Producing pellets from torrefied herbaceous biomass in a commercial capacity

Research Project

by

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Abstract

The global temperature rise has pushed governments more into finding ways to reduce CO_2 emissions, by increasing the use of renewable fuels. Lignocellulosic biomass (wood, forest residues, agricultural residues) is a renewable fuel that has still not been utilized to its full potential. That is because it lacks properties such as homogeneity, high volumetric energy density and low moisture that its main fossil fuel competitor -coal- has. Therefore, a type of pre-treatment is needed for these disadvantages. Torrefaction, a process of heating biomass (200-300 ^oC) in an inert environment (no combustion), provides a product with improved physicochemical properties: reduced hydrophobicity, easier grindability, homogeneity, reduced microbial activity and increased calorific value. However, it is not enough to torrefy the biomass, as it needs to be densified in order to be utilized. With the process of pelletizing, a type of densification, torrefied biomass becomes compact and can be considered as bio-coal. While pelletizing is influenced by many parameters (moisture content, die pressure, torrefaction temperature etc), herbaceous types of biomass such as wheat straw, hay, reeds and grass, still cannot produce quality pellets without the addition of a binder. Typical organic binders such as starch, lignin and sawdust can either be expensive or biologically degrade in storage conditions. That is why addition of plastic binder should be considered. The aim of this report was to investigate how quality torrefied pellets from herbaceous biomass can be produced with the addition of plastic binder in a commercial capacity. Wheat straw and woodchips were used as feedstock, while polyethylene resin was the plastic binder. The biomasses were torrefied at 240 ^oC and 270 ^oC on the Torrgreen facilities, via a pilot-scale packed bed reactor, which recycles the volatiles from torrefaction for inertization and pelletized with a 100 kg/h pellet mill. For the torrefied wheat straw, a design of experiments was formed, to investigate how 4 parameters for pelletizing (torrefaction temperature, moisture content, plastic binder addition and pellet diameter) improved mechanical durability. Results indicated that 5% of plastic improved durability and pellet formation in total, while higher torrefaction temperature weakens the pellets and aggravates pellet formation. The highest durability achieved was 90.3%. Torrefied wood chips were pelletized for reference and showed low durability, mostly due to inability of the pelletizer. Water immersion tests on the straw pellets showed that higher torrefaction temperature increased hydrophobicity a lot, while plastics also had a positive effect. Comparison of the straw pellets produced with conventional wood pellets revealed that the former lack in mechanical durability and are high in ash content, making them incompliant with current standards. Overall, inclusion of plastics in pelletizing of torrefied herbaceous biomass, demonstrated very positive results and improved their quality, while making the operation smoother, but for scaling up and keeping the whole process sustainable a polyethylene (or other plastic) waste stream should be utilized.

1. Introduction

1.1 Background information

Climate change concerns have been rising more and more each year, especially due to the evident extreme climate phenomena occurring more often (U.S. Global Change Research Program, 2018). Due to that, the 21st Conference of the Parties (COP21) in 2015 along with other measures, introduced the Paris Agreement, which obligates the participating countries to take measures to treat climate change and keep the worldwide temperature change below the threshold of 2 ^{o}C (UNFCCC, 2016), while the more recent COP26 at Glasgow set as target to keep the 1.5 ^{o}C within reach (Allan et al., 2021), thus actions need to be taken even quicker. A way to achieve that is to take advantage of renewable energy sources, among them lignocellulosic biomass and in particular agricultural residues, which can contribute to the energy needs of countries that produce them (T. Liu et al., 2014). Currently, the most common type of biomass being used in a large scale is various types of wood (forest and wood industry residues) (Maciejewska et al., 2006) which are co-fired with coal in power plants. Especially in the Netherlands, a total of 2.76 million metric tonnes of wood pellets were imported for co-firing in coal power plants, while they covered approximately 10% of the total renewable energy consumption (USDA, 2021). However, raw wood pellets are not the ideal solid fuel, being inferior compared to coal due to high amount of volatiles, high O/C ratios, high moisture content and low bulk density (Maciejewska et al., 2006; Prins et al., 2007). These disadvantages are increased if instead of wood, other more fibrous biomasses such as hay and straw are used. The fibres obstruct the co-pulverization with coal, while also increase energy demands for grinding (Phanphanich & Mani, 2011). Another important factor that makes raw biomass unsuitable is its biological and microstructural degradation which may occur in storage conditions (Cutz et al., 2021; Kumar et al., 2017). The need for a more uniform, moisture resistant, high energy density biomass-based fuel is essential, in order coal use can be diminished.

Torrefaction is a mild thermal treatment for biomass that takes place in an inert atmosphere and at temperatures 200-300 ^{o}C (J. Li et al., 2012). The process' main advantage is the energy densification achieved, as due to the temperature almost all water content is lost, while low-energy volatiles are also lost, resulting in an overall higher calorific value product (W. H. Chen et al., 2015). Besides that, torrefied biomass becomes hydrophobic and loses all microbial activity, resulting in a fuel that can also be stored long-term without any additional issues (W. H. Chen et al., 2015). In general, Figure 1 contains the most significant upgrades that torrefaction has on the raw biomass. Nevertheless, the degree that a biomass is torrefied plays a huge role on the final product's properties, which will be discussed on the next chapter. Overall, torrefaction is deemed to be the answer to the many issues of untreated biomass cause and can potentially offer a product similar to coal.

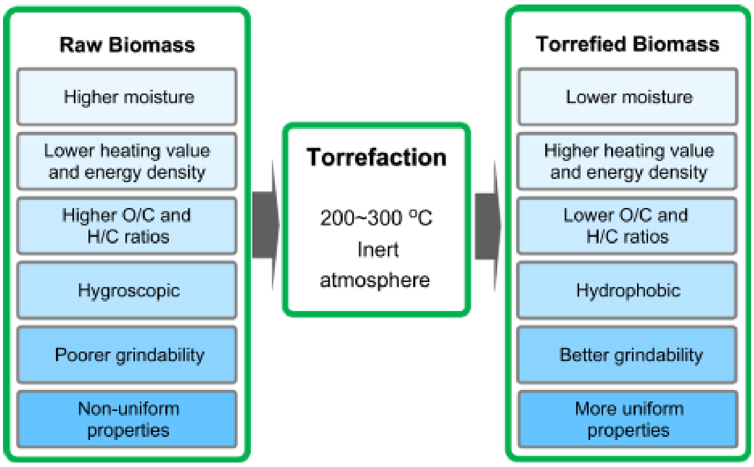

Figure 1. Main advantages of torrefaction on raw biomass's mechanical and thermochemical properties (W. H. Chen et al., 2015).

However, torrefying biomass itself is not enough to create a biofuel. Typically, once biomass has been torrefied it is subjected to a densification process known as pelletization. By pelletizing biomass, the overall feedstock volume reduces, thus increasing the bulk density up to 20 times and the volumetric energy density 4-10 times (H. Li et al., 2012). These advantages ease logistics and reduce transportation costs. Pellet quality is affected by numerous feedstock's factors such as the pre-treatment (drying, torrefying or grinding), its physicochemical properties and the addition of binders (Rudolfsson et al., 2017; Wang et al., 2020). Pelletizing equipment's operational parameters such as die temperature and pressure and pellet press type (Rudolfsson et al., 2017) are also critical to the whole process efficiency.

Currently, most of the studies on pelletizing with torrefied biomass, includes woody biomass feedstock such as pine, birch, bark and spruce. Those feedstocks have given great results, as they naturally contain a larger amount of lignin (compared to other types of biomass), which acts as a binding agent and strengthens the pellet even after torrefaction (W. H. Chen et al., 2021; van der Stelt et al., 2011; Wang et al., 2020). Nevertheless, agricultural residues, which are mostly herbaceous types of biomass, such as wheat straw, hay, reed or grass have only a handful reports regarding torrefaction and pelletization (Agar et al., 2021; Emadi et al., 2017; San Miguel et al., 2022). That is because in general herbaceous biomasses are more challenging for both of these processes. To solve the pelletizing problem, Emadi et al., (2017) attempted adding a small amount of low-density polyethylene (LDPE) plastic to produce torrefied wheat straw pellets and it showed great results. However, they used a single press pelletizer which does not simulate the industrial conditions of a commercial capacity pelletizer.

1.2 Problem statement and research question

Utilizing an agricultural residue waste stream via torrefaction and pelletization has barely been reported and there are still various knowledge gaps around it. The pelletizing part especially, is the one that needs further research. That is because no study has attempted to produce quality pellets commercially from herbaceous biomass by altering various parameters, including the plastic binder addition. Therefore, the goal of this project is to answer the following research question:

1. *"How can high quality torrefied pellets from herbaceous biomass and plastic blends be produced continuously in a commercial capacity?*

Which can be split into three more detailed questions:

1.1 "*What is the effect of moisture content and plastic binder in the pellets' mechanical durability?*"

1.2 "*How do torrefaction temperature and pellet diameter additionally affect the biomass-plastics pellets' properties*?"

