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Evaluation of the Potential of Agricultural Waste Recovery: Energy Densification as a Factor for Residual Biomass Logistics Optimization

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Abstract: The use of residual forms of biomass, resulting from processes of transformation of the agri-food and/or forest industries, presents itself as an alternative with high potential for energy recovery, given the existing availability, both from the perspective of quantities, but also from the perspective of geographic distribution. In this work, samples of four by-products originating from the agri-food industry were collected, namely coconut shells, sugarcane bagasse, cashew nutshells, and palm kernel shells, which were characterized in the laboratory by determining their Thermogravimetric and Elemental analysis, subsequently calculating the High Heating Value, Low Heating Value, Hardgrove Grindability Index, Mass Yield, Energy Yield, and Energy Densification Ratio. The values obtained show the potential to optimize logistical operations related to transportation, demonstrating that energy densification operations, especially if associated with physical densification processes, enable the use of these residual forms of biomass in the replacement of fossil fuels, such as coal.

Keywords: agriculture waste; thermochemical conversion processes; biomass energy; waste recovery

1. Introduction

The need for new forms of energy has given rise to a rapid development of energy technologies from all renewable sources [1]. Those developments led to major technological breakthroughs, which are reflected in greater energy production capacity

and cost reductions [2]. The latter is likely the most important factor considered in the choice of a particular type of energy to be implemented. Decision makers tend to opt either for choosing the cheapest forms or, at the very least, for energy profiles that required less investment vs. production capacity, while allowing permanent cash flow, thus, justifying a return on investment that is acceptable to investors [3].

Energy production, however, is an issue that arguably should be addressed through a strategy-setting approach at the national levels. In the case of European Union countries, the following strategy has, however, been mainly addressed from a purely economic perspective, wherein financial models determine the priority agenda of investments, often disregarding environmental and decentralized variables related to energy production [4]. In reality, energy production is one main topic that should be based on a thorough strategic discussion, since, for highly energy-dependent countries, a greater renewable energy production capacity should lead to significant reductions in the balance of transactions as well as a higher level of sustainability, related to economic decarbonization [5].

Currently, with the paradigm shift caused by environmental issues raised by climate change anticipated adverse effects, there has been a gradual mindset change regarding the issue of managing the energy matrix [6]. Policy makers increasingly show a greater willingness to give priority to alternative renewable energy sources at the expense of traditional sources of fossil origin, such as oil or coal. This new principle is especially valid when it is possible to use renewable sources in direct substitution for fossil sources [7] if coal consumption can be avoided because the installed wind capacity is able to satisfy the needs of the electricity grid, or if the annual hydrological regime is able to keep the hydroelectric power plant system operational [8]. However, this direct relationship between these renewable sources and meteorological factors leads to intermittent production and it is, therefore, not possible to have a constant supply for the power grid without extensive energy storage capacity [9]. For this reason, it is necessary to have dispatchable production points, which are able to ensure this supply when seasonal or daily weather conditions make it necessary [10].

From this perspective, biomass, being a renewable source of energy, can be a viable alternative fuel. At the same time, biomass presents a global availability in practically all latitudes of the globe, except for the poles and some desertic regions, but is widely distributed among the most inhabited regions where energy consumption is more intense [11]. However, present production of usable and available biomass that can be used as a source of energy is mostly carried out far from centers of consumption [12]. For example, residual forms of biomass of agricultural origin are produced far from large urban centers, where energy needs are more pressing, forcing the transport of energy products [13]. This transport, and the subsequent set of associated logistic operations, such as handling or storage, entails a set of costs that can make the operation and the sustainability of the process unfeasible [14].

In the above context, biomass shows distinct advantages, such as its widespread availability, the decentralized ability to be used in gas, liquid, or solid forms the energy potential and the capacity to serve as a counterpoint to the intermittency of other sources. However, biomass shows challenges, which include its low heating value, high moisture content, and low density, coupled with geographical dispersion, all reflected, for higher scale utilizations, in higher transport costs [15]. The sum of these issues makes the use of biomass for energy production often difficult to implement, since the costs associated with all the ancillary operations make the process unfeasible, making it imperative to study variables associated with the logistics process, in addition to variables associated with energy recovery processes [16].

There are several forms of biomass resulting from commercial agroforestry businesses, such as the production of coconut, sugarcane, cashew nuts, or palm oil, which deliver a steady supply of waste that, in the absence of use, can turn into environmental problems through the emission of greenhouse gases (GHGs), either by their combustion

to eliminate the materials, or by rotting due to lack of destination or form of recovery [17]. Thus, the possibility of transporting these materials to a destination where they can be valorized should be a very interesting possibility, were it not for the previously mentioned problems related to the logistical costs associated with these materials [18]. The scale of food processing facilities is generally not large enough to serve as an "end use point" for utility scale production, but it is often of sufficient scale to serve as a location for preprocessing prior to shipment to a larger, centralized conversion facility.

The possibility of materials being subjected to energy densification processes, such as thermochemical conversion processes, eliminating moisture and volatile compounds with low heating value, is associated with a densification process, such as pelletization or briquetting, which can transform waste with little value into readily transportable energy products, capable of functioning as an alternative to coal. This is mainly of the most common types with widespread commercial use, such as sub-bituminous and bituminous coals [19]. Another advantage is that those energy value-added products can be feedstock used directly in coal-fired power plants, without the need for major changes, since they have similar physico-chemical properties with regard to heating value and grindability [20].

In the above context, the aim of the present study was to characterize the properties of four widely used residual biomass feedstocks of agroforestry origin, namely coconut shells, sugarcane bagasse, palm kernel shells (PKS), and cashew nutshells, which are abundant in tropical and subtropical regions, and constitute a reserve with the potential to supply countries that are currently replacing coal as a way to reduce greenhouse gases (GHGs) emissions. Based on the results obtained, it was intended to demonstrate that the energy densification of these materials allows the reduction of logistical costs, and, thus, their transport to places of final use away from the points of production.

