Article
Lignin Pellets for Advanced Thermochemical Process—From a Single Pellet System to a Laboratory-Scale Pellet Mill

Sergio Jaimes Rueda 1, Bruna Rego de Vasconcelos 1, Xavier Duret 2 and Jean-Michel Lavoie 1, * 1

1 Biomass Technology Laboratory, Department of Chemical Engineering and Biotechnological Engineering, Université de Sherbrooke, 3000 Boulevard de l’Université (P3 Pavillon), Sherbrooke, QC J1K 0A5, Canada; sergio.andres.jaimes.rueda@usherbrooke.ca (S.J.R.); bruna.rego.de.vasconcelos@usherbrooke.ca (B.R.d.V.)
2 ReSolve Énergie Inc., Lac-Mégantic, QC G6B 2V9, Canada; xavier.duret@resolveenergie.com
* Correspondence: jean-michel.lavoie2@usherbrooke.ca

Abstract: Lignin pellets were produced using a single pellet system as well as a laboratory-scale pellet mill. The feedstock used in this work was lignin isolated from poplar wood (Populus tremuloides) using a direct saccharification process. An investigation was performed on the influence of the initial moisture content on the dimensions, impact and water resistance, fines content, mechanical durability, calorific value, and ash content, and, finally, the ultimate analysis was performed. These properties were then compared to pellets made from softwood bark using the same pelletization unit. Lignin pellets were then manufactured using four different types of additives (corn oil, citric acid, glycerol, and d-xylose) and ultimately, they were stored in two different conditions prior being tested. In general, manufacturing pellets that were entirely made of lignin generated samples with an overall higher hydrophobicity and higher calorific value. However, the ash and sulfur content of the lignin pellets (1.58% and 0.32% in scenario 2, respectively) were slightly higher than the expected CANplus certification values for Grade A pellets of ≤0.7%, and ≤0.04%, respectively. This study intends to show that lignin could be used to produce this new kind of pellets, pending that the initial material has a low ash and moisture content.

Keywords: lignin pellets; poplar wood; solid biofuels; moisture content; additives

1. Introduction

Despite the most recent environmental concerns related to greenhouse gas emissions combined with the different actions undertaken by numerous governments around the world to reduce carbon intensity, the demand for fossil fuels is still high if not increasing in most countries around the globe. The geopolitical instability around the world will most definitely have an impact on the energy sector and in addition to finding new options to cope for actual supply, many countries are looking for more environmentally friendly solution aiming at a reduced carbon intensity (CI). In recent years, one option for reducing CI has been to increase the use of biofuels, and when it comes to solid biofuels, wood is often the most used raw material, mostly because of its low ash content (as compared to agricultural biomass as an example). This particularity is essential in combustion since ashes are often related to many issues, including an increased level of corrosion in combustion equipment, as reported by Stele et al. [1]. Wood pellets are generally produced from residues of the wood industry, such as sawdust and shavings as well as harvest residues, which have also been considered for this purpose [2]. Wood is essentially composed of three macromolecules, which are hemicellulose, cellulose, and lignin, where the latter is usually embedded within a matrix of the two other and is considered as being the second most abundant biopolymer in nature (after cellulose). Even though it has been the subject of much research throughout the years, its biosynthesis, chemistry, and structure remain a popular subject in the open literature to this day. Lignin is also a natural polymer, which is, contrarily to cellulose, mostly composed of aromatic structures.
which overall involve less oxygen and more carbon. The macromolecule also has been associated with a considerable number of potential applications, such as producing new composite-type polymeric materials to produce bio-based monomeric aromatics, as well as to produce biofuels, including pellets, that can be used as a solid fuel to produce heat and/or power. One of the important aspects of solid biofuels is the quality standards defining the physical and chemical properties of the pellets, of which moisture is often a prevalent parameter. Literature offers many pieces of information aimed at determining the best moisture content in the raw material used to produce pellets because, to the best of our knowledge, no standard value is available. In most biofuels, water content remains a challenge because it affects the energy that can be generated from the fuel downstream. Biomass itself usually contains high levels of moisture, which is why it often goes through a drying process to reach moisture levels below 15% (also recommended by the pelletizing equipment manufacturers). Such a procedure will ensure that the pellet produced will have humidity levels below 10%, as required by certifications such as CANplus [3]. The moisture content influences the self-ignition capacity of the biomass as well as its calorific value because, during combustion, part of the fuel energy is dedicated (or wasted in most cases) to the evaporation of water and, consequently, it is not available for other energy uses. The water content also impacts the durability of the pellets while increasing their bulk density, without forgetting that it can also induce fungi production and/or spore emissions. Literature has shown many studies where pellets were produced using raw material with varying moisture contents. In addition, the dimensions of the pellets is one of the most important parameters since it has a direct impact when feeding such biofuels in combustion chambers. The latter are often designed and optimized based on the values suggested by the CANplus certification [3], which can lead to the deduction that pellets exceeding these specifications can have negative impact at different levels in a combustion system. For solid biofuels, the calorific value corresponds to the amount of energy released during combustion per unit mass, generally expressed in MJ/kg. When it comes to pellets, this parameter varies according to the origin of the biomass used as raw material and according to the CANplus certification, this value should be greater or equal to 16 MJ/kg. Of course, there is a wide array of feedstocks that could be used and could reach such specifications. Ash content is also a crucial parameter when it comes to boiler design and cleaning operation because the combustion of pellets containing significant amount of inorganic will require a more effective and perhaps complex ash removal process. This parameter varies according to the different types of biomass used in the manufacture of pellets, especially for tissues such as bark and grass, which may lead to higher amounts of ash after combustion. The elements that usually contribute the most to ash content are Si, Ca, Mg, K, Na, P, S, Cl, Al, Fe, and Mn, and their overall content is deemed low when comprised between 1% and 2%, according to the CANplus certification [3].