1.3 "*How do torrefied herbaceous biomass pellets compare to conventional wood pellets?*"

Regarding research question 1.1, moisture addition is critical for pelletization after torrefaction, with a level of 10-15% being the optimal for improved pellets (Roberto García et al., 2018; Kumar et al., 2017; Rudolfsson et al., 2017; Stelte et al., 2011). Different plastic binders have been used in pelletization of torrefied biomass (PET, PP, LDPE, MDPE) and most of the studies concluded that a level of 5-10%wt highly improves mechanical properties such as hardness, mechanical durability and tensile strength, while also thermochemical ones such as HHV (Cramwinckel, 2020; Emadi et al., 2017; Peng et al., 2021; Samaksaman et al., 2019). Thus, by testing combinations of the 2 parameters, we expected pellets with improved mechanical durability.

The hypothesis for the 2nd question (1.2) can be answered from the study of Agar et al., 2021, where they pelletized biomasses in pilot-scale and concluded that moderate to high torrefaction temperatures (250-280 ^{o}C) influence negatively durability. However, the report from Rudolfsson et al., 2017 conclude the opposite, but for higher temperatures (290-315 ^{o}C) and the pellets were produced in a single pellet press. The most expected outcome still is the first mentioned, meaning durability will decrease with higher torrefaction temperature. Regarding the pellets' diameter, increasing the diameter would indirectly decrease the pressure applied on the pellet. Wang et al. (2020) mentioned that "high compression pressure" is needed for achieving pelletization from torrefied biomass and producing good quality pellets, thus it is expected that larger diameter pellets will be of worse quality.

Regarding research question 1.3, a design of experiments is going to be formed, resulting in various combinations of pellets in terms of moisture content, plastic binder, torrefaction temperature and die diameter. In general, it is more likely that the wood pellets would be superior in terms of mechanical characteristics due to their higher lignin content and that the energy content of the torrefied pellets is going to be closer to the wood ones, in comparison with raw herbaceous biomass pellets (Agar et al., 2021; Föhr et al., 2017).

2. Theoretical background

2.1 Biomass

Biomass can be shortly defined as the "living and recently dead biological species that can be used as fuel or in chemical production" (Basu, 2010). It comes from botanical or biological sources or both. In general, categories of biomass are agricultural (food grain, seed hulls), forest (trees, sawdust), municipal waste, energy crops (soybean and canola) and biological waste from animals (Basu, 2010; Cramwinckel, 2020). The most abundant of the aforementioned is claimed to be the plant dry matter or lignocellulosic biomass (Ge et al., 2018).

Therefore, the potential of lignocellulosic biomass is of high importance. By the term lignocellulosic, biomass is characterized by its 3 major polymeric components: cellulose, hemicellulose and lignin (Negi et al., 2020). These chemical compounds, based on the type of biomass, are usually found in compositions of 40–60%, 15–30% and 10–25%, respectively (D. Chen et al., 2018).. Cellulose is a linear macromolecular polysaccharide and the primary structural component of cell walls, providing the "skeletal structure of most terrestrial biomass"(Basu, 2010). Hemicellulose belongs to a group of carbohydrates of low polymerization and is also a component of cell walls, but with much less strength and structure compared to cellulose (Basu, 2010). Lignin is a biopolymer that is responsible for the structural strength of the plant, by being a binding agent of the cellulose fibres, holding them together (Basu, 2010). It is found in larger concentrations in woody biomasses (25-30% wt), which are characterized by slow growth and tightly bound fibres, while in herbaceous biomasses that grow faster and have their fibres more loosely packed, the composition ranges below 20% wt (Cramwinckel, 2020; McKendry, 2002; Negi et al., 2020).

2.2 Torrefaction

Torrefaction is a thermal pre-treatment process for lignocellulosic biomass, where it is heated up to 200-300 ^{o}C for a specific amount of time, in an inert atmosphere (absence of oxygen) (W. H. Chen et al., 2015). By not having oxygen, combustion is avoided, while the hemicellulose content of the biomass is consumed. Lignin and cellulose are only slightly decomposed, but they undergo some physicochemical changes (W. H. Chen et al., 2015; Joshi, 2015). In particular, devolatilization is the first decomposing process that occurs and affects only hemicellulose, especially in lower temperatures close to 200 ^{o}C. At these temperatures only depolymerization occurs on cellulose and lignin, while devolatilization above 250 ^{o}C. The physicochemical processes are shown more analytically in **Figure 2**.

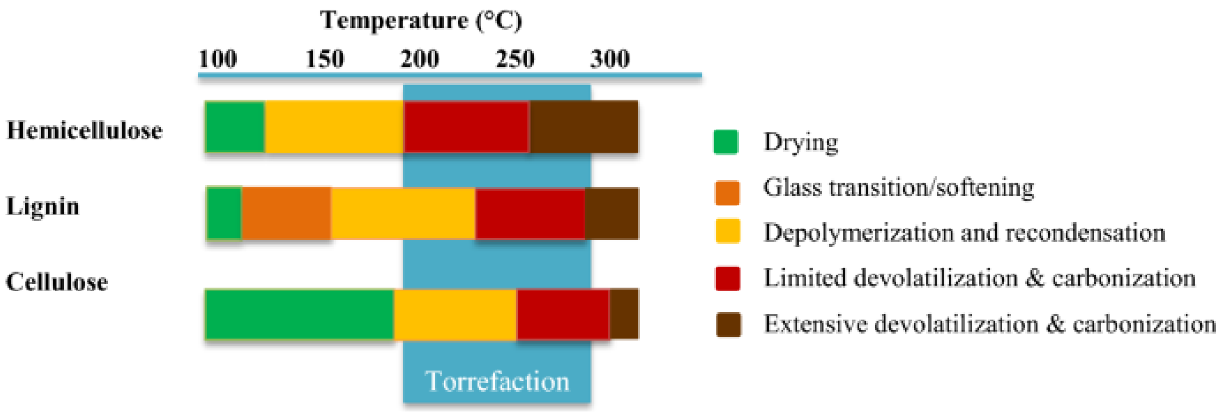

Figure 2. Basic stages of torrefaction and their effect on hemicellulose, cellulose and lignin (Negi et al., 2020).

2.2.1 Advantages of torrefaction

The effect of torrefaction produces a biomass with very low moisture content, higher heating value, lower atomic H/C and O/C ratios, much enhanced grindability and reactivity, improved hydrophobicity and eliminated biological activity (W. H. Chen et al., 2015; Joshi, 2015; van der Stelt et al., 2011). These advantages make torrefied biomass a solid biofuel that can resemble coal much more than raw biomass, as can be seen in the van Krevelen diagram in **Figure 3**.

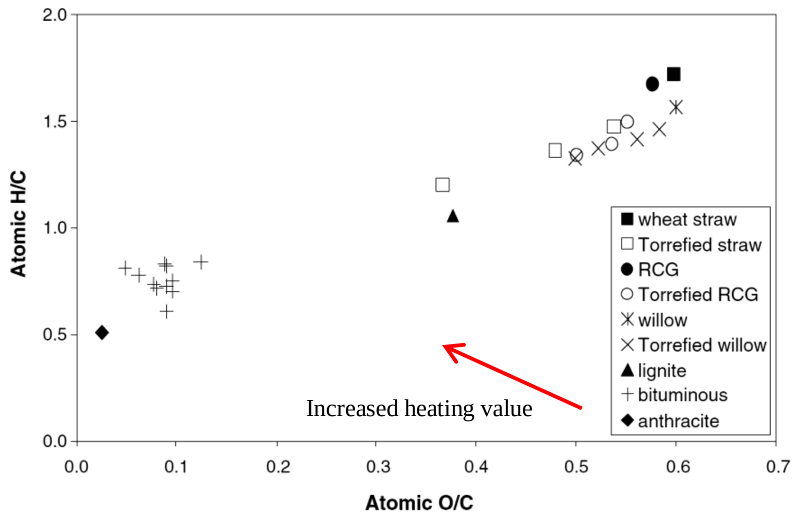

Figure 3. Comparison of coal's atomic H/C and O/C ratio with raw and torrefied biomass. (Bridgeman et al., 2008)

The van Krevelen diagram helps in categorizing types of fossil fuels, such as kerogen, petroleum, lignite and bituminous coal (Levin et al., 2019). Therefore, by only having the elemental (ultimate) analysis of a biomass feedstock, it can quickly be estimated how good of a fuel in terms of heating value it would be. In general, raw biomasses tend to have high H/C and O/C ratios, meaning high oxygen and hydrogen amounts, which translates to more volatiles and lower heating value (Basu, 2010).

2.2.2 Effect of torrefaction temperature and time

Temperature and residence time are the two most critical parameters that determine the final product's parameters (Negi et al., 2020). In particular, torrefaction can be categorized in light, mild and severe based on the operational temperature ranges, which are 200-235, 235-275 and 275-300 ^{o}C respectively (W. H. Chen et al., 2015; W. H. Chen & Kuo, 2011). Longer residence times decreases the mass and energy yields but increases the volumetric energy density of the product (Negi et al., 2020), which is referred as the energy content per unit of volume.

The three different torrefaction operating regimes have increasingly larger impact on the biomass. Higher temperatures result in higher heating values, while the mass loss (volatiles) is also higher, while for lower temperatures it is the opposite (Bergman et al., 2005). The effects on lignin, cellulose, hemicellulose and the can be seen in **Table 1**.

