energies

Review

A Review on the Effects of Pretreatment and Process Parameters on Properties of Pellets

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Abstract: The development and utilization of biomass can not only address the demand for low-carbon energy and reduce environmental pollution, but can also facilitate the achievement of carbon neutrality. However, there are many factors justifying the case for low utilization of agricultural residues. These factors could be well controlled by producing top-quality pellets. Production of pellets is generally accompanied by the problems of high energy consumption and serious mold wearing. To eliminate these deficiencies, pretreatment has attracted scholars' attention. In this review, the effects of four pretreatments on the properties of pellets were assessed. Thermal pretreatment can improve the hydrophobicity of pellets, and optimize their properties, while degradation of diverse extractives is noteworthy. Hydrothermal pretreatment improves the physical properties of pellets, through the increase of polar functional groups on the surface of the biomass. Ultrasonic vibration-assisted (UV-A) pelleting produces pellets under low pressure without a heating process; however, it is still not applied to large-scale production. Supercritical fluid extraction can achieve the graded utilization of extracts and bioactive substances in biomass, and the residues can be subsequently utilized as pellet feedstock. Mild hydrothermal treatment is a promising approach to improving the quality of agricultural pellets. Additionally, the effects of process parameters on the physical and chemical properties of pellets should be systematically analyzed.

Keywords: biomass; agricultural residues; pretreatment; process parameters; properties of pellets

Citation: Li, W.; Wang, M.; Meng, F.; Zhang, Y.; Zhang, B. A Review on the Effects of Pretreatment and Process Parameters on Properties of Pellets. *Energies* **2022**, *15*, 7303. https://doi.org/10.3390/en15197303

Academic Editor: Alberto Coz

Received: 6 September 2022
Accepted: 29 September 2022
Published: 4 October 2022

1. Introduction

The non-renewable nature of fossil energy, and humanity's excessive dependency on it, has created challenges to the sustainable development of the global economy. This unrestrained consumption of fossil energy is causing ever more serious deterioration of the ecological environment through, for example, air pollution and greenhouse gas emission. Development of renewable and clean energy is the top priority worldwide. The potential of agriculture for bioenergy utilization has been confirmed. Sugar cane, maize, rice, wheat, soybeans cassava, and sugar beet were the leading crops in 2020. The production of crop residues has increased from 1589 Mt in 1960–1961 to 5280 Mt in 2020–2021 [1,2]. In 2020, 47% of the total production of crop residues globally belonged to Asia, followed by the rates of 29%, 16%, 7%, and 1% in America, Europe, Africa, and Oceania, respectively [2].

Biomass is the only renewable resource that can participate in repository and logistics, accompanied by its great energy potential. Biomass accounts for about 90% of all renewable energy resources in Lithuania [3]. In India, around 686 M tons of residue are accounted for by the top 10 crops [4]. The amount of crop residue has been stable at 800 M tons in each year of the past 10 years in China [5]. In order to improve the rural fuel supply and protect the rural ecological environment, the Chinese government has implemented a number of policies to support the utilization of agricultural residues, promoting the comprehensive utilization of agricultural straw resources. The comprehensive utilization rate of agricultural residues in China continuously improved from 2010 to 2020, as illustrated in Figure 1. In 2020, that rate reached 87.6%, in which the utilization rates of fertilizer, fodder, energy and fuel, stroma, and raw materials were 62.1%, 15.4%, 8.5%, 0.7%, and 1.0%, respectively. Compared to 2010, the proportion of fertilizer utilization has gradually increased, whereas the proportion of fuel utilization has decreased, because direct combustion of agricultural residues was prohibited. Crop residues have been also used as animal feeds, bedding materials, domestic fuels, roof thatching, and packaging materials in India [6]. The utilization rate of crop residues as fertilizers is over 65% in the USA, Japan, and Canada, and around 20% in Korea [7]. Lebanon, Pakistan, Syria, Iraq, Israel, and Tanzania and other African countries use crop residues as feed for animals [8]. India's National Policy on Biofuels-2018 recommended the increased usage of biofuels in the energy and transportation sectors of the country during the upcoming decade [9]. Poland is planning to produce at least 80% of its total energy from renewable sources, and more than 75% of its biomass energy will be from crop residues [10]. The American Energy draft proposed that cellulosic ethanol production should account for 3% of total fuel ethanol demand by 2012 and 44% by 2022 [11]. The 2015 alternative energy development plan in Thailand focused on the development of the share of renewable energy in final energy consumption, from 11.9% in 2014 to 30.0% in 2036 [12,13]. The comprehensive utilization rate of agricultural residues in China will reach 94% in 2030, and it will be fully applicable after 2040 [14].

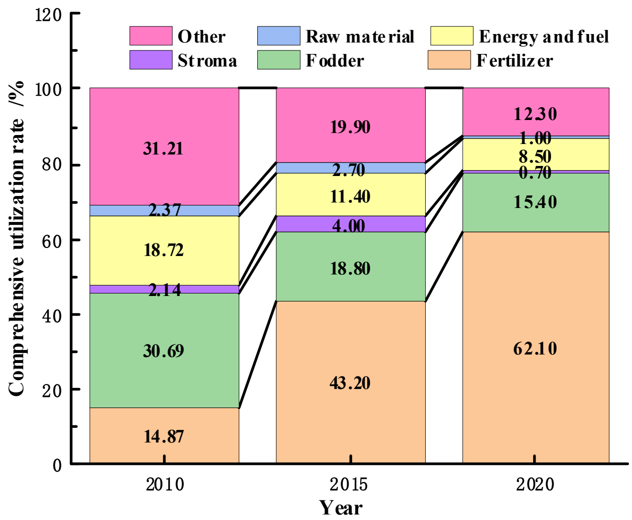

Figure 1. The comprehensive utilization of crop straw in 2010, 2015, and 2020.

High moisture content, volatile matter and costs of logistics, low-density raw materials, the melting temperature of ash [15], slagging, fouling, corrosion of heat exchangers [16], and the non-uniform size of biomass particles [17] are the main reasons for the low utilization of agricultural residues. Furthermore, hydrophilic agricultural residues are highly favored, which may be attributable to their noticeable hemicellulose level, strong seasonality, and high dispersion, resulting in difficulty establishing a large-scale and stable raw material supply system. Therefore, accumulation of biomass dust, microbial degradation of humid conditions, and internal heat-induced ignition are consequences, mainly originating from the long-term repository and overseas logistics of hydrophilic agricultural residues [18].

For energy production, utilization of biomass as pellets was suggested, because a more cost-effective biofuel could be obtained, versus the direct application of non-modified biomass residues [19]. Pellets possess unique benefits, involving reduced volume, maximized density, unified shapes, optimized repository costs, easier deposition, dust reduction, and attenuation of transport costs [20]. In addition, in terms of their composition and combustion, pellets are friendly to the environment, due to their reduced contents of ash, sulfur, arsenic, and heavy metals, achieved by selecting appropriately the composition of the raw materials [21–24], and by proper pretreatments during the forming process [25–28]. Although much research has shown that there is no net addition of CO_2 generation during the combustion of pellets [29–31], one fact needs highlighting: the CO_2 equivalent generated by pellets was not considered, including the residues and greenhouse gas from biomass harvesting, transportation, drying, and transformation into combustion [32]. According to the results of Pelletier et al. [32], the effects of pollutant emissions (such as CO_2, aromatic compounds, CO, etc.)—especially that from incomplete combustion process—on the potential for global warming are significant. In order to analyze the emissions of pellets accurately, it is necessary to associate carbon neutrality with life cycle analysis and combustion efficiencies. Fortunately, since 2007, China has concentrated on the development of biomass power generation, biogas, solid fuels, and biomass liquid fuels, and emphasized improving the utilization efficiency of biomass to promote bioenergy in 2016. In China, on the basis of China's Intended Nationally Determined Contribution, the emission of CO_2 will peak in 2030, followed by carbon neutrality in 2050 [33]. Regarding the environmental performance of pellets, they will be outstanding alternatives to coal production [33,34].