Furthermore, several methods can be considered to determine the resistance to impact of the densified materials which often revolve around dropping a solid biofuel sample several times from a previously established height onto a floor material of specific composition. It is necessary to record the mass of the sample before performing the drop tests, after which the samples are sieved and weighed. This method is also known as “drop resistance” or “shattering resistance”. Although many tests are used to determine the shattering resistance of densified material, there is, to the best of our knowledge, no standard method. As an example, many researchers [4–7] used a specific shattering resistance test to determine the durability of biomass pellets and briquettes. Pellets were dropped from a known height of 1.85 m onto a metal plate four times. The impact resistance of the pellets was then defined as the percentage of the initial weight retained after dropping. Richards [8] introduced the impact resistance index (IRI) based on the number of drops and the number of new pieces generated by the drops. Lindley et al. [9] used a similar method to determine the impact strength of briquettes where each briquette was dropped from a height of 1.0 m onto a concrete surface. This procedure was performed ten times, after which the percentage of weight loss was calculated.
When it comes to water resistance in wood pellets, Lee et al. [10] used three types of commercial wood pellets that were exposed to water. The parameters evaluated were the durability, density, and porosity before and after exposing them to water. Overall, it was found that when the pellets were in direct contact with water, they began to degrade. Richards [8] used a briquette and placed it in water to determine its resistance against water absorption and disintegration. The briquette was immersed in water for 30 min and pressure was applied at 10 min intervals. After this, the briquette was removed from the water, wiped to remove the excess moisture from the surface, and weighed. This study led to the introduction of the water resistance index (WRI), which allows better comparison between samples. Lindley et al. [9] also used briquettes to determine their water resistance and, in this case, the samples were submerged in 25 mm of water at 27 °C for 30 s, after which the percentage of weight gain in the briquettes was calculated and recorded.

There are different studies on the influence of storage time on the physical properties of pellets. For example, Dyjakon and Noszczyk [11] analyzed the influence of storage at −28 °C on the mechanical durability of commercial pellets made from different biomass. They found that the freezing and subsequent defrosting process caused a decrease in the mechanical durability as compared to normal storage conditions at 20 °C. In addition, pellet storage at freezing temperature did not affect their mechanical durability. Lee et al. [12] conducted a study and investigated the effects of storage configuration, storage time, storage temperature, and wood pellet quality on the net calorific value over a 6 months period. They showed that the storage time increased by 1% and 2% (in net calorific value) when closed storage was selected.

Looking at pelletization as a pre-treatment method, Takada et al. [13] exposed the influence of lignin on the durability of the pellets as well as on the use of pellets as a raw material for bioconversion. The work also suggested that during the granulation process, when the glass transition temperature of the lignin is exceeded, it becomes plasticized and acts as a glue, thus improving the durability of the pellet. In addition to lignin, other natural additives were reported for this purpose, such as sugar, starch, and vegetable oils, as well as lignin derivatives such as lignosulfonate and Kraft lignin.

Although the literature on pelletization is abundant, there are practically no studies that show the behavior of lignin (in general) as a solid biofuel, and there are even less when it comes to the fabrication of lignin pellets. In addition, the work carried out on the subject were generally not conclusive, so a possible utilization to develop models or validate experimental results was not possible. In this context, the present work is intended to provide a baseline for the manufacture of lignin pellets using two devices: a single pellet system and a laboratory-scale pellet mill. This study aims to evaluate the influence of the initial moisture content of lignin, as well as the use of four different additives (corn oil, citric acid, glycerol and D-xylose) on the physical and chemical properties of the pellets, such as their dimension, durability, fines content, calorific value, and ash content, all of which will be complemented by elemental analysis (C, H, N, S). These properties were evaluated using the CANplus certification [3] as guideline.

2. Materials and Methods

Finding the optimal manufacturing conditions for the granulation process, such as pressure, temperature, and compression rate (processing parameters) usually requires an extensive set of experiments. For this study, the CANplus certification was used to validate the properties of the manufactured pellets and two scenarios were considered. In scenario 1, four values of initial moisture content in the raw material were randomly chosen and analyzed (% humidity): 8.01% (H1), 26.21% (H2), 30.07% (H3), and 38.83% (H4). However, although random, the values selected could serve as a basis to know how dry the lignin isolated from the saccharification process should be to generate a good quality solid biofuel.

Reference pellets were manufactured in the same pellet mill from softwood bark (mixture of balsam fir and spruce) with 6.57% humidity, and the physical properties were compared with pellets made from poplar wood lignin. In scenario 2, to improve the
physical properties of the pellets, four different types of additives were used. Additionally, they were stored inside the building for 30-day at 22.3 °C (condition 1) while other samples of pellets were stored in a cold chamber for 8 days at 2.0 °C (condition 2) although this test was performed only for those manufactured using a laboratory-scale pellet mill. Finally, with the single pellet system, different conditions of temperature, compression pressure and time were used until the resulting pellets were within the values required by the CANplus certification [3].

2.1. Raw Material

In this study, poplar wood (Populus tremuloides) forest residue was used to produce sugar and lignin at a pilot scale. RéSolve Énergie Inc. (Lac-Mégantic, QC, Canada), provided the purified lignin, which was generated using their proprietary process. The latter is referred to as a “direct saccharification of lignocellulosic biomass”, which is a non-enzymatic chemical process. Sugars were used to produce low carbon intensity (CI) ethanol while the residual lignin was used as raw material to produce pellets using two devices: a single pellet system and a laboratory-scale pellet mill. The lignin samples that were used to produce pellets were mixed with hot water to obtain the different initial moisture contents that were investigated in this work (referred to H1, H2, H3, and H4). To homogenize the moisture level, samples were stored in sealed plastic buckets for 10 days before pelletization. Prior to pelletisation, thermogravimetric analysis (TGA) were performed to characterize lignin degradation as well as to evaluate its thermal stability and degradation kinetics. The material was tested on a range of temperatures to identify the optimal operating one which is in this case the temperature where lignin would get softer while degrading as minimally as possible. This TGA also allowed measuring the mass variation of the sample as a function of temperature which in this case ranged from room temp to 1000.0 °C (10.0 °C/min) using a 90.0 mL/min flow of nitrogen. The lignin used before the sample’s conditioning is shown in Figure 1a while the general block diagram of the ReSolve process is shown in Figure 1b.

**Figure 1.** (a) Poplar wood-based lignin produced at pilot scale using ReSolve process and (b) block diagram of the ReSolve process, which is the source of the lignin used in this work.