Table 1. Torrefaction temperature regimes effect on hemicellulose, cellulose and lignin. (W. H. Chen et al., 2015)

Classification	**Light**	**Mild**	**Severe**
Temperature (°C)	200-235	235-275	275-300
Consumption			
Hemicellulose	Mild	Mild to severe	Severe
Cellulose	Slight	Slight to mild	Mild to severe
Lignin	Slight	Slight	Slight
Liquid colour	Brown	Brown dark	Black

2.3 Pelletizing of biomass

Biomass, raw or torrefied, cannot be utilized without a densification treatment. That is because of its very low bulk density (40-200 kg m^{-3}) (H. Li et al., 2012) and the low volumetric energy density (1.4-5 GJ/m^3) (Joshi, 2015; Maciejewska et al., 2006), which makes it difficult to handle and to replace common fuels such as coal (800 kg/m^3 and 18-24 GJ/m^3). Densification is defined as "the compacting process of a material under specified conditions" (Gilvari et al., 2019), which can be split into pelletizing and briquetting based on the equipment used, which affects the size of the final product (Bhattacharya et al., 1989).

2.3.1 Pelletizing and briquetting principles

The densification process of biomass is considered complex and is influenced by various chemical and physical phenomena. In particular, Bhattacharya et al., 1989 had reported that the lignin contained in biomasses, which softens around 130-190 °C acts as an "internal glue" factor, which leads to bonding. They also claimed that water presence can reduce the softening point of lignin at around 100 °C. However, they also add that some studies contradict each other, concluding that the interlocking of fibers while also different chemicals converting under high pressure and temperature also play a role in the densification process.

More recent studies (Lope G. Tamil, 1996; Tumuluru, Wright, et al., 2012) argue that essentially, densification is achieved by the formation of solid bridges. These occur form through "chemical reactions and sintering solidification, hardening of the binder or melted substances or crystallization of the dissolved materials". It is also mentioned that indeed water or other liquids can enhance bonding of the biomass through interfacial forces and capillary pressures.

Table 2. Comparison of differences between briquettes and pellets (Alamsyah et al., 2015; Chaloupková et al., 2018; Tumuluru, Wright, et al., 2012).

Type of densification	**Diameter (mm)**	**Length (mm)**	**Equipment**
Briquetting	10-70	25-300	Briquette presses (Hydraulic/Mechanical piston press, screw extruder, roller press)
Pelletizing	3-10	18-25	Pellet mills (ring die and flat die)

2.3.2 Pelletization parameters

Pelletizing operational changes can have a strong effect on the produced pellet. These can be temperature, die pressure, biomass moisture content, pellet diameter and pellet press type (Rudolfsson et al., 2017).

Temperature plays a very important role to the process as it affects the lignin softening point which aids pellet formation. It increases by the friction created between the roller and the die, as seen in **Figure 4**. The more time the pelletizer operates the higher temperature it can reach. The addition of biomass instantly drops the temperature, but then rises as the friction increases (Gilvari et al., 2019; H. Li et al., 2012). Overall, higher temperatures in the range of 200-230 ^oC have been reported to achieve high durability and hardness for torrefied pellets, while at least 150 ^oC should be exceeded (Gilvari et al., 2019). However, the conclusions from those studies are mostly originated from single or laboratory scale pelletizers, which tend to have big variations compared to industry scale ones, as for example there could be high moisture loss that gives brittle pellets (Gilvari et al., 2019; Segerström & Larsson, 2014).

Compression pressure is also an operational parameter with an effect on the pellet's hardness and density. In particular, higher pressures tend to lead to increased density, but the effect on hardness is mixed (Gilvari et al., 2019). Optimum pressures differ for differently treated materials, as for example 60 MPa work better for gasified palm kernel shell (Bazargan et al., 2014), while 128 MPa are optimum for wood biochar (Hu et al., 2016). Furthermore, compression pressure's effect is indirectly observed in the pellet or briquette diameter. Essentially, under the same operating conditions if the diameter is increased, the pressure is reduced as the same force is being applied to higher area.

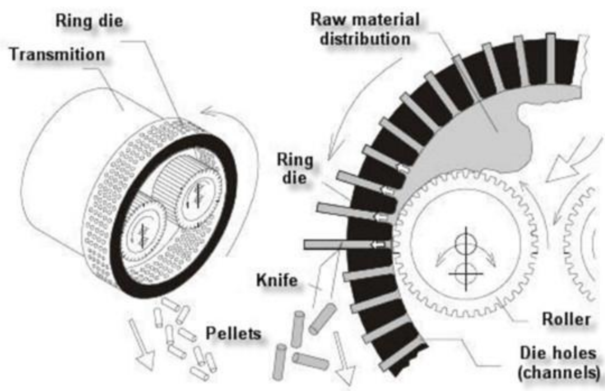

Figure 4. Rotating die pellet mill[1]

Moisture content is another factor considered to be key for the densification process. Both early and recent studies, have concluded that water content facilitates heat transfer, while also enhances bonding and induces the flowability of lignin (Bazargan et al., 2014; Bhattacharya et al., 1989; Gilvari et al., 2019). It has also been universally reported that moisture content initially aids the product strength and durability, but after a point it decreases (Gilvari et al., 2019). In terms of values, 13-17% has been mentioned to be the ideal window for production of pellets, while a general 10-20% is also acceptable (Bhattacharya et al., 1989; Roberto García et al., 2018; Kumar et al., 2017). A way to intensify the effect of water in pelletizing is by conditioning the biomass with steam, which due to its higher temperature and gaseous form, enhances lignin softening in a larger degree, resulting in better pellets (C. Liu & Wyman, 2005; Tumuluru, Wright, et al., 2012).

2.3.3 Torrefied pellets and binder addition

Multiple studies have concluded that torrefied biomass is more difficult to densify while it tends to become more brittle (Gilvari et al., 2019; Rudolfsson et al., 2017; Wang et al., 2020). Only a few reports have shown results with mechanical durability similar to the raw wood pellet ones (above 98%), while the majority of the values lie in the range of 85-95% (Gilvari et al., 2019).

[1] Obtained from https://www.macreat.com/wood-pellet-making-machine/

Consequently, various binders have been tested experimentally to increase durability. Some of the more common ones are lignin and starch, while other types of raw biomasses can also be used such as sawdust, peanut shells, grape pomace etc (Dai et al., 2019; Roberto García et al., 2018). However, these solutions can be either expensive (starch) or deteriorate the properties that make torrefied pellets more desirable, such as hydrophobicity and low microbial activity (if the binder is raw biomass). Therefore, the binders currently used still don’t provide long term solutions.

That is why, plastics as binder could be a way to solve the issues above while also enhance other properties of the torrefied pellets. Only a few studies have reported results from including plastics in pelletization (Cramwinckel, 2020; Emadi et al., 2017; Samaksaman et al., 2019), where it showed that durability, HHV and tensile strength were increased significantly. These studies nonetheless, included only single press pelletizers where temperature and pressure were controlled. Thus, there is still a knowledge gap if this procedure would be feasible in commercial pelletizer.

3. Materials and methods

All the torrefaction experiments and analyses took place at Torrgreen B.V. and the Dredging Lab at Technical Universiteit Delft.

3.1 Materials

Two types of biomasses were used for the experiments, that is wheat straw and wood chips. Wheat straw is a type of herbaceous biomass which is mostly considered as waste, in fact the largest agricultural residue in Europe[1]. However, its potential energy is not utilized, while it could be the base for a solid biofuel via producing torrefied pellets. Wood chips on the other hand, are a woody biomass, most commonly used as a commodity solid fuel, while also producing excellent quality pellets. Moreover, they are currently the most used source for torrefying pellets (Wild & Calderón, 2021). As a result, a comparison between the 2 biomasses can give a better overview of how far off are the herbaceous torrefied biomass pellets.

Figure 5. Wood chips (left) and wheat straw bales (right) that were used for the torrefaction runs.

[1] Reported in an article uploaded on the EU Research and Innovation Magazine: https://ec.europa.eu/research-and-innovation/en/horizon-magazine/wheat-straw-waste-could-be-basis-greener-chemicals

Regarding the use of plastic, Polyethylene resin in granular form was acquired from Resinex and produced by DOW Chemicals. Its specs are shown in Table 3. Although it is not categorized as LDPE or HDPE, its properties indicate that it has the density of LDPE, but with a melting temperature of HDPE.

Table 3. Polyethylene resin's properties.

Plastic name	Diameter (μm)	Density (g/cm^3)	Vicat Softening Temperature (°C)	Melting Temperature (°C)
DOWLEX™ 2629UE Polyethylene Resin	800	0.935	119	124

3.2 Torrefaction pilot scale reactor

The torrefaction reactor used for all the experiments is a fixed bed pilot-scale, built and designed by Torrgreen B.V. It consists of a blower, the fixed bed reactor, a flare for volatile combustion and a propane combustor to provide heat. It is designed for both drying and torrefying biomass, by using an automation system with 3 different operations: drying, torrefaction and cooling. The reactor is a stainless-steel hexagonal barrel with a volume of 160L. The cooling liquid is water, being sprayed by nostrils which create a misting effect, for optimal evaporative cooling. The water was pumped via a Karcher high pressure pump. Nitrogen was used as inertization gas. For straw, the reactor was loaded with biomass up to 70-75% of its volume to avoid the "ratholing" effect caused by the difficulty in packing herbaceous biomasses. For woodchips, the reactor was filled halfway due to their higher bulk density. Two runs at 240 °C and 270 °C were tested for both the biomasses. For straw, the residence time was 10 minutes, while for woodchips 30 minutes, due their difference in structure. Below the flowchart of the setup can be seen. For a picture of the setup, **Figure 20** at Appendix depicts the system better.