2. Materials and Methods

2.1. Sample Collection and Preparation

Samples of sugarcane bagasse, cashew nut shells, coconut shells, and PKS from Costa Rica, Brazil, Malaysia, and Nigeria, respectively, were collected in Portugal, as part of several samples that arrived for viability tests in March 2020. The origins of the materials were selected based only on its availability. With the exception of sugarcane bagasse, all other materials were destined for energy recovery in Europe, so samples were sent in significant quantities so that buyers could carry out laboratory tests to validate the products. Sugarcane bagasse feedstock, produced in the laboratory following the procedure described by Nunes et al. (2020), was used [21] with some variations in the tasks of sugarcane juice extraction and drying. The sugarcane samples were purchased and were subsequently washed to remove any dirt or contamination. Thereafter, they were cut into fragments and squeezed, as shown in Figure 1. In the end, the squeezed fragments were washed, so that any excess sugar still present could be removed, simulating the industrial process, which works under running water. Then, all materials were dried in a laboratory oven for 24 h at a temperature of 90 °C.
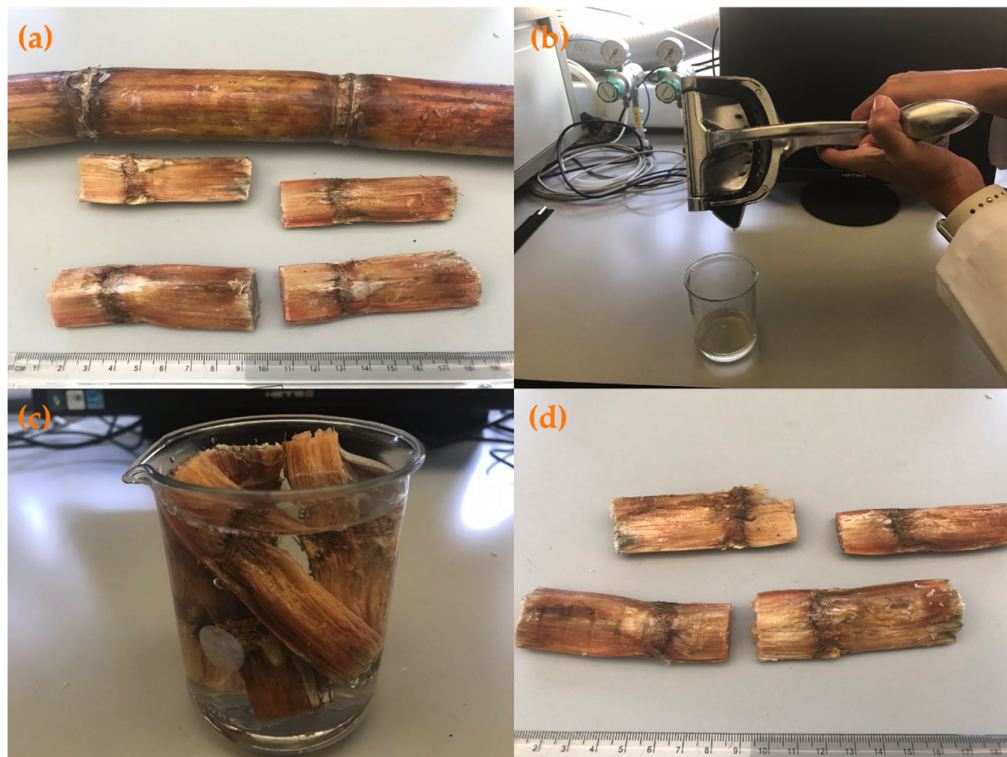
Figure 1. Process of preparing sugar cane samples to obtain bagasse. (a) Cutting sugar cane fragments, (b) juice extraction, (c) washing the sugarcane bagasse, and (d) dry fragments of sugarcane bagasse.

After drying, three samples of approximately 300 g, of each biomass feedstock, were weighed. Each set of three samples was subjected at thermal treatments of heating at 300 °C and 400 °C and without heating. Heat treatments followed the procedure presented by Ribeiro et al. (2018) [22] and were also previously used by other authors, e.g., Viana et al., 2018, and Sá et al., 2020 [23,24]. Biomass samples were compacted to cylinder specimens, which were wrapped in aluminum foil. The cylinder specimens were wrapped in aluminum foil in such a way so that the foil was tightened as much as possible for limiting the amount of air trapped to a minimum. Then, the wrapped specimens were placed inside a ceramic muffle furnace consisting of a metallic monobloc, covered with refractory bricks, with kaolin insulation. The furnace is heated by a system of electrical resistance coils, according to two sequential programs established to achieve process temperatures of 300 °C and 400 °C shown in Table 1.

Table 1. Correspondence of the four programmable levels with the different phases, depending on the temperature and residence time.

Phases	Temperature (°C)	Residence Time (minutes)	Temperature Range	
			300 °C Treatment	400 °C Treatment
Heating	T_{room} (18 °C) to T_1	30	18–180	18–180
Drying	T_1 to T_2	60	180–300	180–400
Torrefaction	T_2	90	300	400
Cooling	T_2 to 50 °C	Enough to cool and safely remove the samples	300–50	400–50

2.2. Elemental Analysis

The elemental composition of the samples was analyzed in accordance with standard EN 15104: 2011, Solid Biofuels–Determination of Total Content of Carbon, Hydrogen and

Nitrogen–Instrumental Methods. The oxygen content was thereafter estimated by a weight difference according to Equation (1).

$$w(O) = 100 - w(C) - w(H) - w(N) - w(S) \tag{1}$$

where $w(O)$ is the oxygen content (%), $w(C)$ is the carbon content (%), $w(H)$ is the hydrogen content (%), $w(N)$ is the nitrogen content (%), and $w(S)$ is the sulphur content (%). The sulfur amount was considered nil for all biomass samples. Samples were tested in duplicate.

2.3. Proximate Analysis

Thermogravimetric analysis (TGA) was conducted in accordance with the standards EN 14775: 2009, Solid Biofuels—Determination of Ash Content, EN 15148: 2009, Solid Biofuels—Determination of Volatiles Content, and EN 14774-3: 2009, Solid Biofuel—Determination of Moisture Content. Samples were tested in duplicate.

