest VM (70.1%), and the lowest FC (17.1%) and ash (6.6%) due to decomposition at a lower temperature (225 °C) than in torrefaction (275 °C). Blend pellets exhibited a reverse pattern to torrefied pellets with increasing hydrochar composition (VM, 60.8–63.2%; FC, 18.9–20.8%; and ash, 9.5–12.3%).

Volatile matter in biomass includes substances that vaporize at lower temperatures, like water, organic compounds, and certain inorganic elements. Both torrefaction

and hydrothermal carbonization decrease volatile matter. Torrefaction involves heating biomass to remove moisture, releasing volatiles including water, which break down into gases and solid char. Higher temperatures break down organic compounds, reducing volatile matter. Similarly, hydrothermal carbonization removes water and, at high temperatures and pressures, breaks down complex organic compounds, leading to reduced volatile matter (Medina-Ramos *et al.*, 2014).

In Figure 3, it is clear that the hydrochar pellet has a lower fixed carbon content compared to the torrefied biomass pellet. As a result, the inclusion of hydrochar as a binder in pellet production decreases the fixed carbon content. The addition of 10% hydrochar decreased the fixed carbon content of blend pellets with no significant difference from the torrefied biomass pellet. Moreover, a higher proportion of volatile matter was observed with increased binder content, which made ignition of the pellets easier (Chen *et al.*, 2019).

Figure 3 shows the ash content of the pellets. Torrefied biomass pellets had the highest ash content (12.7%), while hydrochar pellets had the lowest (6.6%). The ash content of torrefied biomass pellet was notably 49.3% higher than that of hydrochar pellet. Using hydrochar as a binder significantly reduced the ash content of the pellet, offering a promising solution to address high ash content in pelletized fuel. High ash content can lead to lower heating value and potentially reduce combustion system efficiency. Biomass tends to have high ash content due to inorganic minerals and elements absorbed from the soil during growth. These minerals include potassium, sodium, calcium, and silica (Cheruyiot, 2011). The increase in torrefied biomass ash content stems from volatile component removal and is a concentration effect. Torrefaction extracts organic compounds, leaving a denser mass of solid components, including minerals and inorganic substances, contributing to ash content. As torrefaction reduces total mass while retaining minerals, the proportion of minerals in the final product rises, resulting in higher ash content. Torrefaction raises ash content in treated biomass, while hydrochar exhibits lower levels. Hydrochar, produced through hydrothermal carbonization, involves high temperatures and pressures with water, causing minerals like potassium, sodium, and calcium to dissolve, which reduces ash content. Additionally, the removal of volatile compounds during the process reduces overall material mass, but despite the concentration effect the ash content is lowered.

The results of the heating value determinations are shown in Figure 4. The HHV and LHV of the torrefied biomass pellet were 19.38 and 18.25 MJ/kg, respectively,

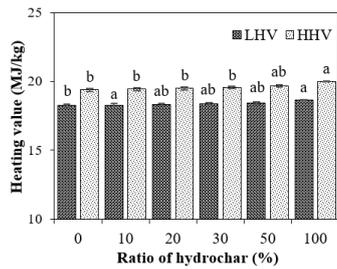


Figure 4. LHV and HHV of torrefied biomass, hydrochar, and blend pellets (Significant differences ( $p \leq 0.05$ ) between samples are indicated by different letters.)

while the energy contents of hydrochar pellet in the same order were 20.00 and 18.64 MJ/kg. With the addition of hydrochar (for 10% and 50%), the HHV and LHV of blend pellets increased from 19.45 and 18.29 MJ/kg to 19.69 and 18.44 MJ/kg, respectively. However, there was no significant change in heating value with the addition of hydrochar, likely because the hydrochar was derived at a lower temperature than the torrefied sample.

### 3.4 Morphological features of pellets from torrefied biomass blended with hydrochar

Figures 5(a-f) depict cross-sectional surface characteristics of torrefied biomass, hydrochar, and blend pellets. Specifically, Figure 5(a) shows the cross-section of torrefied biomass pellets, processed at 275 °C without a binder. The surface appears rough and non-uniform with scattered elongated cracks. These features result from moisture and natural binder loss during thermal treatment, leading to particle compaction and adhesion solely through compressive force, without the usual binding agent present in the torrefied biomass.

Figure 5(f) depicts a cross-sectional image of hydrochar pellet. The interfaces between hydrochar particles exhibited a relatively smooth appearance, and they demonstrated a comparable level of homogeneity when compared to the torrefied biomass pellets. This particular conditioning method helps preserve hydroxyl groups (-OH) and natural binders, resulting in a softer texture compared to torrefied biomass. When pelletized, the hydrochar particles exhibit plasticity and readily form connections due to the presence of highly polar functional groups retained after

conditioning. The formation of these connections is made possible through the interaction of multiple forces, such as electrostatic attraction, hydrogen bonding, van der Waals forces, mechanical interlocking, and viscous forces (Islam *et al.*, 2019), which act between adjacent particles during compression.

The addition of hydrochar (10-50%) resulted in significantly smoother cross-sectional surfaces of the pellets, as shown in Figures 5(a-f). The hydrochar can fill the gaps between the irregular-shaped torrefied biomass particles, resulting in fewer voids being observed. This indicates an improved interconnection among the torrefied particles, suggesting enhanced cohesion facilitated by the binder. This agrees with Reza *et al.*, (2014), who found that pine pellets torrefied at 300 °C exhibit numerous cracks and a noticeable absence of binding. When 10% or more hydrochar is added, the cracks diminish in size. Further, addition of 25% hydrochar reduces the particle gap even more, resulting in pellets that are 93% more durable. The increased inclusion of hydrochar in the pellets leads to a reduction in void space, allowing for the formation of solid bridge-type bonds.