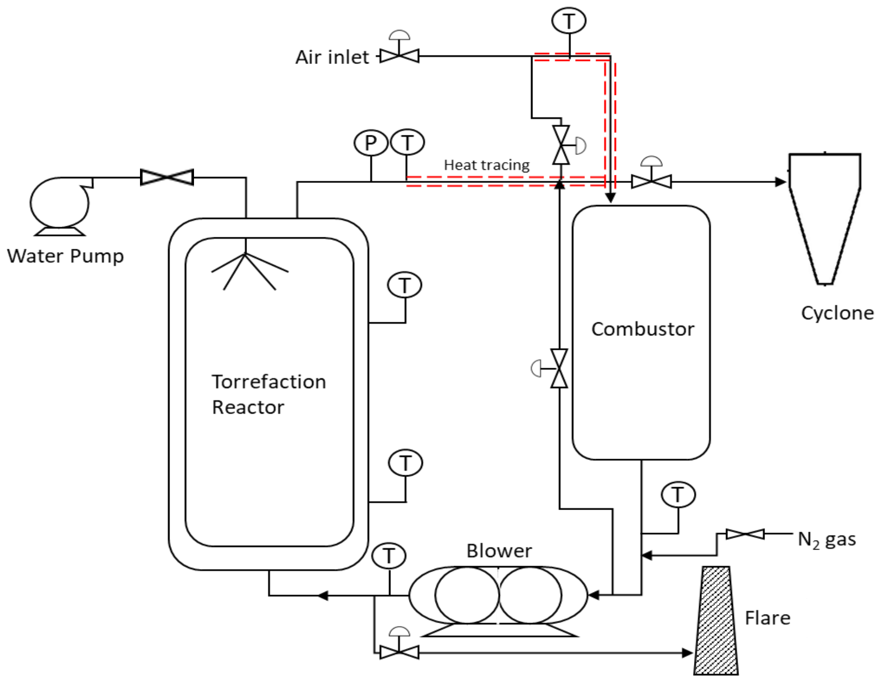

Figure 6. Torrefaction setup flowchart. Initially for drying, air enters from the top, is heated in the combustor and then led to the reactor. In torrefaction mode, the system closes to atmosphere, with the valve leading to the flare

opening occasionally based on the pressure built up from the volatile production. In cooling mode, the system remains closed but bypasses the combustor until temperature drops.

3.3 Moisture content

The moisture content (MC) was measured for each raw and torrefied biomass before and after the torrefaction runs. The method used was according to the standard EN 14774-2 by drying the samples in an oven at 105 ± 2 ^oC for 24hr and measuring the mass difference. For the torrefied pellets a Biopoint Moisture Analyzer by Supertech Agroline ApS was used which has the EN 55013-1:2017 standard applied.

3.4 High Heating Value

High heating value (HHV) was determined for every type of pellet made. A HAMCO 6C bomb calorimeter was used for all the measurements. Initially, 0.8-1.1g of pellets were weighed in a Kern ADJ 200-4 scale, after being dried. Before the measurements, the bomb had been standardized with benzoic acid of known calorific value and the following equation was used for calculations:

$$HHV = \frac{\Delta T * W - CV_{th} * w_{th} + CV_{wire} * w_{wire}}{w_{bio}} * 0.004184$$

Where,

ΔT (^oC): Temperature difference before and after the calorimeter run.
W (cal/^oC) : Water equivalent, calculated when testing with the standard substance, benzoic acid.
CV_{th} (cal/g): Calorific value of cotton thread used for ignition assistance.
w_{th} (g): Weight of cotton thread.
CV_{wire} (cal/g): Calorific value of nichrome wire used for ignition.
w_{wire} (g): Weight of nichrome wire.
W_{bio} (g): Weight of biomass/pellet sample.

The number 0.004184 being multiplied with the fraction is used for converting cal/g to MJ/kg. The final value of the equation represents the HHV of the biomass on a dry basis. The measurements were executed as duplicates for each pellet.

3.5 Pelletizing setup and experiments

A rotating die pellet mill, model KL150 B/C, was used for the pelletizing experiments. To monitor the die temperature, a thermocouple was mounted on the outer shell around the die as seen on **Figure 7**. Moreover, in order to control the die's pressure on the roller, compression springs were placed in the hole of the bolts that are used to increase or decrease the pressure applied. Finally, in order to aid pelletizing, a small pump was installed to add droplets of water onto the hot die to create steam. The blue pipe seen on Figure 7 is the one coming from the pump.

Regarding the experimental procedure, before each pelletizing run the biomass was firstly grinded to 1mm by a custom-made hammer mill and then brought to the desired moisture content, after being conditioned for 24hr. Two different moisture contents were tested, 10% and 15%. Moreover, the amount of plastic that was investigated was 5%, thus the samples were mixed before entering the pelletizer with the plastic powder. Initially, a small amount of biomass was poured in the hopper and used in order the die to get up to operating temperature. In addition, two different pellet diameters were tested, 6mm and 8mm.

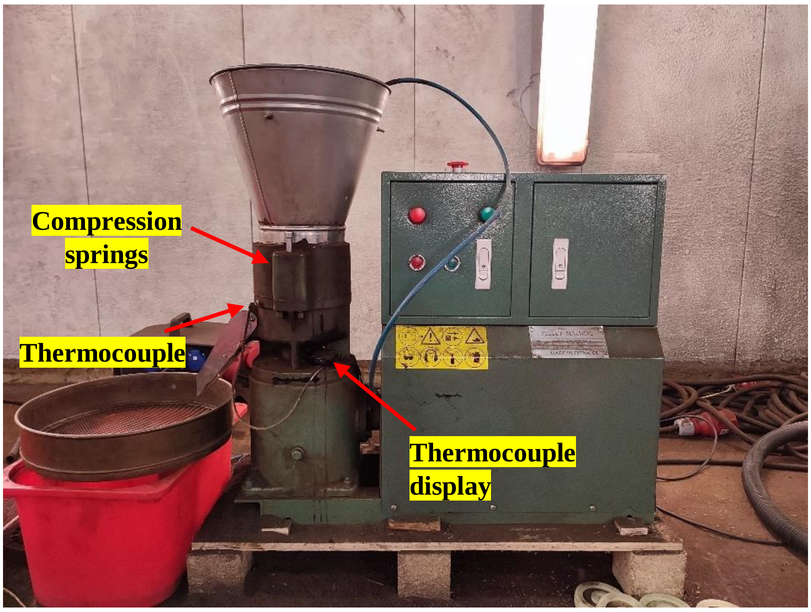

Figure 7. Pelletizing setup (the attached thermocouple is seen with the display of temperature).

3.6 Mechanical durability

Mechanical durability (MD) was measured according to the EN15210-1 standard, where 500 ± 5g of pellets sample were weighed, after being initially sieved in a 3.15mm sieve to separate any smaller pieces/fines. Then the sample was placed in the tumbler used in the Dredging Lab at TU Delft and operated at 50rpm for 10 minutes, meaning 500 total rotations. After the end of the run, the remaining pellets where again manually sieved in a 3.15mm sieve to remove any excess fines and the remainder was weighed. The mechanical durability was measured via the following calculation:

$$Mechanical\ Durability\ (\%) = \frac{M_{si}}{M_{in}} * 100$$

Where:

M_{in} : Pellet mass before entering the tumbler (g)

M_{si} : Sieved pellet mass after the run (g)

The sample size was 1kg, thus the measurements were duplicates.

3.7 Water immersion tests

Water immersion tests were performed to measure the moisture uptake of the pellets. It is an indirevt way of investigating the extend of torrefaction and effectively the increase in hydrophobicity of the pellets. As there is no standard method for this measurement, a similar method that Wang et al., 2020 used in their report was selected. In particular, a small number of pellets was weighed and then immersed in a beaker filled with water, with approximately 1:10 weight ratio of pellets to water. Then, 2 immersion times were tested, 1hr and 4hr. When the pellets were removed from the water, they were patted dry with an absorbing paper and weighed again. The weight difference of the pellets gave the overall moisture uptake.

3.8 Design of experiments

A design of experiments (DOE) was performed in order to observe the multiple effects of different parameters on the torrefied pellets. The method used was Taguchi's methodology, which is suitable for problems containing a lot of parameters and the focus is on improving a product's quality[1]. In this case, the target for improving quality is MD, by altering 4 factors and each one having 2 levels: 240 and 270 °C torrefaction temperature, 10 and 15% MC, 0 and 5% binder and 6 and 8 mm pellet diameter. According to Taguchi, depending on the number of factors and levels, an orthogonal array can be formed, which contains all the adequate combinations of parameters and their levels to optimize the target value. Based on that, the appropriate array is the L8 one, which results in 8 different samples. By using the add-in software XLSTAT in Excel, the orthogonal array was created with random combinations of parameters as shown in **Table 4**.

Table 4. Taguchi's L8 array calculated randomly by XLSTAT.

Observations	Tor. Temperature (°C)	Moisture (%)	Plastic (%w/w)	Pellet diameter (mm)
Obs1	240	10	0	6
Obs2	240	10	5	8
Obs3	240	15	0	8
Obs4	240	15	5	6
Obs5	270	10	0	8
Obs6	270	10	5	6
Obs7	270	15	0	6
Obs8	270	15	5	8

To identify the different samples the following structure was used: (Tor. Temp/ MC%/ Plastic%/ Diameter). For example, Obs1 from **Table 4** would be referred to as (240/10/0/6). After measuring each sample's MD, a parametrization was performed with a goal of maximizing it.