2.4. Determination of Heating Value

The heating value (HV) of a fuel was defined by Moran and Shapiro (2002) as being equal to its combustion enthalpy module [25]. The distinction of a high heating value (HHV) and a low heating value (LHV) is that, while the former is obtained when all the water formed in the combustion is liquid, the latter is obtained when steam is formed. The difference between a high heating value (HHV) and LHV is equivalent to the energy necessary for vaporization of the water formed in the combustion [26]. A high heating value (HHV) was calculated from elemental analysis results using Equation (2) presented by Channiwala and Parikh (2002) [27].

$$HHV = 0.3491 \times C + 1.1783 \times H - 0.1034 \times O - 0.0151 \times N - 0.0211 \times Ashes + 0.1005 \times S \tag{2}$$

whose validity range is $0\% \leq C \leq 92.25\%$, $0.43\% \leq H \leq 25.15\%$, $0.00\% \leq O \leq 50.00\%$, $0.00\% \leq N \leq 5.60\%$, $0.00\% \leq S \leq 94.08$, $0.00\% \leq Ashes \leq 71.4\%$, and 4.75 MJ/kg $\leq HHV \leq 55.35$ MJ/kg. Normally, for solid fuels, the value determined in the laboratory is the HHV of the dry material. The contents of moisture and ash are also relevant because these components cause high variations in the HV of biomasses [26]. A low heating value (LHV) of the dry material was calculated from the HHV and the elemental analysis results, where the enthalpy of water vaporized during combustion is discounted [28]. Equation (3) can be used for calculating LHV [27].

$$LHV = HHV - m_{H_2O} \times \Delta H_{H_2O_{vap}}(25\ ^{\circ}C) \tag{3}$$

where $\Delta H_{H_2O_{vap}}(25\ ^{\circ}C)$ is the enthalpy of water vaporization at 25 °C.

The mass of water formed during combustion is calculated using Equation (4).

$$m_{H_2O} = 9 \times H \tag{4}$$

where H is the hydrogen content in the dry solid fuel [26,27].

2.5. Material Grindability

Grindability is associated with the capacity of a material to be crushed or ground [29]. This property is a quality parameter commonly used in ores, coke, and coal to estimate yields and energy spent on milling processes [30]. Quantitatively, biomass grindability can be determined by using the Hardgrove Grindability Index (HGI) with adaptations, and, for lower HGI values, a greater energy will be required to grind the material. Bridgeman et al. (2010) used a method adapted from the British standard BS1016-112, Determination of Hardgrove Grindability Index of Hard Coal, released in 1995, to quantify the grindability of torrefied miscanthus [31]. Equation (5) predicts the HGI using proximate analysis results [32].

$$HGI = 102.69 + 4.227 \times S_{total} - 1.634 \times V - 0.569 \times A - 0.237 \times M \qquad (5)$$

where V, A, M, and S_{total} refer to volatiles, ash, moisture, and sulphur.

2.6. Specific Density and Energy Density

Lastly, the energy densification ratio (EDR), mass yield (MY), and energy yield (EY) were calculated by Equations (6)–(8) [33].

$$EDR = \frac{HHV_{dried\ treated\ biomass}}{HHV_{dried\ raw\ biomass}} \qquad (6)$$

where HHV $_{dried\ torrefied\ biomass}$ and $HHV_{dried\ raw\ biomass}$ refers to HHV of torrefied biomass and dried raw biomass, respectively.

$$MY\ (\%) = \frac{w_{dried\ treated\ biomass}}{w_{dried\ raw\ biomass}} \times 100 \qquad (7)$$

where $w_{dried\ torrefied\ biomass}$ is the mass of dried torrefied biomass and $w_{dried\ raw\ biomass}$ is the mass of dried raw biomass, and

$$EY\ (\%) = MY \times EDR \qquad (8)$$

where MY is the mass yield ratio (%) and EDR is the energy densification ratio.

3. State-of-the-Art

3.1. Framework

A significant range of biomass feedstocks that can be used as fuel, if properly pre-processed, for improving hindrances previously mentioned, such as high moisture content, lower heating value, and lower density, as has been widely reported over recent decades in many studies of biomass for energy applications, e.g., by Garcia et al. (2016), by Thorenz et al. (2018), Nunes (2020), or Nunes et al. (2020) [11,34–36]. There is also a wide variety of studied biomasses, ranging from those common in European latitudes, such as the maritime pine (*Pinus pinaster*), as reported by the studies by Enes et al. (2019), by Álvarez-Álvarez (2018), or by Viana et al. (2018) [23,37,38], to others available in other latitudes, as shown by Ratnasingam et al. (2015) or Ioras et al. (2012), wherein the energy properties of rubberwood from Malaysia are analyzed [39,40], or by Hytönen et al. (2019) who studied rubberwood, from Southern Thailand [41].

Many other studies were carried out on different types of technologies for converting and enhancing biomass, such as the work of Kim et al. (2014) wherein hydrothermal carbonization of anaerobically digested sludge for solid fuel production and energy recovery is analyzed [42], or the work presented by Lee et al. (2017) about pyrolysis of agricultural residues aiming biochar production [43], or the work of Sheth and Babu (2010) where the topic of hydrogen production from biomass gasification is addressed [44]. Many more studies are available, among which the works of Mohan et al. (2016), Lu and Ren (2016), Mohan et al. (2019), and Dahiya et al. (2018), on the diverse forms of recovery of energy and chemicals, e.g., biorefinery techniques [45–48].

A wide range of biomass studies have also been investigated for reintroduction of waste, resulting from forest management and agricultural production operations aiming to integrate these operations, under the new perspective of the circular economy, in new supply chains and value creation wherein waste is a source of additional income generation [49]. There are also works that approach this theme from different perspectives and are usually associated with different sectors of activity. For example, Rudi et al. (2017) analyzed the layout of a biomass value chain specifically for the Upper Rhine region [50], while Umar et al. (2013) analyzed the value chain of an activity sector, which is, in this case, the palm oil biomass renewable energy industry in Malaysia [51], demonstrating, in any case, the interest in the valorization of this type of material and their integration in the supply chains of biomass for energy.

3.2. Agricultural Residual Biomass

3.2.1. Sugarcane Bagasse

Sugarcane bagasse is a type of biomass that originated from residues of sugarcane cultivation widely available around the world [21]. For this reason, it is also one of the most studied forms of biomass, mainly due to the fact that this feedstock is associated with the production of bioethanol, a biofuel used to replace gasoline with wide dissemination, e.g., in Brazil and India [52–54]. The need to increase the efficiency and productivity of processes related to the recovery of sugar as much as possible from sugarcane bagasse has become a top scientific priority reflected by the huge number of published studies. For example, Rabelo et al. (2017) presented a paper on biorefineries for the production of bioethanol, methane, and heat from sugarcane bagasse [55], while Macrelli et al. (2012) carried out a technical-economic evaluation of the production of second generation bioethanol from sugarcane bagasse [56], which is in line with the study presented by Ramadoss and Muthukumar (2015) that addressed the topic of increasing productivity using dual salt on the pretreatment of sugarcane bagasse with hydrogen peroxyde [57]. However, perhaps the form of energy recovery most associated with sugarcane bagasse is direct combustion and conversion to charcoal [21]. In this perspective, there are several studies that characterize samples of sugarcane bagasse from different sources, highlighting the works originating in Brazil, such as those by Morais et al. (2017), Novotny et al. (2015), or Zandersons et al. (1999) [58–60], works originating in India, such as those by Shukla and Kumar (2017), Shanmukharadhya and Sudhakar (2007), or Varshney et al. (2019) [61–63], or even works originating from other sources, such as the Caribbean or Southeast Asia region, such as the works by Gongora and Villafranco (2018), by Quintana et al. (1998), Yevich and Logan (2003), or Arbex et al. (2000) [64–67].