According to the European Biomass Association's statistics, in 2017 the global consumption of densified biomass fuels was about 32 Mts, of which 75% were mainly consumed as wood pellets in Europe [35]. Noticeable growth has been reported globally in wood pellet demand over the past decade, especially in Western countries, and in South Korea and Japan [36]. In 2018, 34 Mts of pellets were demanded globally, and demand is expected to reach 69 Mts by 2025 [37]. In 2019, the rate of pellet production was as high as 40.5 Mts worldwide. Globally, this rate peaked in Europe (55%), followed by America (30%) [38]. In 2020, on the basis of data released by the World Bioenergy Association, biomass global supply rose as high as 55.6 EJ, of which solid biomass had the greatest proportion (85%) [39]. The energy potential of woody biomass was confirmed, in particular, when higher energy content was densified. The outstanding properties of non-woody pellets have absorbed scholars' attentions, of which woody chips were found to have the greatest level of advantageousness, followed by herbaceous biomass, owing to its remarkable compressibility [40]. As waste recycling could positively influence carbon reduction, a variety of biomass residues were suggested for the purpose of pelletizing [40].

Pretreatment of biomass residues may improve fermentable sugar yield, biogas yield, and the properties of biochar; however, different pretreatment technologies have recently been utilized to produce pellets. The main objective of the current review was to figure out the effects of four pretreatment methods on pellet properties. The merits and demerits of these methods were described, and the effects of operational parameters were systematically analyzed.

2. Integrating Pretreatment Approaches with Pelletization

The various molding processes of pellets can mainly be divided into normal temperature molding, hot-pressing molding, and carbonized molding. The traditional methods mainly include ring-die pelleting, flat-die pelleting, screw extrusion, piston pressing, and roll pressing [41]. Normal temperature molding is mainly utilized in ring-die pelleting and flat-die pelleting [42]. The equipment has a simple structure, and its maintenance is user-friendly. Compared to normal temperature molding, hot-pressing molding can remarkably attenuate compression energy consumption, improve product performance, and prolong the service life of key components [42,43]. However, the use of external heat resources increases the power consumption. Carbonized molding partly improves the compressibility of biomass, and reduces die wears; at the same time, it is difficult to pellet without additives, because the surface viscosities of biomass decrease after carbonization [44,45]. Due to the natural recalcitrance of biomass, the physicochemical structures need to be modified for its application [46]. There are many pretreatment methods, including physical (milling and grinding), physicochemical (steam pretreatment, hydrothermolysis, and wet oxidation), chemical (alkali, dilute acid, oxidizing agents, and organic solvents), biological, and the combination of these [47]. Chemical pretreatment methods are often used to degrade lignocellulose and cellulose, remove lignin from biomass, and enhance hydrolysis [48]. Biological treatment is safe, environmental friendly, less energy intensive, and cost effective [31,49]. Despite several studies showing that it can improve physical quality [50–52], this method requires a long processing cycle. To convert crop residues into high-quality pellets, four pretreatment approaches integrated with pelletization are discussed in this review: thermal pretreatment, hydrothermal treatment, ultrasonic vibration-assisted pelleting, and supercritical fluid extraction.

2.1. Thermal Pretreatment Integrated with Pelletization

Thermal pretreatments, such as torrefaction and pyrolysis, can improve the hydrophobicity of pellets, and optimize their physical and chemical properties, making them worthwhile for research purposes. As a thermal conversion approach, torrefaction is advantageous for the purpose of improving the energy density of biomass, mainly via the heating of biomass to reach moderate temperatures under atmospheric pressure, while no oxygen is required [53]. After torrefaction, most celluloses and lignin remain, and carbohydrates and lipids could be residually found in biodried products, functioning as outstanding lubricants and binders in the pelletization [27].

Torrefaction is the most influential treatment for improving pellet quality, owing to its function in improving the physicochemical properties of biomass [54–56]. Torrefied pellets exhibit a greater density, attenuated moisture contents, elevated calorific values, enhanced efficiency, and reduction of microbial growth, particularly in humid conditions, versus untreated-wood pellets, facilitating long-term repository and overseas logistics [45,57–59]. During in situ torrefaction and densification, at a high temperature, a viscous molten lignin is formed, facilitating migration, reducing the gap between particles, and forming stable hydrogen bonds using hemicellulose and lignin, leading to the production of pellets with maximized hardness and density, and minimized specific energy consumption [60]. The complementary effects of torrefaction and co-pelletization improve the characteristics of pellets made from biomass and cater bean cake, with high heating values, low moisture absorption, and outstanding combustion characteristics [28]. It has been proposed to reduce the energy required for pelletization when torrefied biomass is co-pelletized, which could be achieved by the smooth extrusion resultant from the oil content of mustard meal unctioning as a lubricant during densification [59]. Xia et al. [61] reported that the density and calorific value were elevated by 7.99% and 15.01%, respectively, while the strength was reduced by 11.33% after torrefaction. Chen et al. [60] showed that 250 °C was an optimum temperature for rice straw and rice husk torrefaction, with densities of 1236.84 and 1277.50 kg m^{-3} under 150 MPa pressure, respectively. Reduced moisture adsorption (59%), elevated carbon content (range, 21–25%), attenuated oxygen content (35–46%), and increased heating value (22%) were noted as the outstanding characteristics of torrefied pellets versus other precursors [62].

On the basis of Sarker et al.'s findings [63], the following parameters should be set for optimizing torrefaction of fuel pellets: microwave power (10 min, 90 g feeding load, 250 W); relaxed density (maximum, 1090 kg m^{-3}); durability (83%); and mechanical strength (0.55 MPa). Noticeable improvement of energy content and hydrophobicity was confirmed via the optimized torrefaction, which could be attributed to the degradation of the lignocellulosic structure. Ma et al. [27] revealed the pellet density and energy consumption trend with increasing torrefaction temperature, as shown in Figure 2. However, Siyal et al. [64] pointed out the elevated friction coefficient of densification, via degrading a variety of extractives in the biomass torrefaction; simultaneously, some hemicelluloses were decomposed, which reduced the contents of the hydroxyl groups, decreased the strength of the hydrogen bonds during densification, and attenuated the quality of the pellets.

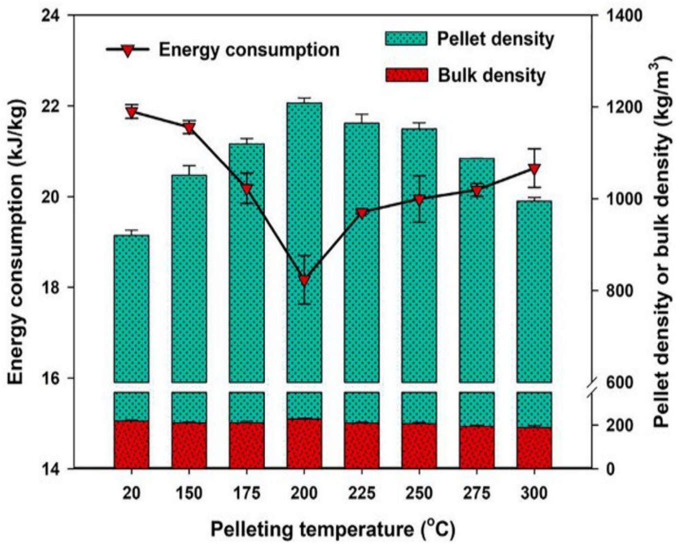

Figure 2. Pellet density and energy consumption for pelletization of untreated and torrefied BPs. Adapted with permission from Ref. [27]. 2022, Elsevier.

Cheng et al. [65] concentrated on the emission of particulate matter originating from agricultural biomass combustion, and non-oxidative and oxidative torrefactions, which could result in the noticeable elevation of the yield of PM_{10}. It was confirmed that the variation of the main composition of PM_1 from KCl to K_2SO_4 could be attributed to the torrefaction-induced noticeable release of Cl. The presence of O_2 caused remarkable levels of Ca and K in PM_{1-10}, in which alkali and alkaline-earth metals could be transformed into coarse particles, as illustrated in Figure 3.