4. Results and Discussion

4.1 Torrefaction runs

Two torrefaction runs were completed, producing torrefied wheat straw and woodchips in two different forms. As it can be seen from **Figure 8**, the biggest difference between the two torrefied samples is their colour, as the 270 °C one is darker and almost completely black, confirming the theory mentioned in Chapter **2.1**. Moreover, while the torrefied samples were much more brittle than the raw one, the 270 one was even softer than the 240.

[1]https://eng.libretexts.org/Bookshelves/Industrial_and_Systems_Engineering/Book%3A_Chemical_Process_Dynamics_and_Controls_(Woolf)/14%3A_Design_of_Experiments/14.01%3A_Design_of_Experiments_via_Taguchi_Methods_-_Orthogonal_Arrays

Figure 8. Straw (left) and woodchips (right) torrefaction products after the 240 °C and 270 °C runs.

4.1.1 Effect of torrefaction on elemental composition

The torrefied samples were sent to ALS Inspection labs for proximate and ultimate analysis. The results can be found at *Appendix*. Torrefaction upgrades the fuel quality of each biomass by removing a part of the volatiles that have low HHV, thus reducing the amount of oxygen which leads to a larger percentage of carbon. **Figure 9** and **Figure 10** indicate the effect of torrefaction, as in both cases carbon is increased up to 5.8% and 7.9% respectively.

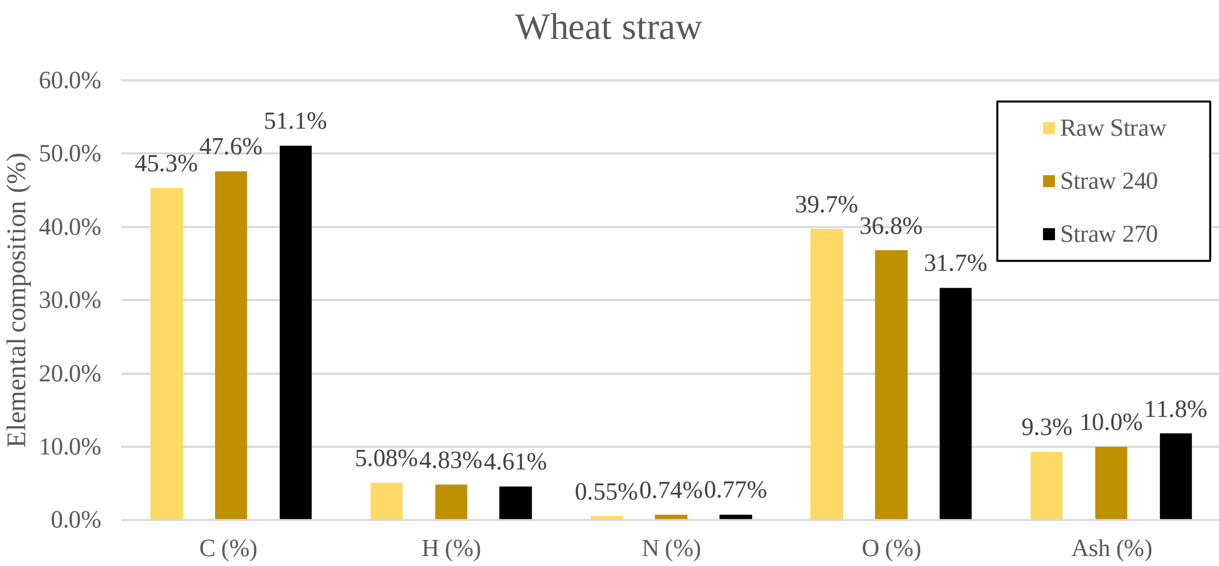

Figure 9. Percentage difference in elemental composition for wheat straw after torrefaction.

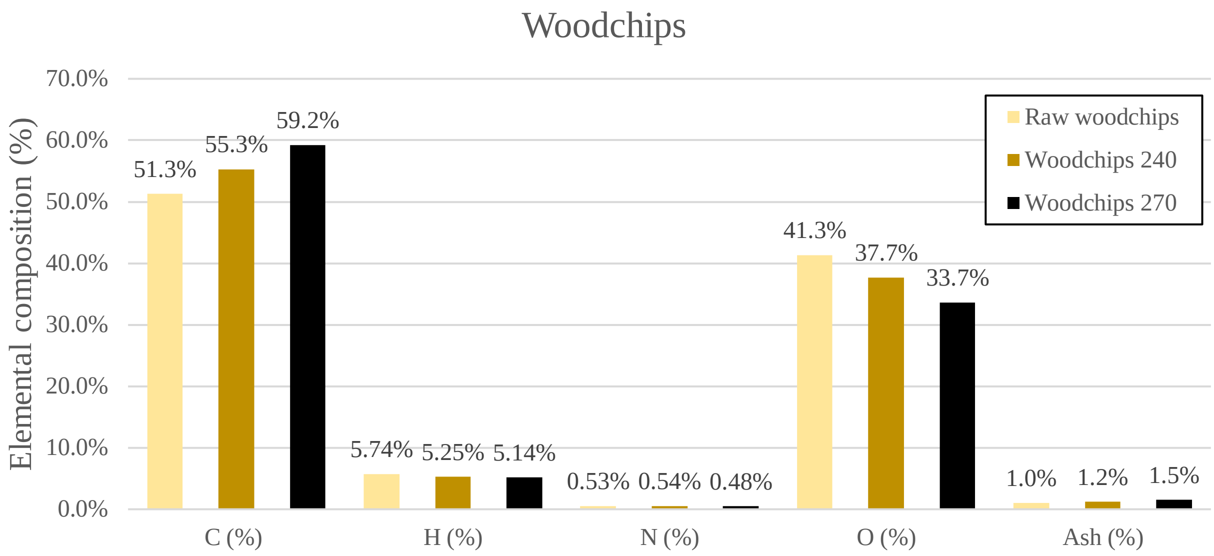

Figure 10. Percentage difference in elemental composition for woodchips after torrefaction.

Overall, woodchips contain a higher carbon percentage which is always desirable if torrefied biomass is competing with coal. The biggest difference between the 2 biomasses is the ash content, as wheat straw contains almost 10 times more than the woodchips. This is generally expected from herbaceous biomasses and especially straw, due to their higher uptake of nutrients during growth (Tumuluru, Hess, et al., 2012).

Another way to classify and compare the elemental composition of biofuels to conventional is the van Krevelen diagram, which combines the H/C and O/C ratios. It can be seen that torrefied woodchips and wheat straw at 270 ^{o}C come quite close to the ratios of lignite, a commodity solid fuel that is mostly used for electricity generation. However, the torrefied products are still far off fuels such as bituminous coal and anthracite that are heavily carbonized. This is due to the -still- high percentage of volatiles contained after torrefaction.

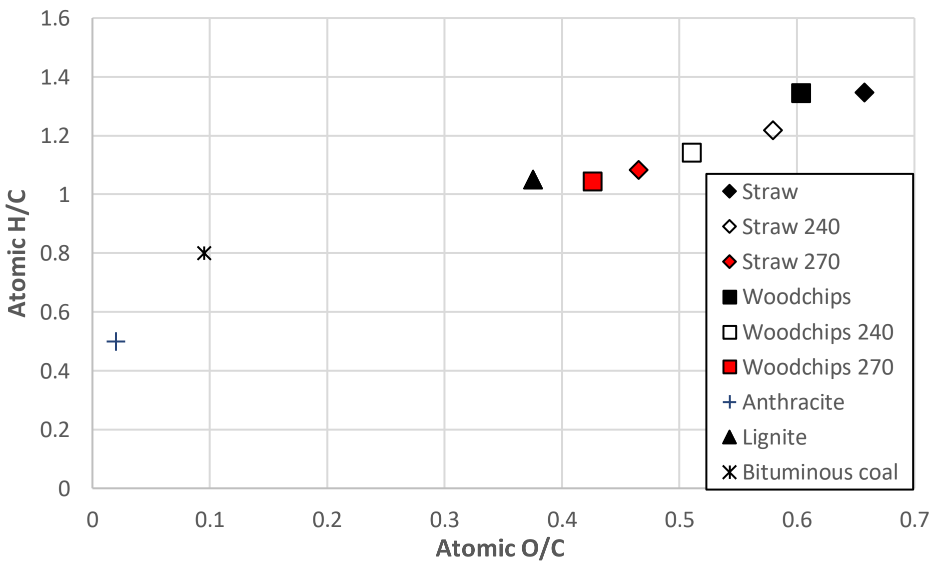

Figure 11. Van Krevelen diagram for the torrefied biomass samples and comparison with commodity fuels.