2.2. Experimental Setup

2.2.1. Single Pellet System

Pellets were individually produced in a piston-cylinder type system that was designed and assembled at the Biomass Technology Laboratory (BTL) machine shop located at the Université de Sherbrooke (Sherbrooke, QC, Canada). The system, although strongly inspired by models used in the literature [14–16] involved two modifications:

- A temperature control and heating system were implemented on the system;
- A sensor was added on the bench to monitor compression pressure during the process.
These modifications allowed for a better understanding of the pelletization procedure while providing more data on the system behavior. The lignin compression system consisted of a classical metal piston-cylinder, where the latter has a 6.5 mm diameter and 80 mm in length, made of hardened steel and packed with heating elements and thermal insulation. The cylinder channel allowed the introduction of the lignin from the tom while a removable base allowed recovering the pellets after pressing. A general overview of the design of the experimental bench is shown in Figure 2a while the actual setup is presented in Figure 2b. The hydraulic individual press was mounted on a Chicago Pneumatic CP86200 test machine (Chicago, IL, USA), making it possible to control the force of the piston inside the cylinder during the lignin compression process.

![Figure 2](image)

**Figure 2.** (a) 3D drawing of the experimental system designed to produce individual pellets and (b) picture of the actual experimental bench located at the Biomass Technology Laboratory in Sherbrooke, Quebec, Canada.

Heating was supplied to the cylinder using a heating tape that was wrapped around the compression chamber, and the temperature was monitored directly using a thermocouple. During the manufacture of one pellet (as can be seen in Figure 3), the system was preheated to reach the desired temperature (77 °C), after which it was maintained stable, showing a temperature variation of ±3 °C. The single pellet system was employed to determine the influence of the compression chamber temperature on the formation of a single lignin pellet. The compression chamber of the single pellet system was fed with sequential samples of 0.26 g lignin per layer, producing pellets with three layers with a 0.78 g average mass. The lignin sample was compressed to a maximum pressure of 12 MPa, a value that was confirmed using a 50 kN load cell located at the base of the cylinder. The force supplied by the hydraulic press was 21.57 kN, and the compaction pressure of the sample under this force was calculated considering the area of the cylinder-piston system and yielded a pressure of 650.17 MPa. The compression pressure was kept stable for 2 min, then the base of the cylinder was removed after which the pellet was pressed out at the same compression velocity. This pelleting procedure resulted in a product with a shiny structure, where each layer was exposed to the same pressure for the same period. The pellet was cooled down and stored in an air-tight plastic bags at room temperature (22.3 °C), after which physical and chemical parameter measurements were performed. The reproducibility of the experimental conditions was facilitated by an exhaustive cleaning (involving brushing and air blowing the compression chamber) between samples.
Figure 3. Pellet production process using the single pellet system employed at the Biomass Technology Laboratory.

2.2.2. Laboratory-Scale Pellet Mill

Pellets were also produced in a laboratory-scale pellet mill, although the system was slightly modified to fit the expectations of this work. More specifically, to maintain constant heat on the system, a silicone rubber heating tape equipped with a time percentage dial control (BSAT) and a thermal insulation tape was positioned around the die. The previously-mentioned heating tape system was a model BSAT051006 from BriskHeat (Columbus, OH, USA) and by adding it to the unit, it allowed the supply of heat to the flat die during the pelletization process, thus providing more data on the thermal behavior of the manufacturing process. The pellet mill itself was a Buskirk Engineering laboratory-scale unit (Ossian, IN, USA) model PM605, and is depicted in Figure 4. It was equipped with a variable-speed electric drive, although the extrusion rate used to manufacture the lignin and softwood bark pellets was 1920 RPM. The flat die used in the present experiments had a hole diameter of 6.35 mm and a length of 38.1 mm, providing a compression ratio of 6.

Before pelletization, the system was preheated for 1 h, after which the pellet mill was fed using approximately 1.00 kg of raw material. Heat was supplied to the system by conduction using the previously mentioned silicone rubber heating tape positioned around the flat die. The temperature was set between 86.1 °C and 132.1 °C and led to an operating temperature ranging between 61.1 °C and 77.1 °C inside the roll-die system. The
temperature was monitored using a pocket thermal camera, model PTi120, from Fluke (Everett, WA, USA). After pelletization, a sieve with round holes (3.15 mm diameter) was used to sieve the solid biofuel that was produced, according to the ISO 3310-2 method [17]. The lignin pellets were then cooled and stored in a closed plastic bag at room temperature (23.5 °C), after which physical and chemical parameters measurements were performed. Figure 5 shows the lignin pellet production procedure using the continuous laboratory-scale pellet mill.

![Lignin Pellet Production Process](image)

**Figure 5.** Pellet production process using the laboratory-scale pellet mill operated at the Biomass Technology Laboratory.

The reproducibility of the experimental conditions was facilitated by an exhaustive cleaning between samples (involving brushing and air blowing in the roll-die system). The temperature of the heating tape was selected in light of the Thermogravimetric Analysis (TGA) results of the poplar wood lignin. The operating temperature range was from room temp to 1000.0 °C/min at 10.0 °C/min using a 90.0 mL/min nitrogen flow. The TGA (TA Instruments Q500, New Castle, DE, USA) experiments were performed at the Biomass Technology Laboratory (BTL).

The different moisture content values were selected based on previous studies that were made for this type of biomass, as well as on work performed to manufacture pellets by numerous authors [1,18–27]. Furthermore, the temperature value for the pelletization process was selected based on previous studies on lignin, such as those performed in different publications [28–40].

### 2.3. Properties of Pellets

#### 2.3.1. Moisture Content

Moisture content (MC) was measured before and after granulation, as suggested by the ISO 18324-1:2015 method [41]. According to this procedure, samples are dried in an oven at 105 °C until reaching constant mass. The mass loss induced from the drying process is then interpreted solely as a loss of humidity. Water content was then calculated using the mass of water reported on the mass of dry matter. This part of the testing was performed using an HT1000 industrial oven (The Grieve Corporation, Round Lake, IL, USA). Four initial moisture contents were analyzed and the final moisture content of the pellets was shown to be highly dependent on the conditions of the granulation process, especially the initial moisture content of the lignin but also the die temperature and extrusion rate, as well as compression pressure.
2.3.2. Dimensions

The dimensions of the lignin pellets were measured using a digital electronic micrometer (iGaging 35-040-025-I; San Clemente, CA, USA) with an accuracy of 0.001 mm, and the results were reported for lignin pellets manufactured at different moisture content. The measurements for the diameter and length of the pellets were performed according to the ISO 17829:2015 method [42] and are shown below:
- For the length, each end of the micrometer was positioned at both ends of the pellets;
- For the diameter, three measures were gathered: the first one in the center as well as two more from each end of the pellets.