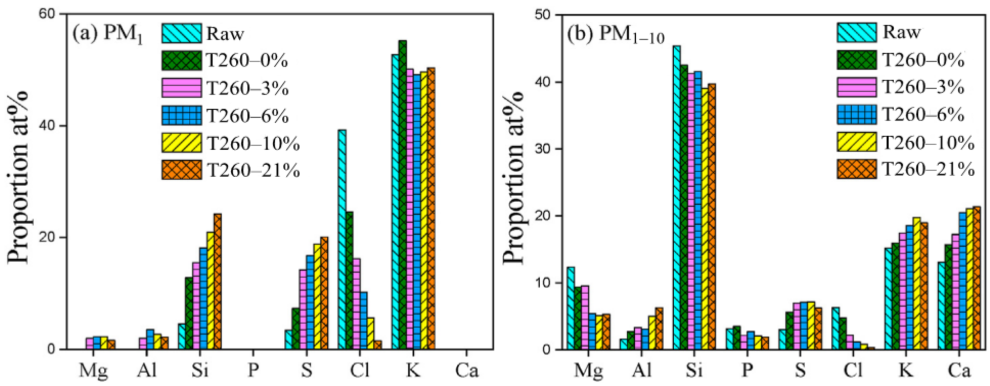

Figure 3. Chemical compositions of PM_1 and PM_{1-10}, from the combustion of raw and torrefied cotton stalk pellets at 260 °C with different oxygen content. Adapted with permission from Ref. [65]. 2022, Elsevier.

Once densification was completed, torrefaction could raise the porosity of the raw wood pellets that would be associated with the volatilization of the hemicellulosic components from the lignocellulosic matrix and the attenuated moisture content of the pellets [66]; the shape or integrity of the wood pellets, however, would not be changed. Kambo et al. [54] indicated that torrefaction had the disadvantage of being energy-intensive, owing to the high friction in the die channel, and produced pellets that tended to have low durability. Generation of brittle dust by torrefaction was confirmed, which could reduce the density of the biomass, and ultimately influence handling, repository, and shipment [63].

In comparison with torrefaction, biomass can be decomposed into gas, liquid, and solid products via pyrolysis under an inert atmosphere, in which biomass is promptly heated (temperature, ≥300–650 °C) [57]. There are two pathways to producing pyrolysed pellets:

pyrolysis before pelletization, and pelletization-induced pyrolysis [67,68]. Numerous researchers suggested the applicability of biomass in torrefaction and pyrolysis before the release of water in pelletization and partial volatilization, which was more difficult than the production of pellets using raw materials under the same conditions, resulting in greater pressure and temperature to obtain pellets with the same quality [63,67,69]. Pelletization-induced pyrolysis, which can be utilized for the purpose of producing binder-free pellets, has attracted scholars' attention. Siyal et al. [67] presented the mass and energy yields of pyrolysed furfural residue pellets (PYFRPs), pyrolysed sewage sludge pellets (PYSSPs), and pyrolysed furfural residue–sewage sludge pellets (PYFRSSPs), respectively, as presented in Table 1.

Table 1. Physical and mechanical characteristics of pellets. Adapted with permission from Ref. [67]. 2022, Elsevier.

Quality Parameters	**Temperature (°C)**					
	200	**250**	**300**	**450**	**650**	**850**
			Particle density (g/cm^3)			
Raw FRPs			PYFRPs			
1.38 ± 0.01						
[a]RT-15 min	1.309 ± 0.01	1.25 ± 0.01	1.16 ± 0.01	1.188 ± 0.01	1.141 ± 0.00	1.119 ± 0.00
RT-30 min	1.29 ± 0.02	1.248 ± 0.01	1.166 ± 0.01	1.172 ± 0.01	1.146 ± 0.01	1.128 ± 0.01
RT-45 min	1.279 ± 0.01	1.24 ± 0.02	1.169 ± 0.02	1.169 ± 0.01	1.154 ± 0.01	1.146 ± 0.02
Raw SSPs			PYSSPs			
1.517 ± 0.02						
[a]RT-15 min	1.459 ± 0.01	1.409 ± 0.02	1.354 ± 0.01	1.259 ± 0.01	1.209 ± 0.01	1.181 ± 0.07
RT-30 min	1.448 ± 0.02	1.406 ± 0.02	1.349 ± 0.01	1.254 ± 0.01	1.201 ± 0.02	1.178 ± 0.07
RT-45 min	1.446 ± 0.01	1.4 ± 0.02	1.334 ± 0.01	1.25 ± 0.01	1.193 ± 0.01	1.167 ± 0.01
Raw FRSSPs			PYFRSSPs			
1.296 ± 0.02						
[a]RT-15 min	1.237 ± 0.02	1.221 ± 0.01	1.08 ± 0.01	1.043 ± 0.01	1.007 ± 0.01	1.004 ± 0.01
RT-30 min	1.222 ± 0.01	1.198 ± 0.02	1.071 ± 0.02	1.041 ± 0.01	1.006 ± 0.01	0.993 ± 0.01
RT-45 min	1.219 ± 0.01	1.194 ± 0.01	1.064 ± 0.01	1.039 ± 0.01	0.998 ± 0.01	0.989 ± 0.02
			Volumetric energy density (kJ/m^3)			
Raw FRPs			PYFRPs			
29.3 ± 0.00						
RT-30 min	28.66 ± 0.17	29.66 ± 0.23	29.86 ± 0.16	30.35 ± 0.15	30.12 ± 0.17	30.18 ± 0.16
Raw SSPs			PYSSPs			
12.38 ± 0.00						
RT-30 min	12.26 ± 0.24	11.36 ± 0.13	10.17 ± 0.14	8.92 ± 0.15	8.34 ± 0.11	7.88 ± 0.15
Raw FRSSPs			PYFRSSPs			
26.56 ± 0.00						
RT-30 min	25.17 ± 0.13	25.08 ± 0.14	24.14 ± 0.17	23.85 ± 0.14	23.84 ± 0.17	23.56 ± 0.10
			Strength (N/mm^2)			
Raw FRPs			PYFRPs			
5.92 ± 0.79						
RT-15 min	5.435 ± 0.90	5.147 ± 0.46	3.81 ± 0.86	1.891 ± 0.25	1.394 ± 0.08	1.267 ± 0.03
RT-30 min	5.238 ± 0.78	4.542 ± 0.59	3.35 ± 0.76	1.751 ± 0.41	1.197 ± 0.24	1.025 ± 0.05
RT-45 min	4.892 ± 0.64	4.368 ± 0.82	3.04 ± 0.48	1.63 ± 0.45	1.101 ± 0.07	0.859 ± 0.11
Raw SSPs			PYSSPs			
5.238 ± 0.22						
RT-15 min	4.297 ± 0.06	3.263 ± 0.14	2.085 ± 0.13	1.114 ± 0.05	1.012 ± 0.05	0.837 ± 0.03
RT-30 min	3.804 ± 0.12	3.104 ± 0.08	1.926 ± 0.07	1.038 ± 0.06	0.99 ± 0.07	0.805 ± 0.04
RT-45 min	3.785 ± 0.28	3.059 ± 0.55	1.795 ± 0.24	0.945 ± 0.04	0.95 ± 0.05	0.719 ± 0.04
Raw FRSSPs			PYFRSSPs			
5.68 ± 0.18						
RT-15 min	5.465 ± 0.34	4.985 ± 0.02	3.711 ± 0.07	2.218 ± 0.09	1.909 ± 0.23	1.798 ± 0.1
RT-30 min	5.159 ± 0.65	4.94 ± 0.21	3.403 ± 0.04	2.145 ± 0.06	1.836 ± 0.08	1.655 ± 0.03
RT-45 min	5.013 ± 0.32	4.833 ± 0.09	3.23 ± 0.04	2.104 ± 0.11	1.821 ± 0.04	1.566 ± 0.04

[a]RT is residence time.

2.2. Hydrothermal Treatment before Pelletization

Under aqueous conditions, hydrothermal pretreatment can be defined as a correlation of physical and chemical pretreatment processes [70]. It increases the number of polar functional groups on the surface of the biomass. Numerous studies have reported that the chemical structure of biomass contributes to particle bonding, and further affects the quality of the pellets. Anukam et al. [71–73] and Liu et al. [74] elucidated the multiple polar functional groups in the structure of biomass, via enhancing electrostatic attraction that could create intermolecular bonding, such as hydrogen bonding and van der Waal's forces. Sylvia et al. [75] attempted to utilize macromolecular composition for the purpose of predicting process settings, particularly for high pellet durability; Orthogonal Partial Least Squares Projections to Latent Structures models were established via macromolecules, die compression ratio, and feedstock moisture content. Escalated values of fixed carbon content and heating, hydrophobicity, mechanical strength, and reduced ash content resulted from hydrothermally pretreated biomass versus raw (untreated) biomass [76]. Compared with untreated biomass, cotton stalk and sawdust after hydrothermal treatment at 200 and 300 °C increased the density, compressive strength, and heating value of pellets by 9.15–27.3%, 114.0–241.3%, and 5.1–59.0%, respectively. With the increase of the hydrothermal temperature from 200 to 260 °C, the yield of the charcoal briquette made from cotton stalk and sawdust increased, and the calorific value remained stable, while the combustion characteristic became worse [77].