4.1.2 Effect of torrefaction on HHV

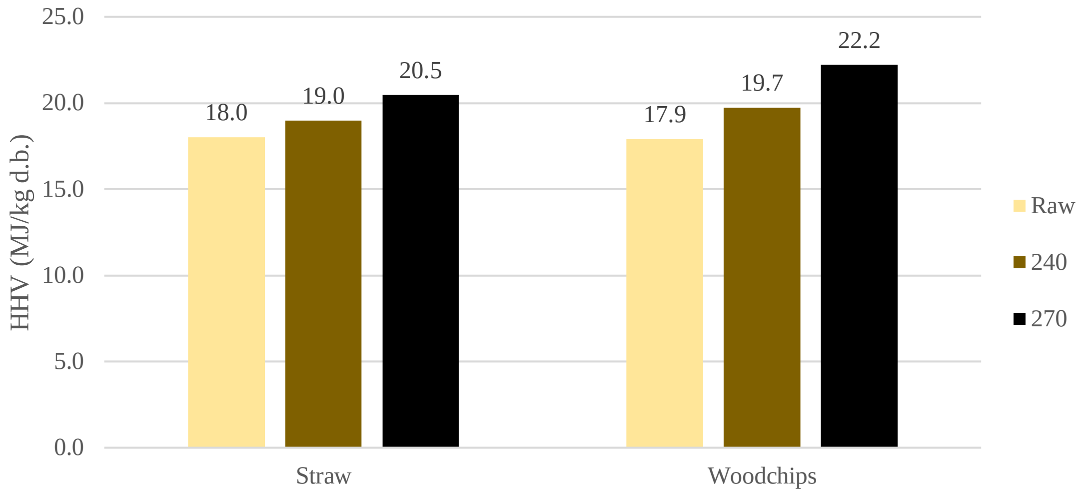

Figure 12. Effect of torrefaction on HHV on dry basis to wheat straw and woodchips.

The increase in calorific value is expected due to the decrease in volatiles composition and the percentage increase of carbon (see **Figure 9** and **Figure 10**). The difference between the values does not seem very significant (around 1 MJ/kg for straw), however **Figure 12** depicts the HHV on dry basis. **Figure 13** shows the more accurate effect that torrefaction has on the calorific value. In particular, the increase is larger, if the samples' moisture is also included. For straw, the raw moisture content was 10.4% but only because it was left out for a long period of time. When the bales first arrived in the facilities, the moisture was around 20%, which would make its calorific value 14.4 MJ/kg and the difference with the torrefied straw much bigger. As mentioned, one of torrefaction's advantages is that even if the biomass is stored for a period of time, due to its increased hydrophobicity, the moisture content will always remain low and below 10%. Thus, the overall effect on the HHV is more impactful, even with the 240 ºC run.

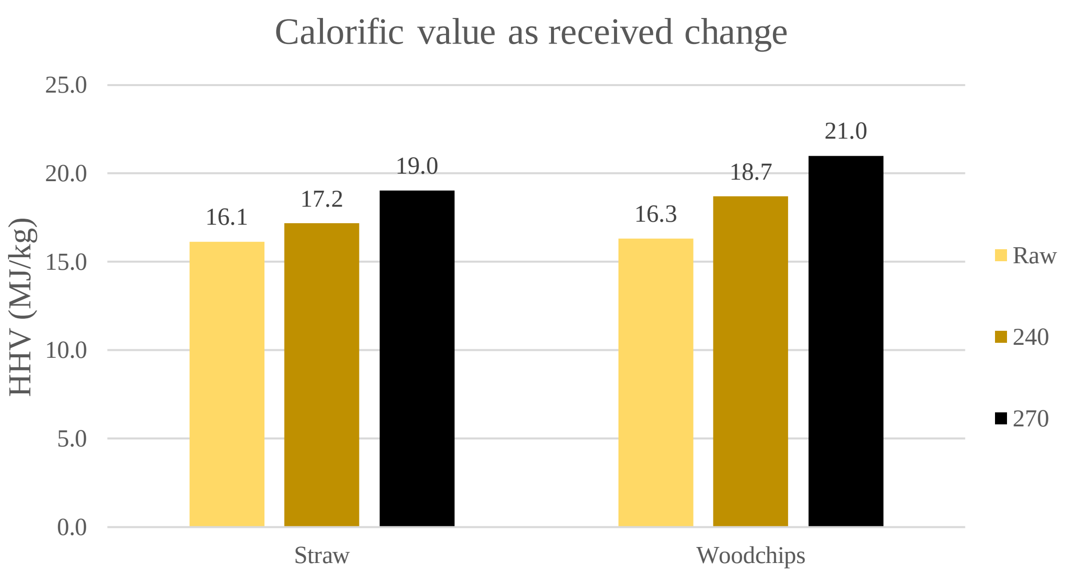

Figure 13. Calorific value change on samples as received.

4.2 Pelletizing results

Multiple pelletizing tests were executed to tune the pelletizer and find optimal operational parameters for achieving pellet production. It was noticed that the difference of the thermocouple display and the actual die's actual temperature was around 30-40 °C lower. Moreover, it was found that during pelletizing with plastics, pellets were soft below a certain temperature, meaning that lignin was not reaching its glass transition point. That temperature was at 85-95 °C (thermocouple display), meaning inside the die it was 115-125 °C, which is also very close to the plastic's softening and melting temperature.

4.2.1 Mechanical durability results

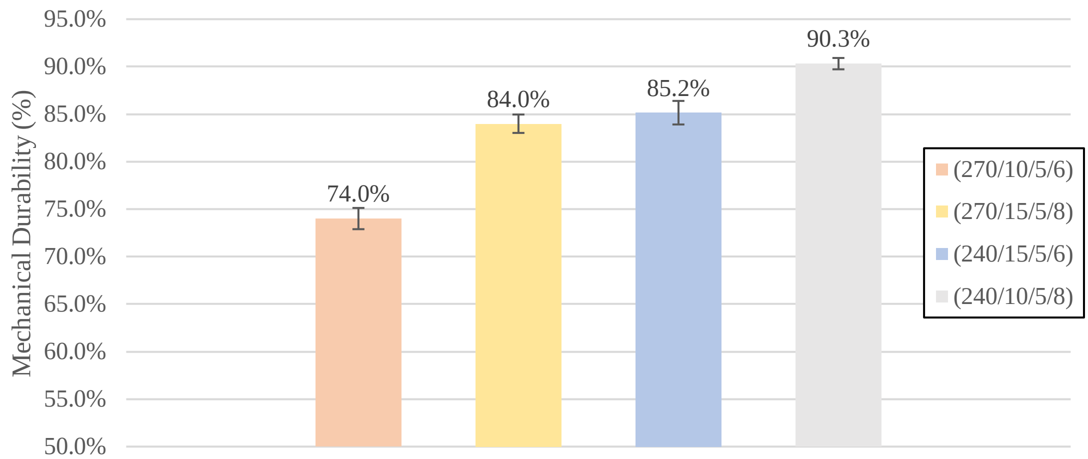

Figure 14. Mechanical durability results on the torrefied wheat straw pellets that contain plastic.

The results on the wheat straw torrefied pellets indicate a huge effect of the plastics on the final product. First of all, pellets were able to form during pelletization with the addition of plastic powder, which is a positive sign for future use. Moreover, the highest durability was achieved on the (240/10/5/8) sample, while overall the 240 °C samples showed better durability. The 8mm torrefied pellets seem to be more durable than the 6mm ones, but there is not a high certainty to that. However as seen in **Table 5**, the only known torrefied straw pellet production in a commercial capacity had showed high durability on a 8mm pellet, which further supports the claim.

In the *Durability results* part on the Appendix, the importance of the binder clearly appears in the cases of the two samples that couldn't pelletize at all, which are the ones at 270 °C and no plastic. The main reason for that could be the quality of the pelletizer itself, but still even 5% makes a distinct difference. Moreover, the much-increased brittleness of the 270 °C biomass might have played a role.

Pelletizing with raw and torrefied woodchips was also attempted, as a reference point. The results for various combinations of woodchips can be seen in **Figure 15**. Not all the possible samples were created, as only some reference points were chosen. Moreover, the inclusion of raw woodchips was placed in order to "calibrate" in a sense the pellet mill that was used.

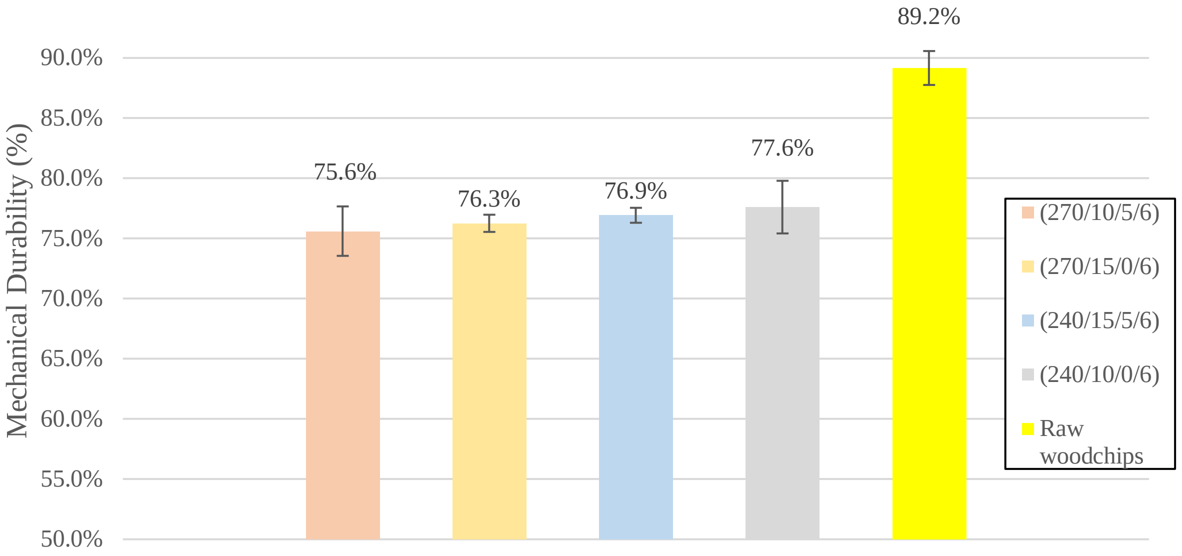

Figure 15. Mechanical durability of torrefied and raw woodchip pellets.