3.2.2. Coconut Shells

Coconut shells are another form of biomass that has a high availability and geographic distribution, and it is already commonly used for value added goods and services such as energy production, as well as for the production of high-quality biochar widely used for conversion into activated carbon materials, used in preparation of filters and adsorbent products [68–71]. Energy uses have also been studied by Tsai et al. (2006), where the products resulting from the rapid pyrolysis of rice straw, sugarcane bagasse, and coconut shells in an induction-heating reactor are described, or in the work of Abrahim and Homenauth (2019), for evaluating the energy potential of the different varieties of coconuts in Guyana, or the work of Kumar et al. (2003), by analyzing the opportunities for using coconut shells in Sri Lanka [72–74]. Studies on the most efficient technologies for the energy recovery of coconut shells are also available. Sundaram and Natarajan (2009) carried out experimental work with pyrolysis, similarly to works conducted by Said et al. (2015), Rout et al. (2016), or Ali and Naebulharam (2017) [75–78]. With regard to torrefaction, there are also some articles, such as those of Chen and Kuo (2010), Tanchuling and De Leon (2018), or Hilmiyati et al. (2018) [79–81], presenting satisfactory results in conversion yields and product quality, when compared with similar conversion processes of common biomasses used in Europe and in the USA [82].

3.2.3. Cashew Nut Shells

Of the materials covered in the present study, cashew nut shells are perhaps the feedstock with the smallest quantities globally available. However, it can still be considered an abundant product, with world production divided by several countries, namely Vietnam, India, Ivory Coast, Philippines, Benin, Tanzania, Mali, Guinea-Bissau, and Brazil [83]. The energy uses of the biomass generated by the exploitation of the cashew nuts have been documented for a long time, namely with the work of Nindi (1991), on the uses of the cashew nut shells for the production of biochar in Tanzania, or Ramachandra

(1998), on the energy recovery of agricultural waste in the Indian state of Karnataka [84,85]. However, only more recently, studies on the energy recovery of this waste have become more common, such as with pyrolysis tests and the characterization of the resulting products by Ábrego et al. (2018), or by the valorization through gasification carried out by Sharma and Kaushal (2020) [86,87]. The main difficulty encountered in handling this material is related to the amount of oil that normally impregnates the product [88,89], and the irritant and allergenic properties of the oil that can cause dermatitis and related problems [90].

3.2.4. Palm Kernel Shells

The industry associated with the production of palm oil is of the utmost importance in agroforestry sectors in tropical countries, and also the one with the highest productivity [91]. Palm oil is a very broad-spectrum feedstock with uses ranging from food and biofuel production to cosmetics, soap, or detergents [92]. In fact, this constant demand for palm oil, given its versatility as a raw material, has also led to the expansion of planted forests of *Elaeis guineensis* coming into conflict with native forests in different parts of the world, as shown by numerous studies. For example, Petrenko et al. (2016) presents the ecological impacts of this expansion in Indonesia, while de Almeida et al. (2020) assess the long-term changes in the landscape caused by the expansion of palm cultivation in the Brazilian Amazon [93,94]. The use of palm oil in the production of biofuels, mainly liquids, is widely covered in the literature, as shown by the available publications, such as the works of Benjumea et al. (2008), by Mekhilef et al. (2011), by Al-Widyan and Al-Shyoukh (2002), by Ong et al. (2011), by Crabbe et al. (2001), or by Noiroj et al. (2009) [95–100]. However, the palm oil extraction process also results in a set of residues, including oil palm trunks (OPT), oil palm fronds (OPF), empty fruit bunches (EFB), palm pressed fibers (PPF), palm kernel shells (PKS), and palm oil thousand effluent (POME), which have been studied over the years, in order to find a solution for their elimination and/or recovery [101]. The use of thermochemical conversion pyrolytic technologies has already been addressed such as by Sabil et al. (2013), Assadullah et al. (2014), Faizal et al. (2018), Jaafar and Ahmad (2011), or by Rentizelas and Li (2016) [102–106]. The use of direct combustion has also been widely studied in the works of Pawlak-Kruczek et al. (2020), Odusote and Muraina (2013), Okoroigwe and Saffron (2012), or by Suheri and Kuprianov (2015) [107–110]. Currently, the use of residues from the palm oil industry, mainly PKS, is widely used in power generation, with a steady market installed in Southeast Asia, for supplying biomass to countries such as Japan and South Korea [111–114].

3.3. Biomass Thermochemical Conversion Technologies

Biomass thermochemical conversion technologies are processes wherein, by manipulating the temperature with an atmosphere poor or without oxygen, biomass undergoes structural changes that improve its energy properties, increase its grindability, and upgrade its physical storage capacity, due to an increase in hydrophobicity, and resistance to biotic agents [115,116]. As previously mentioned, there are several works on the thermochemical conversion of biomass residues of the most different provenances. However, the upscaling of conversion from laboratory to industrial production scale is an unchartered territory, since, currently, there are no large-scale operative production units, which allow confirmation of the promising experimental data made available in recent decades. Some examples are the works by Nunes (2020) presenting a study on the grindability of agroforestry wastes, aiming its use as an alternative to coal in coal-fueled power plants, by Gil et al. (2015), which present a study on the grindability and combustion behavior of coal and torrefied biomass blends, by the study from Yu et al. (2019), that analyzes the improvement of energy density and grindability of wood pellets by dry torrefaction, or by the study from Sá et al. (2020), that analyzes torrefaction in a perspective of pre-treatment of biomass for eliminating halogenated products such as chlorine [11,24,117,118]. The principles of thermochemical degradation can be also

utilized for assessing the chemical structure of biomaterials, through instrumental analysis of the products released by biomass pyrolysis. In this context, analytical techniques combining pyrolysis with gas chromatography (GC) and mass spectroscopy (MS), forming the Py-CG-MS system, can be used to identify the degree of thermochemical conversion of biomass and biochar, allowing to (i) upgrade the standards of quality evaluation of these materials, (ii) track the termed "combustion continuum" fundamental in the dynamics of environmental black carbon and carbon sequestration, and (iii) deepen the insight about the true technological potential of these byproducts.