2.3.3. Impact Resistance

The impact resistance was conducted following the guidelines of the ASTM D440-86:2002 method [43] and was applied to pellets with the varying humidity content. Each sample contained approximately 48.0 g of pellets. The bags to store the pellets were 170 mm × 129 mm sealable bags made from a 100% cotton canvas material and sold by Baotongle. Each bag was dropped 10 times from a 2.0 m height. The mass of the bag prior to and after the experiments was determined using a precision electronic scale (model Pokerty). Then, Equation (1) was used to determine the percentage of weight loss for the resistance to impact experiments.

\[ P = 100 \times \frac{W_1 - W_2}{W_1}, \]  

where \( P \) is the percentage of weight loss, \( W_1 \) is the weight of lignin pellets before shattering, and \( W_2 \) is the weight of lignin pellets after shattering and sieving.

2.3.4. Water Resistance

The percentage of water adsorbed by the pellets was obtained using water resistance experiments that were performed for each moisture conditions used to produce the pellets. Three samples of approximately 10.29 g (of lignin or softwood bark pellets) were used in this experiment. The samples were immersed in 0.52 L of water for 2 min at 23.5 °C, after which the mass gain percentage was calculated. The mass of the samples was determined using the same precision scale as described in the previous steps. The lignin pellets were then sieved to select the samples that were to be used for the impact as well as the water resistance experiments.

2.3.5. Fines Content

Fines are here defined as the particles of a dimension less than 3.15 mm that are attached to the pellets. To reduce the fines content, after production and cooling, the pellets were passed through sieves. The fines content in poplar wood lignin and softwood bark pellets was determined using the ISO 18846:2016 test method [44].

2.3.6. Mechanical Durability

Durability is an important parameter in the wood pellet industry, and tumbling is one of the most employed method to determine the durability and quality of pellets. Low durability usually leads to a greater tendency towards rupture and, hence, the production of fine particles. A high content of the latter may suggest issues in storage and handling and may also induce health and environmental challenges. To determine the durability of the pellets manufactured in this work, two samples of approximately 100 g each were used. These samples have been set to tumble for 2 min at 50 rpm, after which the pellets were sieved on a 3.15 mm diameter sieve. The durability of the pellets produced was evaluated as described in the ISO 17831-1:2015 test method [45] after which the data were processed using Equation (2).

\[ DU = 100 \times \frac{m_2}{m_1}, \]  

where \( DU \) is the durability, \( m_1 \) is the initial mass of the sample, and \( m_2 \) is the mass of the sample after the experiment.
where $DU$ is the mechanical durability (in %), $m_1$ is the mass of the sieved pellets before the tumbling process (expressed in g), and $m_2$ is the mass of the sieved pellets after the tumbling process (also expressed in g).

2.3.7. Calorific Value

The calorific value was obtained using a model 6400 calorimeter from Parr Instruments (Moline, IL, USA) which is available at the BTL. The method used in this specific case was based on the ISO 18125:2017 method [46]. The calorimeter was calibrated using benzoic acid (recommended as standard) and verified before each run. At first, water was added to the calorimetric pump to generate a saturated vapor phase before combustion. All the water formed from the combustion of hydrogen as well as from the original moisture content was considered as liquid water. For the analysis of the poplar wood lignin and softwood bark pellets, the first step involved the preparation of the sample. This part included the sample mass measurement and its subsequent introduction into the equipment, which was then closed and pressurized with oxygen. This method relies on the combustion of the product from the ignition caused by the electric current (through conductive cables) and oxygen.

2.3.8. Ash Content

The determination of the ash content is based on the measurement of the residual mass recovered after subjecting the samples to a combustion at $550 \pm 10 \, ^{\circ}C$, as described in the ISO 18122:2015 method [47]. The equipment used to determine the ash content in the lignin pellets was a Thermolyne F-A1730 industrial furnace (Sybron Corporation, Dubuque, IA, USA).

2.3.9. Carbon, Hydrogen, and Nitrogen Content

The nitrogen content in the lignin pellets was obtained using the ISO 16948:2015 [48] method where a defined mass sample is burned in a mixture of oxygen and gas, under conditions such that it generates ash and gas combustion products. The mass fractions of carbon dioxide, nitrogen, and water vapor were then quantified.

2.3.10. Sulfur Content

The sulfur content was determined by decomposition using a calorimetric bomb with an excess of oxygen, followed by the absorption of the acidic combustion gases in water. The quantification of both elements was performed following the ISO 16994:2016 standard [49].

2.4. Additives

In this part of the work, four additives were employed to investigate their impact on the downstream pellets, including (i) corn oil, (ii) citric acid, (iii) glycerol, and (iv) d-xylose. Corn oil is obtained from the germ of corn that has been previously separated from the corn grain (maize). It can be used as a raw material to produce biodiesel, as well as for other industrial uses such as balsam, paint, nitroglycerin, etc. The pure vegetable glycerin that was used in this work was clear, odorless, lubricating and emulsifying. This product is hypoallergenic and easily soluble in water, which made it a good candidate to be used as an additive in the raw material. Citric acid powder ($C_6H_8O_7$) is one of the most common preservatives and flavoring additives. It can be found naturally in fruits and vegetables. It can also be obtained at an industrial level as a by-product of the metabolism of sucrose or glucose from the mold Aspergillus niger. Finally, d-xylose (wood sugar, $C_5H_{10}O_5$) is a fundamental component of hemicellulose in lignocellulosic biomass. Xylose can represent 20–30% of the weight of lignocellulosic biomass. Zhang et al. [50], suggested that xylose, along with glucose, can be converted into liquid biofuels (e.g., ethanol or butanol). However, glucose is more often used for ethanol production while xylose is often considered as a side-product of hemicellulose fermentation. In this work 330 mL of pure vegetable glycerin derived from non-GMO palm equivalent to 22% of the raw material was used, and a mixture of 60 g of d-xylose in 100 mL of water were separately mixed with poplar wood lignin. Each additive used was blended for approximately 10 min.
to provide a uniform distribution in the raw material. The mixtures were stored in plastic buckets inside the laboratory at room temperature (an average of 22.3 °C). It is expected that these four products, used as bio-additives with the raw material, could act as co-binders and lubricants to reduce the energy required to produce lignin pellets at laboratory-scale.