The values for the samples tested are lower than the ones reported or expected from literature (**Table 5**), as even torrefied woodchips have demonstrated mechanical durability values above 90%. Also, the raw woodchip pellets value of 89.2% is quite low compared to the commercial value of at least 95%, while plastic did not seem to affect the durability in this case. This could be explained by the higher amount of lignin usually contained in woodchips (McKendry, 2002), leading to different polymer-to-polymer interactions. Besides that, during pelleting with woodchips (raw and torrefied), the pelletizer seemed to struggle to produce them, almost leading to its failure. This, combined with the results of durability indicate that the pelletizer played a bigger role to the final product than expected. The original use stated for this pellet mill did not mention torrefied biomass and was directed for animal feed pellets more.

Table 5. Comparison of this report's mechanical durability of various torrefied feedstock pellets with literature.

Biomass type	Mechanical durability (%)	Torrefaction Temperature (^oC)	Pellet diameter (mm)	Additive	References[1]
Wheat straw	90.3	240	8	5% PE	Current report
	85.2	240	8	-	
	84.0	270	8	5% PE	
	94.5	250	8	-	(Agar et al., 2021)
Poplar	95.6	250	8	-	
	87.5	280	8	-	
Pine	98.0	280	6	20% Glycol	(R. García et al., 2020)
	91.8	250	8	-	(Föhr et al., 2017)
	86.5	315	8	-	(Rudolfsson et al., 2017)

4.2.2 Water immersion results

There were 4 types of pellets selected, that had all the different factors and their levels included. While a statistical correlation cannot be extracted from the results, an indication about the behaviour of these characteristics is expected.

[1] The data taken for this table used only reports that produced pellets from bench/industrial scale pelletizers for more accurate comparison.

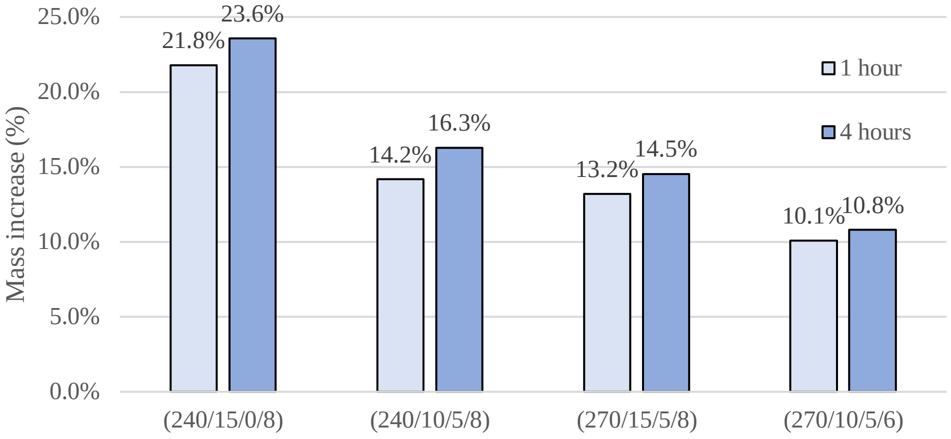

Figure 16. Moisture uptake for 4 different types of torrefied wheat straw pellets.

From **Figure 16** it is observed that the biggest moisture uptake capacity was on the (240/15/0/8) sample, while the lowest on the (270/10/5/6). Furthermore, the other sample of 270 °C showed low moisture uptake. As seen from **Figure 17** the 240 °C pellets with no plastic were swollen even after 1 hour of immersion, while the 270 °C with plastic and small diameter were almost intact after 4 hours.

Figure 17. Results after 1 hr immersion test on the (240/15/0/8) sample (left) and after 4hr on the (270/10/5/6) sample (right).

These results indicate first and foremost that more intense torrefaction increases hydrophobicity significantly. They also demonstrate the increase of torrefaction degree from 240°C to 270°C, as most likely the hemicellulose on the 240°C samples was still present (based on how swollen one sample was). In addition, the presence of plastic, shows a likely correlation to lower moisture uptake capacity, as the two more comparable samples (240/15/0/8) and (240/10/5/8) show a relatively large difference of about 7%. This should be expected to a degree as plastics (including PE) are hydrophobic (Silvia et al., 2019). That observation could be important in the overall decision about whether to include plastics into pelletization. There might also be a small indication of a better hydrophobicity at 6mm pellets, but this cannot be said with certainty. Consequently, higher hydrophobicity means higher degree of torrefaction, thus indeed 270 °C torrefied wheat straw is quite more brittle compared to the 240°C one (see **Figure 14**), which can partly explain why pelletizing it without any binder did not have any success.

4.2.3 Effect of higher plastic content

During the project, other biomasses were also torrefied and pelletized. One of them was hay, which was torrefied at 220 °C. The product was pelletized with different amounts of plastics, to have a more informative view on what happens above 5%. The 6mm die was used, while the measured moisture content was 12%. The figure below shows a high increase in durability with the increase of the polyethylene resin. The biggest effect is spotted between the 5% and 10% concentrations, where durability increased from 84.3 to 95.9% which is close to wood pellets values (Craven et al., 2015). From 10% to 20% the difference was almost insignificant, with only 1% increase. This happens due to the fact that from a point above, durability cannot increase much more, hence the Moreover, the HHV increase with plastic is something expected, as a typical calorific value for polyethylene plastic is around 43 MJ/kg (Panda et al., 2010).

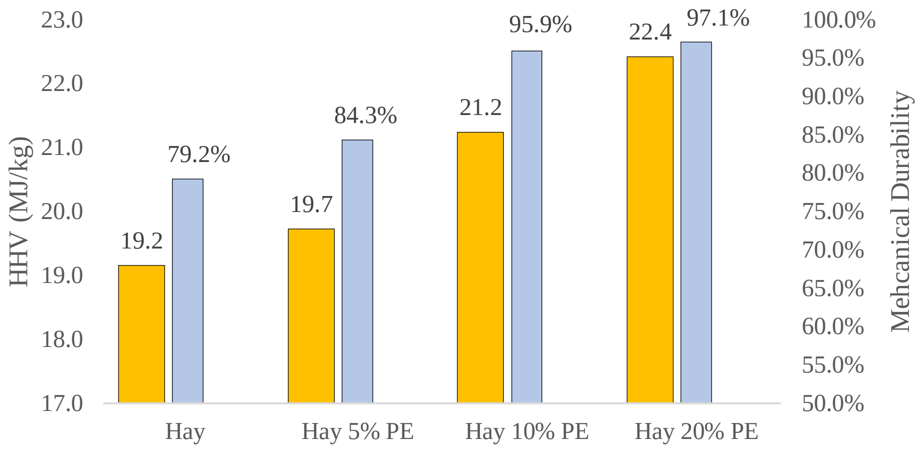

Figure 18. Plastic concentration effect on torrefied hay pellets' mechanical durability and HHV on dry basis.

Figure 19. Pictures of torrefied hay pellets. On the left, there are the ones with 5% PE and on the right with 10% PE.

From the pictures above, a noticeable difference can be seen. With 5% plastic, the pellets remained “shiny” on the outside, a very common characteristic which indicates that lignin has reached its glass transition point (Bhattacharya et al., 1989). However, with 10% plastic this shininess is gone. Although it might seem that the pellets aren’t strong, the results show otherwise. The co-existence of polyethylene and lignin might result into the glass transition point of the latter to change, thus reducing the shininess, but the hardness of the plastic keeps the pellet strong and durable.

4.2.4 Comparison with existing standards

Wood pellets demand and production is growing, especially in EU (Craven et al., 2015). From house use and industrial heating to power generation, their applications are plenty. However, their main issue its their raw material, which is wood – not always a waste, which can lead to high grinding costs as well as a decrease in quality over time due to hygroscopicity (Phanphanich & Mani, 2011). Torrefied pellets from agricultural waste would solve both of these problems, especially the moisture uptake one. However, as of today due to the few industrial installations of torrefaction plants and units, there is still uncertainty regarding the standards around torrefied pellets (Agar et al., 2021; Gilvari et al., 2019). Every country has its own regulations for pellets or briquettes, thus comparison is not easy. For example, in some countries such as Italy additives are banned for A1 classification, while Austria has a limit of 2% (García-Maraver et al., 2011). The European standard committee CEN/TC 335 developed a more generic technical specification called CEN/TS 14588:2004 that includes various classifications and relevant terminology (García-Maraver et al., 2011). The overall table with these classifications is shown at the *Appendix*. For a comparison between the pellets made at the Torrgreen facilities and conventional wood pellets, **Table 6** was constructed which includes the best wheat straw pellet made in terms of durability and where it falls in classification regarding to the CEN/TS 14588:2004 standard.