Steam pretreatment, as an extensively employed hydrothermal pretreatment that leads to structural disruption and lignin relocation, enhances pellet durability and heating values. Attenuating the necessity of size reduction, noticeably elevating pellet durability via relocating the plant cell wall lignin to the fiber surface, and strengthening binding among particles can result from pre-steamed biomass [78]. Tang et al. [79] found that lignin modification was the most precious consequence of steam treatment, while the role of particle size distribution in the enhancement of the pelletization process and pellet quality was not highly emphasized. Takada et al. [80] also pointed out that lignin redistribution could be originated from pre-steaming, and might enhance pellet durability. However, no change of lignin distribution by pelletization within the cell wall was figured out, and the original ease of hydrolysis could be retrieved via subsequent refining, as illustrated in Figure 4. Remarkably improved pellet durability resulted from addition of lignosulfonates to pelletization, which positively influenced subsequent cellulose hydrolysis; this could be attributed to the function of lignosulfonates as a surfactant, and to the attenuated unproductive binding of enzymes to the lignin.

In biomass densification, the function of lignin and sugar as a natural binder was confirmed, especially under high temperature and pressure, and this may upgrade the fuel pellet quality and retain hydrophobicity [81]. The pretreatment of steam-exploded biomass (temperature, 180 °C; pressure, 0.9 MPa) was accompanied by 13% reduction of the energy required [82]. A different result was obtained by Tang et al. [83], in which steam explosion did not influence the compression energy of ground poplar. However, steam explosion would affect the higher heating values of pellets. Compared with five different steam pretreatment approaches (acid, auto-catalyzed, neutral sulfite, acid sulfite, and deacetylated), the autocatalytic steam explosion and SO_2 steam explosion were appropriate for producing pellets for thermochemical application. Deacetylation or neutral sulfonation was not recommended for producing pellets, due to the low lignin recovery and the preserved amount of hemicellulose [83], as illustrated in Figure 5. Steam explosion caused noticeable reduction of the levels of ash and silicon in wheat straw [66].

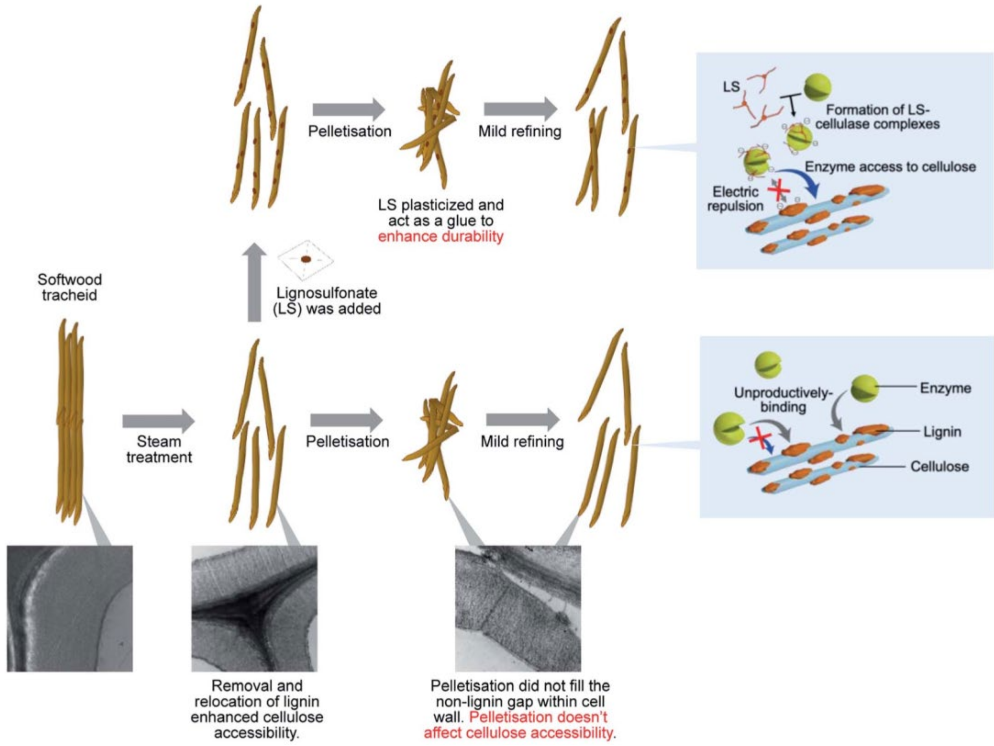

Figure 4. The influence of steaming, pelletization and lignosulfonate addition on pellet durability and cellulose hydrolysis. Adapted with permission from Ref. [80]. 2020, Royal Society of Chemistry.

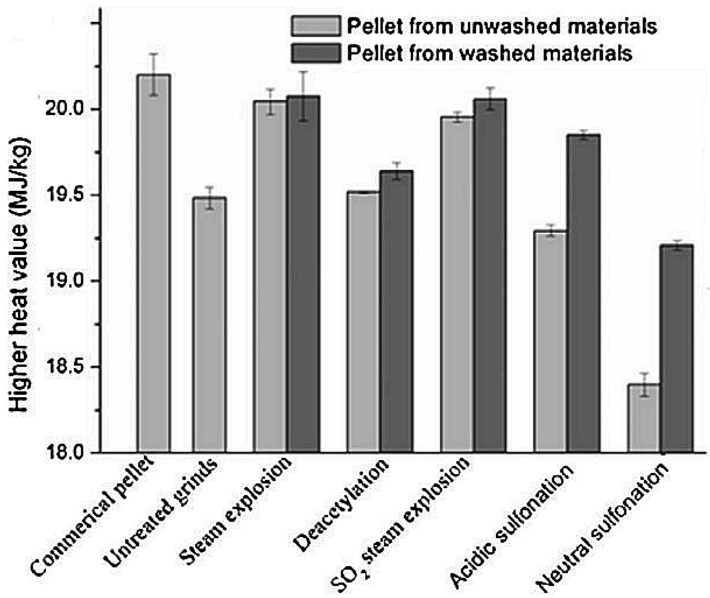

Figure 5. Comparison of the HHV of commercial pellet, raw pellet, and pretreated pellet. Adapted with permission from Ref. [83]. 2018, Elsevier.

Regarding hydrothermal treatment, it is feasible to noticeably reduce the requirements of processing conditions, including temperature and reaction time, versus those of the torrefaction process, particularly when the same contents of mass and energy yield are applied [54]. In order to escalate the quality of agricultural pellets and fuel properties, mild hydrothermal treatment was suggested, owing to its potential in industrially manufacturing pellets [76].

2.3. Ultrasonic Vibration-Assisted Pelleting

Scholars have attempted to develop an ultrasonically vibrating tool for the purposes of compressing unheated biomass and producing pellets via ultrasonic vibration-assisted (UV-A) pelleting [84]. An ultrasonic vibration generation system actuated by a pneumatic loading system consists of three major components: a power supply (converting 60-Hz electrical power into 20-kHz); a converter (converting high-frequency electrical energy into mechanical motion); and a pelleting tool [41], as shown in Figure 6. High frequency-vibrated particles result from the ultrasonic vibration, and the ultrasound energy can mainly be absorbed consequently as heat generation [42]. Due to heat release from ultrasonic vibration, the lignin becomes soft, and bonds together with the cellulose and the hemicellulose [85], resulting in UV-A pelleting-produced biomass pellets without high temperature steaming, under high pressure, with binder materials [86,87]. UV-A pelleting exhibits lower pelleting force, shorter pelleting time, and less swelling compared to no ultrasonic vibration [88].