3.4. Biomass Supply Chains

3.4.1. Short Distance Supply Chains

Short distance supply chains are those shorter than 50 km, or when, even if the distance is larger, only one transportation mode is used. The methodology of short distance supply chains is used for describing the process flowsheet from biomass harvesting to the stage of availability for conversion or recovery, eventually including the last stage of final consumption after conversion/recovery [119–121]. Several steps can be identified for a short distance supply chain for a biomass torrefaction unit, shown in Figure 2, where the different stages of the supply chain are listed, as well as the set of tasks associated with each stage.
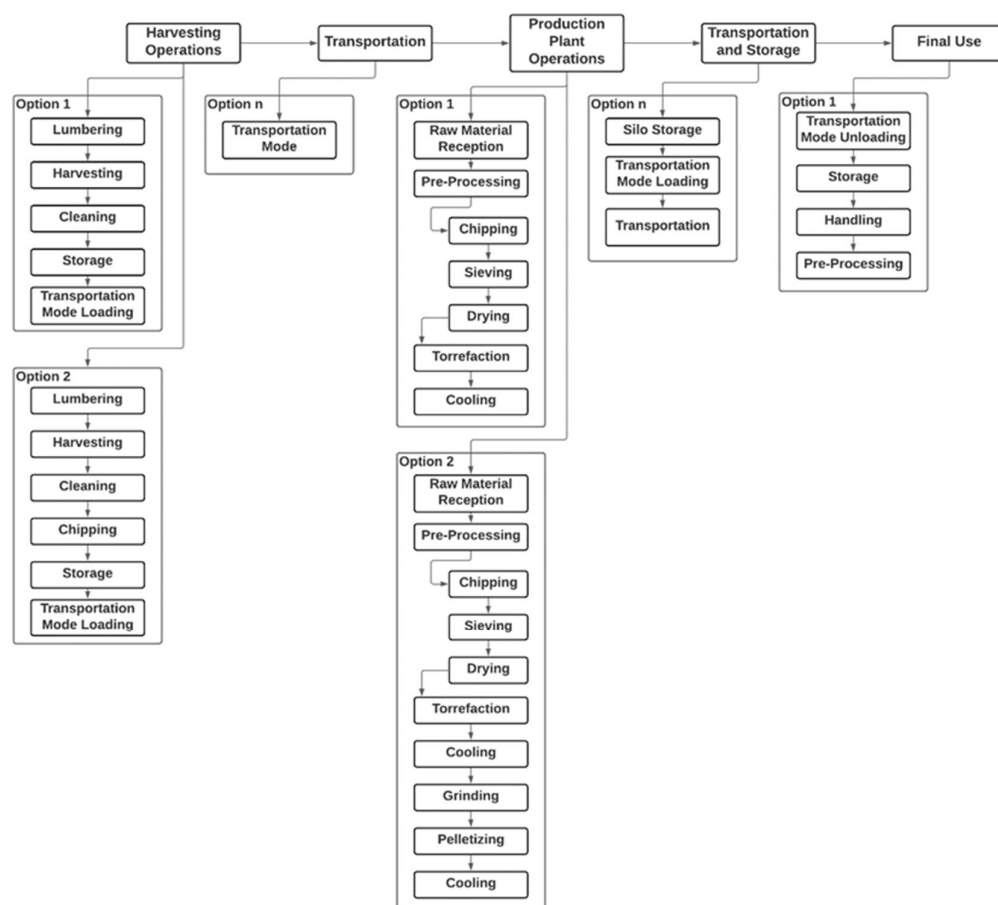

Figure 2. Definition of the different steps and options that may be present in a short distance supply chain.

Based on the definition of the different steps and respective tasks, it is possible to calculate the costs associated with the entire operation, providing the sequence of options followed during the process, since the model allows the combination of all different options and tasks between them. The total costs associated with the supply chain are calculated using Equation (9).

$$TOC = HOC + TC + PPOC + TSC + FUOC, \tag{9}$$

where TOC are Total Operation Costs, HOC are Harvesting Operation Costs, TC are Transportation Costs, PPOC are Production Plant Operation Costs, TSC are Transportation and Storage Costs, and FUOC are Final Use Operation Costs.

Each of the costs associated with each step are calculated using Equations (10)–(14).

$$HOC = Op_n \text{ Costs } \left(\sum_{i=1}^{n} T_n = T_1 + \cdots + T_{n-1} + T_n \right), \tag{10}$$

$$TC = Op_n \text{ Costs } \left(\sum_{i=1}^{n} T_n = T_1 + \cdots + T_{n-1} + T_n \right), \tag{11}$$

$$PPOC = Op_n \text{ Costs } \left(\sum_{i=1}^{n} T_n = T_1 + \cdots + T_{n-1} + T_n \right), \tag{12}$$

$$TSC = Op_n \text{ Costs } \left(\sum_{i=1}^{n} T_n = T_1 + \cdots + T_{n-1} + T_n \right), \tag{13}$$

$$FUOC = Op_n \text{ Costs } \left(\sum_{i=1}^{n} T_n = T_1 + \cdots + T_{n-1} + T_n \right), \tag{14}$$

where Op_n costs are the costs associated with each of the five options and T_n corresponds to the sequence of the n tasks incorporated to pursue each option. This model includes all operations of a logistical nature, even those associated with the production process. In this way, the model intends to exhaustively describe all operations in the process, with the objective of optimizing the design and operation of the logistical process.

The great difference between the model presented above in relation to other studies, namely those presented by Pavlou et al. (2016), Perrin et al. (2017), Paolotti et al. (2017) or Acuna et al. (2019), lies in the fact that these authors do not include the logistics components that occur inside the biomass recovery units in their analyses, dealing only with the logistical processes that occur outside of biomass recovery [122–125]. However, it is understood that, similarly to the treatment that is given in, e.g., automotive industry, short-distance operations should be evaluated, aiming to eliminate unnecessary operations and to achieve cost reductions [126–129].

3.4.2. Long Distance Supply Chains

Long distance supply chains are defined by including different types of transportation systems in their route, usually multimodal, with transfer units, or, in a simpler situation for being longer than 50 km, the distance usually accepted for the viability of non-densified biomass transportation [130,131]. These supply chains are associated, for example, with the supply of solid biofuels to coal-fired power plants, which can use wood pellets in a cofiring system [132]. For example, currently, North American wood pellet producers have their preferred markets in Europe and Asia (Japan and South Korea), with supply routes designed for delivering their predefined products [133–136].