Table 6. Comparison of Torrgreen pellets with current standards and wood pellets.

Parameters	CEN/TS 14588:2004[1]	Torrefied wheat straw pellet	Torr. Pellet Classification	Conventional wood pellet[2]
Moisture content (%)	≤ 15	8.8	M10	≤ 10
Mechanical Durability (%)	≥ 90	90.3	DU90	≥ 97.5
HHV (MJ/kg d.b.)	-	20.3	-	≥ 20.0
Bulk density (kg/m^3)	-	≥ 650*	-	651
Ash content (%)		≤ 9.1**	A6.0+	≤ 0.7
Pellet diameter (mm)	≤ 25	8	D08	6
Additives (%)	-	5	-	0

*Estimation by using containers of known volume and filled with pellets
**The value is max. 9.1% (from proximate analysis) due to the addition of plastic which theoretically would reduce it

The comparison between the wood pellet and the torrefied wheat straw one indicates that the latter cannot be yet considered comparable with the former. That is because two of its properties are very far from the wood pellets' ones, meaning mechanical durability and ash content. Mechanical durability is one of the most critical but there is still room for improvement and the fact that pellet can fit into a classification is rather important. Regarding ash, it is a property that comes with the type of biomass feedstock and in general herbaceous biomass tend to have higher ash content as mentioned above. A way to overcome that issue would be to wash away minerals and then treating it with acid or alcohol (Pottathil et al., 2012), but that may affect fuel properties. Thus, torrefied pellets from herbaceous biomass would not be a feasible solution for domestic use or industrial boilers.

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[1] European Standard Committee CEN/TC 335 including specifications for solid biofuels and analytical techniques

[2] Craven, J. M., Swithenbank, J., Sharifi, V. N., Peralta-Solorio, D., Kelsall, G., & Sage, P. (2015). Hydrophobic coatings for moisture stable wood pellets. *Biomass and Bioenergy*, *80*, 278–285.

5. Conclusions

Converting waste biomass into a solid biofuel is crucial for transitioning to renewable fuels and energy. Torrefaction is proven to enhance fuel properties of biomass, but the densification process that follows is of equal importance. Three biomasses were torrefied at the Torrgreen facilities: wheat straw, woodchips and hay. For the densification process, pelletizing was chosen as the most suitable, as briquetting is cannot work with torrefied material. The pellets that were produced, combined different parameters and one of them was the addition of plastic (PE) binder. The biomass which the pelletizing was focused on was wheat straw, while woodchips were used for comparison and hay was used for additional experiments. It was the first time a report showcased that pelletizing of torrefied herbaceous biomass with plastic addition can be achieved in a larger than lab-scale pelletizer. The main conclusions of the experiments are:

- Plastic binder concentration of 5% increased the durability of torrefied wheat straw pellets. For the 270 ^{o}C biomass it was the only way to produce pellets.
- Increasing plastic content also showed an increase of pellet hydrophobicity, however with no statistical certainty.
- High torrefaction temperature had a negative effect on durability, with the value of 240 ^{o}C being the more suitable in order to both get pellets and achieve an increase in HHV.
- While there is need for moisture addition on torrefied biomass before pelletization, different moisture levels did not seem to affect durability that much compared to the other parameters.
- The 8mm diameter pellets had better durability than the 6mm ones.
- Statistical analysis could not be executed due to high inaccuracies caused by the 2 samples that did not pelletize.
- Comparison of conventional wood pellet with the ones produced at Torrgreen, showed that with the existing standards, herbaceous biomasses' main problem is the high ash content and the relatively low durability.
- The quality of the pellets was negatively affected by the type of pelletizer used, which was clear while pelletizing with woodchips. Thus, pelletizer type is also a critical parameter for acquiring quality torrefied pellets.

The decision of including a small amount of plastic in pelletizing of torrefied herbaceous biomass is multifaceted. On one hand it provides advantages that could contribute to torrefaction commercialization, such as the increase in durability, HHV and hydrophobicity. On the other hand, in order for this process to be sustainable and cost-saving, plastics have to come from a waste stream. In this report, virgin PE powder was used, which is very expensive (7000€/tonne) for an industrial process. Ideally, a mixed PE waste stream would be a feasible solution, but still those plastics should be milled to a suitable size. Moreover, the torrefied pellets with plastics should be directed for gasification, in which gaseous pollutants such as dioxines and polycyclic aromatic hydrocarbons (PAHs) would be broken down due to very high temperatures. Thus, it is limiting in terms of other uses such as domestic heating.

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Appendix

Torrefaction runs

Figure 20. Torrefaction setup at Torrgreen facilities.

Table 7. Raw and torrefied proximate analysis[1].

Biomass type	HHV (MJ/kg d.b[2])	MC (%)	FC (%)	Volatiles (%)	Ash (%)
Straw	18.02	10.4	15.5	65.8	8.3
Straw 240	18.98	9.5	19.9	61.5	9.1
Straw 270	20.47	7.0	26.3	55.7	11.0
Woodchips	17.91	8.9	15.0	75.4	1.0
Woodchips 240	19.74	5.3	21.6	72.0	1.1
Woodchips 270	22.21	5.4	26.8	66.4	1.4

Table 8.Raw and torrefied ultimate analysis[3].

Biomass type	C (%)	H (%)	N (%)	O (%)	Ash (%)
Straw	45.3	5.08	0.55	39.70	9.3
Straw 240	47.6	4.83	0.74	36.80	10.0
Straw 270	51.1	4.61	0.77	31.69	11.8
Woodchips	51.3	5.74	0.53	41.31	1.0
Woodchips 240	55.3	5.25	0.54	37.69	1.2
Woodchips 270	59.2	5.14	0.48	33.65	1.5

Durability results

Table 9. Mechanical durability of various samples.

Sample (Tor.Temp/MC/Plastic%/Diameter)	Mechanical durability (%)
Wheat straw	
(240/10/0/6)	75.00 ± 1.51
(240/10/5/8)	90.32 ± 0.60
(240/15/0/8)	84.05 ± 0.58
(240/15/5/6)	85.15 ± 1.24
(270/10/0/8)	0.00
(270/10/5/6)	74.01 ± 1.12
(270/15/0/6)	0.00
(270/15/5/8)	83.99 ± 0.97
Woodchips	
(240/10/0/6)	77.61 ± 2.19
(240/15/5/6)	76.92 ± 0.62
(270/10/5/6)	75.61 ± 2.05
(270/15/0/6)	76.26 ± 0.71
(-/17/0/6) *	89.17 ± 1.41
Hay	

[1] For the proximate and ultimate analysis, the samples were sent to ALS Inspection B.V. labs
[2] d.b = dry basis
[3] Ultimate analysis is on d.b.

(220/12/0/6)	79.23 ± 1.52
(220/12/5/6)	84.33 ± 0.95
(220/12/10/6)	95.93 ± 0.59
(220/12/20/6)	97.07 ± 0.43

*This sample refers to raw woodchips with 17% initial moisture

Common standards

Table 10. Austrian standards for energy pellets ÖNORM M 7135.(García-Maraver et al., 2011)

Parameter		Limit values
Physical	Diameter (mm)	4–10
	Length (mm)	<5D
	Particle density (kg/dm^3)	<1.12
Mechanical	Durability (%)[a]	<2.3
Chemical	Moisture content (%)	<10
	Ash content (%)	<0.5
	Heating value (kcal/kg)	>4302
	N (%)	<0.3
	S (%)	<0.04
	Cl (%)	<0.02
	Additives (%)	<2

Table 11. Classification of parameters included in CEN/TS 14588:2004 published by the European Standard Committee CEN/TC 335 (García-Maraver et al., 2011).

Parameter	Classification
Size (diameter and length) (mm)	D06: $D \leq 6 \pm 0.5$ and $L \leq 5D$
	D08: $D \leq 8 \pm 0.5$ and $L \leq 4D$
	D10: $D \leq 10 \pm 0.5$ and $L \leq 4D$
	D12: $D \leq 12 \pm 1.0$ and $L \leq 4D$
	D25: $D \leq 25 \pm 1.0$ and $L \leq 4D$
Moisture content (%)	M10: ≤10%
	M15: ≤15%
	M20: ≤20%
Ash content (%)	A0.7: ≤0.7%
	A1.5: ≤1.5%
	A3.0: ≤3%
	A6.0: ≤6%
	A6.0+: >6%
N (%)	N0.3: ≤0.3%
	N0.5: ≤0.5%
	N1.0: ≤1%
	N3.0: ≤3%
	N3.0+: >3%
S (%)	N0.05: ≤0.05%
	N0.08: ≤0.08%
	N0.1: ≤0.1%
	N0.2+: >0.2%
Cl (%)	CL0.03: ≤0.03
	CL0.07: ≤0.07
	CL0.1: ≤0.1
	CL0.1+: >0.1
Durability[a]	DU97.5: ≥97.5
	DU95.0: ≥95
	DU90: ≥90
Fines content (%<3.15 mm)	F1.0: ≤1%
	F2.0: ≤2%
	F2.0 +: >2%
Bulk density (kg/m^3)	Recommended value should be included by manufacturer
Heating value (kcal/kg)	Recommended value should be included by manufacturer
Additives	Binding materials and ash inhibitory should be included in the label

[a] Durability has been defined in terms of the percentage of whole pellets after testing.