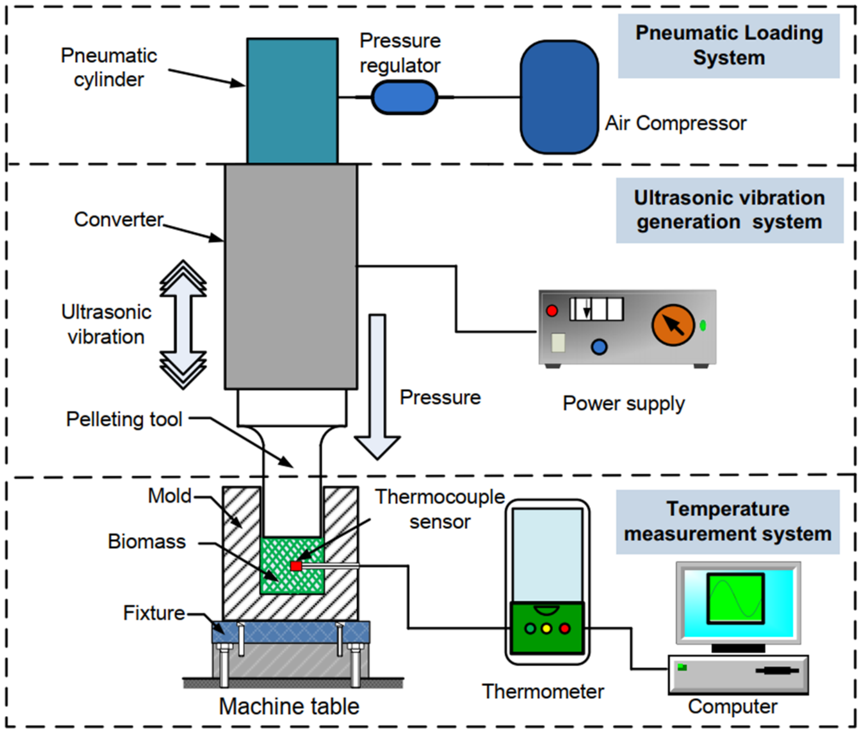

Figure 6. Schematic illustration of the ultrasonic pelleting unit. Adapted with permission from Ref. [41]. 2016, Elsevier.

A one-dimensional (axial) model of UV-A pelleting has been developed, using wheat straw to predict the influences of ultrasonic power and pelleting pressure on pellet density. Regardless of moisture content and particle size, the model prediction agreed well with the reported experimental results [85,89]. Response surface methodology was also employed, to predict the density and durability of the pellets in UV-A pelleting of sorghum stalks [90]. According to the models and confirmed experiments, durability (1239 kg m^{-3}) and pellet density (93%) were respectively measured under specifically defined conditions (i.e., pelleting time (44 s), ultrasonic power (50%), and pressure (42 psi)). However, the influence of water content was not considered. Fan et al. [91] pointed out that ultrasonic power, cylinder pressure, moisture content, and particle size significantly affected pellet density, and developed a predictive model for pellet density.

There are three problems with a single ultrasonic unit: firstly, the thickness of the pellets is limited—that is, subjected to the heat transmission from one side; secondly, the top and bottom densities are inconsistent, influencing pellet durability; finally, the low efficiency and occasional occurrence of carbonization [92]. Hence, two sandwich-type ultrasonic vibrators (Figure 7) were designed to eliminate the abovementioned shortcomings [93]. Consistency inside the pellet was retained in the compression, because both the upper and the lower surfaces simultaneously received ultrasonic vibration. The density and durability were noticeably improved.

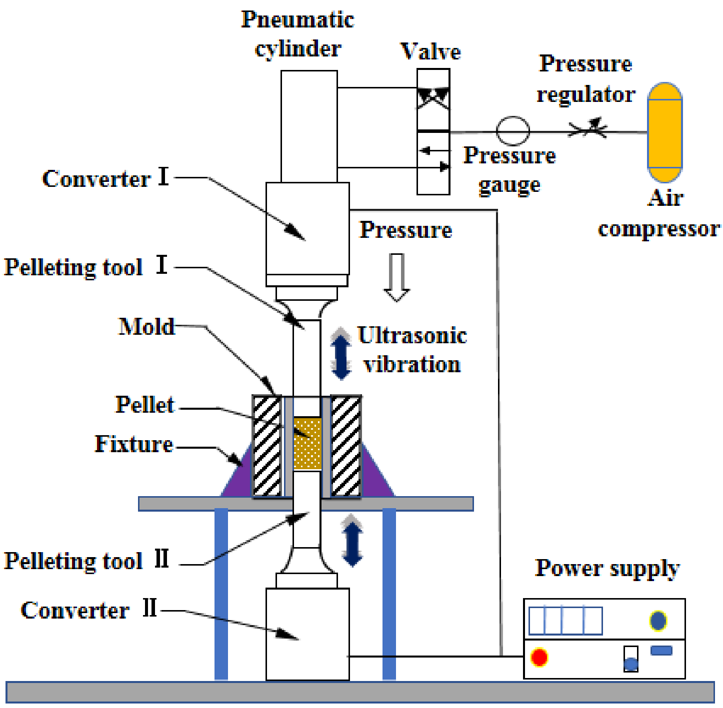

Figure 7. Schematic illustration of the dual ultrasonic pelleting unit.

2.4. Supercritical Fluid Extraction

No direct utilization of lignocellulosic biomass as energy feedstock was suggested. Extracts and the bioactive substances present in them should first be obtained, and production residue could be subsequently used as energy feedstock [26]. Supercritical carbon dioxide extraction ($scCO_2$) could be advantageous to expand the type of materials, which may result in production of pellets and quality improvement of the nominated features of the solid biofuel [26].

$ScCO_2$ had been demonstrated to remarkably reduce off-gassing (CO, CO_2, CH_4) from sawdust pellets, accompanied by insignificant influences on durability, calorific values, and density [94]. The O_2 level slightly decreased from 20% to 19.3%, while the CO level after storage was less than 2×10^3 ppmv, which was approximately 85% lower than the reference level. The same trend of emissions of CO_2 and CH_4 could be found, which were respectively reduced by 85% and 94%, as illustrated in Figure 8.

Hot-water extraction operated at 165 °C for approximately 2.5 h could result in the elevation of the energy levels of the pellets, while the ash content in willow was reduced by less than 1% [95]. After pretreatment of yellow birch trees and sugar maple via hot water (175 and 200 °C, 30 min) in a batch reactor, the energy level and density of the pellets yielded to within 30% and 40%, respectively. The rise of compressive strength as high as $\geq$ three times was confirmed, and friction in the die was noticeably reduced [96]. The water resistance was also improved, as shown in Figure 9.

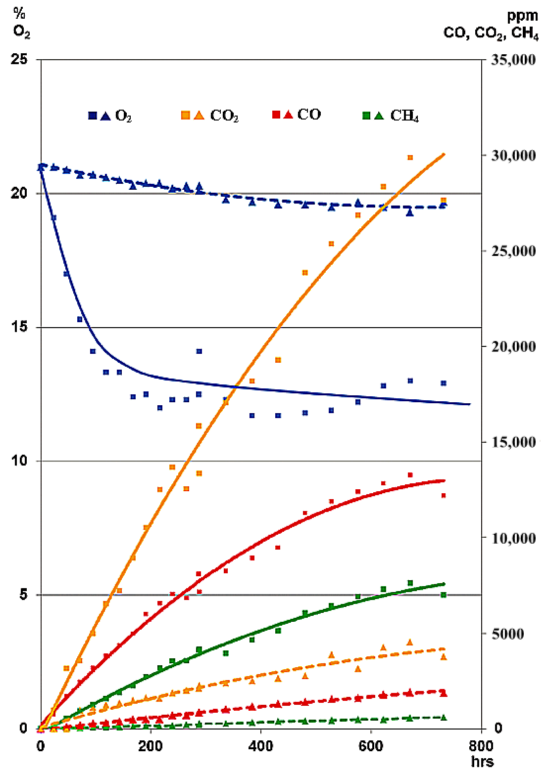

Figure 8. Concentration measured during off-gassing experiment. $scCO_2$-extracted (dotted line) and non-extracted pellets (solid line, reference); 30 days storage in 19 L test cylinder at 23 °C. Adapted with permission from Ref. [94]. 2016, Royal Society of Chemistry.