3. Results and Discussion

3.1. Properties of a Raw Material

The thermogravimetric curve (Figure 6—green line) represents the mass loss (percentage), as a function of temperature. The derivate weight change curve (blue line) corresponds to the first derivative of the thermogravimetric curves and shows the variation in mass over time, recorded as a function of temperature. The thermogravimetric analysis shown in Figure 6 also presents three evident stages of decomposition: the first, at a temperatures of approximately 100 °C, can be attributed to the loss of water through desorption (considered a thermal event of physical origin) which could be considered has a non-significant mass loss. The second, occurring at higher temperatures, is attributed to the decomposition of the lignin pellets. The third, at temperatures above 370 °C, is due to the decomposition of the remaining lignin, which started at lower temperatures and continued up to 500 °C. Additionally, according to the thermogravimetric analysis, poplar wood lignin showed a decrease in its thermal stability, exhibiting a glass transition temperature (Tg) at 90.68 °C.

![Figure 6. Thermogravimetric analysis of poplar wood lignin in an inert atmosphere.](image)

3.2. Thermogravimetric Image of the Experimental Setup

In light of the information gathered from the TGA, specific attention was given to the temperature inside the roll-die system employed for the production of the pellets. An overview of the temperature found inside the experimental setup was obtained using a pocket thermal camera, as can be seen in Figure 7.

![Figure 7. Thermographic image of the system used to produce lignin pellets and both lignin and softwood bark pellets.](image)

3.3. Pellets Properties from a Single Pellet System

To gather this information, the single pellet system was used and enabled a rapid and inexpensive testing at a smaller scale. Results show that, according to the CANplus certification, the optimal lignin pellets were manufactured at 79.1 °C and 12 MPa, conditions that were kept stable for 2 min. Figure 8 provides an example of the pellets that were produced from poplar wood lignin, while Table 1 shows reports on their physical and chemical properties.
Figure 7. Thermographic image of the system used to produce (a) lignin pellets and (b) both lignin and softwood bark pellets.

Figure 8. Poplar wood lignin pellets produced from a 6.14% moisture content feedstock and using a single pelletizing system.

Table 1. Presents the average values and standard deviation for the properties of 40 pellets from poplar wood lignin.

<table>
<thead>
<tr>
<th>Property</th>
<th>Poplar Wood Lignin</th>
<th>CANplus Certification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter (mm)</td>
<td>6.25 ± 0.04</td>
<td>6 ± 1 or 8 ± 1</td>
</tr>
<tr>
<td>Length (mm)</td>
<td>20.01 ± 4.09</td>
<td>3.15 &lt; L ≤ 40</td>
</tr>
<tr>
<td>Moisture content (%)</td>
<td>6.14 ± 0.47</td>
<td>≤10</td>
</tr>
<tr>
<td>Mechanical durability (%)</td>
<td>99.16 ± 0.03</td>
<td>≥98.0</td>
</tr>
<tr>
<td>Fines content (%)</td>
<td>0.19 ± 0.14</td>
<td>≤1.0</td>
</tr>
<tr>
<td>HHV (MJ/kg)</td>
<td>19.26</td>
<td>≥16.5</td>
</tr>
<tr>
<td>Ash content (%)</td>
<td>1.97 ± 1.16</td>
<td>≤1.2</td>
</tr>
</tbody>
</table>

The results showed that the properties of the pellets depended on the initial moisture content of the lignin. In addition, to obtain pellets that comply with the certification requirements, it is necessary to guarantee an initial moisture content between 9 and 12% in the original lignin feedstock. Table 1 shows that the physical properties are within the values required by the CANplus certification, except for the ash content.

The conditions used in the single pellet system where then applied to a laboratory-scale pellet mill, and the results are presented in the following section.
3.4. Pellets Properties from a Laboratory-Scale Pellet Mill—Scenario 1

For scenario 1, four cases of initial moisture content were analyzed (% humidity): 8.01% (H1), 26.21% (H2), 30.07% (H3), and 38.83% (H4). Poplar wood lignin and softwood bark pellets manufactured through the laboratory-scale pellet mill are both shown in Figure 9.

![Poplar wood lignin pellets](image)

**Figure 9.** Scenario 1: poplar wood lignin pellets produced with an original feedstock having a moisture content of: (a) 8.01 wt%, (b) 26.21 wt%, (c) 30.07 wt%, (d) 38.83 wt% and softwood bark pellets (e) 6.57 wt%.

Results for the physical properties, both for lignin and softwood bark pellets manufactured using the laboratory-scale pellet mill, are depicted in Table 2.

**Table 2.** Physical properties of the poplar wood lignin and softwood bark pellets produced using the laboratory-scale pellet mill following scenario 1.

<table>
<thead>
<tr>
<th>Property</th>
<th>Poplar Wood Lignin</th>
<th>Softwood Bark</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>H1</td>
<td>H2</td>
</tr>
<tr>
<td>Moisture content (%)</td>
<td>8.01 ± 0.62</td>
<td>26.21 ± 1.80</td>
</tr>
<tr>
<td>Diameter (mm)</td>
<td>6.39 ± 0.04</td>
<td>6.40 ± 0.05</td>
</tr>
<tr>
<td>Length (mm)</td>
<td>12.47 ± 5.67</td>
<td>12.26 ± 5.67</td>
</tr>
<tr>
<td>Mass (g)</td>
<td>0.41 ± 0.21</td>
<td>0.45 ± 0.59</td>
</tr>
<tr>
<td>Mechanical durability (%)</td>
<td>94.80 ± 0.28</td>
<td>93.78 ± 0.90</td>
</tr>
<tr>
<td>Fines content (%)</td>
<td>1.45 ± 0.11</td>
<td>2.90 ± 0.09</td>
</tr>
<tr>
<td>HHV (MJ/kg)</td>
<td>23.17</td>
<td>19.57</td>
</tr>
<tr>
<td>Ash content (%)</td>
<td>1.11 ± 0.49</td>
<td>1.37 ± 0.11</td>
</tr>
</tbody>
</table>