4. Results

4.1. Elemental Analysis

The results obtained in the elementary analysis are shown in Table 2, including *p*-values for two-way ANOVA with n = 2 for the effect of feedstock type and thermal treatment type. This information was added as well in Tables 3, 4, and 5.

Table 2. Results obtained in the elemental analysis (average values shown).

Materials	Test	C (wt%)	H (wt%)	N (wt%)	O (wt%)
Sugarcane bagasse	As received	47.30	6.56	0.601	45.54
	300 °C	78.60	6.46	0.381	14.56
	400 °C	77.50	7.59	0.305	14.61
Cashew nutshells	As received	53.50	6.39	0.435	39.68
	300 °C	69.50	5.19	0.962	24.35
	400 °C	76.70	3.49	1.060	18.75
PKS	As received	52.00	5.43	0.322	42.25
	300 °C	58.50	3.56	0.603	37.34
	400 °C	56.00	2.90	0.613	40.49
Coconut shells	As received	46.70	5.35	0.869	47.08
	300 °C	70.10	3.60	1.120	25.18
	400 °C	74.10	3.28	1.030	21.59
p value: feedstock		0.0232	0.0174	0.0013	0.0042
p value: thermal treatment		<0.001	0.5446	0.5944	<0.001

The choice of feedstock impacts elemental composition for all measured elements, with cashew nutshells and PKS having higher carbon, lower nitrogen, and lower oxygen contents. Hydrogen content is highest in bagasse and cashew nutshells. Thermal treatment impacts carbon content and oxygen content in all samples, even though there is not a consistent trend when comparing the 300 °C and 400 °C treatment. This is likely due to preferential volatilization of oxygen-rich compounds from the biomass, which is not enhanced by temperatures above 300 °C. For example, in the case of sugarcane bagasse, wherein, after a rise in carbon content from 47.30% to 78.60% for the test carried out at 300 °C, a slight reduction was observed at 400 °C. A similar situation is observed for the PKS, where, after a rise from 52.00% to 58.50%, there is a decrease to 56.00%. In the case of cashew nutshells and coconut shells, values of consecutive rise of 53.50%, 69.50%, and 76.70% and 46.70%, 70.10%, and 74.10%, respectively, were obtained. There is a tendency similar to carbon for the hydrogen contents in sugarcane bagasse with the values evolving from 6.56% to 6.46% and 7.59%. In the remaining materials analyzed, there was a downward trend in the levels of hydrogen content. Nitrogen has a tendency of content increase with temperature for cashew nutshells and PKS samples, while there is a downward trend in the content in the case of sugarcane bagasse. In coconut shells, there was an increase from 0.869% to 1.120% followed by a decrease to 1.030%. The oxygen content shows a downward trend in cashew and coconut shells, from 39.68% and 47.08% without heating, to 24.35% and 25.18% at 300 °C, and to 18.75% and 21.59% at 400 °C, respectively.

4.2. Proximate Analysis

The results obtained in the proximate analysis are shown in Table 3.

Table 3. Results obtained in the proximate analysis (average values shown).

Materials	Test	Moisture (wt%)	Volatiles (wt%)	Ash (wt%)	Fixed Carbon (wt%)
Sugarcane bagasse	As received	3.31	82.25	1.36	16.38
	300 °C	3.79	27.10	4.39	68.52
	400 °C	3.67	25.97	5.38	68.65
Cashew nutshells	As received	5.85	81.32	2.04	16.65
	300 °C	3.55	48.85	7.24	43.92
	400 °C	5.24	22.77	7.42	69.81
PKS	As received	2.65	67.86	9.80	22.34

	300 °C	4.54	27.90	15.32	56.78
	400 °C	4.59	20.80	18.76	60.44
	As received	6.50	70.72	6.38	22.91
Coconut shells	300 °C	3.02	27.60	11.21	61.20
	400 °C	3.75	20.84	12.81	66.35
p value: feedstock		0.2426	0.0023	<0.001	0.1223
p value: thermal treatment		0.3184	<0.001	<0.001	<0.001

Moisture content does not show a statistically significant variation with respect to either feedstock or thermal treatment. Moisture presents a generalized downward trend in all materials, except for sugarcane bagasse, wherein the values are, respectively, 3.31%, 3.79%, and 3.67%. In the case of cashew nutshells, there was a decrease from 5.85% to 3.55%, but followed by an increase to 5.24% for the test carried out at 400 °C, likely related to the fact that the samples were not stabilized in the desiccator after being removed from the muffle. The volatile content shows a downward trend in all samples. The ash content shows an upward trend in all samples, related to the concentration of non-volatile materials and directly proportional to the mass losses which were, for 300 °C and 400 °C, and for each of the materials in the order of Table 1, of 60.16% and 73.63%; 37.77% and 76.67%; 35.79% and 75.46%; and 59.46% and 64.72%, respectively. The fixed carbon content shows an upward trend in all analyzed samples.

4.3. Determination of Heating Value

The results calculated for HHV and LHV are shown in Table 4.

Table 4. Values calculated for high heating value (HHV) and low heating value (LHV).

Materials	Test	HHV (MJ/kg)	LHV (MJ/kg)
Sugarcane bagasse	As received	19.45	16.85
	300 °C	33.45	30.89
	400 °C	34.37	31.36
Cashew nutshells	As received	22.05	19.52
	300 °C	27.69	25.64
	400 °C	28.78	27.39
PKS	As received	19.97	17.82
	300 °C	20.42	19.01
	400 °C	21.38	20.23
Coconut shells	As received	17.59	15.47
	300 °C	25.86	24.43
	400 °C	27.21	25.92
p value: feedstock		0.0442	0.0823
p value: thermal treatment		0.0181	0.0190

HHV and LHV are both impacted by feedstock and thermal treatment (at a 95% level of confidence). A generalized upward trend was observed in the values of HHV and LHV with increasing test temperature in all materials. The maximum values for HHV and LHV were related with sugarcane bagasse samples at 400 °C with 34.37 MJ/kg and 31.36 MJ/kg, respectively. PKS samples showed the lower values of HHV and LHV for 300 °C and 400 °C with 20.42 MJ/kg, 21.38 MJ/kg, 19.01 MJ/kg, and 20.23 MJ/kg, respectively.

4.4. Material Grindability

The results of grindability, calculated for the HGI, are shown in Table 5.