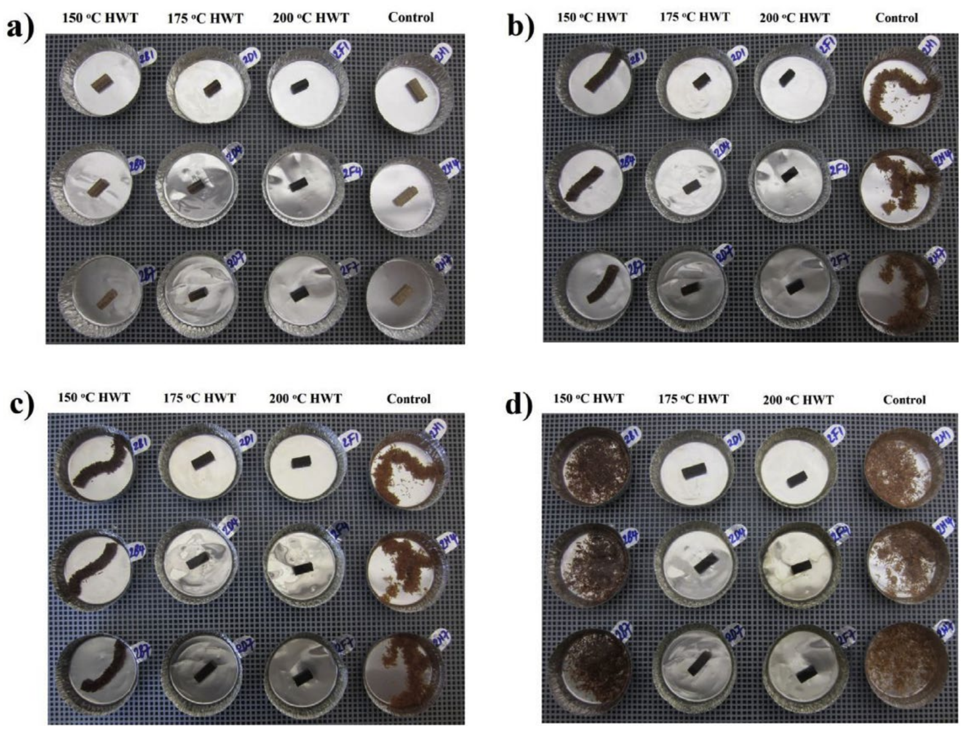

Figure 9. Water immersion tests: appearance of the pellets before being immersed in water (**a**), and after immersion for 5 min (**b**), 24 h (**c**), and 1 week (**d**) in distilled water at 23 °C. Adapted with permission from Ref. [95]. 2017, Elsevier.

3. Effects of Process Parameters (PPs) and Characteristics of Raw Materials on the Quality of Pellets

The quality of pellets could influence their transportation, storage, and applicability, as well as the industrial development. During storage, micro-particles in pellets may absorb moisture in the air, providing materials or conditions that may play a connecting role between particles, thereby causing the dispersion, deformation, and expansion of pellets, affecting their quality and disposal capacity, and reducing their market competitiveness. The factors affecting quality of pellets mainly include two aspects: physicochemical characteristics, including structural composition, particle size, moisture content, etc.; and PPs (e.g., pressure, temperature, and additives).

3.1. Effects of Biomass Types

Agricultural residues have physical and chemical diversities, resulting in great differences to their molding characteristics. Lignin is the most important feedstock component of lignocellulosic biomass [75]; its thermosetting properties are noteworthy, and it can be utilized as a binder material for the purpose of producing pellets in biomass particles [97]. However, its noticeable dependency on moisture content and temperature may hinder its broad utilization. In the pelleting process, greater energy consumption could result from a low lignin level in the agricultural biomass [98]. Incorporation of the physical and chemical properties of agricultural and forestry residues can form complementarity, and improve the quality of pellets.

Numerous scholars have recently targeted blends of biomass for pellets. Ma et al. [99] studied the physical properties of fuel produced by electro-osmotic sludge and corn stover. With the increase of sludging of raw materials, the physical properties of the shaped fuel particles were improved. Brand et al. [100] obtained a blend containing 75% Pinus spp. shavings and 25% rice husk, resulting in the generation of the pellets with the greatest features for energy generation. The pellets obtained from birch sawdust (50% mass) and pea waste (50% mass) had the highest mechanical strength and specific density [101]. The blending rates of solid wastes from rose oil processing, pine bark, and lignite coal powder had significant influences on density, abrasive resistance, water resistance, and the impact resistance of pellets [102]. Nosek et al. [103] assessed the possibility of the use of spent coffee grounds (including a large number of organic compounds) as fuel. The results showed that 100% of spent coffee grounds and 50% spent coffee grounds mixed with 50% sawdust did not reach required strength and durability. Yub Harun et al. [104] pointed out not only that pellets with a greater quality could be attained via incorporation of woody biomass and agricultural biomass, but also that less energy was expected to be consumed for the purpose of compaction versus merely pelleting of woody biomass.

The blends of different biomasses can not only improve physical properties, but can also be advantageous for combustion characteristics. Physical properties, combustion, and emissions of pellets from the blends of Faba Bean Waste and Potato Peels were investigated [25]. The density of pellets was found to range from 1226.22 to 1349.79 kg m^{-3} based on different volumetric ratios of feedstock. The lower calorific value of dried fuel pellets ranged from 15.27 to 16.02 MJ kg^{-1}. Pellets showed efficient combustion capability, and were low in pollutant emissions [25]. Chojnacki et al. [105] pointed out that the addition of 10–30% (dry mass) of apple, carrot, and red beet root pomace to barley straw increased the density and hardness of the pellets. Meanwhile, the addition of 30% carrot or red beet root pomace resulted in the noticeable elevation of ash content, and the mild reduction of calorific values compared to no addition. Pellets made from a mixture of rice husks with wheat straws obtained the highest calorific value (4301.10–4573.50 kcal/g) and a reduced amount of ashes (11.43–13.06%), improving the quality and combustion characteristics of the pellets [23]. Solid fraction of digestate alone, and mixed with sawdust and grain straw to produce pellets, were analyzed [106]. The results revealed that solid fraction of digestate still had energy potential, and would be a valuable substrate for production of solid biofuels, because of similarly low heating values and ash contents. If pellets from hazelnut and olive groves could be mixed with other types of wood, the weaknesses of low bulk density and high ash content could be overcome, and it would be possible to obtain top-quality pellets with outstanding profitability [107]. Blends of abundant available agricultural biomass and woody biomass would not only result in better mechanical properties, but would also be advantageous in satisfying the market for pellets [108].

3.2. Effects of Moisture Contents

A proper amount of moisture in the feedstock is as an important factor in the pelletizing process, whereas excessive moisture contents weaken hydrogen bonds and van der Waals forces, due to increasing the distance between particles [109]. In earlier studies, the optimal moisture that could be used in the compaction process of biomass was reported to be within 7.8–15% [110]. Fresh raw materials generally require a lower moisture content (around 7–8%), whereas stored materials require approximately 11–13% [111,112].

Styks et al. [20] pointed out that the moisture and pressure of the materials have significant influences on the density and mechanical durability of the pellets, depending on the tested materials. With the increase of moisture content (8%–14%), the durability of pellets from Miscanthus decreased while, for Sylphium and Sida, the opposite finding was attained, and the durability increased. According to their results, moisture content of 8% for Miscanthus and Sylphium, and moisture content of 11% for Sida were utilized under pressure of 262 MPa to obtain high-quality pellets [20]. The increase in moisture content from 10% to 15% for ground greenhouse melon residues reduced friction during compression, and facilitated the pelletizing process, resulting in a higher production rate of pellets [113]. High-moisture pelleting was tested by Tumuluru et al. [98] on switchgrass and 2-inch top pine residue blends. The results indicated blending ratios of 1:3 and 1:1, and that the bulk density and durability of the pellets exceeded 550 kg m^{-3} and 95%, respectively, with a moisture content of 20% (w.b.), whereas the moisture content of the pellets was 10.6%, which was slightly above the ISO-17225 standard. Pure beech and pine pellets were assessed in a lab-scale disc mill, to investigate the grind ability characteristics. The moisture content of the pellets did not influence the shape of the milled particles, in terms of circularity and elongation ratio [114].

Quality standards for solid biofuels require moisture content below 10% (pellet class A1) and 12% (briquette class A1). Pellets with a moisture content greater than 10–12% wb (wet basis) may be susceptible to fungal growth, resulting in their decomposition [76]. The upper moisture limit of lower quality classes never exceeds 15% [20]. Three types of commercial wood pellets sprayed by liquid water caused localized swelling of pellets, which dislodged particles from the surface; the pellet durability dropped from 99.5% to 97.5% when the moisture content of pellets increased to 10% wb, and the bulk density was reduced by 27.54% when the adsorbed moisture content was equal to 15% wb [115]. Cutz et al. [37] showed that 1 month storage at 40 °C at a relative humidity (RH) of 85% caused significant degradation of pellets, including creation and expansion of cracks. Pellets with a higher number of cracks at the surface, and within the structure, were more sensitive to degradation during storage, slowing down the bioenergy transition. Tong et al. [116] reported that high humidity (80% RH or above) would result in a higher degradation for pine wood pellets, followed by 95% RH for recycled wood pellets; however, they were stable between 20% RH and 60% RH.