The results from Table 2 show that, as expected, high moisture content in the initial feedstock usually induces low durability of the lignin pellets. Such a condition also leads to the production of a high fines content, which, as mentioned earlier, can lead to different problems at a larger scale. Overall, the results showed that the moisture content is an important parameter for the granulation process, both for the lignin and for the wood
pellets. The calorific value of the solid biofuels, corresponding to the amount of energy released during combustion per unit mass (expressed in MJ/kg) is known to vary for pellets according to the origin of the feed. According to the CANplus certification, independent of the type of feedstock used to produce the solid fuel, the calorific potential must be equal or greater to 16 MJ/kg. Some research published in the literature [51,52] reported a calorific potential between 18.56 and 19.38 MJ/kg for poplar wood. However, in contrast to these publications, this work focuses only on the lignin fraction of this biomass, which should, hence, allow pinpointing the direct impact of this macromolecule on the calorific value of the biomass. Due to the polymeric behavior of lignin, the pelletization process induces a thermal softening that promotes particle agglutination [1,53,54]. Therefore, when the operation is performed at 61.6 °C, a better cohesion occurs between the lignin macromolecules, which overall leads to a solid biofuel with a higher calorific value (see Table 1). Ash content is also an important parameter when it comes to boiler design (or selection) and cleaning operation. Hence, the combustion of solid biofuels with high ash contents would logically require a more regular and effective ash removal process. This parameter, of course, varies according to the different types of biomass used to produce the pellets. In addition, ash is an abrasive material that, in the long term, can cause additional problems such as corrosion of the metallic components of the boilers. The inorganic components that contribute the most to the ash formation are Si, Ca, Mg, K, Na, P, S, Cl, Al, Fe, and Mn. Different authors [51,52,55,56] have reported that poplar wood has a higher natural ash content, reported to be between 0.52 and 1.33% as compared to softwood bark, which was reported to contain between 0.3 and 0.7% of inorganics according to Melin [57]. In addition, they also claimed that this ash content could change according to the variety of the hybrid used, fertilization practices, poplar wood maturity, and soil contamination where the original feedstock was planted. However, according to the CANplus certification [3], the lignin pellets that had low ash contents (pellets produced from an 8.01 wt% humidity content—as shown in Table 1) could eventually be suitable for thermal conversion (including combustion) due to the potential low ash accumulation, slagging, or corrosion in the boilers.

The results of the impact resistance of lignin and softwood bark pellets are shown in Table 3. After being dropped, the softwood bark pellets showed a weight loss of 0.61 ± 0.17 wt%, while the lignin pellets produced from the samples having the lowest moisture content (8.01 wt%) had a weight loss of 3.71 wt%. The loss was still lower when compared to the lignin pellets that were produced from the material with the highest moisture content (38.83 wt%), for which the mass loss reached 21.65 wt%. Overall, results showed that softwood bark pellets had very high impact resistance compared to the lignin pellets.

Table 3. Average weight loss for pellets following the impact resistance test (pellets were dropped from a 2.0 m height onto a concrete floor) for scenario 1.

<table>
<thead>
<tr>
<th>Property</th>
<th>Poplar Wood Lignin</th>
<th>Softwood Bark</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>H1</td>
<td>H2</td>
</tr>
<tr>
<td>Moisture content (%)</td>
<td>8.01 ± 0.62</td>
<td>26.21 ± 1.80</td>
</tr>
<tr>
<td>Initial weight before dropped (g)</td>
<td>47.86 ± 0.06</td>
<td>47.27 ± 0.16</td>
</tr>
<tr>
<td>Final weight after dropped (g)</td>
<td>46.08 ± 0.10</td>
<td>43.85 ± 0.26</td>
</tr>
<tr>
<td>Weight loss (g)</td>
<td>3.71 ± 0.17</td>
<td>7.23 ± 0.38</td>
</tr>
</tbody>
</table>

Overall, the lignin pellets with the 8.01 wt% humidity content produced the strongest samples as compared to the one produced with the material with a higher water content. Table 2 reports on the fines content, both for the lignin and for the softwood bark pellets produced during this project. These results are presented as average values (over 3 repetitions) and indicate the fines generated after the impact resistance test (i.e., after 10 drops). According to the CANplus certification [3], this value can be equal or less than 1.0 wt%.
The results show that the high moisture content of the lignin impacted on the durability as well as the production of fines for the produced pellets. More specifically, pellets containing 38.83 ± 0.37 wt% humidity led to the production of 9.87 ± 0.66 wt% fines. However, for lignin pellets with an 8.01 ± 0.62 wt% humidity content, the production of fines after the impact resistance experiments was 1.45 ± 0.11 wt%. For comparison purposes, when it comes to softwood bark pellets (to 6.57 ± 0.15 wt%), the percentage of fines after 10 drops was 0.17 ± 0.04%. The fines content is an important parameter because high values could impact on the storage and transport of the pellets. For larger quantities, the amount of fines could be related to health hazards (due to the inhalation of dust), without forgetting the potential for explosion. During combustion, the fine content also has an influence on the combustion behavior of the fuel, and it can induce plugging or even damage the ventilation system.

Among the other parameters that could negatively affect the quality of the pellets is their short, medium, and long-term exposure to rain. Other aspects to consider would be the humidity conditions during shipping, handling, and storage. To address this concern, water resistance was tested for pellets made both from lignin and from softwood bark. Results showed that none of the softwood bark pellets maintained their integrity while being submerged in water at room temperature for more than 2 min. The pellets swelled on contact with water, then disintegrated after a few seconds. This shows that softwood bark pellets are brittle on contact with water, as shown in Figure 10e. In contrast, lignin pellets (regardless of the moisture content), presented good resistance when submerged in water, as shown in Figure 10a–d. Table 4 shows the pellets’ mass before and after being submerged in water for 2 min, as well as the percentage of water absorbed.

![Figure 10. Picture of the poplar wood lignin pellets after 2 min submerged in water for pellets with an original moisture content of (a) 8.01 wt%, (b) 26.21 wt%, (c) 30.07 wt%, (d) 38.83 wt%, as well as softwood bark pellets (e) 6.57 wt%.

Note that the softwood bark pellets showed a 205.64 wt% mass increase over their original weight after being immersed in water. In opposition, lignin pellets showed a maximum of 28.87 ± 0.21 wt% water absorption. This indicates that the lignin pellets manufactured in this study have a higher hydrophobic potential compared to softwood
bark pellets. Therefore, it is foreseeable that such pellets could be stored outdoors, which would realistically reduce the capital expenditure (CAPEX) of any pellet-based heating system relying on such solid biofuel.