Table 5. Values calculated for the hard-grove grindability index (HGI).

Materials	Test	HGI
Sugarcane bagasse	300 °C	55
	400 °C	56
Cashew nutshells	300 °C	18
	400 °C	60
PKS	300 °C	47
	400 °C	57
Coconut shells	300 °C	51
	400 °C	60
p value: feedstock		0.3049
p value: thermal treatment		<0.001

HGI is not impacted by feedstock choice but is affected by thermal treatment ($p<0.05$). The calculated values were all greater than 50, with the exception of the value obtained for cashew nutshells at 300 °C and for PKS at 300 °C. In the remaining results, the values were distributed between 51, for coconut shells at 300 °C, and 60, for coconut shells at 400 °C.

4.5. Specific Density and Energy Density

The results calculated for EDR, MY, and EY are shown in Table 6.

Table 6. Values calculated for EDR, MY, and EY.

| Materials	Test	EDR	MY (%)	-	EY (%)
Sugarcane bagasse	300 °C	1.08	26		28.5
	400 °C	1.10	31		33.7
Cashew nutshells	300 °C	1.08	62		67.2
	400 °C	1.05	23		24.5
PKS	300 °C	1.07	64		72.0
	400 °C	1.07	25		26.4
Coconut shells	300 °C	1.06	41		42.9
	400 °C	1.05	35		37.1

The results obtained for EDR with the tests carried out at 300 °C between 1.06 for coconut shells and 1.08 for sugarcane bagasse and cashew shells. For the tests carried out at 400 °C, the values ranged between 1.05 for cashew and coconut shells, and 1.10 for sugarcane bagasse. MY values, except for sugarcane bagasse, showed a downward trend, in line with the mass losses presented. EY values followed the same trend as MY, rising for all materials, except for sugarcane bagasse.

5. Discussion

The main objectives of the characterization of the selected biomasses were the evaluation of their potential for replacing fossil fuels such as coal and possible logistical gains due a densification of these feedstocks, allowing for a financial feasibility of the transportation to more distant locations from the origin. Many studies are available on processes of physical densification of biomass, namely on the form of pellets and briquettes, such as the works of Bhattacharya et al. (1989), wherein an analysis of the pioneering state-of-the-art is presented, of Li and Hiu (2000), which address the issue of

high-pressure densification of wood residues, of Tumuluru et al. (2010), presenting a technical review on biomass processing, with emphasis on the optimization of the densification process, of Panwar et al. (2011), presenting a characterization of briquettes made from residual biomass, of Obidziñski (2014), addressing the pelletization of waste biomass, or the most recent works of Bajwa et al. (2018), Zhang et al. (2020), or Nunes (2020), where priority is given to balancing between the technological properties of densified biomasses vs. their potential to replace coal in energy production [11,137–142].

It was concluded that physical densification alone was not capable of solving all logistical problems of biomass conversion, especially these related with properties such as density and heating value, which allow the materials to become transportable over long distances [143–148]. Moreover, it was found that, with some residual biomasses, transport and use away from the origin would be possible only in addition to the physical densification, an energy densification through thermochemical conversion technologies such as torrefaction would be carried out (e.g., Uslu et al. (2008), Van der Stelt et al. (2011), and Chen et al. (2015) [19,149,150]. Thermochemical conversion technologies show interesting advantages from the point of view of improved logistical properties because they promote energy densification, as can be easily proven from the results obtained in this study. As can be seen in Figure 3, the thermo-chemical conversion processes of biomass promote energy densification, since their products show increases between 28% and 70% of the available energy per unit of mass.
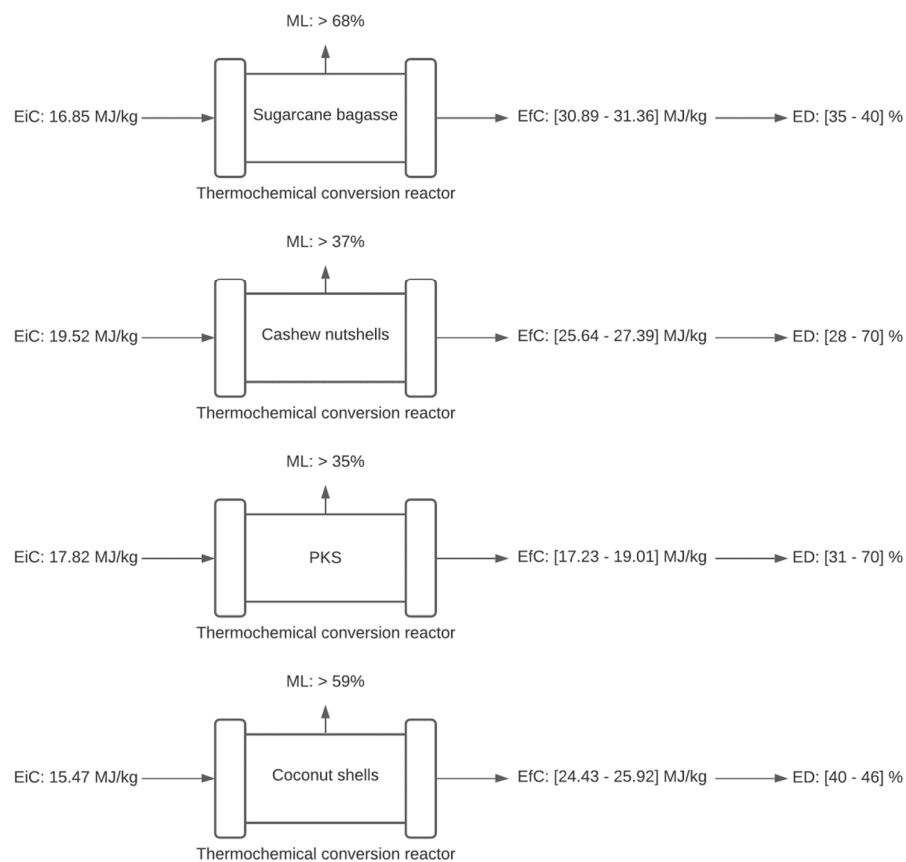
Figure 3. Mass and energy balance (EiC–Initial Energy Content, ML–Mass Loss, EfC–Final Energy Content, ED–Energy Densification).