3.3. Effects of Particle Size

Association of quality of pellets with particle size has been confirmed, and that it may affect a variety of features (i.e., flowability, compression, friction in the pelletizer die, and contact between the adjacent particles) [117]. The great function of the distribution feedstock particle size was emphasized, which might influence pellet quality [118]. Generally, smaller particle sizes of raw materials and higher densities of pellets, due to bonds between the smaller particles, are accompanied by higher levels of energy per unit mass [99,108]. Finer-size particles improve the pellets' performance, because finer-size particles account for a higher surface area, reduce the space between particles, and increase the interparticle forces, including the van der Waals force and the capillary forces [119]. Yilmaz et al. [113] pointed out the association of the reduced ground material particle size with the elevation of density and the durability of pellets in the pelleting of woody biomasses and their blends. Hettiarachchi et al. [109] attempted to interlock particles and minimize voids via utilization of smaller particles that could fill in gaps, in which van der Waals forces and hydrogen bonds resulted. The reduction of the specific energy consumption of pellets, which were produced by garden waste, from 141.2 to 100.2 kWh ton^{-1}, was outlined via minimizing biomass size from coarse (>25.4 mm) to fine (6.25 mm) [120].

Contrarily, an extremely high amount of fine particles (smaller than 0.5 mm in diameter) in the raw materials increased the cost of raw material treatment, and had a negative influence on friction and pellet quality [121]. Top-quality pellets could be produced from agricultural biomass under the optimal particle sizes of 3 mm, 2 mm, 1 mm, 500 μm, 250 μm, and <250 μm, which had proportions of <1%, <5%, 20%, 30%, 24%, and 20%, respectively [122]. Highly and lowly durable pellets could be attained from large and small particles, respectively [118]. The feedstock particle size distribution did not affect the sorption behavior, while it could influence the emission of CO and organic carbon [118].

3.4. Effects of Pressure and Temperature

The density of fuel particles increased as the forming pressure was elevated within a certain range. When the pressure reached a certain value, the density increase with the pressure was no longer significant, because the density of the fuel particles approached the density of the cell wall particles [99].

Compaction pressure and moisture content had noticeable influences on elastic springback value, which could decrease density, while their influence on durability was not confirmed [123]. This indicated the necessity of optimizing the pellet production, including proper particle arrangement, bonding through binders, plasticization [123], and mixing with higher xylan-content materials [124]. With the elevation of molding pressure, the hydrophobicity consequently rose, highlighting that the risen pressure was correlated with the reduction of spacing and gaps between particles, thereby restricting moisture absorption [60,125].

It is obvious that the glass transition temperature of biomass components can be influenced by temperature, which may facilitate particle bonding in pelleting [98]. For lignin, its glass transition temperature was outlined in a particular range (60–140 °C), which may have been dependent on the moisture content of the biomass [126]. Jewiarz et al. [127] indicated that the melting of plastics in refuse-derived fuel could be completed at 120 °C. With the rising of temperature, the components could be melted, and their solidification was associated with cooling, in which the resultant chemical modifications could highlight their outstanding properties for producing pellets [72]. The binding of particles could be enhanced via smoothing protein and lignin at relatively high pressure and temperature, in which compaction and density were accordingly strengthened. When sugarcane bagasse moisture content was regarded as constant, the increase of pellet density from 0.8344 to 1.2112 kg m^{-3} was outlined when the temperature was elevated from 100 to 180 °C, highlighting the noticeable role of temperature in pellet density [128]. The rising durability and density of the microalgal pellets was affected by temperature versus herbaceous and woody biomasses, because microalgae contain almost no cellulose, hemicellulose, and lignin [40].

A well-melted shell on the outer surface of the pellets improved their durability and bulk density. The friction between mold and biomass had a marginal effect on the temperature profiles [97]. The temperature at the center of the pellet was always higher than the temperature at the top of the pellets, and slightly higher than that at the bottom, with ultrasonic power from 40% to 70% [86]. At lower pressure and temperature, pellets formed from furfural residues and sawdust had a higher expansion tendency after extrusion from the die, and during storage [117].

3.5. Effects of Additives

In addition to using the physical and chemical complementary characteristics of different raw materials to improve pellet performance, additives are mainly used to promote formation of pellets. Additives, mainly in the form of a liquid or solid, strengthen the cohesive force between the particles, by producing solid bridges or inducing a chemical reaction [44], and improve the properties of the pellets, especially hardness and durability [45]. There are more than fifty binders that are used for biomass pelletization, including organic and inorganic binders [44]. García et al. [129] used the solid biochar obtained from pyrolysis of eucalyptus (PEc) at 700 °C as an additive, and glycerol as a lubricant, to produce enhanced pine sawdust pellets (PIN). According to their results, pellets made up from a mixture of 90% PIN and 10% PEc were competitive compared to raw PIN pellets. During the roasting process, apparent cracks could be formed when MgO was added and the volume was reduced; further addition of CaO could reduce the generation of cracks and improve the density, in which the phase of briquettes remarkably changed during the roasting stage rather than the preheating stage [130]. Saletnik et al. [34] assessed the effects of sunflower oil sprayed on wood pellets, on the physicochemical parameters. The results indicated that the amount of ash was reduced, that the durability had slightly risen, and that treatment with waste oil at a weight rate of 12% resulted in a 12–16% increase in the caloricity of the wood pellets. Due to the similar physical and chemical properties of bentonite, the components of some industrial solid waste and the application potential of pellet additives were summarized, to improve the quality of the pellets, decrease the cost, save energy, and reduce pollution [131].

Different types of additives can reduce the emission of SO_2, NOx, and particulate pollutants, improve the ash fusion, and prevent ash slagging. Ji et al. [132] showed that the addition of SiO_2 into briquette fuel was beneficial to the formation of calcium silicate complexes, which could reduce the emission of SO_2, while Al_2O_3 increased the release of SO_2 and $NO_{x.}$ Nosek et al. [16] introduced paper sludge as an additive to produce straw pellets. The positive effects had already risen from 1020 to 1260 °C after the addition of 10% sludge, while the calorific value decreased with the increase of sludge content in the mixture. High concentrations of K, Si, P, and Na decreased the ash melting temperature, and high concentrations of Ca, Mg, and Al increased the ash melting temperature of pellets made from buckwheat hulls as additives [15]. The addition of an adequate amount of $NH_4H_2PO_4$ during torrefaction can clearly decrease the PM_1 emission, at the cost of increasing PM_{1-10} emission [133]. The transformation of inorganics during torrefaction and the PM_1 reduction mechanism are illustrated in Figure 10. Table 2 summarizes the effects of additives on pellets.

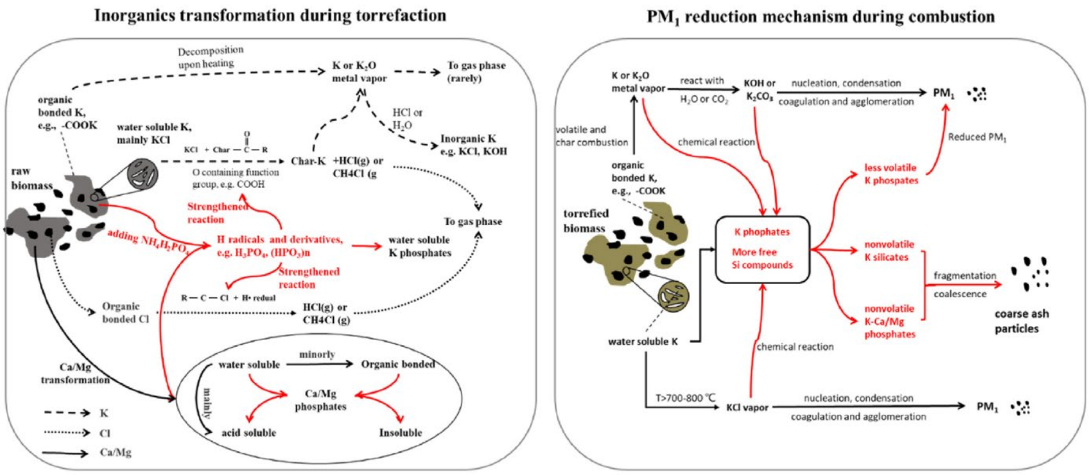

Figure 10. Schematic diagram of inorganics transformation during torrefaction, and PM_1 reduction mechanism during combustion. Adapted with permission from Ref. [132]. 2022, Elsevier.