**Table 4. Scenario 1: mass of pellets before and after being immersed in water for 2 min as well as the percentage of water absorbed.**

<table>
<thead>
<tr>
<th>Property</th>
<th>Poplar Wood Lignin</th>
<th>Softwood Bark</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>H1</td>
<td>H2</td>
</tr>
<tr>
<td>Moisture content (%)</td>
<td>8.01 ± 0.62</td>
<td>26.21 ± 1.80</td>
</tr>
<tr>
<td>Initial weight before immersion (g)</td>
<td>10.11 ± 0.03</td>
<td>10.23 ± 0.02</td>
</tr>
<tr>
<td>Final weight after immersion (g)</td>
<td>12.30 ± 0.03</td>
<td>12.58 ± 0.02</td>
</tr>
<tr>
<td>Water absorbed (%)</td>
<td>21.62 ± 0.15</td>
<td>23.05 ± 0.03</td>
</tr>
</tbody>
</table>

3.5. Pellets Properties from a laboratory-Scale Pellet Mill—Scenario 2 (Additives)

The additives that were used had an important role in the physical and chemical characteristics of the pellets because they acted as binders for the raw material. To improve the properties of lignin pellets, four different bio-additives were employed in the feedstock, including (i) corn oil, (ii) citric acid, (iii) glycerol, and (iv) d-xylose. For this scenario, the poplar wood lignin pellets manufactured through the laboratory-scale pellet mill are shown in Figure 11.

![Figure 11. Poplar wood lignin pellets produced using a laboratory-scale pellet mill with (a) pure lignin, (b) corn oil, (c) citric acid, (d) glycerol, (e) d-xylose.](image)

Immediately after the manufacturing process, measurements were taken to obtain the physical and chemical properties of the pellets, and the results are depicted in Table 5. It was found that the additives increased the ash content by an average value of 0.49%. In addition, d-xylose improved the mechanical durability of the pellets by 0.5%, while the other three additives had a negative impact on this parameter. Finally, the ultimate analysis provided a fraction (by weight) of the elemental constituents of the biomass. The
main elements that were analyzed were carbon and hydrogen, although nitrogen and sulfur concentrations were also analyzed. For the elemental analysis, the used additives significantly increased the carbon fractions but reduced the sulfur fractions. The high sulfur content in pure lignin pellets could be associated to the high concentration sulfuric acid that was used in the biomass pre-treatment process. As for the fines content, the four additives that were used in this work significantly increased this value to a mean value of 0.61%.

Table 5. Physical and chemical properties for the manufacturing process for pellets manufactured from poplar wood lignin combined with additives (Scenario 2). The results show an average value for 91.0 g of pellets approximately, expressed on dry basis.

<table>
<thead>
<tr>
<th>Property</th>
<th>Poplar Wood Lignin</th>
<th>Corn Oil</th>
<th>Citric Acid</th>
<th>Glycerol</th>
<th>D-Xylose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter (mm)</td>
<td>6.33 ± 0.02</td>
<td>6.34 ± 0.03</td>
<td>6.32 ± 0.03</td>
<td>6.06 ± 0.09</td>
<td>6.18 ± 0.08</td>
</tr>
<tr>
<td>Length (mm)</td>
<td>16.82 ± 2.54</td>
<td>12.42 ± 3.67</td>
<td>19.22 ± 6.11</td>
<td>18.40 ± 2.76</td>
<td>17.1 ± 4.38</td>
</tr>
<tr>
<td>Moisture content (%)</td>
<td>13.17 ± 0.35</td>
<td>8.41 ± 0.14</td>
<td>7.09 ± 0.28</td>
<td>6.05 ± 0.46</td>
<td>9.12 ± 0.41</td>
</tr>
<tr>
<td>Mechanical durability (%)</td>
<td>93.81 ± 1.62</td>
<td>93.77 ± 0.97</td>
<td>91.20 ± 0.70</td>
<td>89.44 ± 1.84</td>
<td>94.31 ± 0.48</td>
</tr>
<tr>
<td>Fines content (%)</td>
<td>0.29 ± 0.02</td>
<td>0.62 ± 0.09</td>
<td>0.88 ± 0.35</td>
<td>1.56 ± 0.07</td>
<td>0.56 ± 0.06</td>
</tr>
<tr>
<td>HHV (MJ/kg)</td>
<td>22.62</td>
<td>27.40</td>
<td>22.11</td>
<td>23.17</td>
<td>22.59</td>
</tr>
<tr>
<td>Ash content (%)</td>
<td>1.58 ± 0.12</td>
<td>1.26 ± 0.15</td>
<td>1.13 ± 0.15</td>
<td>0.94 ± 0.16</td>
<td>1.06 ± 0.13</td>
</tr>
<tr>
<td>Ultimate analysis</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon (%)</td>
<td>53.56 ± 1.25</td>
<td>62.20 ± 0.25</td>
<td>54.66 ± 0.09</td>
<td>55.98 ± 0.43</td>
<td>54.24 ± 1.15</td>
</tr>
<tr>
<td>Hydrogen (%)</td>
<td>5.53 ± 0.28</td>
<td>6.72 ± 0.09</td>
<td>5.25 ± 0.05</td>
<td>6.13 ± 0.13</td>
<td>5.18 ± 0.14</td>
</tr>
<tr>
<td>Nitrogen (%)</td>
<td>0.00 ± 0.00</td>
<td>0.00 ± 0.00</td>
<td>0.00 ± 0.00</td>
<td>0.00 ± 0.00</td>
<td>0.00 ± 0.00</td>
</tr>
<tr>
<td>Sulfur (%)</td>
<td>0.32 ± 0.06</td>
<td>0.00 ± 0.00</td>
<td>0.15 ± 0.03</td>
<td>0.15 ± 0.02</td>
<td>0.15 ± 0.02</td>
</tr>
</tbody>
</table>

In literature, some previous work has reported on the ultimate analysis values for poplar wood, although the actual work shows slightly higher values for carbon and sulfur content than those previously reported [52,54–56]. Despite the difference, it is worth mentioning that, in the other work, poplar wood biomass was used, while in this study, only the lignin fraction from this feedstock was employed. In this work, the additives acted as lubricants and increased the rate of lignin pellet production. However, to further improve the physical properties of the lignin pellets, two different storage conditions were used. For the first one, the pellets were stored inside the laboratory building at 22.3 °C for 30 days, while for the second, the samples were stored for 8 days in a cold room (also located at the laboratory) at 2.0 °C. Once the storage period was completed, measurements were carried out to obtain the physical properties of the lignin pellets, such as their moisture content, mechanical durability, and fines content. All samples were stored in sealable plastic bags. For the first condition (see results in Table 6), the process led to an improvement of the properties of the pellets, namely on their mechanical durability and fines content. However, the use of citric acid, glycerol, and d-xylene increased the final moisture content more than for the pellets made with pure lignin or when using corn oil as an additive. Nonetheless, all the pellets made using additives in the feedstock are within the values required for moisture content by the certification [3] except for d-xylene.