### 3.5 Diameter, density, and compressive strength of blend pellets

Figure 6 displays the diameter and length of torrefied, hydrochar, and blend pellets. Torrefied pellets had an average diameter of 8.64 mm. Blend pellets with 10-50% hydrochar content met the ISO/TS 17225-8:2016 standard for 8 mm fuel pellets (allowing 7-9 mm diameter), with diameters ranging within 8.36-8.47 mm. Different binder proportions led to decreased radial expansion of blend pellets compared to torrefied biomass pellets, but with no significant change in average diameter.

Figure 6 shows that torrefied biomass pellets (0% hydrochar) were the longest at about 17.8 mm, while hydrochar pellets (100% hydrochar) were the shortest at around 13.9 mm, deviating from the target size of 8 mm in diameter and 10 mm in length. Increased pellet length post-extrusion indicates expansion behavior. Torrefied biomass pellets expanded more due to the torrefaction having reduced natural binding agents. Hydrochar pellets, on the other hand, exhibited lower expansion due to strong bonding between hydrochar particles. The lignin in hydrochar softens during pelletization at 130 °C, maintaining structural integrity (Venegas-Vasconez *et al.*, 2023).

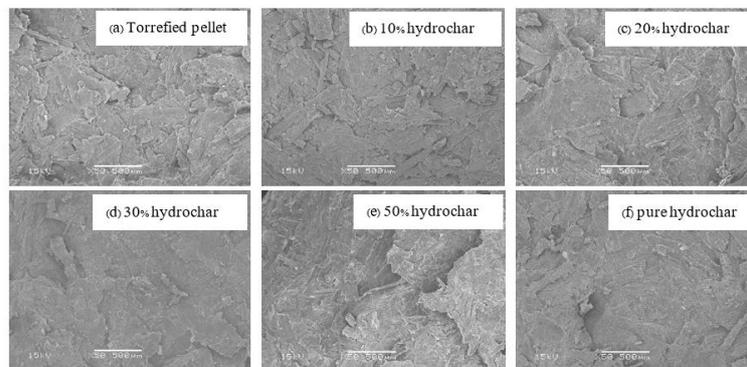


Figure 5. SEM images of torrefied biomass, hydrochar, and blend pellets

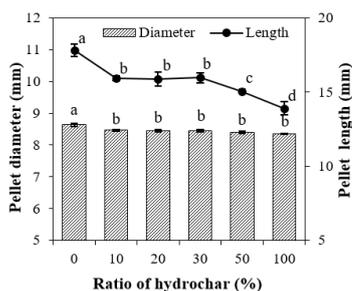


Figure 6. Diameter and length of torrefied biomass, hydrochar, and blend pellets (Significant differences ( $p \leq 0.05$ ) between samples are indicated by different letters.)

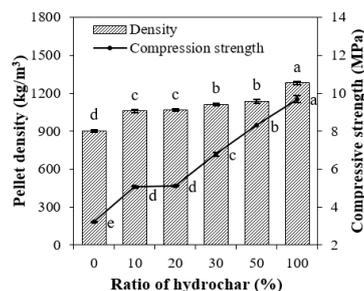


Figure 7. Pellet density and compressive strength of torrefied biomass, hydrochar, and blend pellets (Significant differences ( $p \leq 0.05$ ) between samples are indicated by different letters.)

Figure 7 displays the single pellet density and compressive strength of torrefied biomass, hydrochar, and blend pellets. The average density of single torrefied biomass pellets was  $901.0 \text{ kg/m}^3$ . However, the addition of hydrochar (10-50%) resulted in a significantly higher single pellet density, ranging from 1,059.2 to 1,135.5  $\text{kg/m}^3$ , within the acceptable standard range of 1,000–1,400  $\text{kg/m}^3$  for single pellet density (Stelte *et al.*, 2011). At a significance level of 0.05, there was no difference in pellet density between 10% and 20% binder cases. Also, 30% and 50% cases showed similarity.

Torrefied biomass pellets exhibited the lowest compression strength (3.2 MPa) due to gaps between biomass particles. In contrast, hydrochar pellets demonstrated over three times higher strength (9.7 MPa). The addition of hydrochar (10-50%) resulted in a significant increase in compression strength, ranging from 5.1 to 8.3 MPa. Compared to torrefied biomass pellets, the blend pellets had compression strength increased by 58% (10% hydrochar) and 160% (50% hydrochar). However, no significant difference was found between the 10% and 20% binder cases. Optimal binder performance requires a blend proportion of hydrochar exceeding 20%.

The increased mechanical strength observed in hydrochar pellets can be attributed to the formidable binding forces present within the pellets. These forces are primarily a consequence of phenolic polymers (lignin structures) acting as natural binding agents, as well as the substantial presence of polar functional groups on the surface of hydrochar, leading to strong hydrogen bonding. Furthermore, the removal of extractive compounds originally found in the raw biomass further enhances this strength. The compression strength of the pellets is affected by the amount of lignin in the solid products (Surdi de Castro *et al.*, 2021).

#### 4. Conclusions

Hydrochar enhances pellet cohesion, acting as a binder by forming bridges between biochar particles. This leads to a 160% increase in pellet compressive strength when blended at 50%. Additionally, hydrochar has higher heating value and lower ash content compared to torrefied biomass, and can be produced at lower temperatures. Blending hydrochar as a binder improves pelletization and enhances mechanical and energy properties of torrefied biomass pellets. Future research should explore torrefied biomass pellet

production at different die temperatures, considering their correlation with lignin softening temperature.

#### Acknowledgements

The authors sincerely acknowledge the Department of Physics, Faculty of Science and Technology, Rajabhat Mahasarakham University, for their invaluable research support.

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