From a perspective of cost analysis associated with long-distance maritime transport, of the transatlantic type, where the vessels used can transport around 60,000 tons of biomass pellets, it is possible to quantify a reduction in transport costs per unit of energy, which can vary between 35% and 52%. In the case of smaller boats that carry pellets within the European space, with capacities of around 7500 tons, the cost reductions are smaller. However, they are still quite interesting, ranging from 20% to 40%. In other words, thermochemical conversion technologies present themselves as enhancing the competitive logistical advantages of biomass products, associated with energy transport. In addition, the torrefied biomass products show other advantages, presented in the works of Ciolkosz et al. (2011), Kambo and Dutta (2014), Chen et al. (2018), or Zhang et al. (2020). These advantages are related to the hydrophobicity of materials, which allow their storage in less demanding situations, which can be arranged outdoors, similarly to coal. Moreover, their storage period can even be prolonged indefinitely, since these products do not react to biological activity [151–154]. This is an advantage, especially for short distance supply chains, since torrefied or carbonized biomass can also be used as a raw material for other processes, namely for the production of hydrogen by gasification [155–157].

From the point of view of the aptitude to combustion of the biomasses, there is some agreement of the results obtained in this work with the results from other previous works. In the case of sugar cane bagasse, the results of this work are concordant with those by Nunes et al. (2020), wherein the feasibility of using sugarcane bagasse subjected to carbonization of biomass at different temperatures was demonstrated, for applications of energy recovery from biomass products [21].

There are several references regarding torrefaction and pyrolysis of coconut shell, e.g., Chen and Kuo (2010), which concludes that severe torrefaction is not recommended to pretreat biomass due to a high percentage of mass loss and due to the possible difficulty of densifying torrefied products posteriori [79]. In the work presented by Nasution and Limbong (2017), it is shown that the average yield is 38.20% for a process temperature of 348 °C, which agrees with the results obtained in the present work, where, for 300 °C, a 41% mass yield was obtained, and, for 400 °C, a 35% mass yield was obtained [158]. PKS, perhaps the most studied product, of all those analyzed here, is the product that also presents the most interesting results, mainly for tests carried out at 300 °C, resulting in products presenting a MY of 64% and a 72% EY.

Regarding cashew nut shells, there are currently no studies available that can serve as a point of comparison. However, the results obtained were good, especially mass yield, which is 62%, and EY, which is 170% relative to tests carried out at 300 °C. These values showed a significant potential technological upgrading of the feedstock with thermochemical conversion. In Table 7, the LHV of commercial coals, used for the production of electricity, is shown with values ranging between 16.1 MJ/kg and 33.4 MJ/kg. This LHV range is similar to the LHV range between 19.01 MJ/kg and 31.36 MJ/kg found for the biomass samples analyzed in this work.

Table 7. Approximate lower heating values (LHV) of standard grades of coal (adapted from References [159,160]).

Coal Grade	LHV (MJ/kg)
Anthracite	30.1
Semi-Anthracite	32.1
Low-volatile Bituminous	33.4
Medium-volatile Bituminous	32.2
High-volatile Bituminous A	30.5
High-volatile Bituminous B	28.3
High-volatile Bituminous C	25.0
Subbituminous B	21.3
Subbituminous C	20.8
Lignite	16.1

Another important characteristic of biomass torrefied products is grindability evaluated in this work through the HGI calculation. This characteristic, which defines the greater or lesser difficulty in grinding a fuel, and, consequently, the amount of energy spent in the fuel grinding, which must be injected into the furnace. This is one of the stages of the energy production process wherein the constraints for replacing coal by biomass products normally occur. Most commercial coals show HGI values ≥50 [161], whereas fuels such as non-thermally processed biomass have HGI values <50 [162], indicating that the amount of energy needed to pulverize the products from torrefaction or pyrolysis is similar to that of coals. This quality upgrading delivered through the thermochemical conversion turns biomass, in its different forms, as an alternative to the use of coal for energy production. This was previously shown by authors such as Nunes et al. (2014), Proskurina et al. (2017), Nunes (2020), or Sher et al. (2020) [11,163–165].

However, despite the evident advantages found in the use of forms of biomass, especially those that fit into the residual forms, due to its enormous availability and low cost, the logistical disadvantages remain, associated with its low density and with the significant distances between the origin to the point of consumption. These challenges can still be overcome through a physical densification process such as palletization added to energetic densification by torrefaction, carbonization, or pyrolysis [166]. Economic issues related to the transportation of biomass take on an often-decisive role with regard to the use and recovery of these materials, especially those that are considered residual. The costs associated with transport vary widely from case to case, namely due to the issues related to the density, inherent to the type of material, but also to the distances to which the materials have to be transported. It is, at this point, that the energetic densification of materials assumes a decisive role, since it can enhance their transport over greater distances, especially intercontinental transport, creating a true value chain for waste materials, in a perspective of circular economy. This value chain, if possible, will allow products that, until now, have no use, or at least do not have a use that values waste completely, to be incorporated into a global supply chain, serving as alternative, traditional fossil fuels. This path presents itself as a true tool for mitigating climate change, by contributing to the reduction of GHG emissions.

6. Conclusions

The use of residual biomass as an energy alternative to the use of fossil fuels presents itself as a possibility that assumes increasing importance. In fact, the availability of these materials, combined with their dispersion, makes them very viable alternatives. However, despite these apparent competitive advantages, in the vast majority of cases, it is necessary to use technologies that promote energy densification, while improving other properties, such as grindability, the reduction of humidity, or hydrophobicity. Thermochemical conversion technologies play an important role in this context, since they allow obtaining

products with optimized combustible properties from residual biomasses while improving the perspectives of logistics involved in the transport process between the locations of production and consumption in which the latter is associated with energy production. Overall, the results were very positive with EY values ranging from 24.5%, for samples of cashew nutshells processed at 400 °C, to 72%, for PKS samples processed at 300 °C. At the same time, there is an increase in the values calculated for the HGI, which, in all situations, approach, or exceed, the value of 50, considered to be the most common value found in commercial coals. These results clearly showed the existence of a significant increase in the energy density of the products from thermochemical conversion processes such as torrefaction or pyrolysis. An additional optimization of the logistic transport processes is possible if the thermochemical conversion is associated with physical densification processes, such as pelletization or briquetting. This possibility allows the creation of a value-added chain for waste materials in a real perspective of a circular economy, at the same time that it contributes to the creation of an alternative to fossil fuels. However, further studies are needed, mainly related to the combustion of residual biomasses in their different states of thermochemical conversion, to verify the potential for the occurrence of corrosive, fouling, and slagging phenomena, as well as the combustion stability when used in co-firing processes with coal.

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