Table 6. Experimental results after a 30-day storage time at 22.3 °C (first condition) for pellets manufactured from poplar wood lignin and additives (Scenario 2).

<table>
<thead>
<tr>
<th>Property</th>
<th>Poplar Wood Lignin</th>
<th>Corn Oil</th>
<th>Citric Acid</th>
<th>Glycerol</th>
<th>D-Xylene</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture content (%)</td>
<td>3.90 ± 0.15</td>
<td>5.21 ± 0.08</td>
<td>7.48 ± 0.12</td>
<td>9.23 ± 0.09</td>
<td>14.90 ± 0.08</td>
</tr>
<tr>
<td>Mechanical durability (%)</td>
<td>95.70 ± 0.00</td>
<td>94.49 ± 0.11</td>
<td>91.97 ± 5.08</td>
<td>94.67 ± 0.72</td>
<td>95.79 ± 0.86</td>
</tr>
<tr>
<td>Fines content (%)</td>
<td>0.51 ± 0.00</td>
<td>0.30 ± 0.08</td>
<td>0.41 ± 0.25</td>
<td>0.44 ± 0.03</td>
<td>0.19 ± 0.05</td>
</tr>
</tbody>
</table>

For the second condition (storage at a lower temperature—see results in Table 7), the pellets made of pure lignin, corn oil, and citric acid maintained their moisture content
within the values required by the CANplus certification (less than 10%). Results show that there was also a reduction in the fines content and a significant improvement in the mechanical durability of the pellets. The addition of more citric acid hindered the pellet manufacture process by blocking the pellet mill, probably due to poor lubrication. In addition, the final product seemed extremely dry, which affected the mechanical durability of the lignin pellets. On the other hand, corn oil acted as a good lubricant during the pelletizing process and produced the best lignin pellets, both in terms of moisture and durability values.

**Table 7.** Experimental results from an 8-day storage time at 2.0 °C (condition 2) for pellets manufactured from poplar wood lignin and additives (scenario 2).

<table>
<thead>
<tr>
<th>Property</th>
<th>Poplar Wood Lignin</th>
<th>Corn Oil</th>
<th>Citric Acid</th>
<th>Glycerol</th>
<th>D-Xylose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture content (%)</td>
<td>2.15 ± 0.04</td>
<td>9.78 ± 0.77</td>
<td>8.05 ± 0.41</td>
<td>10.11 ± 0.60</td>
<td>11.99 ± 0.17</td>
</tr>
<tr>
<td>Mechanical durability (%)</td>
<td>97.69 ± 0.09</td>
<td>97.07 ± 0.01</td>
<td>95.46 ± 1.85</td>
<td>96.89 ± 0.44</td>
<td>97.66 ± 0.25</td>
</tr>
<tr>
<td>Fines content (%)</td>
<td>0.16 ± 0.00</td>
<td>0.32 ± 0.05</td>
<td>0.24 ± 0.16</td>
<td>0.20 ± 0.00</td>
<td>0.13 ± 0.02</td>
</tr>
</tbody>
</table>

From these results, it was discovered that the storage process for scenario 2 and condition 2 (additives) caused an increase of the physical parameters of the pellets compared to the first condition (without additives) immediately after the manufacturing process. Furthermore, the results showed that the pellets made from pure lignin or with additives in the lignin could potentially be stored both indoors and outdoors, but to increase the solid biofuel performance, a temperature and time-controlled storage environment could be beneficial.

**4. Conclusions**

In this work, the manufacture of pellets made from the lignin macromolecule extracted from poplar wood biomass was reported, along with an assessment of their properties. From the two scenarios considered in this work, the first one investigated different initial moisture contents that were used to produce pellets which were then evaluated for their physical and chemical properties. These results were compared to other research in the field as well as softwood bark pellets manufactured with the same apparatus at laboratory-scale. The results showed that moisture content is an important factor to produce high-quality lignin pellets since it has a direct influence on their mechanical durability, fines content, ash content, as well as on their calorific value. Ultimately, the lignin pellets manufactured with a 8.01% moisture content presented a mechanical durability of 94.80%. Pure lignin pellets had an overall inferior mechanical durability when compared to softwood bark pellets produced at 6.57% humidity, which in this case led to a 99.48% mechanical durability, a property that was improved with storage conditions. However, for lignin pellets (at a 8.01% moisture content), the calorific value property presented the best result of 23.27 MJ/kg, as compared to softwood bark pellets (at 6.57% moisture content) with 18.24 MJ/kg. Furthermore, when the production of solid biofuel from lignin is intended, low moisture and ash content of the raw material is required. The second scenario addressed in this work concerned the use of four types of bio-additives in the lignin feedstock, which ultimately translated into minor benefits of the physical properties of the pellets. Result showed that the most significant improvement in terms of moisture, durability, and fines content could be observed after storage. However, the additive with the best potential in this work was corn oil, which improved the downstream moisture content (8.41%) and calorific value (27.40 MJ/kg) while reducing the ash (1.26%) and sulfur content (0.00%). These results showed that the high mineral content of lignin could possibly be reduced by preparing the feedstock with a neutral wash, after the biomass pre-treatment. The results presented in this work suggest that there could be opportunities to produce lignin pellets as a side stream from a sugar-oriented biorefinery. Nonetheless, more research is needed to improve the physical and chemical properties of this type of solid biofuel. This study aims to be a first step...
towards the industrial production of lignin pellets, which could eventually be referred to as “third-generation” pellets.


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**Conflicts of Interest:** The authors declare no conflict of interest.

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