Table 2. Effects of additives on pellets.

Feedstock	Additives	Effects	Ref.
Jack pine, balsam fir and black spruce	Starch, lignosulphonate, and pyrolytic lignin	Starch and lignosulphonate could not link torrefied fibers, making pellets easy to crush by hand. Only 15% of pyrolytic lignin produced pellets with good durability.	[45]
Oat hull	Proline, lignin and sunflower oil	Pellets with lignin content ≥15% and proline content ≥5% had the highest density, durability, and hardness. Adding sunflower oil increased the HHV, and decreased the ash content, density, durability, and hardness of the pellets.	[58]
Canola residue	Mustard meal, lignin and Pyrolysis-derived bio-oil	Mustard meal significantly improved the physical and mechanical properties of the pellets. A combination of lignin, mustard meal, and bio-oil produced the best quality pellet from torrefied biomass, with 100% durability and 1.2 MPa tensile strength.	[63]
Furfural residue and sawdust	Synthetic resin	The particle density of the wood pellets was increased and, after storage time of 2 weeks, remained the same as the initial particle density. Die temperature and specific energy consumption were decreased.	[117]
Cornstalk	Phosphorus-based additive ($NH_4H_2PO_4$)	Enhanced the removal of O and the tension of C; effectively reduced the mass and energy losses during torrefaction; decreased the absolute content of Cl and S in the torrefied fuel.	[132]
Maize straw	Calcium phosphate monobasic and ammonium dihydrogen phosphate	Ash fusion temperatures and fusion phenomena were greatly improved by adding $NH_4H_2PO_4$ and $Ca(H_2PO_4)_2$.	[134]
Cotton straw, rice straw, maize straw, pine sawdust, and poplar wood	Fugu coal (bituminous coal) and inorganic additives ($CaCO_3$, CaO, K_2CO_3)	Fugu coal improved the calorific value, and the potassium-based additives had a higher improvement than the calcium-based ones.	[135]
Algal biomass	Iron-based additives (Fe, Fe^{2+} and Fe^{3+})	The inhibitory effects of Fe^{2+} and Fe^{3+} were comparable, but better than those of Fe. Increased the concentration of the iron-based additive load, which had a significant inhibitory effect.	[136]
Corn stalk, rice husks and their blends	Low-rank coal	Promoted the release of alkali chlorides while inhibiting the vaporization of Ca, Mg, and Fe. The slagging was efficiently inhibited.	[137]
Olive-cake and white-wood	Coal pulverized fuel ash and kaolin powder	Increased the flow temperature of the ash compositions. Significantly reduced sintering and clearly inhibited KCl release. Al–Si additive use should be restricted to high K, high Cl biomass.	[138]

3.6. Effects of Holding Time

Holding time can offset the spring-back effect of biomass grinds [83], while it depends on different PPs and characteristics of materials. Li et al. [139] reported a 5% increase in density when holding time was prolonged from 0 to 10 s, without more significantly prolonging holding time. Holding time as a factor from 2 to 4 min was investigated, by four-factor Box-Behnken experimental design and response surface methodology, to predict density, specific energy consumption, and the radial maximum pressure of bagasse pellets [128]. Their results showed that holding time was significant to pellet density, whereas it had no noticeable influence on radial maximum pressure. Holding time-associated studies were only undertaken at the laboratory scale because of low production efficiency.

3.7. Effects of Pelleting Parameters

Pelleting parameters directly affect the friction force and the required compression force in the compression process, and determine the density and quality of pellets. To date, studies on pelletizers have mainly concentrated on the optimization of hole size and shape, which could improve the quality, reduce the wearing of the mold, and prolong the service life of the mold. Regarding the serious wearing and short service life of mold, a scheme of using three-taper combined dies, with taper angles of 6°, 6.5°, and 7°, was put forward [140], as illustrated in Figure 11. Compared with a single taper die, the service life was noticeably improved, and the energy consumption was reduced.

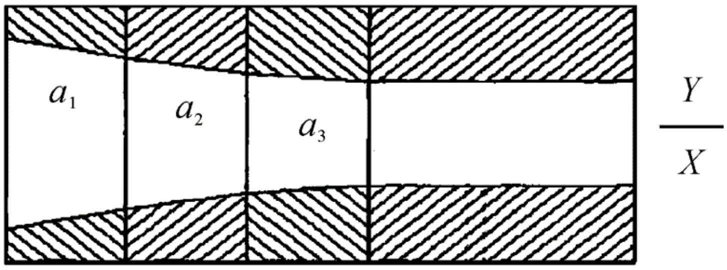

Figure 11. Structure of three-taper combined dies. Adapted with permission from Ref. [140]. 2020, Journal of Chongqing University of Technology.

The unit, bulk, tapped densities, and tensile strength were enhanced when the length-to-diameter ratio (compression ratio) increased from 6 to 10 via a flat die pellet mill. Higher compression ratios would result in higher external load, wedging, and compressive force, which might indicate that the feedstock was packed inside the pressing channel at a high pressure [36]. A compression ratio of 7.0–8.0 for pine wood pellets was recommended, whereas it depended on biomass chemical composition. The compression ratio was the highest for holocellulose (9.828), followed by the raw material (6.860), the extractive-free (5.167), and Klason lignin (3.265) fractions [141]. With extension of the thickness of the die, materials exposed at high temperatures for a longer period could generate meltdowns of plastics easily, and form top-quality pellets [127].

Although the influences of numerous PPs on the quality of pellets could be clarified, the results from laboratory-and pilot-scale researches are questionable, in terms of industrial production of pellets [114,142]. In order to rapidly and accurately acquire high-quality pellets for industrial purposes, quantitative models are required [143].

4. Summary and Recommendations

This review presents an insight into the effects of pretreatment methods and PPs on the properties of pellets. Torrefaction is an efficacious treatment to improve pellet quality, especially calorific value and off-gassing. However, the thermal pretreatment is energy-intensive; thus, the economic benefits and market competitiveness of pellets produced by this approach need to be further studied. Mild hydrothermal treatment, with a noticeably lower energy consumption compared to the torrefaction, is potentially applied in the industrial manufacturing of pellets. UV-A pelleting cannot be applied to timely transfer the heat generated by ultrasonic vibration, causing the reduction of pellet quality when more raw materials are molded, and it is difficult to produce continuously for the purpose of improving production efficiency. The equipment and technologies of continuous production should be further upgraded. Regarding $ScCO_2$ pretreatment, it may operate in diverse conditions, due to the different structures of crop residue, resulting in unstable pellet quality using the same $ScCO_2$ pretreatment method.

In the existing literature, biomass types, moisture contents, particle size, pressure, temperature, and additives were profoundly investigated. However, the effects of holding time and pelleting parameters were not comprehensively described. Additionally, interactions of parameters between pretreatment and molding were not clarified. Extensive research is required, to eliminate the abovementioned deficiencies.

Author Contributions: Conceptualization, W.L.; methodology, B.Z.; validation, W.L.; writing—original draft preparation, W.L.; writing—review and editing, M.W. and F.M.; supervision, Y.Z.; project administration, B.Z.; funding acquisition, W.L. and B.Z. All authors have read and agreed to the published version of the manuscript.

Funding: This work was funded by the Natural Science Talent Support Plan of Heilongjiang Bayi Agricultural University, grant number ZRCPY202010; ZRCPY202106, the Scientific Research Start-up Plan, grant number XYB2014-10; XDB202001, the Postdoctoral Scientific Foundation of Heilongjiang Province in China, grant number [LBH-Z19218] and the Educational Department of Liaoning Province, grant number LJKZ0690.

Data Availability Statement: Not applicable.

Acknowledgments: The authors would like to acknowledge permissions for all copyrighted table and figures from Elsevier and the Royal Society of Chemistry.

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

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